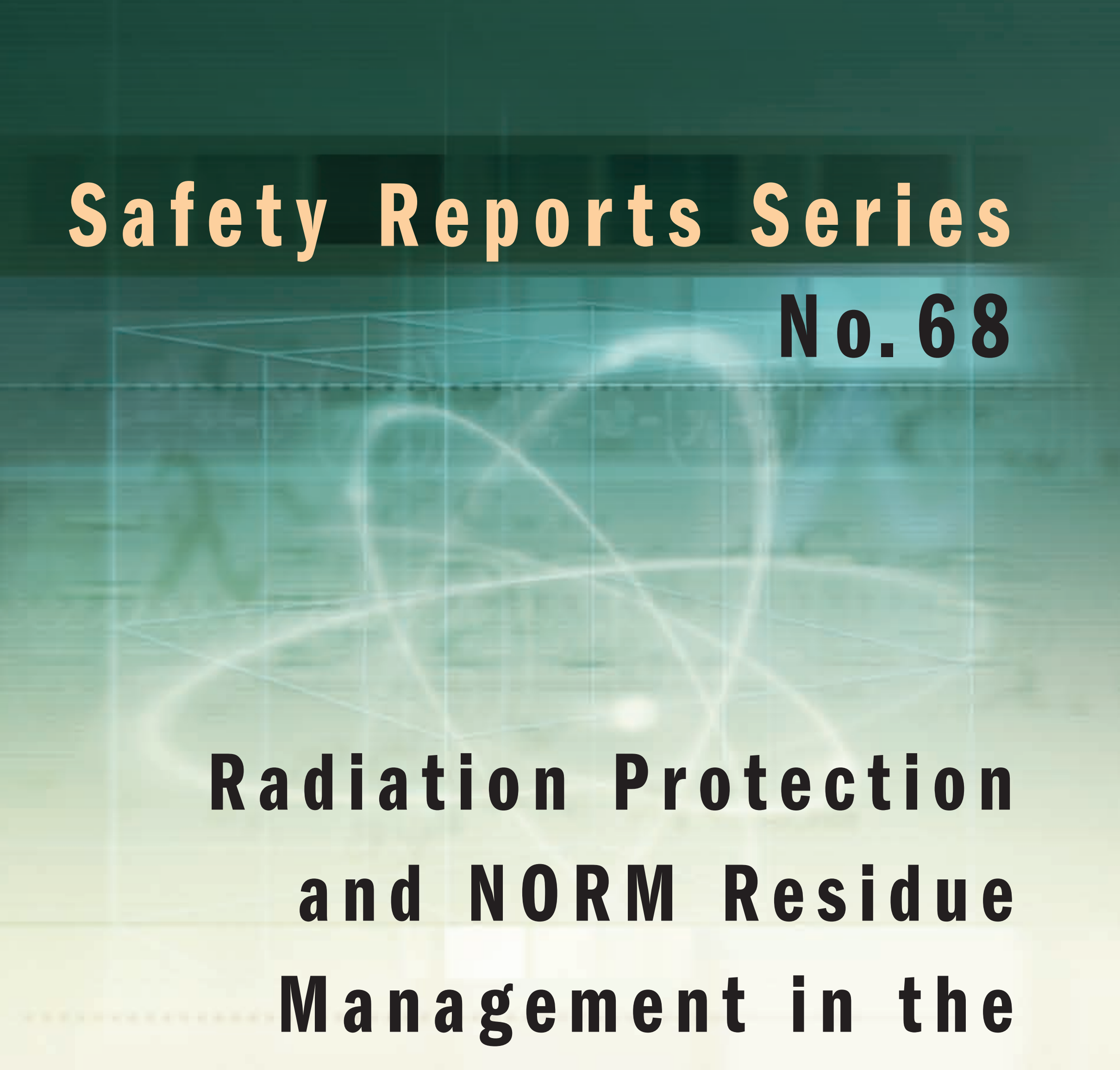

Safety Reports Series

No. 68

Radiation Protection and NORM Residue Management in the Production of Rare Earths from Thorium Containing Minerals

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RADIATION PROTECTION AND NORM RESIDUE MANAGEMENT IN THE PRODUCTION OF RARE EARTHS FROM THORIUM CONTAINING MINERALS

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SAFETY REPORTS SERIES No. 68

RADIATION PROTECTION AND NORM RESIDUE MANAGEMENT IN THE PRODUCTION OF RARE EARTHS FROM THORIUM CONTAINING MINERALS

INTERNATIONAL ATOMIC ENERGY AGENCY
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FOREWORD

The IAEA Fundamental Safety Principles (IAEA Safety Standards Series No. SF-1), together with the International Basic Safety Standards for Protection against Ionizing Radiation and for the Safety of Radiation Sources (Safety Series No. 115), set out the principles and basic requirements for radiation protection and safety applicable to all activities involving radiation exposure, including exposure to natural sources of radiation. The Safety Guides on Occupational Radiation Protection in the Mining and Processing of Raw Materials (IAEA Safety Standards Series No. RS-G-1.6) and Management of Radioactive Waste from the Mining and Milling of Ores (IAEA Safety Standards Series No. WS-G-1.2) provide guidance on the control of exposure of workers and members of the public to naturally occurring radioactive material (NORM) in industrial activities involving the exploitation of minerals. This guidance applies whether or not minerals are exploited for their radioactivity content. The exploitation of thorium containing minerals for rare earths production constitutes one of several industry sectors for which the radioactivity content of the minerals and raw materials involved is high enough to warrant consideration by a regulatory body concerning the possible need to control exposures to workers and members of the public.

This Safety Report has been developed as part of the IAEA's programme for the application of its safety standards in the fields of radiation, transport and waste safety. It is a compilation of detailed information on the processes and materials involved in the exploitation of thorium containing minerals for rare earths production and on the radiological considerations that need to be taken into account by a regulatory body when determining the nature and extent of radiation protection measures to be taken. This is consistent with the graded approach to regulation, within which the application of safety standards requirements is commensurate with characteristics of the practice or source and with the magnitude and likelihood of exposures.

The IAEA wishes to express particular acknowledgement to the contributions made by P.M.B. Pillai and N. Tsurikov. The IAEA officers responsible for this publication were D.G. Wymer and P.P. Haridasan of the Division of Radiation, Transport and Waste Safety.

CONTENTS

1. INTRODUCTION

1.1. BACKGROUND

The rare earths comprise a chemically coherent group of 15 elements with atomic numbers 57 to 71, together with the elements yttrium and scandium, which have similar chemical properties. Rare earths are essential elements for a large number of applications of considerable technological, environmental and economic importance. The minerals used as commercial sources of rare earths contain elevated concentrations of radionuclides in the thorium decay series and, to a lesser extent, in the uranium decay series. Details of these radionuclides are provided in Appendix I. In one such mineral, monazite, thorium concentrations are sufficiently elevated to warrant its being used as a commercial source of thorium. During mineral processing operations, radionuclides may become mobilized and migrate to dusts, scales and other process residues, leading to the possibility of significant radionuclide activity concentrations in these materials even when concentrations in the feedstock mineral are low. Isotopes of radium in particular may become concentrated in scales.

The radioactivity content of mineral feedstocks and process residues creates a possible need to control exposure to workers and members of the public in accordance with the IAEA Safety Standards (the Standards), in particular the Fundamental Safety Principles [1], the International Basic Safety Standards for Protection against Ionizing Radiation and for the Safety of Radiation Sources (the BSS) [2], the Regulations for the Safe Transport of Radioactive Material (the Transport Regulations) [3] and relevant Safety Guides [4, 5]. In many of these materials, activity concentrations of radionuclides in the ^{232}Th decay series (and to a lesser extent the ^{238}U series) are such that, in terms of the Safety Guide on Application of the Concepts of Exclusion, Exemption and Clearance [6], they would be considered for inclusion within the scope of regulation as naturally occurring radioactive material (NORM).

The Safety Report on Assessing the Need for Radiation Protection Measures in Work involving Minerals and Raw Materials [7] confirms that the exploitation of thorium containing minerals for rare earths production constitutes one of several minerals related industry sectors likely to warrant radiation protection measures through the system of regulatory control for practices. That Safety Report emphasizes the particular relevance of the graded approach to the regulation of practices to ensure that any radiation protection measures applied to the industrial processes involved and to the management of NORM residues arising from those processes are commensurate with the characteristics of the operation and with the magnitude and likelihood of the exposures. This implies the need for a thorough understanding of all processes, process materials, associated radiological risks to workers and members of the public and practicalities involved in any protective measures that might need to be considered.

1.2. OBJECTIVE

The objective of this Safety Report is to provide detailed information that will assist regulatory bodies in implementing a graded regulatory approach to the protection of workers and members of the public against exposures associated with the exploitation of thorium containing minerals for rare earth production. This information will also serve as the basis for creating a common understanding between regulatory bodies and other stakeholders such as operators, workers and their representatives, as well as health, safety and environmental professionals, of the radiological aspects of various processes involved and the ways in which these aspects can be addressed appropriately and effectively.

While heavy-mineral sands are a minor source of rare earths production, the mining and beneficiation of these sands for the purpose of producing commercially important heavy minerals such as zircon and ilmenite[1] is an important worldwide industry in its own right. The information provided in this report concerning the mining and beneficiation of heavy-mineral sands is therefore relevant to those involved in the mineral sands industry, even when such deposits are not exploited for rare earths production. In this regard, this report serves as a supplement to safety reports dealing with the zircon and zirconia industries [8] and the titanium dioxide and related industries [9].

1.3. SCOPE

This report provides detailed information on the exploitation of thorium containing minerals for rare earths production, including the processes involved, the management of NORM residues arising from such processes, the radiological characteristics of various process materials (feedstocks, products, by-products and residues), exposure pathways to workers and members of the public,

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[1] Heavy minerals are normally defined as those with densities exceeding 3000 kg/m^3. The heavy minerals of commercial importance are zircon, the titanium bearing minerals ilmenite, rutile and leucoxene and the rare earth bearing minerals monazite and xenotime.

exposure levels, annual effective doses, and examples of good practice with respect to monitoring techniques and practical measures to reduce doses.

Information on the decommissioning of monazite processing facilities is also provided. In addition, the report describes experience gained in dealing with so-called legacy issues from past monazite processing operations that were not conducted in accordance with good radiation protection and environmental management practices.

For each major step in the production of rare earths, the report includes an assessment of regulatory implications in terms of standards for radiation protection and management of radioactive waste as well as requirements contained in the Transport Regulations [3].

1.4. STRUCTURE

This report contains 11 sections, including this introduction. Section 2 provides a general overview of the rare earths industry, including a description of the rare earth elements, their commercial uses and their sources and production. Section 3 summarizes application of the standards to industrial activities involving exposure to natural sources, including application of the transport regulations. The next two sections address the mining and beneficiation of two major hard rock rare earth deposits: Bayan Obo ore from China, the world’s largest source of rare earths (Section 4) and the newly exploited Mount Weld deposit in Australia (Section 5). The next two sections address the mining and beneficiation of placer deposits comprising mineral sands (Section 6) and residues from exploitation of alluvial tin deposits in south-east Asia (Section 7). Other sources of rare earths, including ion adsorption clay deposits in China, are addressed in Section 8. Chemical treatment of the various types of rare earth concentrates derived from the beneficiation of ores is addressed in Section 9. The separation and purification of individual rare earths are addressed in Section 10. Finally, Section 11 addresses the manufacture and use of rare earth products. The report is supplemented by 11 appendices in which more detailed numerical data pertaining to the production of rare earths are presented.

2. OVERVIEW OF THE INDUSTRY

2.1. RARE EARTH ELEMENTS

The 15 rare earth metallic elements with atomic numbers 57–71, also referred to as the lanthanide elements (or ‘lanthanides’), are lanthanum, cerium, praseodymium, neodymium, promethium, samarium, europium, gadolinium, terbium, dysprosium, holmium, erbium, thulium ytterbium and lutetium. They are represented by the single square for lanthanum (La) in the main part of the Periodic Table and listed in a separate sub-table below the main grouping (see Fig. 1). Except for promethium (atomic number 61), which is radioactive and does not occur in significant quantities in nature owing to its relatively short half-life, the rare earth elements are in fact not especially rare — each is more abundant than silver, gold or platinum. The metal yttrium (atomic number 39) is included among the rare earth elements as it occurs with the lanthanides in natural minerals and has similar chemical properties. The metal scandium (atomic number 21) also has properties similar to those of the lanthanides and may occur in rare earth minerals, but is found in a range of other minerals as well. It is rarely, if at all, considered for recovery from rare earth minerals and no provision is made for avoiding or separating it during the processing of such minerals. Further information on rare earths can be found in Appendix II and Refs [10, 11].

Rare earths are normally classified into two subgroups: the ‘light rare earths’ are those lanthanides with atomic numbers in the range 57–63 (La to Eu)

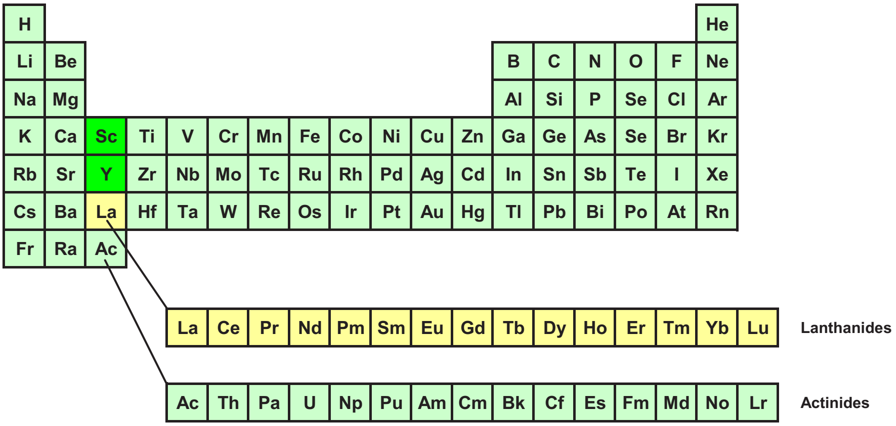

FIG. 1. Periodic table showing the positions of the lanthanides and the elements Sc and Y.

while the ‘heavy rare earths’ are those lanthanides with atomic numbers 64–71 (Gd to Lu) together with Y and Sc, which have similar properties in spite of their low atomic weights. Lanthanides in the light rare earths subgroup are generally more abundant than those in the heavy rare earths subgroup and are more easily extracted. Lanthanides with atomic numbers in the range 62–64 (Sm, Eu and Gd) are sometimes referred to as the ‘middle rare earths’.

2.2. COMMERCIAL USES

A summary of the commercial uses of mixed and individually separated rare earths is provided in Sections 2.2.1 and 2.2.2, respectively. More detailed information can be found in Refs [12, 13].

2.2.1. Mixed rare earths

Because the rare earths have similar chemical properties, they are difficult to separate. Initial commercial uses, which included lighter flints, carbon arc cores for lighting, polishing compounds and additives to glass and ceramics, were therefore based on mixtures of several rare earths. Even now, though the uses of individually separated rare earths account for the highest commercial value, mixtures of rare earths continue to account for the largest quantities used. Current important applications of mixed rare earths include:

(a) Components of fluid cracking catalysts in the refining of crude oil — for example, the addition of 1–5% rare earth chloride to a zeolite catalyst increases its cracking efficiency;
(b) Components of catalytic converters in automobiles in order to stabilize the gamma–alumina support matrix and enhance the oxidation of pollutants;
(c) Metallurgical additives in various ferrous and non-ferrous metal alloys, including alloying agents to desulphurize steel, nodularizing agents in ductile iron and alloying agents in Ni and Co superalloys, hydrogen absorption alloys and alloys based on elements such as magnesium, aluminium and titanium;
(d) The production of ‘mischmetal’ alloy, comprising 51–53% Ce, 22–25% La, 15–17% Nd, 3–4% Pr, 2–3% Sm, 3% Tb, 3% Y and 5% Fe, which is used:

   - (i) As a component of nickel metal hydride rechargeable batteries, which are steadily replacing nickel cadmium batteries for powering portable electronic equipment such as laptop computers and mobile telephones and which are now finding application in the growing market for hybrid drive motor vehicles (10–12 kg REO per battery[2]);
   - (ii) In the production of spheroidal graphite cast iron and various alloys (such as high strength low alloy steel, electrical resistance alloys, alloys for permanent magnets, creep resistant magnesium alloys, aluminium alloys, steel coating alloys, bronzes, lighter flints), as a cermet binder in hard metals and as a chemical reductant in process metallurgy;
   - (iii) In reversible hydrides for hydrogen energy systems;

(e) Various applications in the glass and ceramics industries, including glass polishing components (which are more effective than the traditional rouge), colouring agents, decolouring agents, UV absorbing and anti-browning agents, additives to structural ceramics such as stabilized zirconia and silicon nitride, additives to optical glasses and lenses and as a minor component of certain special glasses.

2.2.2. Individually separated rare earths

Individually separated rare earths are used in relatively small quantities, but their commercial applications are characterized by a high degree of technological sophistication and their use is expanding rapidly. The following applications, while in some cases making use of rare earths mixtures, also make use of individually separated rare earths:

(a) Samarium–cobalt permanent magnets are used in industrial, military and aerospace applications, while less costly neodymium–iron–boron magnets are used in starter motors, windscreen wiper motors and other mechanisms in motor vehicles, medical magnetic imaging, industrial motors, computer disk drives, camera motors and various electronic devices. Some permanent magnets also incorporate Ce or Pr. Other magnetism related applications include magneto-restrictive materials and other special magnetic materials (Pr, Nd, Sm, Gd, Tb, Dy, Ho), magneto-optical recording (Gd, Tb, Dy),

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[2] REO denotes rare earth oxides. Quantities of rare earth elements, irrespective of the actual chemical compounds in which they are incorporated, are generally expressed as equivalent REO.

magnetic refrigeration systems (Gd, Dy, Er), garnets (Gd, Ho, Lu, Y) and garnet bubble domain memory devices (Gd);

(b) Metallurgical applications include the production of steel and nodular iron (Ce), advanced alloys (La, Ce, Nd, Tb, Er, Y, Sc), chemical reductants (La, Ce), and cermet binders (Ce, Y);

(c) Applications in the glass industry include polishing components (La, Ce), colouring agents (Ce, Pr, Nd, Ho, Er), decolouring agents (Ce, Nd, Er), UV absorption agents (Ce, Pr, Nd) and additives to optical glasses and lenses (La, Ce, Nd, Gd, Y);

(d) Applications in luminescent materials (commonly known as phosphors) include colour television and other visual display screens (Ce, Eu, Tb, Lu, Y, Sc), X ray equipment (La, Eu, Gd, Tb, Tm, Yb, Lu) and fluorescent lamps (La, Ce, Eu, Gd, Tb, Dy, Y, Sc);

(e) In the ceramics industry, applications include engineering ceramics (Ce, Y), dental ceramics (Ce, Y), ferro-electric and electro-optic ceramics (La), ceramic capacitors (La, Ce, Pr, Nd), mixed oxide catalysts (La, Ce, Nd, Y), solid electrolytes (Tb, Y), heating elements (La), thermionic emitters (La), refractories (Gd, Ho, Sc), abrasives (La, Ce), ceramic glazes (La, Ce, Nd, Er) and ceramic pigments (Pr);

(f) Applications in the nuclear industry include neutron absorbers (Sm, Dy, Eu, Gd, Dy, Ho), burnable poison (Gd, Dy) and tubing (Y);

(g) Other applications include lasers (La, Pr, Nd, Sm, Gd, Tb, Dy, Ho, Er, Tm, Yb, Y, Sc), fibre optics (La, Nd, Ho, Er, Yb), superconductors (La, Gd, Lu, Y), microwave components (La, Nd, Y), stress gauges (Yb), pigments (La, Ce), La–Ni–H batteries (La) and reversible hydrides for hydrogen energy systems (La, Ce, Pr, Nd).

2.2.3. Commercially available forms of rare earths

A variety of rare earth compounds and metal products is commercially synthesized from extracted rare earths. The compounds include mixed or individual rare earth chlorides and other halides, nitrates, carbonates, oxides, acetates, oxalates and sulphates. Metals can be mixed to form products such as didymium (a mixed Nd and Pr metal used in the production of magnets) and mischmetal.

Purities from 96 to 99.9999% are available for most rare earths. High purity rare earth metals are marketed in the form of sponges, lumps, ingots, crystals, rods, wires, chips, powders, sheets, foils, plates, sputtering plates and custom cast and machined shapes. Alloys including mischmetal, rare earth silicide and ferrocerium are available in a variety of ingot shapes and sizes. Rare earth magnet alloys are marketed in ingot form and crushed ribbon, or may be purchased as mixed oxides for powder metallurgical purposes.

Processing advances in recent years have removed markets for some relatively low specification rare earth products. For instance, a 90% cerium oxide product used mainly by the glass industry for many years has been supplanted by higher grade cerium products with little or no price increase.

2.2.4. Worldwide consumption of rare earths

A breakdown of the 2006 consumption of rare earths by application and geographical region is provided in Table 1. While catalysts, magnets, metal alloys, polishing and glass each account for a significant share of worldwide consumption by weight (together accounting for 80% of the total amount), most of the market value (70%) is associated with magnets and phosphors, these being the two main applications involving individually separated (and thus higher value) rare earths.

2.3. SOURCES AND PRODUCTION QUANTITIES

Rare earths are found in primary deposits associated with igneous intrusions and associated veins, dikes and pegmatites and in secondary deposits of beach, dune and alluvial placers. While more than 200 minerals are known to contain rare earths at concentrations exceeding 0.01%[3], the principal minerals from which rare earths are sourced commercially are:

(a) Bastnäsite, $(Ce,La,Y)(CO_3)F$, a fluorocarbonate occurring in carbonatites and related igneous rocks, with a rare earth content of 58–75% REO;
(b) Monazite, $(Ce,La,Nd,Y,Th)PO_4$, occurring in heavy-mineral sand deposits, vein type deposits in granite and low grade tin ores from south-east Asia, with a rare earth content of 35–78% REO;
(c) Rare earth bearing clay, an ion adsorption type of ore formed by lateritic weathering of igneous rocks, with a rare earth content of 0.05–4% REO;
(d) Xenotime, YPO_4, occurring with monazite in heavy-mineral sands and tin ores, with a rare earth content of 54–65%;

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[3] Throughout this report, mineral concentrations expressed as percentages refer to mass concentrations.

TABLE 1. WORLDWIDE CONSUMPTION OF RARE EARTHS IN 2006 BY APPLICATION AND REGION *(from Ref. [14])*

	Consumption (t REO)						Relative consumption	
	China	Japan, and south-east Asia	USA	Europe	Other	Total	By weight	By value
Catalysts	6500	3500	6000	5000	500	21 500	20%	6%
Magnets	14 000	5000	750	500	250	20 500	19%	37%
Metal alloys	10 250	4000	1500	1000	250	17 000	16%	11%
Polishing	7000	4500	1000	1000	500	14 000	13%	3%
Glass	7250	3500	1000	1000	250	13 000	12%	3%
Phosphors	4500	2750	500	500	250	8500	8%	33%
Ceramics	2000	2000	1000	500	Negligible	5500	5%	4%
Other	6500	1000	250	250	Negligible	8000	7%	3%
Total	58 000	26 250	12 000	9750	2000	108 000	100%	100%

(e) Loparite, $(Ce,Ca,Na)_2(Ti,Nb)_2O_6$, a titanate related to perovskite (and hence also referred to as niobium perovskite) which occurs in alkaline igneous rocks, with a rare earth content of 28–37% REO.

Detailed data on the rare earth, thorium and uranium content of rare earth deposits (expressed as oxides) are presented in Appendix III. A summary of the data for commercially exploited deposits is given in Table 2. The levels of thorium and uranium in rare earth deposits, while depending on the type of mineral and its region of occurrence, generally exceed the worldwide median values for soil by up to 200 times in the case of thorium and up to 30 times in the case of uranium.

Examples of individual rare earth concentrations in rare earth minerals are provided in Appendix IV and can be summarized as follows:

(a) Bastnäsite and monazite contain mostly the light rare earths, predominantly Ce, La and Nd. The Y content is low;

TABLE 2. RARE EARTH, THORIUM AND URANIUM CONTENT OF RARE EARTH DEPOSITS

Deposit	Country of origin	Concentration (%)		
		REO	ThO_2	U_3O_8
Bastnäsite–monazite ore	China	1–10	0.04–0.07	0.0002
Bastnäsite ore	China, USA	0.5–12	0.02–0.1	0.002–0.004
Rare earth bearing clay	China	0.05–1	0.005	0.005
Heavy mineral sands	Various	0.1–5	0.0006–0.4	0.0003–0.006
Rare earth ore, principally supergene monazite[a]	Australia	4–25	0.075 (max. 0.18)	0.003 (max. 0.006)
Loparite ore	Russian Federation	0.1		
Soil (median values) for comparison purposes [15]	Worldwide		0.0008[b]	0.0003[b]

[a] Some zones also contain churchite, a hydrated yttrium phosphate mineral.

[b] Derived from activity concentration data assuming that 1 g of natural uranium contains 12 350 Bq of U-238 and 1 g of natural thorium contains 4057 Bq of Th-232.

(b) Ion adsorption clays vary widely in composition, containing mostly La and Nd but with some deposits also exhibiting significant concentrations of other heavy rare earths having even atomic numbers. The Ce content is characteristically low (0.3–5.5%), while the Y content is generally high;

(c) Xenotime has significant concentrations of both the heavy and light rare earths, especially those having even atomic numbers, and the Y content is high;

(d) Loparite contains mostly the light rare earths La, Ce and Nd. Concentrations of the heavy rare earths and Y are very low.

Information on rare earth mineral resources is presented in Table 3. Nearly 70% of proven reserves of rare earths are located in just three countries: China, the Russian Federation and the United States of America. Production of rare earths since 1950 is shown in Fig. 2. In the early years of production, modest amounts of rare earths were produced from various monazite bearing deposits and as minor components of uranium and niobium extraction. By 1966, however,

TABLE 3. RESOURCES OF RARE EARTHS

	Rare earths, including yttrium but excluding most scandium [18]		Yttrium [19]	
	Reserves (t REO)	Contribution to total (%)	Reserves (t Y_2O_3)	Contribution to total (%)
Australia	5 200 000	5.9	100 000	18.4
Brazil	48 000	0.05	2200	0.4
China	27 000 000	30.7	220 000	40.4
Russian Federation	19 000 000	21.6	0	0
India	1 100 000	1.3	72 000	13.2
Malaysia	30 000	0.03	13 000	2.4
Sri Lanka	0	0	240	0.04
USA	13 000 000	14.8	120 000	22.0
Other countries	22 000 000	25.0	17 000	3.1
Total (rounded)	88 000 000	100	540 000	100

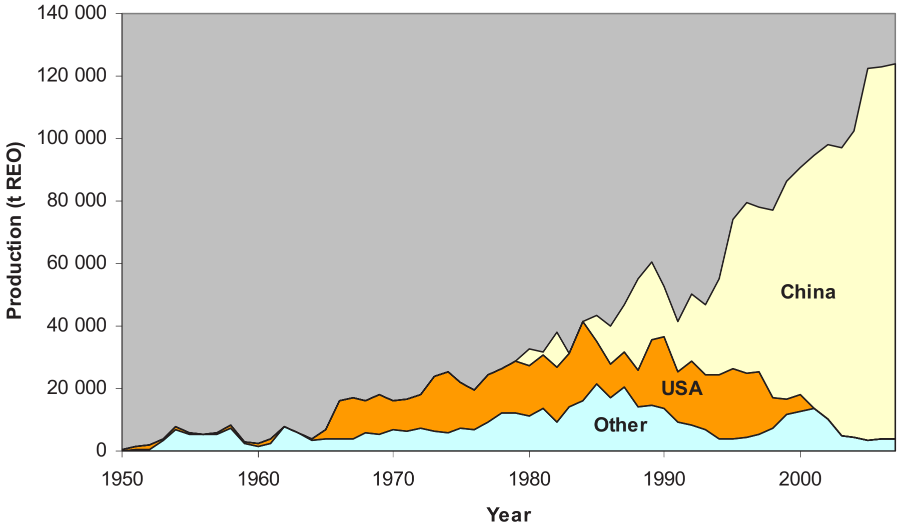

FIG. 2. Worldwide production of rare earths, compiled from data given in Refs [16, 18, 20, 21].

most rare earths production was being sourced from the Mountain Pass mine in California, USA, where a carbonatite intrusion containing significant concentrations of the light rare earths hosted mainly by bastnäsite and related minerals was exploited. Mountain Pass remained the dominant source of rare earths until the mid-1980s, at which time production from China started to increase dramatically. Most Chinese production comes from the Bayan Obo deposit in the Inner Mongolia region (a complex ore containing commercially significant concentrations of rare earths, iron and niobium hosted principally by bastnäsite and a thorium deficient form of monazite), from deposits of rare earth bearing ion adsorption clay in southern China and from bastnäsite in Sichuan Province. In 2004, total Chinese production was 98 000 t REO [16], of which 59% came from the Bayan Obo deposit and 26% came from ion adsorption clay deposits [17]. The ion adsorption clay deposits of southern China are the source of most of the world's yttrium production.

Between 2000 and 2007, operations at Mountain Pass were suspended, while concern about radioactivity has led to a decline in production from many monazite based sources associated with heavy-mineral sands. This has left China as the main source of rare earths production. Total world production in 2008 is estimated to have been 124 000 t REO [18]. A breakdown of this total, shown in Table 4, reveals that 97% of this came from China. The data in Table 4 also indicate that almost 99% of the world production of yttrium came from China in 2007. It has been predicted that in 2012, worldwide demand for rare earths will be

TABLE 4. PRODUCTION OF RARE EARTHS IN 2008

	Rare earths, including yttrium but excluding most scandium [18]		Yttrium [19]	
	Production (t REO)	Contribution to total (%)	Production (t Y_2O_3)	Contribution to total (%)
Brazil	650	0.5	15	0.2
China	120 000	97.1	8800	98.9
India	2700	2.2	55	0.6
Malaysia	380	0.3	4	0.04
Other countries	Data not available		0	0
Total (rounded)	124 000	100	8900	100

between 180 000 and 190 000 t, of which about 130 000 t (70%) is expected to come from China [14].

2.4. PRODUCTION PROCESS

2.4.1. Mining

Primary deposits are mined by conventional opencast and underground methods using, for hard rock deposits, drilling and blasting. Secondary (placer) deposits are mined by dredge mining methods, as well as by manual surface collection and mechanized dry mining methods using conventional earthmoving equipment.

2.4.2. Physical beneficiation

After mining, hard rock deposits are first crushed and ground and then beneficiated to produce a mineral concentrate, usually by means of hot froth flotation. Placer deposits are beneficiated using wet gravity separation followed by dry processes such as magnetic and electrostatic separation to produce various types of mineral concentrates. Physical beneficiation increases rare earths content and usually also concentrations of the radioactive components. The thorium content in mineral concentrates varies from less than 0.1 to about 10%, while the uranium content varies from very low values to 1%. Monazite has the highest thorium content, while bastnäsite (the largest commercial source of rare earths) contains relatively low concentrations of both thorium and uranium.

2.4.3. Chemical processing

Rare earth mineral concentrates are chemically processed to extract intermediate groups of mixed rare earth compounds. Chemical treatment of mineral concentrates derived from hard rock deposits may start with roasting in air (calcining) to drive off carbon dioxide and oxidize cerium to the tetravalent state. This is in many situations followed by treatment with hydrochloric acid to dissolve non-cerium rare earths, yielding a marketable cerium concentrate which can be used directly as a low value product (for instance, for glass polishing) or further separated into high purity individual rare earths. Alternatively, high temperature direct chlorination is a universal ore treatment process that easily integrates with subsequent process steps. The process produces an anhydrous rare earth trichloride product that is well suited for the production of mischmetal.

Chemical processing of mineral concentrates derived from placer deposits is usually accomplished using sodium hydroxide. The caustic attack process produces a mixed rare earth thorium hydroxide cake suitable for further processing with a variety of methods to separate thorium and individual rare earths, as well as a marketable by-product, trisodium phosphate.

2.4.4. Extraction and purification of individual rare earths

Individual rare earth compounds are produced from mixed rare earth chloride using methods such as selective oxidation, selective reduction, fractional precipitation, fractional crystallization, solvent extraction and ion exchange. The compounds produced include chlorides, fluorides, oxides, carbonates and nitrates. The production of rare earth metals with purities generally in the range of 98–99% is achieved using various reduction routes. Higher levels of purity, while sometimes achievable with reduction methods, are normally obtained through a sequence of refining processes.

2.4.5. Manufacture of rare earth products

Rare earth products are manufactured using a variety of processes, depending on the nature of the product and degree of purity required. Some rare earth products may be manufactured directly from mineral concentrates such as bastnäsite and monazite. Others are produced as end products of chemical processing or as intermediates in the preparation of rare earth metals. Many rare earth products, particularly those of a more sophisticated nature, can only be obtained by further processing of intermediates or metals.

3. GENERAL RADIATION PROTECTION CONSIDERATIONS

3.1. APPLICATION OF THE STANDARDS TO INDUSTRIAL ACTIVITIES INVOLVING EXPOSURE TO NATURAL SOURCES

3.1.1. Scope of regulation

Paragraph 2.5 of the BSS [2] states that “Exposure to natural sources shall normally be considered as a chronic exposure situation and, if necessary, shall be subject to the requirements for intervention …”, meaning that in such circumstances exposure does not fall within the scope of regulation in terms of the requirements for practices. However, there are some industrial activities giving rise to exposure to natural sources that have the characteristics of practices and for which some form of control in accordance with the requirements for practices may be more appropriate. Paragraph 2.1 of the BSS states that “The practices to which the Standards apply include … practices involving exposure to natural sources specified by the [regulatory body] as requiring control …”. This exposure includes “public exposure delivered by effluent discharges or the disposal of radioactive waste … unless the exposure is excluded or the practice or the source is exempted” (BSS, para. 2.5(a)). The exploitation of thorium containing minerals for rare earths production is identified in Ref. [7] as being among those industrial activities likely to require consideration by the regulatory body in this regard.

The Safety Guide on Application of the Concepts of Exclusion, Exemption and Clearance [6] states that it is usually unnecessary to regulate (as a practice) material containing radionuclides of natural origin at activity concentrations below 1 Bq/g for radionuclides in the uranium and thorium decay series and below 10 Bq/g for ^{40}K.[4] The Safety Guide states that the aforementioned values may be used in the definition of the scope of national regulations or to define radioactive material for the purpose of such regulations, as well as to determine whether material within a practice can be released from regulatory control.

3.1.2. Graded approach to regulation

Where the activity concentration values specified in Ref. [6] are exceeded, a graded approach to regulation as a practice is adopted in accordance with the requirements of the BSS (paras 2.8, 2.10–2.12 and 2.17) and the guidance given in Ref. [6]. Application of the graded approach to the regulation of operations involving exposure to NORM is described in Refs [4, 7] and is summarized in Sections 3.1.2.1–3.1.2.3.

3.1.2.1. Initial assessment

An initial assessment is made of the process in question, the materials involved and the associated exposures. For industries engaged in the processing of NORM, the exposure pathways to workers and members of the public that are most likely to require consideration are those involving external exposure to gamma radiation emitted from bulk quantities of process material and internal exposure via the inhalation of radionuclides in dust. Internal exposure via the inhalation of ^{220}Rn (thoron) and its progeny emitted from process material may also need to be considered during the exploitation of minerals containing relatively high concentrations of thorium, such as monazite and xenotime, especially where fine grained residues and/or enhanced radium levels are present and ventilation is poor. Internal exposure via ingestion is unlikely to require consideration under normal operational circumstances.

The assessment of the effective dose received by an individual involves summing the personal dose equivalent from external exposure to gamma radiation in a specified period and the committed equivalent dose or committed effective dose, as appropriate, from the intake of radionuclides in the same period. The assessment method is described in more detail in Ref. [4].

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[4] These criteria do not apply to radon, residues in the environment and commodities such as foodstuffs, drinking water and construction materials, which are normally treated as chronic exposure situations and subject to the requirements of intervention, nor do they apply to materials in transport, the criteria for which are specified separately in the Transport Regulations (see Section 3.1.3).

3.1.2.2. Regulatory options

The four basic options open to the regulatory body, in ascending order of degree of control, are as follows:

(1) The regulatory body may decide that the optimum regulatory option is not to apply regulatory requirements to the legal person responsible for the material. The mechanism for giving effect to such a decision could take the form of an exemption. For exposure to NORM, an exemption is likely to be the optimum option if the material does not give rise to an annual effective dose received by a worker exceeding about 1–2 mSv, i.e. a small fraction of the occupational dose limit [22], bearing in mind that the dose received by a member of the public in such circumstances is likely to be lower by at least an order of magnitude [7].

(2) Where a regulatory body has determined that exemption is not the optimum option, the minimum requirement is for a legal person to formally submit a notification to the necessary regulatory body of the intention to carry out the practice. As in the case of a decision to grant an exemption, this is an appropriate option when the maximum annual effective dose is a small fraction of the applicable dose limit, but it provides the added reassurance that the regulatory body remains informed of all such practices.

(3) Where the level of exposure to NORM is such that neither exemption nor the minimum regulatory requirement of notification is the optimum regulatory option, the regulatory body involved may decide that a legal authority has to meet additional (but limited) obligations to ensure that exposed individuals are adequately protected. These obligations would typically involve measures to keep exposures under review and to ensure that working conditions are such that exposures remain moderate, with little likelihood of doses approaching or exceeding the dose limit.[5] The mechanism for imposing such obligations on a legal person is the granting of authorization in the form of a registration [4].

(4) Where an acceptable level of protection can only be ensured through the enforcement of more stringent exposure control measures, authorization in the form of a licence may be required [4]. This is the highest level of the graded approach to regulation and its use for practices involving exposure

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[5] For situations in which workers are exposed to gamma radiation and radionuclides in inhaled dust, Ref. [22] states that, "Control, if considered necessary, would include the use of methods to suppress or contain any airborne dusts and general radiological supervision".

to NORM is likely to be limited to operations involving significant quantities of material with very high radionuclide activity concentrations.

3.1.2.3. Control measures for authorized practices

A detailed account of the control measures that may be appropriate for authorized practices involving work with minerals and raw materials is provided in Refs [4, 5]. In terms of the graded approach to regulation, the nature and extent of such measures will be commensurate with type of practice and levels of exposure, but will generally entail the establishment of some form of radiation protection programme with suitable provisions for monitoring and dose assessment at a more detailed level than in the initial assessment referred to in Section 3.1.2.1.

Specific radiological measures in the workplace, such as control of the occupancy period or even shielding may sometimes be appropriate to minimize external exposure to NORM. Materials with relatively low activity concentrations give rise to modest gamma dose rates (typically no more than a few microsieverts per hour), even on contact. In such cases, discouraging access, for example by storing materials in mostly unoccupied areas, may be sufficient. In areas containing materials with relatively high activity concentrations, physical barriers and warning signs may be necessary.

Exposure to airborne dust is likely to be controlled already in many workplaces through general occupational, health and safety (OHS) regulations. Control of air quality for the purpose of minimizing dust levels may also help to reduce radon and thoron concentrations. Therefore, the extent to which existing OHS control measures are effective in minimizing workers' radiation exposure is something that a regulatory body would first need to establish before deciding to impose additional control measures for purely radiological reasons. In some workplaces, existing OHS control measures alone may provide sufficient protection against internal exposure. In other workplaces, additional control measures specifically for radiation protection purposes may become necessary for achieving compliance with the standards. Engineered controls are the favoured option, with working procedures and, finally, protective respiratory equipment should be considered only when further engineering controls are not effective or practicable.

Complete containment of material is often impractical, especially where large quantities of low activity concentration materials are involved. Spills and the spread of materials outside a specific area are often of no radiological significance unless substantial and persistent airborne dust levels result. Prevention of resuspension of dust is therefore likely to be the most effective approach. Specific measures to control surface contamination only become meaningful when materials with higher activity concentrations are present.

Worker awareness and training are particularly important for supporting the introduction of local rules and for creating an understanding of the precautions embodied in such rules. Individual employee work practices may exacerbate dust generation and, in some cases may completely negate the effect of any engineering controls installed. There may be deficiencies in the way in which equipment maintenance tasks are undertaken, implying the need for periodic review to determine if improvements are possible. The general standard of housekeeping and spillage control also needs to be kept under regular review. Even when low activity concentration materials are handled, a reasonable standard of housekeeping may be necessary to ensure that dust resuspension is adequately controlled. Very high standards would generally be required in process areas where highly active material such as monazite is handled.

3.1.3. Applicability of the Transport Regulations to material in transport

3.1.3.1. Basic criteria

The safety requirements for material in transport are set out in the Transport Regulations [3]. The transport of material, in its natural or processed state, associated with the production of rare earths from thorium containing minerals may or may not fall within the scope of the Transport Regulations, depending on the activity concentration of a material. The Transport Regulations apply only if the activity concentration of a material exceeds ten times the activity concentration for exempt material.[6] For individual radionuclides of natural origin, the activity concentrations for exempt material are shown in Table 5.

3.1.3.2. Mixtures of radionuclides

For mixtures of radionuclides, the activity concentration for exempt material is:

$$X_{\mathrm{m}} = \frac{1}{\sum_i \frac{f(i)}{X(i)}} \qquad (1)$$

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[6] These activity concentration criteria apply only to materials containing radionuclides of natural origin that are either in their natural state, or have been processed for purposes other than extraction of the radionuclides, and which are not intended to be processed for the use of radionuclides.

TABLE 5. ACTIVITY CONCENTRATIONS FOR EXEMPT MATERIAL IN TRANSPORT

	Activity concentration (Bq/g)	Progeny included in secular equilibrium
Th-232 decay series		
Th_{nat} (Th-232)	1	Ra-228, Ac-228, Th-228, Ra-224, Rn-220, Po-216, Pb-212, Bi-212, Tl-208 (0.36), Po-212 (0.64)
Th-232	10	—
Ra-228	10	Ac-228
Th-228	1	Ra-224, Rn-220, Po-216, Pb-212, Bi-212, Tl-208 (0.36), Po-212 (0.64)
Ra-224	10	Rn-220, Po-216, Pb-212, Bi-212, Tl-208 (0.36), Po-212 (0.64)
U-238 decay series		
U_{nat} (U-238)	1	Th-234, Pa-234m, U-234, Th-230, Ra-226, Rn-222, Po-218, Pb-214, Bi-214, Po-214, Pb-210, Bi-210, Po-210
U-238	10	Th-234, Pa-234m
Th-234	1000	Pa-234m
U-234	10 or 100[a]	—
Th-230	1	—
Ra-226	10	Rn-222, Po-218, Pb-214, Bi-214, Po-214, Pb-210, Bi-210, Po-210
Pb-210	10	Bi-210, Po-210
Po-210	10	—
K-40	100	—

[a] The applicable value depends on the chemical form of the material.

where

X_m is the activity concentration for exempt material;
$f(i)$ is the fraction of activity concentration of radionuclide i in the mixture;
$X(i)$ is the activity concentration for exempt material of radionuclide i;

and the condition for application of the Transport Regulations (see Section 3.1.3.1) is:

$$\sum_i x(i) > 10 X_m \tag{2}$$

where *x(i)* is the activity concentration of radionuclide *i* in the mixture.

Combining Eqs (1) and (2) and making the substitution:

$$f(i) = \frac{x(i)}{\sum_i x(i)}$$

provides the condition for application of the Transport Regulations as:

$$\sum_i \frac{x(i)}{X(i)} > 10 \tag{3}$$

3.1.3.3. Material with decay chains in equilibrium

For materials in which the radionuclides in each of the uranium and thorium decay series are (or are deemed to be) in equilibrium, the values of activity concentrations for exempt material for U_{nat}, Th_{nat} and ^{40}K (see Table 5) can be used to derive the conditions for application of the Transport Regulations, with the progeny of ^{238}U and ^{232}Th automatically being taken into account. Eq. (3) then becomes:

$$\frac{x(U_{nat})}{1} + \frac{x(Th_{nat})}{1} + \frac{x(^{40}K)}{100} > 10 \tag{4}$$

3.1.3.4. Material with decay chain segments in equilibrium

Available data on the radionuclide composition of a material may indicate that equilibrium conditions do not prevail throughout the decay chains but that it may be possible to treat a material as a mixture of decay chain segments, each of which is assumed to be in equilibrium. In such cases, however, the available data and/or the information in Table 5 may not always be sufficiently detailed to determine unequivocally whether the Transport Regulations apply, in which case a conservative estimate may have to be made by assigning the highest individual radionuclide activity concentration in each decay chain or chain segment to all radionuclides in that decay chain or chain segment. Further information on how to proceed when insufficient data are available on individual radionuclide activity concentrations is provided in the Transport Regulations.

3.2. EXPOSURE TO GAMMA RADIATION

The main radionuclides contributing to gamma exposure are ^{228}Ac, ^{212}Pb and ^{208}Tl from the ^{232}Th decay series and ^{214}Pb and ^{214}Bi from the ^{238}U decay series. The highest gamma energy (2614 keV) is associated with ^{208}Tl. In the mining and beneficiation of rare earth minerals, exposure to gamma radiation arises mainly from accumulations of large amounts of mineral concentrates or residues. In the chemical processing of mineral concentrates, dose rates are generally highest near process tanks, filters and residue stockpiles. For workers, workplace monitoring or individual monitoring techniques, or a combination of both, are used.

3.3. EXPOSURE TO RADIONUCLIDES IN INHALED DUST PARTICLES

Airborne dust particles arise from the resuspension of contamination on floors and other surfaces, releases from operations and the conveying of minerals. For inhalation of such particles by workers in the rare earths industry, exposure to radionuclides in the thorium decay series is the main concern in regards to radiation protection. In situations where radionuclide activity concentrations in the materials being handled are low, as in the case of bastnäsite, it is important to recognize that the silica content of the airborne dust is likely to be of greater concern for occupational health than the radionuclide content.

3.3.1. Monitoring techniques for workers

In the rare earths industry, as with other NORM industries, routine determination of radionuclide intake by workers is in most cases achieved using techniques based on air sampling. This approach is consistent with the findings of an investigation into monitoring strategies and methods for optimization of internal exposures of workers to NORM, carried out for the European Commission [23]. One of the conclusions of that investigation was that, "Air sampling, rather than biological sampling (or whole body counting) is the best way of assessing doses and providing ALARA information."

Bioassay techniques are sometimes used, but their application requires specialist knowledge and facilities if they are to yield useful information and even then the results obtained may be subject to large uncertainties. The applicability of bioassay techniques for routine use in the rare earths industry is therefore limited. However, they can be useful for confirming conclusions drawn from monitoring programmes based on personal air sampling, particularly where estimated intake corresponds to an effective dose approaching or exceeding the applicable dose limit, and for clarifying the biokinetics of inhaled material. Bioassay techniques that have been investigated for determination of thorium intake include the measurement of thoron in breath, direct in vivo counting and the measurement of thorium in samples of excreta and blood. Each of these techniques has advantages and disadvantages. Using the LUDEP computer code [24] for implementing the ICRP respiratory tract model [25] and assuming an aerosol particle activity median aerodynamic diameter (AMAD) of 5 μm and lung absorption class S, it has been determined that long term (tens of years) inhalation of ^{232}Th at a rate of 1 Bq/a results in an accumulated lung burden of 0.16 Bq [26]. This value is consistent with the result of a calculation reported in Ref. [27], which showed that a continuous chronic intake of ^{232}Th at a rate of 1 Bq/d for 32 years results in a lung burden of 55.9 Bq.

3.3.1.1. Monitoring techniques based on air sampling

Guidance on the use of techniques based on air sampling for the monitoring of workers is provided in Ref. [4]. Such techniques involve the drawing of air through a filter to capture dust particles, which are then analysed by measuring the activities of alpha emitting radionuclides in the thorium and uranium decay series. The use of gross alpha activity measurements to determine intake is subject to the following considerations:

(a) In materials that have not been chemically processed, the equilibrium of the thorium and uranium decay chains is unlikely to be significantly disturbed, allowing equilibrium to be generally assumed for freshly generated airborne dust particles. However, as discussed in the Annex to Ref. [4], some radon and thoron may escape from the dust particles when they are analysed in the laboratory after a delay of some days. The resulting depletion in radon and thoron leads to a corresponding depletion in short lived radon or thoron progeny. For minerals with extremely low thoron and radon emanation coefficients, such as the heavy minerals zircon and monazite, 100% retention of thoron and radon can be assumed. On this basis, 1 Bq of ^{232}Th captured on a filter corresponds to 6 Bq of measured gross alpha activity, while 1 Bq of ^{238}U corresponds to 8.32 Bq of measured gross alpha activity. For other minerals, the retention can be expected to be in the range 50–100% and it would seem reasonable to assume 75% retention as being typical. On this basis, 1 Bq of ^{232}Th captured on a filter corresponds to 5.25 Bq of measured gross alpha activity, while 1 Bq of ^{238}U corresponds to 7.54 Bq of measured gross alpha activity.

(b) In materials that have been subject to chemical processing, equilibrium conditions in airborne dust particles can no longer be assumed and analysis may have to include the measurement of certain individual decay progeny.

Two basic types of air sampling techniques are currently in use: stationary air sampling (also known as workplace or static air sampling), in which a sampling device remains at a fixed location in a workplace, and personal air sampling, in which a sampling device is attached to a worker in a position such that the air sample is reasonably representative of the air breathed by the worker. In the rare earths industry, radionuclide intake by workers is in most cases determined by personal air sampling. The use of stationary air sampling can result in dust inhalation doses being significantly underestimated, sometimes by several orders of magnitude, particularly in workplaces where the resuspension of dust by worker activities is a significant factor [23].

Because of the difficulties in applying personal air sampling to every exposed worker all of the time, monitoring strategies usually involve the assignment of workers to work categories that reflect the general nature and scope of work activities. In many workplace situations involving exposure to dust containing radionuclides of natural origin, such exposure is not uniform within a work category since a worker may, during the course of a work shift, spend time in different exposure environments. This is said to be particularly true for workers involved in the separation of heavy-mineral sands [28]. A further complication arises in accounting for the wearing of respiratory protective equipment.

Air sampling equipment and techniques have been reviewed in terms of their applicability to radiation protection in workplaces involving exposure to radionuclides in airborne dust [23]. Some of the findings of that review are summarized in the following:

(i) Air samplers are designed to follow a specific particle size sampling convention based on industrial hygiene sampling criteria and therefore typically underestimate the true ambient aerosol and thus the activity inhaled. The degree of underestimation depends on the AMAD and geometric standard deviation (GSD) of the ambient aerosol and on the type of sampler used. A correction factor can be applied to minimize the degree of underestimation of the airborne activity concentration but this does not remove all of the uncertainty because the AMAD and GSD vary with the location, time and circumstances of dust production and can therefore never be known precisely.

(ii) Aerosol particle size distribution also has a significant effect on dose coefficient, leading to an additional source of uncertainty when assessing the effective dose due to inhalation of particles. The dependence of dose coefficient on AMAD is particularly strong for particles of lung solubility class S. When assessing the effective dose, it is important to select a sampler with an efficiency that follows as closely as possible the AMAD dependency of relevant dose coefficients.

(iii) Knowledge of the lung absorption class is important because it is needed for determining not only the most appropriate dose coefficient but also the type of sampler to best minimize errors arising from incomplete knowledge of particle size distribution.

(iv) The preferred type of sampling for minimizing dose assessment errors is inhalable sampling for particles in lung absorption class F and thoracic sampling for particles in lung absorption classes M and S. While particles of class M or S are likely to be encountered in many NORM industries, for instance in the processing of heavy-mineral sands, it should be pointed out that thoracic samplers are presently not as widely available as inhalable samplers and are often not suitable for alpha counting owing to the dust particles being collected on foam rather than flat filters.

(v) Where particle size distribution is not known, the assignment of an AMAD of 5 μm has been found to reasonably minimize errors in assessing effective dose, whatever the true value of AMAD, sampling type, radionuclide and lung class.[7] The assignment of a GSD of 2.5 is recommended by the ICRP for aerosols with an AMAD above 1 μm when the actual value is unknown [25]. Sampling efficiency correction factors, as referred to in (i), for a GSD of 2.5 are shown in Table 6.

The activity inhaled by workers, as determined from the alpha counting of dust samples collected on filters, may be underestimated if there is significant alpha particle self-absorption in large particles or in multilayers or agglomerates of smaller particles deposited on a filter. This possibility was investigated for monazite dust [29]. It was concluded that self-absorption effects could be minimized by restricting dust loadings on filters to less than 1 mg/cm^2. With the

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[7] The measurement of AMAD requires specialized equipment such as a cascade impactor.

TABLE 6. SAMPLING EFFICIENCY CORRECTION FACTORS FOR ESTIMATING THE AIRBORNE ACTIVITY CONCENTRATION OF PARTICLES WITH A GSD OF 2.5 [23]

AMAD (μm)	Correction factor		
	Inhalable sampler	Thoracic sampler	Respirable sampler
1	1.04	1.05	1.11
5	1.18	1.41	2.5
10	1.31	2.1	5.6

use of a 37 mm diameter filter and a typical volume of air of 1 m^3 sampled in a shift, this restriction would be met if the dust concentration did not exceed 10 mg/m^3.

Various types of filter mediums and sampling cassettes are available. Where dust concentration is relatively low (say, about 1–2 mg/m^3) and sampling is undertaken over a 4–6 h period, the choice of filter medium and cassette is not likely to be critical. However, when dust concentration is relatively high (more than about 3 mg/m^3) and sampling is undertaken for a period of 8 h or more, the selection of equipment requires more careful consideration. For some types of filter medium, such as PVC, part of the sample may be lost as a result of dust not fully adhering to the surface. For some types of monitoring cassette, dust may adhere to the inside wall, requiring it to be removed by washing and added to the material collected on the filter prior to radiometric analysis.

3.3.1.2. Measurement of thoron in breath

The measurement of thoron in breath is generally regarded as the most sensitive of the various bioassay techniques available for determining thorium intake [30–33]. The thoron contained in exhaled breath is used as a measure of the ^{224}Ra, and hence the ^{232}Th, contained in the lung. Exhaled thoron activity is expressed as an activity of the freely emanating ^{224}Ra parent which would support the thoron concentration measured at a subject's mouth. The technique has been used on workers at the Bayan Obo rare earths mine, China [32] and on several groups of workers involved in the separation of heavy-mineral sands [34, 35]. It provides a relatively inexpensive and portable means of detecting moderate levels of inhaled thorium in the body. However, the technique has the following disadvantages:

(a) The measurements have to be taken after a layoff period from active work following intake to account for the clearance of activity in the upper airways and the possible presence of short lived thoron progeny. The layoff period has to be at least 12 h but preferably 72 h to allow for seven half-lives of ^{212}Pb.

(b) The measurements require knowledge of the relationship between exhaled thoron, expressed as the emanating ^{224}Ra equivalent activity at the mouth, and the lung burden of thorium. This relationship, referred to as the thoron emanation rate, appears to depend on the nature of the thorium contamination, thereby making it important to calibrate breath measurement against in vivo measurements of thorium lung burden [36]. The calibration procedure requires workers with thorium lung burdens that are high enough to be detected by the in vivo gamma counting technique. Various estimates of the thoron emanation rate are reported in the literature:
   (i) The emanation rate is taken to be 9% in Ref. [30];
   (ii) For former thorium refinery workers, a value of 9.2% is reported [37];
   (iii) For workers in Australian heavy-mineral sand separation plants, the thoron emanation rate was determined experimentally by comparing exhaled thoron activity with the lung burden determined by in vivo counting. The mean value was found to be 4.7% in an initial study involving six workers [34] and 3.7% in a follow-up study involving 19 workers [35];
   (iv) In Ref. [37], a thoron emanation rate of 10% was contemplated. However, it was thought that this might underestimate the thorium lung burden because there may not have been time for equilibrium between ^{232}Th and ^{212}Bi to be established owing to exhalation of ^{220}Rn and to intermediate members of the decay chain, particularly ^{228}Ra and ^{224}Ra, having left the lung [38]. Consequently, based on the experience of the Argonne National Laboratory [39], a correction factor of 2 was applied [26], giving an overall thoron emanation rate of 5%, similar to the value of 3.7% reported in Ref. [35] (see (iii) above);
   (v) In Ref. [40], thorium chest burdens determined by thoron in breath measurements were compared with those determined by whole body counting. It was concluded from this comparison that the thoron emanation rate of 9% determined in previous studies is applicable only in the case of long term exposure situations in which a substantial portion of the thorium has been translocated from the lung to other organs. In the case of short exposure periods, it was concluded that most of the activity would be confined to the lungs and that a thoron emanation rate of 20% would seem more realistic.

The wide variation in these values illustrates why the use of the thoron in breath techniques is of limited value for dose assessment.

Two basic methods for measuring thoron in breath are reported:

(1) The first method, as described for instance in Refs [30, 34], is based on the so-called double filter system. Air from the lung is exhaled into a cylinder fitted with filters at both ends. Exhaled thoron decays during its transit and the progeny are collected on the exit filter. After a delay of 5 h to allow the progeny to decay, the alpha activity on the filter is measured using alpha counting.

(2) The second method, as described for instance in Refs [32, 41], is derived from the experience of the Argonne National Laboratory [42]. The method is based on electrostatic collection onto a negatively charged Mylar disc of ^{212}Pb, 85–88% of which is positively charged. After the collection period, the alpha decays can be measured using low level alpha spectrometry [33].

3.3.1.3. Direct in vivo counting

Thorium in the body can be measured by direct in vivo counting of major gamma energy peaks 0.911 MeV of ^{228}Ac and 2.61 MeV of ^{208}Tl, both radionuclides being progeny of ^{232}Th. Two types of measurement geometry are used: chest counting (static geometry) to measure radioactivity in the thorax region (see, for instance, Ref. [41]) and whole body counting (static or scanning geometry) to measure radioactivity in the subject from head to toe. In vivo whole body counting has been undertaken on some workers in mineral sand separation plants and monazite processing plants [34, 35]. However, due to the limited sensitivity of conventional counting techniques, incremental thorium intake from prolonged exposure can be detected by such techniques only when they are substantial and not when they result from the low airborne dust contamination levels usually encountered in the rare earths industry. Use of the technique in routine operations characterized by these more moderate intakes requires expensive, low background installations. Here again, a layoff period from active work of 72 h is advised to avoid interference from short lived thoron progeny inhaled or plated out on the body of an exposed individual.

3.3.1.4. Measurement of thorium in excreta

Techniques based on the sampling of excreta (see, for instance, Ref. [30]) may also suffer from limited sensitivity, owing to the low solubility of most types of thorium containing material inhaled. Interpretation of the dosimetric significance of measurements conducted on excreta samples is difficult and depends on the biokinetic model used, as demonstrated, for instance, in Refs [43, 44]. Alpha spectrometry and spectrophotometry are two commonly used techniques for low level determination of thorium intake by workers, but with new developments in other measurement techniques, such as neutron activation analysis (NAA) and inductively coupled plasma mass spectrometry (ICP–MS) of urine samples, it is now possible to achieve substantially lower limits of detection [45–50]. Faecal sampling has been conducted on workers in various thorium related industries in Australia [51], Brazil [52] and India [49]. Measurements of thorium in faeces are potentially very sensitive to recent exposures of thorium because the amount of thorium excreted, following a constant level of intake, reaches a steady value within days. However, while faecal measurements are reported to have application to the determination of both short term and long term intake [51], doubts have been raised as to their usefulness for determining long term intake [28]. It is pointed out in Ref. [53] that faecal sampling does not appear particularly useful for long term chronic intake, since 82% of inhaled Th is cleared very rapidly ($T_{½}$ <1 d) from the respiratory system.

3.3.1.5. Measurement of thorium in blood

Investigations of the concentration of thorium in the blood serum of Western Australian heavy-mineral sands workers have been conducted [48, 54], while investigations of the concentrations of thorium in both blood serum and clot of thorium plant workers have been conducted in India [55]. It is suggested in Ref. [54] that blood testing is a more reliable technique than urine testing, since variations in the intake of fluids such as water and alcohol, or loss of water through perspiration, cause significant differences in the amount of urine excreted.

3.3.1.6. Sensitivity of monitoring techniques

A comparison of the lower limits of detection for various techniques used to determine thorium intake is shown in Appendix V. The minimum detectable annual doses typically achievable by various monitoring techniques have been calculated [23]. The results for ^{232}Th series radionuclides in equilibrium are shown in Table 7 for air sampling techniques and Table 8 for bioassay techniques.

TABLE 7. TYPICAL SENSITIVITY OF AIR SAMPLING TECHNIQUES IN DETERMINING DOSES DUE TO INTAKE OF ^{232}Th SERIES RADIONUCLIDES [23]

Lung absorption class	Minimum detectable annual effective dose (mSv)					
	Personal sampling (2 L/min)			Static sampling (20 L/min)		
	Alpha	Beta	Gravimetric	Alpha	Beta	Gravimetric
Sampling duration of one week						
F	0.1	2.6	0.065	0.01	0.26	0.0065
M	0.034	0.83	0.021	0.0034	0.083	0.0021
S	0.031	0.76	0.019	0.0031	0.076	0.0019
Sampling duration of one day						
F	0.52	13	0.33	0.052	1.3	0.033
M	0.17	41	0.11	0.017	4.1	0.011
S	0.16	38	0.097	0.016	3.8	0.0097
Sampling duration of one hour						
F	2.6	64	1.6	0.26	6.4	0.16
M	0.85	21	0.53	0.085	2.1	0.053
S	0.78	19	0.49	0.078	1.9	0.049

Note: The values for alpha counting (low background) and beta counting (high background) are based on a background count of 0.01 and 0.1 counts/s, respectively, and on a counting time of 1 h. The values for gravimetric analysis are based on an assumed activity concentration of Th-232 of 10 Bq/g.

3.3.2. Calculation of effective dose

3.3.2.1. *Dose coefficients for individual radionuclides*

Dose coefficients (values of committed effective dose per unit intake of activity) for the inhalation of radionuclides by workers and members of the public are specified in a database compiled by the International Commission on Radiological Protection (ICRP) [56]. The values are based on a model for the respiratory tract described in Ref. [25]. The dose coefficients for workers quoted in Ref. [56] cover a wide range of AMADs. Most of the values for AMADs of 1 and 5 μm also appear in Table II–III in Schedule II of the BSS [2]. Dose

TABLE 8. TYPICAL SENSITIVITY OF BIOASSAY TECHNIQUES IN DETERMINING DOSES DUE TO INTAKE OF ^{232}Th SERIES RADIONUCLIDES [23]

Lung absorption class	Minimum detectable annual effective dose (mSv)			
	Lung counting twice per year (Th-232)	Whole body counting twice per year (Ra-228)	Urine sampling 2–12 times per year (Th-232)	Faecal sampling 4 times per year (Th-232)
F	—	>20	0.2–57 (2 samples per year)	3–30
M	>20	>20	0.2–63 (4 samples per year)	0.1–12
S	19–27	>20	6–1500 (2 samples per year)	0.05–7

Note: The value ranges of minimum detectable dose correspond to the range of detection limits reviewed in literature applied to the mean dose coefficient measured for the considered monitoring frequency. The considered monitoring frequency is the minimum value which ensures that uncertainty about the time of intake will not lead to a relative dose estimate error significantly greater than a factor of three.

coefficients for members of the public are specified in Ref. [56] for a default AMAD of 1 µm and also appear in Table II–VII in Schedule II of the BSS [2].

It is pointed out in Ref. [23] that in most NORM industries the radioactive components of the material involved, and thus of their aerosol particles, are contained within a matrix of non-radioactive elements and their compounds. These matrices determine the solubility of the particles and it is therefore appropriate to choose the same lung absorption class for all radionuclides contained within them. Many of these materials are highly resistant to chemical attack and their dust particles are obviously lung absorption class S. This is said to apply to materials such as heavy-mineral sands (including, notably, monazite [48]) and radium rich process scales. However, lung absorption class S may not be appropriate for materials that have been subjected to vigorous chemical treatment, as occurs in processes for extracting rare earths from mineral concentrates. In such cases, it may be more appropriate to determine the lung absorption class experimentally for the particular material involved.

Table 9 sets out inhalation dose coefficients applicable to workers in industrial operations involving low solubility airborne dust particles. The coefficients relate to lung absorption class S to the extent possible and cover all radionuclides with significant dose contributions. In the few instances where a value for class S is not available, the value provided is that corresponding to the next fastest lung absorption class for which information is available.[8] Most of the values in Table 9 are taken from Ref. [56]. In some cases (denoted by figures in italics), values for lung absorption class S are not available from this source and data are quoted instead from the results of other calculations [23, 57] similarly based on the ICRP respiratory tract model described in Ref. [25]. Table 10 sets out the inhalation dose coefficients applicable to workers in industrial operations involving medium solubility airborne dust particles such as those that might occur in chemical processing facilities. The coefficients relate to lung absorption class M and are taken from Ref. [56] or, for figures in italics, from Ref. [23].

Many assessments of worker doses reported for NORM industries are based on the inhalation dose coefficients quoted in Table II–III in Schedule II of the BSS [2] for AMADs of 1 or 5 µm, since these coefficients have been agreed upon by international consensus and continue to provide the basis for regulation in accordance with the Standards. However, the BSS do not provide coefficients for ^{228}Ra, ^{224}Ra, ^{226}Ra, ^{210}Pb and ^{210}Po in lung absorption class S, with the result that, for low solubility particles, the coefficients for class M (or F in the case of ^{210}Pb) have to be used instead. For medium solubility particles, the coefficients for lung absorption class F have to be used for ^{210}Pb. In addition, the dose coefficients for ^{228}Th provided in Tables 9 and 10 are revised values that were incorporated into the ICRP database [56] after the publication of the BSS. For 5 µm AMAD low solubility particles with a ^{232}Th activity concentration 5–10 times that of ^{238}U (typical of bastnäsite and monazite) and with decay progeny in equilibrium, use of the BSS dose coefficients instead of those in Table 9 implies a 7–8% underestimation of total inhalation dose. Considering the many uncertainties associated with any dose assessment of this nature, these differences are of no great consequence. For medium solubility particles, the use of the BSS dose coefficients instead of those in Table 10 makes very little difference, irrespective of the degree of equilibrium.

Derivation of the dose coefficients for ^{226}Ra quoted in the ICRP database [56] and the BSS [2] is based on the assumption that ^{222}Rn produced in the respiratory tract does not have the same biokinetic behaviour as its parent ^{226}Ra, but rather is removed to the environment at a rate of 100 d^{-1} (see Annex B of Ref. [58]). The derivation in Ref. [23] of corresponding ^{226}Ra dose coefficients for lung absorption class S quoted in Table 9 is based on this same assumption. It is suggested in Ref. [23] that if the radon removal rate is very much lower than 100 d^{-1} and remains so when a particle is inhaled, alternative dose coefficients for ^{226}Ra in lung

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[8] The use of dose coefficients for lung absorption classes other than class S in these few instances may lead to some underestimation of the dose.

TABLE 9. RADIONUCLIDE SPECIFIC INHALATION DOSE COEFFICIENTS FOR WORKERS (LOW SOLUBILITY DUST PARTICLES)

	Dose coefficient for specified AMAD (µSv/Bq)[a]				
	0.3 µm	1 µm	3 µm	5 µm	10 µm
Thorium decay series					
Th-232	32	23	17	12	8.1
Ra-228	3.3 (M)	15	14	11	7.1
Th-228	45	37	33	25	18
Ra-224	3.3 (M)	2.9 (M)	3.1 (M)	2.8	1.3 (M)
Series in equilibrium (dose per unit activity of the parent radionuclide)	83.6	77.9	67.1	50.8	34.5
Uranium decay series					
U-238	10	7.3	7.2	5.7	3.5
U-234	12	8.5	8.6	6.8	4.1
Th-230	18	13	10	7.2	5.2
Ra-226	4.4 (M)	8.7	8.7	6.9	4.2
Pb-210	0.76 (F)	5.2	5.3	4.3	2.7
Po-210	3.9 (M)	3.9	3.5	2.7	1.7
Ac-227 (from U-235 series)[b]	88	66	52	47	27
Series in equilibrium (dose per unit activity of the parent radionuclide)[b]	53.1	49.6	45.7	35.8	22.6

[a] The dose coefficients quoted are those corresponding to lung absorption class S, except where specified in parentheses as M or F. Values shown in italics are taken from Ref. [23] (rounded to two significant figures) or from Ref. [57] in the case of ^{224}Ra. All other values are taken from Ref. [56].

[b] The natural abundance of U-235 is 0.711%, resulting in an activity concentration in natural uranium of 0.046 relative to that of U-238. Consequently, the relative contributions to the inhalation dose from the U-235 decay series radionuclides are insignificant except for that from Ac-227. The dose coefficient for Ac-227 is multiplied by 0.046 in calculating the dose coefficient for the full series.

TABLE 10. RADIONUCLIDE SPECIFIC INHALATION DOSE COEFFICIENTS FOR WORKERS (MEDIUM SOLUBLITY DUST PARTICLES)

	Dose coefficient for specifiedAMAD (µSv/Bq)[a]				
	0.3 µm	1 µm	3 µm	5 µm	10 µm
Thorium decay series					
Th-232	54	42	37	29	18
Ra-228	3.3	2.6	2.2	1.7	0.98
Th-228	36	30	28	22	12
Ra-224	3.3	2.9	3.1	2.4	1.3
Series in equilibrium (dose per unit activity of the parent radionuclide)	96.6	77.5	70.3	55.1	32.3
Uranium decay series					
U-238	3.8	2.6	2.2	1.6	1.1
U-234	4.4	3.1	2.7	2.1	1.4
Th-230	51	40	35	28	17
Ra-226	4.4	3.2	2.8	2.2	1.5
Pb-210	0.76 (F)	1.0	0.93	0.74	0.45
Po-210	3.9	3.0	2.8	2.2	1.1
Ac-227 (from U-235 series)[b]	260	210	180	150	90
Series in equilibrium (dose per unit activity of the parent radionuclide)[b]	80.2	62.6	54.7	43.7	26.7

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[a] The dose coefficients quoted are those corresponding to lung absorption class M, except where specified in parentheses as F. Values shown in italics are taken from Ref. [23] (rounded to two significant figures). All other values are taken from Ref. [56].

[b] The natural abundance of U-235 is 0.711%, resulting in an activity concentration in natural uranium of 0.046 relative to that of U-238. Consequently, the relative contributions to the inhalation dose from the U-235 decay series radionuclides are insignificant except for that from Ac-227. The dose coefficient for Ac-227 is multiplied by 0.046 in calculating the dose coefficient for the full series.

absorption classes S and M are needed in order to avoid an underestimation of dose. However, for typical rare earth minerals, in which the ^{232}Th decay chain predominates over the ^{238}U decay chain, there is no strong case for using such alternative dose coefficients since the degree of underestimation of the total dose from all radionuclides, calculated from Ref. [56], is only a few per cent.

3.3.2.2. Overall dose coefficients for typical rare earths process materials

Overall dose coefficients, expressed in terms of ^{232}Th activity, for low and medium solubility dust particles of typical rare earths process materials, can be deduced from Tables 9 and 10 assuming decay chains in equilibrium and any particular ratio of ^{232}Th activity concentration to ^{238}U activity concentration. However, when carrying out dose assessments on the basis of air sampling measurements, the measured quantity is usually gross alpha activity. Therefore, it is better to express dose coefficients in terms of gross alpha activity measured on a sampling filter, requiring a relationship between gross alpha activity and ^{232}Th activity be established. This relationship, shown in Fig. 3, depends not only on the relative contributions to gross alpha activity from the ^{232}Th and ^{238}U decay chains but also on the degree to which thoron and radon are retained within the dust particles captured on a sampling filter, as explained in Section 3.3.1.1. Overall dose coefficients expressed in terms of gross alpha activity retained on a sampling filter are shown in Fig. 4 for general mineral dust (assuming

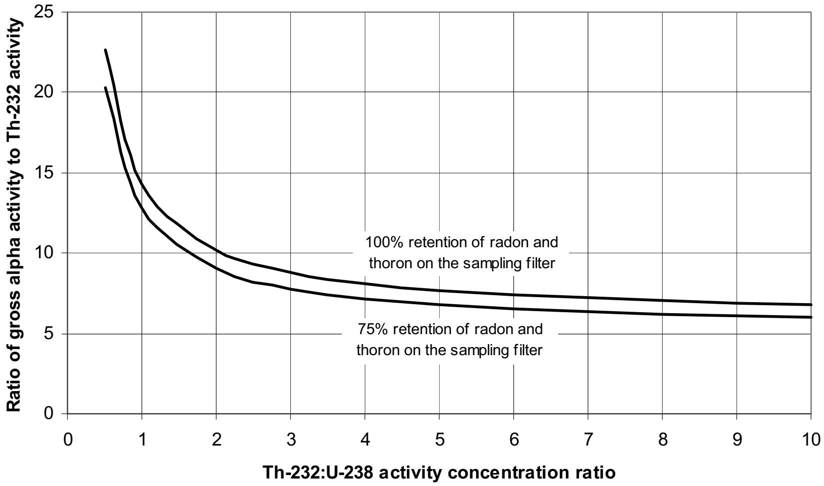

FIG. 3. Relationship between gross alpha activity and ^{232}Th activity in dust particles captured on a sampling filter.

75% thoron and radon retention) and Fig. 5 for heavy-mineral dust (assuming 100% thoron and radon retention).

3.4. INHALATION EXPOSURE OF WORKERS TO THORON AND RADON

3.4.1. Monitoring of exposures

Practical experience has demonstrated that thoron and radon activity concentrations in heavy-mineral sands and associated operations are low and, in such circumstances, occasional confirmatory monitoring would generally be all that is required. Area monitoring techniques focusing on poorly ventilated areas, enclosed vessels and storage areas for thorium-containing materials are normally sufficient.

In industries in which materials containing significant levels of thorium are handled, such as monazite processing, regular monitoring of thoron progeny is likely to be required. Again, area monitoring using grab sampling techniques is

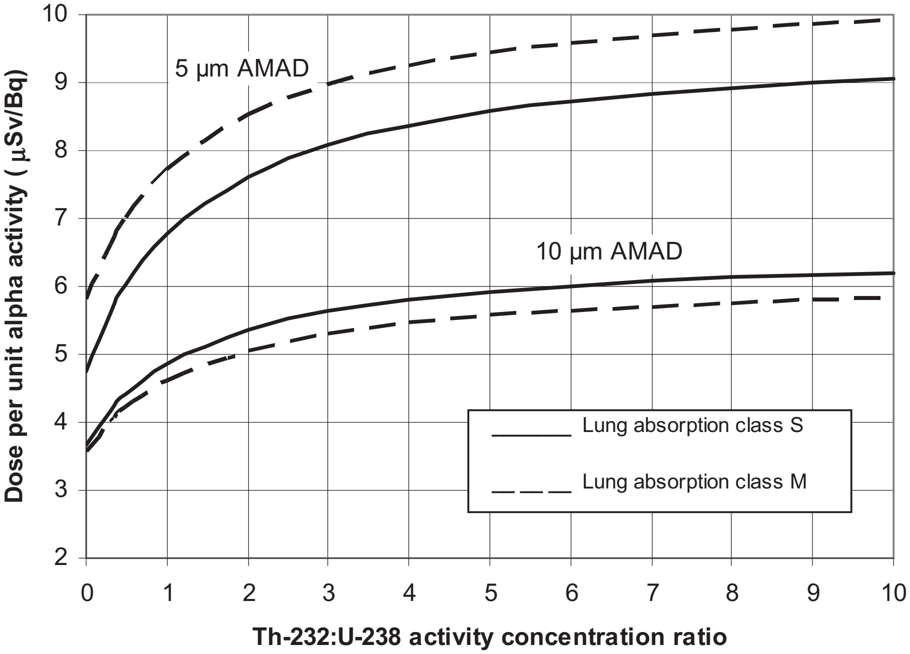

FIG. 4. Dose coefficients for gross alpha activity, assuming 75% retention of radon and thoron on a sampling filter.

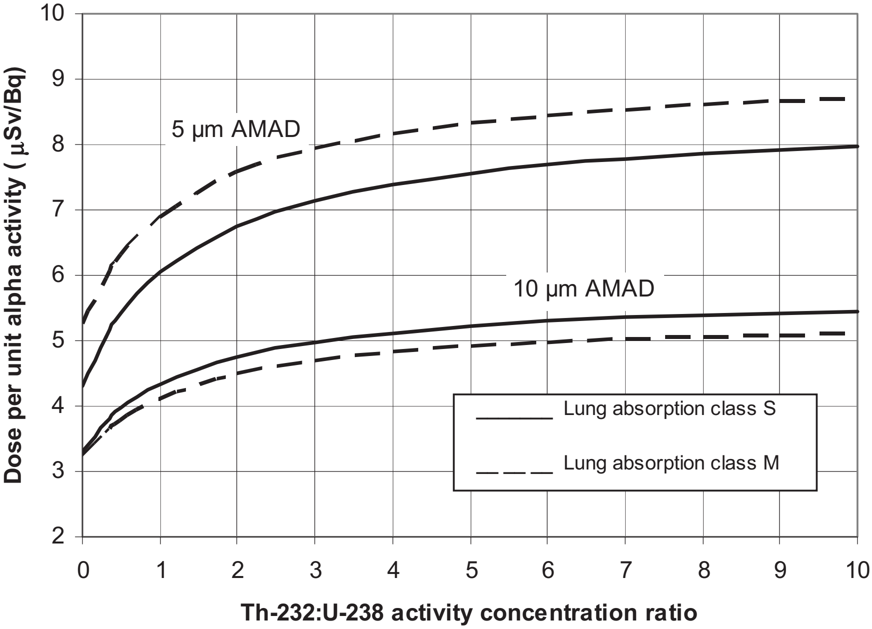

FIG. 5. Dose coefficients for gross alpha activity, assuming 100% retention of radon and thoron on a sampling filter.

commonly employed. It is likely that progeny will be severely out of equilibrium with a parent. Primarily because of the short half-life of thoron compared with that of some of its progeny, there is often a considerable difference between the spatial distribution of the gas and its progeny in enclosed workplaces, making an assessment of thoron equilibrium more difficult than for radon. Measurement of progeny rather than a parent gas is regarded as the easier and more appropriate approach, although it has been pointed out that concentrations of both thoron gas and its progeny may need to be measured in close proximity to sources containing high concentrations of thorium, where localized thoron concentrations can be very high [59].

Of the thoron progeny, only ^{212}Pb and ^{212}Bi make any significant contribution to total potential alpha energy. The contribution per becquerel of ^{212}Pb is 0.0691 µJ, while the contribution per becquerel of ^{212}Bi is 0.00656 µJ. The contribution per becquerel of the parent radionuclide ^{220}Rn is very small in comparison — nearly two orders of magnitude lower than that of ^{212}Bi. Since the majority of potential alpha energy is provided by ^{212}Pb, measurement of activity concentrations of this radionuclide in the air (in becquerels per cubic metre) usually allows a good estimate to be made of total potential alpha energy concentration (PAEC) (in joules per cubic metre).

Various measuring techniques are available for the measurement of thoron and its progeny in workplaces. The counting methods for radon and its progeny described in Refs [60, 61] can in principle be adapted for thoron and its progeny, with certain limitations. Some continuous monitoring instruments can also handle thoron and its progeny. For individual monitoring of workers, an integrating device based on the principle of nuclear track detection is capable of measuring exposure to thoron progeny. Details of some of the more commonly used measurement techniques are given below:

(a) For assessment of exposure to thoron progeny using the method described in Ref. [62], a known volume of air is drawn through a filter paper for a sampling period of less than 60 min (usually 10 min). The filter paper is subsequently counted for alpha activity after a delay period of 5–17 h after the end of sampling. By this time, ^{212}Pb attains transient equilibrium with its alpha emitting progeny ^{212}Bi. The ^{212}Pb activity is calculated after correcting for decay. At any one time, the amount of alpha activity on the filter paper and the decay characteristics exhibited depend on the quantities of the two radionuclides present. A conversion factor, which is dependent upon the delay period, converts alpha disintegrations per minute to thoron progeny concentration in joules per cubic metre. The activity of each radionuclide can be determined by conducting a sequence of counts at various intervals after sampling and solving relevant radioactive ingrowth and decay equations. A simple two count method is described in Ref. [59].

(b) For thoron gas, a passive alpha track detector technique has been developed which measures both radon and thoron [63]. The technique, based on the electret ion chamber generally used to measure radon, has been modified to measure thoron [64].

(c) Thoron gas can be directly estimated using the modified scintillation method, which involves the determination of the total alpha disintegrations from thoron and its immediate progeny ^{216}Po in a known volume of air [60, 65, 66]. Filtered air is collected in a scintillation cell and counted for 10 min. From the count, the number of thoron atoms and thoron activity are calculated. The method is quick, but limited in accuracy at concentrations below 400 Bq/m^3.

(d) Another method for measuring thoron and radon concentrations makes use of a double filter sampler [60]. This method can also be used for measuring thoron in breath to determine thorium intake by workers (see Section 3.3.1.2). The inlet filter collects all suspended particles including thoron and radon decay progeny and allows only thoron and radon gases into the chamber along with the sampled air volume. The thoron and radon gases decay partially while in transit through the chamber and most of the decay progeny are collected on the exit filter. This filter is subjected to programmed alpha counting and thoron and radon gas concentrations are back-calculated from the counting data [67, 68]. Double filter samplers of small and large capacities (2.4 and 150 L) have been used for sampling thoron (and radon) in mineral processing plants in India [65, 68]. The LLD is reported in Ref. [65] to be 27 Bq/m^3 for thoron and 0.7 Bq/m^3 for radon.

(e) Because of the different half-lives in the radon and thoron decay chains (see Appendix I), techniques have been developed in which the time differences between pulses from these decay chains in detectors can be used to distinguish between them and measure their activities separately [65, 70].

(f) One type of personal alpha dosimeter records alpha emissions from ^{212}Po separately, which allows for direct measurement of exposures to thoron progeny [71].

3.4.2. Calculation of effective dose

Dose coefficients for the progeny of thoron and radon, derived from numerical data provided in the BSS [2], are given in Table 11. Assuming an annual exposure period of 2000 h, inhalation of thoron progeny at a PAEC of 1 μJ/m^3 will give rise to an annual dose of 0.96 mSv. The corresponding figure for radon progeny is 2.8 mSv.

TABLE 11. DOSE COEFFICIENTS FOR OCCUPATIONAL EXPOSURE TO THE PROGENY OF THORON AND RADON

	Dose coefficient	
	(mSv per mJ·h·m^{-3})	(mSv per mJ)
Thoron progeny	0.48	0.39
Radon progeny	1.4	1.2

4. MINING AND BENEFICIATION OF BASTNÄSITE–MONAZITE ORE AT BAYAN OBO, CHINA

The deposit at the Bayan Obo mine, China, which contains a mixture of bastnäsite and thorium deficient monazite, accounts for nearly half of worldwide rare earths production (see Section 2.3). The excavated ore typically contains 30–35% iron, 5–6% REO and 0.13% niobium.

4.1. PROCESS DESCRIPTION

4.1.1. Main process

The ore is mined from two large open pits using standard mining methods involving drilling and blasting, followed by loading with electric shovels. It is then transported 150 km by rail to ore processing facilities. During beneficiation to recover the rare earth content of the ore, magnetite, fluorite, haematite and niobium oxide are recovered. Iron and steel are produced as co-products. Various schemes for the production of rare earth concentrates are described in literature [13]. They involve crushing, grinding and classification of the ore to 90% <74 µm, followed by the use of techniques such as flotation, magnetic separation, table separation and washing to produce a mineral concentrate containing 65–67% bastnäsite and 14–15% monazite [72]. The REO content is 61% (75% as bastnäsite and 25% as monazite [73]).

The mineral concentrate is subjected to further ore dressing to produce separate bastnäsite and monazite concentrates. This selective separation of bastnäsite and monazite has been the subject of several investigations of flotation processes using a selective collector or selective depressant, with a view to achieving more efficient separation of the less radioactive component bastnäsite. In one such investigation [73], 84.7% of the bastnäsite was recovered in a concentrate that contained 97.2% bastnäsite and 69.5% REO. However, owing to partial inter-growth of bastnäsite and monazite in the ore, the complete separation of bastnäsite from monazite is not achievable and the production of a third mineral fraction, a mixed bastnäsite–monazite concentrate containing 40–60% REO, becomes inevitable [11].

4.1.2. Management of residues

A large amount of waste rock, comprising topsoil, host rock and low grade mineralized rock, is generated during mining and is deposited in piles at the mine site. Sprinklers are deployed to control the generation of dust. A small portion of the low grade mineralized rock is used for road construction and for the embankment of a new tailings pond and plans are in place for more to be used in the future. The topsoil and host rock may be returned to the excavation as backfill when the mining operation is complete. Tailings from the beneficiation process are pumped to a tailings pond for storage. In order to reduce airborne dust, water spraying is applied to wet the surface of the tailings, the tailings pond is covered with waste water from ore processing and trees are planted in the surrounding area.

The production of iron and steel generates blast furnace slag and ferrous slag. The slag is conveyed to a slag dump for storage and about half of it is used to recover iron and to make building materials. Liquid effluents are pumped to the tailings pond. The production of construction materials from blast furnace slag involves mixing it with low activity material such as flyash in a ratio such that the activity concentration is sufficiently reduced to meet the radiological requirements for construction materials. A formula from a manufacturer for making bricks is: 60–70% steel slag, 10–15% blast furnace slag, about 10% flyash and 12–17% cement.

4.2. RADIONUCLIDE ACTIVITY CONCENTRATIONS

The ore is generally considered to contain 0.04% ThO_2 [26], corresponding to a ^{232}Th activity concentration of 1.4 Bq/g, although a higher ^{232}Th activity concentration of 2.6 Bq/g has also been reported [74]. The ^{232}Th activity concentrations in the process materials are shown in Table 12.

4.3. OCCUPATIONAL EXPOSURE

4.3.1. Exposure pathways

It is reported in Ref. [32] that the mine employs nearly 7000 workers, of which about 3000 are exposed to thorium containing airborne dust (2001 data). Elevated thoron concentrations in air are also found. Exposure to gamma radiation is significant only in the mining areas.

TABLE 12. RADIOACTIVITY CONTENT OF MINERAL CONCENTRATES, RESIDUES AND BY-PRODUCTS

	ThO_2 (%)	Radionuclide activity concentration (Bq/g)		Ref.
		Th-232	U-238	
Bastnäsite concentrate	0.15–0.22	*5.3–7.8*	—	[11]
Monazite concentrate	0.17–0.40	*6.1–14*	—	[11]
Bastnäsite–monazite concentrate, 40% REO	0.28	*10*	—	[75]
Bastnäsite–monazite concentrate, 60% REO	0.2	*7.1*	—	[72]
Rare earth concentrate	0.2	*7.1*		[76]
Iron concentrate	0.0073	*0.26*	—	[76]
Concentrates (unspecified)	*0.27*	9.6	0.020 (Ra-226)	[74]
Tailings	*0.028*	1.0	0.022	[74]
Tailings	*0.048*	1.6	—	[76]
Ferrous slag from iron and steel production	*0.014–0.044*	0.5–1.6	—	[76]
Bricks made from slag	*0.006*	0.212	0.051 (Ra-226)	[76]
Cement made from slag	*0.007–0.009*	0.24–0.33	0.02–0.08	[76]

Note: Values in italics are derived, assuming that 1 g of thorium oxide contains 3566 Bq of Th-232.

4.3.2. Gamma dose rates

Absorbed gamma dose rates have been measured at the mining facility over a period of many years and are summarized in Table 13. It can be concluded from these values that the general level of external gamma exposure is moderate in all working areas of the mine.

TABLE 13. GAMMA DOSE RATES AT THE MINE SITE AND ITS SURROUNDINGS

	Absorbed dose rate (μGy/h)	Ref.
Mining areas	2.31	[32]
Mining areas	1	[26]
Mining areas	0.6–2	[76]
Disposal sites	0.4–0.8	[76]
Some areas with low grade mineralized rock	Up to 1.2	[76]
RE plant, tailings pond, sintering plant, iron plant	0.3–0.5	[76]
Local natural background (for comparison)	0.085	[76]

4.3.3. Radionuclides in airborne dust

The dustiest working area is the crushing area. During the period 1982–1991, the average airborne dust concentration was 188.7 mg/m^3, with a range of 9.3–875 mg/m^3 [32]. However, it is noted in Ref. [77] that improvements to ventilation and dust control systems during this period resulted in dust levels in 1991 being 20 times lower than those in 1983, as reflected in the results of a follow-up investigation in 1993–1994, which showed an airborne dust concentration of 1.6–74 mg/m^3 in the crushing area and 1.2–16.4 mg/m^3 in the six other dust-generating areas [26]. Given that the ThO_2 concentration of the ore is 0.04% (see Table 2), these more recently measured dust concentrations correspond to ^{232}Th activity concentrations in air of 2.3–106 mBq/m^3 in the crushing area and 1.7–23 mBq/m^3 in the other dusty areas.

During the period 1983–1994, 1301 measurements of thoron in breath were carried out on 781 mine workers exposed to airborne dust containing thorium, of whom 638 worked in areas with thorium containing dust (1158 measurements) and 143 worked in non-dusty areas (143 measurements) [32]. The average ^{232}Th lung burden determined from these measurements was 1.60 Bq for workers in dusty areas and 0.30 Bq for workers in non-dusty areas. Assuming that a long term intake at a rate of 1 Bq/a gives rise to a lung burden of 0.16 Bq (see Section 3.3.1), the mean long term intake of ^{232}Th can be estimated to be 10 Bq/a in dusty areas and 1.9 Bq/a in non-dusty areas. The distribution of ^{232}Th lung burdens for workers in dusty areas is shown in Table 14, together with corresponding estimates of annual intake. These results show that 92% of

TABLE 14. OVERALL DISTRIBUTION OF THORIUM LUNG BURDEN AND INTAKE FOR 638 DUST EXPOSED WORKERS AT BAYAN OBO MINE *(from Ref. [32])*

Number of workers	Th-232 lung burden (Bq)	Estimated long term intake (Bq/a)
585	<2.19	<14
43	2.22–4.41	14–28
2	4.44–6.63	28–41
2	6.67–8.85	42–55
5	8.89–11.07	56–69
1	11.11	69

workers had lung burdens of less than 2.19 Bq (corresponding to a long term intake of less than 14 Bq/a) and 98% had lung burdens below 4.44 Bq (corresponding to a long term intake of less than 28 Bq/a). Improvements to ventilation and dust control systems during the period under consideration resulted in thorium lung burdens of dust exposed workers in 1991 being three times lower than those in 1983 [77].

In a more specific investigation, conducted during the period December 1993 to August 1994 [26], 136 dust exposed workers were randomly selected from all types of dust generating workplaces. The investigation included an assessment of the average thorium lung burden for workers in the crushing area and each of the other six (less dusty) workplace areas. The results are shown in Table 15. The higher airborne dust concentrations measured in the crushing area are clearly reflected in higher thorium lung burdens.

4.3.4. Thoron and radon progeny

During the period 1982–1991, the average PAEC of thoron progeny in the crushing area was 10 800 Mev/L (1.73 μJ/m^3), with a range of 420–67 700 MeV/L (0.067–10.85 μJ/m^3) [32]. The average PAEC of radon progeny was reported to be about 10 times lower. In a subsequent investigation, the PAEC of thoron progeny in the crushing area during 1991 was reported to be in the range of 270–73 600 MeV/L (0.043–11.79 μJ/m^3) with the PAEC of radon progeny at each measurement location being 6–18 times lower [26]. The PAEC of thoron progeny in the other six (less dusty) types of working area was in the range

TABLE 15. THORIUM LUNG BURDEN AND INTAKE FOR 136 DUST EXPOSED WORKERS AT BAYAN OBO MINE BY WORKPLACE CATEGORY *(from Ref. [26])*

	Number of workers	Th-232 lung burden (Bq)		Estimated mean long term intake (Bq/a)
		Range	Mean	
Crushing	64	0.13–7.26	1.71	11
Lowest exposed group	36	0.13–1.45	0.82	5
Intermediate exposed group	10	1.52–2.18	1.74	11
Highest exposed group	18	2.38–7.26	3.76	24
Other	72	0.11–1.45	0.58	4
Main mine	20	0.11–1.25	0.64	4
Eastern mine	17	0.20–1.32	0.59	4
Train transportation	10	0.13–0.66	0.39	2
Bus transportation	10	0.13–0.92	0.45	3
Technical electricity	8	0.26–1.45	0.68	4
Highway	7	0.20–1.39	0.62	4

of 200–440 MeV/L (0.032–0.070 μJ/m^3). A more recent report [76] shows a thoron progeny PAEC of 226.1 MeV/L (0.036 μJ/m^3) at the disposal site. A summary of these results, together with estimates of corresponding annual exposures (assuming, conservatively, an annual exposure period of 2000 h) are shown in Table 16. Exposure limits specified in the BSS [2] are also included for comparison. All exposures are well below the BSS limits.

4.3.5. Effective dose

Annual effective doses received by workers from exposure to external gamma radiation are reported in Ref. [76]. General workers in areas of elevated gamma dose rates are estimated to receive a dose of 0.24 mSv/a, assuming an annual exposure period of about 2000 h. Workers involved in mining, including trucking of the ore, are estimated to receive a dose of about 1 mSv/a or more, while for workers at disposal sites the estimated dose is 0.7 mSv/a.

TABLE 16. EXPOSURE TO THORON AND RADON PROGENY AT THE BAYAN OBO MINE [26, 32, 76]

	PAEC (μJ/m^3)		Potential alpha energy exposure (mJ·h·m^{-3})			
			Estimated value		Limit [2]	
	Average	Range	Average	Range	Five year average	Single year maximum
Thoron progeny					42	105
Crushing area 1982–1991	1.73	0.067–10.85	3.5	0.13–22		
Crushing area 1991	—	0.043–11.79	—	0.08–24		
Other dusty areas 1991	—	0.032–0.070	—	0.06–0.1		
Disposal sites 1998	0.036	—	0.072			
Radon progeny					14	35
Crushing area 1982–1991 (approximate values)	0.2	0.004–1	0.4	0.01–2		

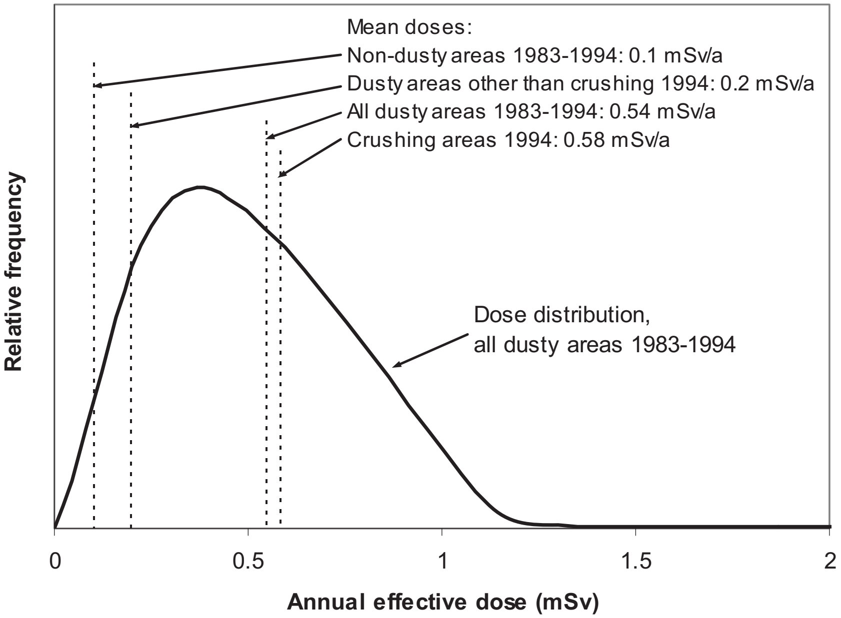

FIG. 6. Dust inhalation doses estimated to have been received by workers at Bayan Obo mine.

The committed effective doses received by workers from the inhalation of thorium containing dust can be estimated by applying the relevant 5 μm AMAD dose coefficient listed in Table 9 to the thorium intake amounts reported in Section 4.3.3. The results are shown in Fig. 6. The mean committed effective dose for the dustiest area (the crushing area) is 0.58 mSv/a and from the dose distribution it can be seen that very few workers are likely to receive a dose of more than 1 mSv, irrespective of the type of dusty area in which they work.

The committed effective doses received by workers from the inhalation of thoron and radon progeny can be estimated by applying the dose coefficients listed in Table 11 to the potential alpha energy exposures reported in Table 16. For the area with the highest thoron and radon progeny concentrations (the crushing area), the mean committed effective dose, assuming an annual exposure period of 2000 h, is 1.68 mSv/a for thoron progeny and about 0.56 mSv for radon progeny.

4.3.6. Measures to reduce doses

Measures to reduce doses and the effect of these measures are reported in Refs [77, 78]. During the period 1983–1991, average airborne dust concentrations in the workplaces were reduced by a factor of 20 (from 1144 to 48 mg/m^3) as a result of improvements to the ventilation and dust control equipment. Other measures to reduce dose included instruction in the use of personal protective equipment and the introduction of job rotation for those few workers having an estimated thorium lung burden higher than 4.44 Bq (about 1.6% of dust-exposed workers, see Table 14). As a result of these dose reduction measures, the average thorium lung burdens of dust exposed workers decreased by a factor of three (from 0.85 to 0.25 Bq).

4.4. PUBLIC EXPOSURE

An assessment of the radiological impact of the mining and beneficiation facility on the public is reported in Ref. [76]. Soil in areas downwind of the mine, including the Bayan Obo city area, was found to be contaminated with dust that had blown in from the mining facility. The activity concentration of ^{232}Th in the upper 10 cm layer of soil was 0.08–0.12 Bq/g. This resulted in absorbed gamma dose rates of 0.1–0.15 μGy/h, with an average value of 0.121 μGy/h — some 50% higher than the normal natural background value (0.085 μGy/h) and 30% higher than the average indoor value (0.0922 μGy/h). The additional dose received by the public as a result of these elevated levels was reported to be 0.044 mSv/a. Thoron progeny concentrations in the city were also elevated, with a reported PAEC of 199.4 MeV/L (0.032 μJ/m^3) [76].

Doses were estimated for individuals living in houses constructed from bricks containing slag residues. The incremental annual effective dose from gamma radiation attributable to the use of such bricks was found to be about 0.2 mSv. Indoor thoron progeny concentrations were reported to be three times normal levels, with an incremental annual effective dose of 0.02 mSv.

4.5. REGULATORY CONSIDERATIONS

The ore and several of the mineral concentrates obtained from it have ^{232}Th activity concentrations greater than 1 Bq/g. Consequently, the mining and beneficiation operations at Bayan Obo would, following Section 3.1.1, need to be considered for regulatory control as a practice. Annual effective doses received by workers from gamma radiation and dust inhalation are each generally less than 1 mSv. However, the dose from inhalation of thoron and radon progeny combined could be as high as about 2 mSv. Annual effective doses received by the public are expected to be a very small fraction of 1 mSv.

In view of the possibility of some workers receiving doses moderately above 1 mSv per year, the appropriate regulatory option might be to require authorization in the form of registration in order to ensure the ongoing monitoring of exposure and that basic measures are taken to keep doses as low as reasonably achievable.

Since it is likely that the thorium and uranium decay chains will be in equilibrium in the ore and rare earth concentrates, Eq. (4) in Section 3.1.3.3 can be used to determine whether the Transport Regulations apply to the transport of these materials. The determining factor in this regard is ^{232}Th activity concentration, since ^{238}U concentration is very low. The ^{232}Th activity concentrations in the ore (about 2 Bq/g) and the bastnäsite concentrate (8–10 Bq/g) are such that the Transport Regulations would not apply. However, the activity concentration of the monazite concentrate (6–14 Bq/g) could bring this material within the scope of application of the Transport Regulations.

5. MINING AND BENEFICIATION OF RARE EARTH ORE AT MOUNT WELD, AUSTRALIA

The Mount Weld rare earth deposit in Australia lies over the centre of the Mount Weld carbonatite, a circular intrusive igneous complex approximately 3 km in diameter located south-east of Laverton in Western Australia. Deposits and zones of rare earth, phosphate and niobium–tantalum mineralization are hosted by layers of weathered carbonatite, laterite and lake sediment over the carbonatite. The principal minerals are apatite, pyrochlore, magnetite, ilmenite, monazite and quartz. The deposits are overlain by 30–50 m of barren lake clay and transported alluvium. The mining and mineral processing operations associated with the Mount Weld deposit are still being developed. The data reported in this section are therefore preliminary, most having been obtained from Ref. [79].

The rare earth, thorium and uranium contents of the ore are given in Table 17. The rare earth content comprises mostly the light rare earths (see Table 93 in Appendix IV).

TABLE 17. RARE EARTH, THORIUM AND URANIUM CONTENT OF MOUNT WELD ORE

	Concentration (%)	
	Average	Maximum
Total REO	17–18	42
ThO_2	0.075	0.18
U_3O_8	0.003	0.006

5.1. PROCESS DESCRIPTION

5.1.1. Main process

During the initial mining campaign in 2007–2008, 409 000 t of ore at approximately 18% REO were mined from an open cut by blasting and power shovels and stockpiled within the mining lease area. The maximum production in future is expected to be about 270 000 t/a. After crushing, the stockpiled ore will be fed by a front end loader into a flotation concentration plant, which is yet to be constructed. This plant will produce approximately 65 000 t/a of rare earth concentrate with an REO concentration of 40% and expected thorium and uranium concentrations of 0.17% ThO_2 and 0.003% U_3O_8. The concentrate will be transported by road to a container handling port for export to Malaysia, where a hydrometallurgical processing plant dedicated to the processing of Mount Weld rare earth concentrate will be constructed in the near future. For transport purposes, the concentrate will be packaged in sealed bags before being loaded into containers.

5.1.2. Management of residues

When the concentration plant is in operation, it is expected to generate about 177 000 t/a of tailings with an REO concentration of about 7–8% and expected thorium and uranium concentrations of 0.05% ThO_2 and 0.003% U_3O_8. These tailings will be deposited in a tailings dam adjacent to the plant as a resource for anticipated further processing in future.

5.2. RADIONUCLIDE ACTIVITY CONCENTRATIONS

Radionuclide activity concentrations in the various process materials can be calculated from ThO_2 and U_3O_8 concentrations provided in Sections 5.1.1 and 5.1.2, assuming that 1 g of ThO_2 contains 3566 Bq of ^{232}Th, that 1 g of U_3O_8 contains 10 470 Bq of ^{238}U and that equilibrium conditions for the ^{232}Th and ^{238}U decay chains in the ore are maintained in the concentrate and tailings. The results are shown in Table 18.

5.3. OCCUPATIONAL EXPOSURE

5.3.1. Exposure pathways

Owing to relatively low radioactivity levels in the process materials, exposure of workers during operation and maintenance of the facility can also be expected to be relatively low. The exposure pathways of possible concern are external exposure to gamma radiation and internal exposure from the inhalation of radionuclides in airborne dust and from the inhalation of thoron. Internal exposure from the inhalation of radon is considered to be insignificant, owing to the very low concentrations of uranium in the process materials.

5.3.2. Gamma dose rates

The results of regular gamma radiation surveys conducted at the mine site are seen in Table 19. Measurements were generally made at a distance of 1 m. On

TABLE 18. ACTIVITY CONCENTRATIONS IN PROCESS MATERIALS ASSOCIATED WITH THE MINING AND BENEFICIATION OF MOUNT WELD ORE

	Radionuclide activity concentration (Bq/g)			
	^{232}Th decay series		^{238}U decay series	
	Average	Maximum	Average	Maximum
Ore	2.7	6.4	0.3	0.6
Concentrate	6.1	—[a]	0.3	—
Tailings	1.8	—	0.3	—

[a] —: Similar to average values.

TABLE 19. GAMMA DOSE RATES ASSOCIATED WITH MINING OPERATIONS AT MOUNT WELD

Survey location	Number of surveys	Dose rate (μSv/h)	
		Average	Range
Inside mining pit	10	1.33	0.10–5.25
Mining pit boundary	13	0.15	0.06–0.24
Ore stockpiles	9	1.82	0.88–2.57
Waste rock storage	8	0.40	0.07–1.82

the basis of expected activity concentrations in the rare earth concentrate, gamma dose rates predicted for the plant are 3.5–4.0 μSv/h at a distance of 1 m from the concentrate.

5.3.3. Radionuclides in airborne dust

Regular dust monitoring at the mine site using personal air samplers started in October 2007 and results are available so far for 179 samples. The gross alpha activity concentration is 3–30 mBq/m^3, with an average of 7 mBq/m^3.

5.3.4. Thoron progeny

Measurement of thoron progeny concentrations inside the mining pit and in the area of the stockpiled ore started in October 2007. Ten minute air samples are taken and alpha activity concentrations on the filter paper are measured within 6–15 h after sampling, in accordance with the method described in Ref. [80]. Results are available so far for 37 samples taken from inside the mining pit and 21 from the ore stockpile area. Thoron progeny concentrations have remained within the range of 0.01–0.06 μJ/m^3 except for one instance when concentrations of 0.4–1.9 μJ/m^3 were detected during calm early morning conditions. Taking all results into account, the average thoron progeny concentration in the operational areas of the site is 0.24 μJ/m^3.

5.3.5. Effective dose

Individual monitoring of workers for gamma radiation using thermoluminescent dosimeters (TLDs) started in January 2008. The doses determined over a one year monitoring period for workers routinely involved in the mining and transport of ore were 0.02–0.31 mSv (typically about 0.13 mSv). Workers occasionally visiting the pit and ore stockpiles received no measureable dose, while a surveyor spending a considerable proportion of the working period in these areas received a dose of 0.32 mSv in a year. The annual dose from exposure to gamma radiation is, on average, 0.02–0.32 mSv and is unlikely to exceed 1 mSv. These doses are, as expected, lower than those predicted from the gamma dose rate measurements described in Section 5.3.2 (which, depending on the locations and occupancy times assumed, could amount to considerably more than 1 mSv).

Once the concentration plant is constructed, the range of doses received from external gamma radiation is expected to rise from 0.02–0.32 mSv to 0.20–0.50 mSv, owing to the higher dose rates expected in the vicinity of the concentrate. However, no significant increase is expected in doses received from inhalation of airborne dust and thoron, except during the crushing operation. The rare earth concentrate is expected to contain sufficient moisture to prevent the generation of dust and to significantly limit the emanation of thoron.

Measurements of airborne dust particle sizes inside the mining pit, based on four samples so far, indicate an average AMAD of 12.8 μm with a range of 9.3–18.7 μm. Assuming a ^{232}Th:^{238}U activity concentration ratio of 10 (see Table 18) and decay chain equilibrium in the airborne dust, the dose per unit measured gross alpha activity for low solubility material with an AMAD of 10 μm is 6 μSv/Bq (see Fig. 4). Annual exposure time in the pit and near ore stockpiles is estimated to be 900 h. For an average dust activity concentration of 7 mBq/m^3 (see Section 5.3.3) and a breathing rate of 1.2 m^3/h, the committed effective dose due to dust inhalation in a year is calculated to be:

$$6\ \mu\text{Sv/Bq} \times 1.2\ \text{m}^3\text{/h} \times 7\ \text{mBq/m}^3 \times 900\ \text{h} = 0.05\ \text{mSv}$$

Since the value assigned to the AMAD of the dust particles (10 μm) is based on only four measurements, it has been suggested that a more conservative value of 5 μm might be more appropriate, in which case the dose per unit measured gross alpha activity would be 8 μSv/Bq (see Fig. 4). This would lead to an estimated annual dose of 0.06 mSv. However, the assumed dust exposure time of 900 h near ore stockpiles is expected to be a considerable overestimate. In practice, the committed effective dose in a year is expected to be about 0.02–0.03 mSv.

Taking the average thoron progeny concentration of 0.24 μJ/m^3 for the operational areas of the mine and the thoron progeny dose coefficient of 0.39 mSv/mJ given in Table 11, and assuming a breathing rate of 1.2 m^3/h and an annual exposure time of 900 h, the annual dose from inhalation of thoron is calculated to be:

$$0.39 \text{ mSv/mJ} \times 1.2 \text{ m}^3\text{/h} \times 0.24 \ \mu\text{J/m}^3 \times 900 \text{ h} = 0.10 \text{ mSv}$$

Taking into account all the above contributions to effective dose, the total effective dose in a year is currently estimated to be generally in the range of 0.15–0.48 mSv, with a maximum of 1.6 mSv. After the commissioning of the concentration plant, this is expected to rise to 0.3–0.7 mSv, with a maximum of less than 3 mSv. Details are provided in Table 20.

5.4. PUBLIC EXPOSURE

With suitable tailings management, it is unlikely that public exposure will be of any concern, owing to low activity concentrations of the process materials and the fact that no chemical processing is carried out at the site. The results of 11 external gamma radiation surveys carried out surroundings at the site show dose rates at background levels (0.05–0.19 μSv/h with an average of 0.12 μSv/h).

5.5. REGULATORY CONSIDERATIONS

The ^{232}Th activity concentrations in the ore, concentrate and tailings are above 1 Bq/g. Consequently, the mining and beneficiation operations at Mount Weld would, under the terms of Section 3.1.1, need to be considered for

TABLE 20. DOSES RECEIVED BY MINE WORKERS AT MOUNT WELD

	Annual effective dose (mSv)			
	Current mining operations		After commissioning of the concentration plant	
	Normal range	Maximum	Normal range	Maximum
External gamma radiation	0.02–0.32	0.9	0.2–0.5	~2
Inhalation of dust	0.02–0.06	0.2	0.02–0.10	0.4
Inhalation of thoron	0.10	0.5	0.10–0.20	0.5
Total (rounded)	0.05–0.40	1.6	0.3–0.8	<3

regulatory control as a practice. Annual effective doses received by workers from gamma radiation, dust inhalation and thoron progeny inhalation are each likely to be considerably less than 1 mSv, although the total dose could approach 1 mSv. Some workers could potentially receive doses of up to 2 or 3 mSv in a year. Annual effective doses received by members of the public are expected to be a very small fraction of 1 mSv.

In view of the possibility of some workers receiving doses moderately above 1 mSv per year, the appropriate regulatory option might be to require authorization in the form of registration, in order to ensure the ongoing monitoring of exposures and basic measures to keep doses as low as reasonably achievable.

Since it is likely that thorium and uranium decay chains will be in equilibrium in the rare earth concentrate, Eq. (4) in Section 3.1.3.3 can be used to determine whether the Transport Regulations apply to the transport of this material. Ignoring the minor component associated with ^{40}K, it can be concluded that, because the sum of the activity concentrations of ^{232}Th (6.1 Bq/g) and ^{238}U (0.3 Bq/g) does not exceed 10, the concentrate will not be subject to the Transport Regulations.

6. MINING AND BENEFICIATION OF HEAVY-MINERAL SAND

Heavy-mineral sand deposits occur underwater or may form part of sea beaches or coastal dunes created by wind action over long periods of time. They may also occur inland in coastal strips up to a few tens of kilometres wide. The highest heavy-mineral concentrations are found in beach deposits (see Fig. 7). These deposits are richest during periods of intense wave action, following which, if left unrecovered, they are likely to become wholly or partly washed away.

Deposits of heavy-mineral sand may occur in populated areas, leading to high levels of natural background exposure due to the relatively high thorium content of the monazite component. Investigations in parts of India [81, 82] have revealed absorbed dose rates of 0.2–3 μGy/h (up to 20 μGy/h in some instances [83]), ^{232}Th activity concentrations in soil of 0.1–1.5 Bq/g and airborne dust activity concentrations of 0.03–0.4 mBq/m^3.

The composition of heavy-mineral sand deposits varies according to location. Data on the composition of various deposits of heavy-mineral sand are

FIG. 7. Beach deposits of heavy-mineral sand.

provided in Appendix VI. The total heavy-mineral content of raw sand is typically in the range 5–50%, while the monazite component (the principal source of rare earths) usually forms less than 1% of the raw sand. In some beach sand deposits such as those on the south and west coasts of India, the heavy-mineral concentrations and monazite concentrations have in the past reached 70% and 4%, respectively [84].

Ilmenite is usually the main product of heavy-mineral sand operations, typically accounting for 45–75% of total mineral production. The other heavy minerals produced commercially are mostly zircon and rutile, each of which typically accounts for 2–20% of total heavy-mineral production. Monazite accounts for only 1–2% of production. Other heavy minerals that may be present include garnet, sillimanite, leucoxene and xenotime. All of these minerals have densities greater than 3000 kg/m^3, hence their designation as heavy minerals. The chemical formulas and densities of these minerals are provided in Table 21.

The rare earth content of monazite is typically 40–70%; detailed data are provided in Appendix VII. The rare earth content of xenotime is about 67% [13].

TABLE 21. HEAVY MINERALS FOUND IN HEAVY-MINERAL SAND

	Chemical formula	Typical density (kg/m^3)
Ilmenite	$FeO \cdot TiO_2$	4540
Zircon	$ZrO_2 \cdot SiO_2$	4680
Rutile	TiO_2	4260
Monazite	$(Ce,La,Nd,Y,Th)PO_4$	5240
Garnet	$(Mg,Mn,Fe,Ca)_3(Al,Cr)_2(SiO_4)_3$	4110
Sillimanite	$Al_2O_3 \cdot SiO_4$	3250
Leucoxene	$Fe_2O_3 \cdot TiO_2$	3500
Xenotime	YPO_4	4660

6.1. PROCESS DESCRIPTION

6.1.1. Main process

The mining process is described in Section 6.1.1.1. The general processes for recovering individual heavy minerals of commercial value are described in Sections 6.1.1.2 and 6.1.1.3. The details of these separation processes may vary, depending on the mineral composition of the raw sand and the need for recovering specific minerals of value.

6.1.1.1. Mining

The collection of beach deposits may involve relatively simple, small scale operations. The deposits are collected manually and heaped for transportation to the beneficiation plant. In some operations, the deposits are scraped and collected by conventional earthmoving equipment such as front end loaders, bulldozers and scrapers and are transported to a central collection point using rail mounted trolleys although some degree of manual handling is also involved. Depending on the requirements, the sand may be dried by spreading in the sun before being transported to the beneficiation plant.

The mining of heavy-mineral sand deposits in dunes is carried out either through manual surface collection or by mechanized dry mining methods using conventional earthmoving equipment. The collected sand is beneficiated at the mine site or transported to the beneficiation plant by trucks. A typical dry mining operation is shown in Fig. 8.

The mining of large, relatively deep coastal deposits (up to 6 m deep) is carried out using power shovels and floating dredges with dragline excavators or bucket suction excavators (‘suction cutters’). A typical dredge mining operation is shown in Fig. 9. A degree of initial concentration is usually performed at the dredge site itself. The material is then transported as slurry through a pipeline to the beneficiation plant.

6.1.1.2. Primary separation

The various heavy minerals are separated from the remaining, low value material (‘gangue’ minerals) using wet separation techniques that rely on differences in density. This process, referred to as the primary separation process, removes up to 90% of the gangue minerals. Typically the process, shown schematically in Fig. 10, is carried out at the mine site and involves the following steps:

FIG. 8. Dry mining operation, showing pre-concentration operations in the foreground.

FIG. 9. Dredge mining operation.

(a) Large rocks and pebbles, shells, roots and other oversize material are removed by screening;

(b) Fine particles known as slimes, mainly clay particles less than 75 µm in size, are removed by hydrocyclones;

(c) The majority of wet separation work involves the use of groups of spiral separators. Sand bearing water at a carefully controlled density is fed into the spiral separators, illustrated in Fig. 11, from a ‘constant density tank’. As the slurry flows down the spirals, the smaller and less dense minerals (essentially silica sand) are thrown to the outside while the larger and darker coloured heavy minerals are guided into offtake holes on the inside of the spirals by splitters (the yellow tabs shown in Fig. 12) and collected as a pre-concentrate with a heavy-mineral content of 80–85%. Various types of spiral groups are used, with considerable recycling of the various streams between the different spiral groups to maximize both the grade and recovery of the mineral products;

(d) The pre-concentrated slurry may then be upgraded to about 90–98% heavy-mineral content using a series of further spirals and wet separation tables.

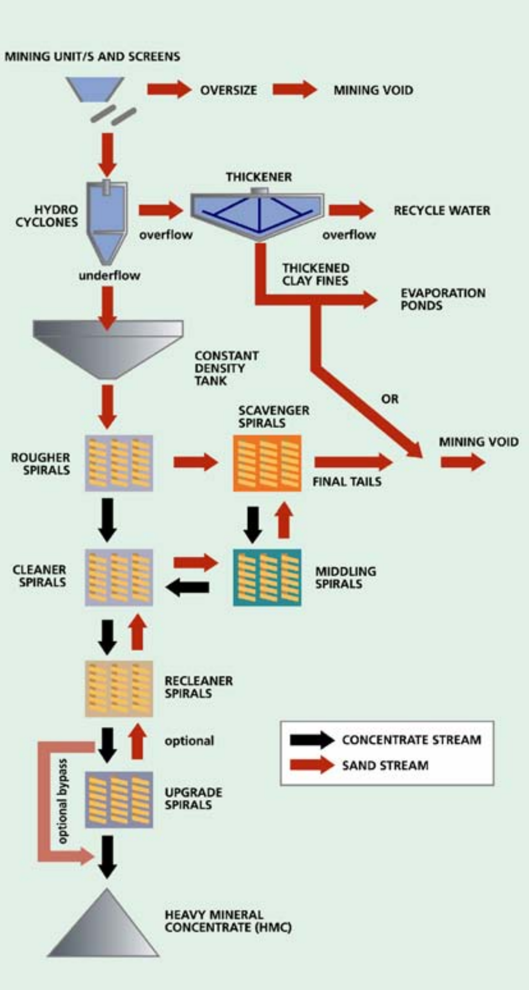

FIG. 10. Schematic representation of the primary separation process (courtesy: Iluka Resources Limited, Australia).

Where heavy-mineral sand is obtained from more than one source (for example, beach deposits and dredging operations), controlled blending of the feedstocks is usually carried out at the pre-concentration stage or the upgrading stage to ensure a consistent composition of the heavy-mineral concentrate. A stockpile of wet heavy-mineral concentrate is shown in Fig. 13.

FIG. 11. Groups of spirals used in the primary separation process.

FIG. 12. Detailed view of a spiral used in the primary separation process.

FIG. 13. A stockpile of heavy-mineral concentrate from the primary separation process.

6.1.1.3. Secondary separation

In the next stage of the processing operation, referred to as the secondary separation process, the separation techniques applied to the heavy-mineral concentrate involve various magnetic and electrostatic separators. The magnetic separation process is usually conducted as a dry technique (as is the subsequent electrostatic separation process), requiring the wet concentrate to be dried before it can be separated into magnetic and non-magnetic fractions.[9] In preparation for the dry magnetic separation process, the heavy-mineral concentrate is usually pumped as slurry to a storage facility and then dried at a temperature of typically 150˚C using oil fired fluidized bed driers. Alternatively, the heavy-mineral concentrate may be sun-dried before being mechanically conveyed to the storage facility.

A typical dry separation process is schematically shown in Fig. 14. The higher iron content of ilmenite gives it a higher magnetic susceptibility than rutile, zircon and monazite, allowing it to be magnetically separated from these minerals early in the process. The rutile and zircon remain together after this initial magnetic separation stage and are subsequently separated from each other electrostatically by taking advantage of the higher electrical conductivity of rutile relative to zircon. Residual magnetic minerals, such as the small amounts of ilmenite that might have been recovered with the rutile, are removed using stronger magnets than used earlier. The small quantities of quartz, kyanite and staurolite contained in the heavy-mineral concentrate have similar magnetic and conductive properties to zircon and follow it when it is separated from ilmenite and rutile. Most of these particles are removed by gravity separation through more spirals. Any rutile that might have slipped through to this point is removed by further electrostatic separation and returned to the rutile circuit. In some operations, fine separation of some minerals is effected by wet tabling and froth flotation. Magnetic separation is used to remove the magnetic monazite from the non-magnetic zircon. Air gravity concentrating table ('air table') separation techniques are used for final cleaning of the zircon. In some cases, traces of iron associated with the zircon particles can be removed by leaching to produce a higher quality product. Where the intention is to chemically process monazite to extract rare earths and/or thorium, the purity of the monazite concentrate may be upgraded to 97–99% using magnetic separation. Where there is no immediate

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[9] New generation wet high intensity magnetic separators are now available that allow magnetic separation of wet concentrate.

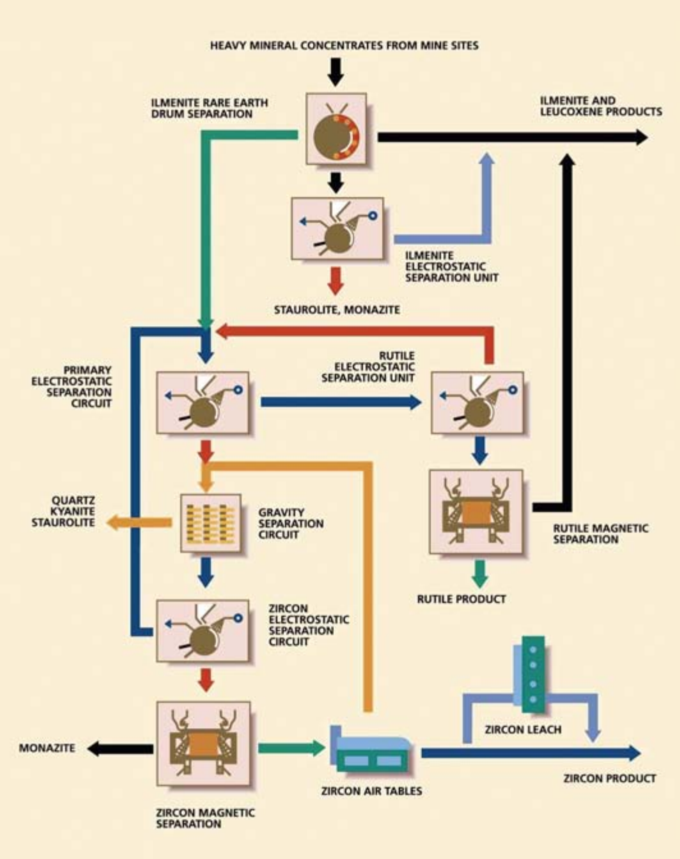

FIG. 14. Schematic representation of the secondary separation process (courtesy: Iluka Resources Limited, Australia).

intention to process the monazite, the purity of the monazite stream may be as low as 30%.

6.1.1.4. Scale of operations

Mining operations and separation plants vary greatly in size and degree of sophistication. Operations in some parts of the world are very small, with a processing capacity of 50–100 t/d. Many of these plants are of single level design, utilizing fewer than 20 units of separation equipment and employing fewer than 20 persons. Manual handling and mineral transport are generally done using shovels, buckets and wheelbarrows. Most worldwide production comes from larger operations. Dry separation plants in some countries have processing capacities of 250–500 t/d and contain several hundred units of separation equipment, while in India and Australia, for instance, the capacities are typically 300–3500 t/d. Such plants are generally of a multilevel design, with the sand being moved around the plant on open conveyors and in bucket elevators and transferred between levels or to equipment by a system of enclosed pipes and chutes.

In India and Australia, a typical heavy-mineral sands operation may directly employ 400–500 workers, starting from the mechanized mining operation and ending with product dispatch. Operations involving the manual mining and transportation of beach sand are more labour intensive. While most of the operations in the dry separation plants are mechanized, considerable numbers of workers are employed for manual operations such as conveying, tabling, product bagging, plant maintenance and recovery of spillage.

6.1.2. Management of residues

6.1.2.1. Residues from the mining and primary separation processes

Most of the solid residues arising from the mining and primary separation processes are in the form of sand tailings. Other constituents include topsoil, peat, clay, clay fines (slimes), shells, organic waste in the form of vegetation, trees and roots, and other screened out material. The total amount of residue generated depends on the concentrations of heavy minerals in the raw sand and the recovery effected in the processes. A typical facility in Australia producing 500 000 t/a of heavy-mineral concentrate generates 6 million t/a of sand tailings and 700 000 t/a of oversize solids [85], corresponding to approximately 930 kg of residues per tonne of raw sand mined, of which 800 kg are in the form of sand tailings. In operations in India, typically 600–700 kg of sand tailings are generated per tonne of raw sand mined [86].

Sand tailings, which are essentially free of heavy minerals (and radioactivity), arise from dredge mining and wet concentration plants, concentrate upgrading plants and beach washing plants. They are generally pumped directly as slurry to the mined out areas as backfill. In dredge mining operations, sand tailings from any initial concentration steps performed at the dredge site are returned directly to the dredged area prior to the dredging of the next site [86, 87].

Topsoil, peat, clay, vegetation and other screened out material are kept at the mine site for topping the refilled areas to facilitate remediation [86]. Oversize solids from dry mining operations may be buried below the sand tailings or overburden, usually at a depth of at least 3 m. Occasionally, if of a suitable quality, they are used for constructing roads on-site. In dredge mining operations, oversize solids from the screening process are discharged directly into the dredging pond [87].

Clay fines (slimes) arising from primary separation processes are normally returned to the mining void, in some cases after drying in evaporation ponds, in others by mixing directly with coarser sand tailings. Some may be incorporated into subsoil as part of the remediation process. In dredge mining operations, a portion of the clay fines is entrained in the tailings, trapped under tailings or pumped to a thickener (to recover some of the associated water) and combined with sand tailings. The remainder is discharged into the dredging pond [86, 87].

Residual water from wet concentration processes contains clay fines and other suspended particles which may be removed by settling. This water is reused within the process; either evaporated in ponds, returned to the mining void or discharged as effluent.

6.1.2.2. Residues from the secondary separation process

Residues from the secondary separation process consist mainly of tailings from the spiral separators (in the same physical form as the original ore), oversize solids, dust extracted from the dry separation plant dust extraction systems, particulate matter collected from stack discharges and clay fines removed from the separation plant water system. Monazite concentrate also constitutes a processing residue if there is no immediate intention of using it as a source of rare earths or thorium. A typical secondary separation plant in Australia processing 500 000 t/a of heavy-mineral concentrate produces 70 000 t/a of tailings, 6000 t/a of oversize solids, 2000 t/a of mill dust and stack particulates and 1000 t/a of clay slimes [85]. The total amount of waste from the secondary separation process is only about 10 kg per tonne of raw sand mined (1%), or about 160 kg per tonne of heavy-mineral concentrate processed, of which 140 kg is in the form of tailings. The amount of tailings generated in Indian dry separation plants is reported to be 70–100 kg per tonne of concentrate processed [86]. The management of residue from the secondary separation process is described in Refs [86, 87] and is summarized below.

The dry separation tailings contain silica sand and any unrecovered heavy minerals. In terms of guidelines for operations in India [86], the tailings are deposited directly into the dredge mining area for recycling, along with fresh feed to the dry mill or, where only surface mining or beach washings collection is practiced, placed in earthen trenches or other suitable storage. The earthen trenches are provided with soil topping and the areas are designated as controlled areas with barriers to prevent public access. In terms of guidelines for operations in Western Australia [87], dry separation tailings are pumped as slurry from the spiral plant to hydrocyclones and onto a stockpile. This stockpile may also serve as a receiving point for other residues, as follows:

(i) Oversize material from the screening of minerals, which is transported directly to the stockpile;
(ii) Dust removed from the dry separation plant dust extraction system, which is transported pneumatically to a holding bin, from where it is transported to the stockpile either by truck in a damp form or as slurry in a pipeline;
(iii) Clay fines (slimes), which are pumped to shallow evaporation dams and, when partially dry, are dug out of the dams and deposited on the stockpile.

The stockpiled materials are then transported to designated mine site disposal areas, usually the mining void. A typical backfilled mined out area is shown in Fig. 15. Tailings, in a wet or dry state, are transported by mechanized means such as pumping, conveyors or trucks, as appropriate. If the tailings contain significant quantities of radioactivity due to residual monazite content, special measures may be needed for transport and disposal. The disposal areas are designed so that they can be covered by a suitable layer of non-radioactive sand tailings and/or overburden.

Monazite circuit tailings are usually collected separately and, where there is no intention of processing them further, are blended with mine sand tailings and disposed of in the mine pit. Dust from the monazite plant is removed using a dedicated dust extraction system and stored in suitable receptacles, for instance, 2 t bulk bags. Subsequently, it is transported to designated dry separation plant tailings disposal areas at mine sites.

In India, monazite concentrate used to be stored in 50 kg bags placed in sheds. These storage facilities were treated as controlled areas with restrictions on access and occupancy. Nowadays, if monazite concentrate is to be used for subsequent extraction of rare earths or thorium, it is stored in earthen trenches at

FIG. 15. Backfilled mined out area.

mine sites in discrete, well-documented locations away from regularly occupied areas. The crude monazite concentrate is pumped as slurry directly into a trench and periodically topped with about 1 m of mineral free sand to reduce radiation fields to levels similar to those naturally encountered in the surrounding area. The trenches are well demarcated with a boundary fence or wall and identified with prominent warning signs. If the monazite concentrate is to be treated as waste, it is blended with mine sand tailings before disposal in a mine pit.

The required cover thickness for disposal areas may vary from 0.5–1 m to more than 3 m, depending on radiological considerations. Cover thicknesses of up to 5 m were proposed in Western Australia in the late 1990s [88]. Current waste management practices for mining and mineral processing in Australia are described in Ref. [89].

Spillage of minerals in dry separation plants is collected mechanically (for instance, by a sweeper, mechanical loader or vacuum cleaner). Dry spillage is recycled back into the process through a dry feed bin. Wet spillage can be fed back into a plant through wet heavy-mineral concentrate or used to retreat feed systems. Alternatively, spillage can be taken back to the mine site as part of dry separation plant tailings or as a concentrator feed that is then spread on the mine face and remined.

In dry separation operations in India, liquid effluents from a plant are reported to arise from mild acidic treatment of sillimanite and zircon in the flotation process. Liquid effluents are treated in a neutralizing pond before being discharged to water bodies [90].

Atmospheric emissions are mitigated by equipping stacks with baghouses or cyclones to trap particulates. Intercepted material is disposed of with the dust extracted from the dry separation plant dust extraction system. Alternatively, stack outlets may be connected directly to the dry separation plant dust extraction system.

6.1.2.3. Remediation of mined and refilled areas

As part of mining site remediation, residues such as topsoil, peat, clay, clay fines, vegetation and other screened out material are incorporated into the topping of refilled areas. These areas are stabilized by planting suitable trees and other vegetation in a continuous reforestation programme to restore ecological balance to the extent possible [82, 91].

6.2. RADIONUCLIDE ACTIVITY CONCENTRATIONS

6.2.1. Heavy-mineral sand

The main source of radioactivity in heavy-mineral sand is the thorium bearing mineral monazite, which is typically present at a concentration of 0.1–1% (see Table 96 in Appendix VI). Activity concentration data for heavy-mineral sand are provided in Table 98 in Appendix VIII.

The ThO_2 content of monazite is typically around 5–6%. Assuming that 1 g of ThO_2 contains 3566 Bq of ^{232}Th, the activity concentration of ^{232}Th in heavy-mineral sand can be expected to be a few tenths of a becquerel per gram, which is consistent with values reported in literature. In some heavy-mineral sand deposits, thorium content is much higher, noting particularly the Manavalakurichi deposit in Tamil Nadu, India. The monazite content of this deposit is 3–4% and the monazite itself contains 9–10% ThO_2. This leads to a ^{232}Th content of the order of 10 Bq/g, some 20 times higher than typical levels.

The uranium content of heavy-mineral sands is typically an order of magnitude lower than the thorium content, leading to ^{238}U activity concentrations that are in most cases a few times lower than ^{232}Th activity concentrations.

6.2.2. Heavy-mineral concentrate

The primary separation process removes most of the (non-radioactive) gangue material, with the result that radioactivity in the heavy-mineral concentrate is higher than that of the raw sand. Activity concentration is generally increased by a factor of 2–15, depending on, among other things, the amount of gangue material contained in the raw sand. This leads to ^{232}Th and ^{238}U activity concentrations in the heavy-mineral concentrate typically ranging from fractions of a becquerel per gram to a few becquerels per gram. Activity concentration data are provided in Table 99 of Appendix VIII.

6.2.3. Monazite

This mineral is the main source of rare earths and exhibits the highest activity concentration of all the process materials associated with the mining and beneficiation of heavy-mineral sand. Activity concentration data are provided in Table 100 of Appendix VIII.

At a typical ThO_2 content of 5–6%, a ^{232}Th activity concentration of around 200 Bq/g can be expected. The majority of activity concentration data reported in literature fall within the range of 140–250 Bq/g. Higher activity concentrations (230–400 Bq/g, corresponding to ThO_2 concentrations of 6–11%) are found in monazite from India, while lower activity concentrations (7–32 Bq/g, corresponding to ThO_2 concentrations of 0.2–0.9%) are found in 'black monazite' from Taiwan, China. Measurements made on Australian monazite imply that the Th decay chain is essentially in equilibrium [92].

Activity concentrations of ^{238}U generally fall within the range of 6–40 Bq/g (corresponding to U_3O_8 concentrations of 0.06–0.4%), with values for India at the upper end of this range. Lower concentrations (5–6 Bq/g, corresponding to U_3O_8 concentrations of 0.05–0.06%) are again found in black monazite from Taiwan, China.

6.2.4. Xenotime

This mineral is also a source of rare earths. Like monazite, it exhibits significantly elevated radioactivity levels (but, in the case of thorium, not to the same extent). Xenotime produced in Australia has a ^{232}Th activity concentration of about 60 Bq/g and a ^{238}U activity concentration of about 50 Bq/g [87].[10]

6.2.5. Other heavy minerals

The radioactivity levels of all the other heavy minerals are low. Activity concentration data are provided in Tables 101–104 of Appendix VIII. Typical ranges of activity concentrations are given in Table 22. Higher activity concentrations are reported in literature, but these are generally associated with incomplete separation of monazite and not representative of material used in commercial applications.

6.2.6. Processing residues

6.2.6.1. Tailings

Activity concentration data for tailings from the primary and secondary separation processes are provided in Table 105 of Appendix VIII. Radioactivity levels in primary separation tailings are close to natural background levels (typically 0.05–0.1 Bq/g for ^{232}Th and 0.01–0.02 Bq/g for ^{238}U). Higher levels are reported for secondary separation tailings (typically 1–20 Bq/g for ^{232}Th and 0.1–10 Bq/g for ^{238}U).

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[10] Australia is the main source of xenotime from heavy-mineral sand separation. Xenotime is also recovered during the processing of tin mining residues in south-east Asia (see Section 7).

TABLE 22. RADIONUCLIDE ACTIVITY CONCENTRATIONS IN HEAVY MINERALS THAT ARE NOT SOURCES OF RARE EARTHS

	Activity concentration (Bq/g)	
	Th-232	U-238
Ilmenite	0.04–2	0.03–0.4
Zircon	0.5–1	1–4
Rutile	0.06–0.4	<0.1–0.7
Garnet	0.2–0.4	—
Sillimanite	0.06–0.15	—
Leucoxene	0.3–5	0.2–1

6.2.6.2. *Oversize solids*

Mass concentrations of thorium and uranium in oversize solids are reported for heavy-mineral sand operations in Australia [87]. Corresponding activity concentrations for the primary separation process are <0.20 Bq/g for ^{232}Th and <0.12 Bq/g for ^{238}U. Corresponding activity concentrations for the secondary separation process are significantly higher, at 0.3–2.6 Bq/g for ^{232}Th and 0.6–1.9 Bq/g for ^{238}U.

6.2.6.3. *Clay fines (slimes)*

Mass concentrations of thorium and uranium in clay fines are reported for heavy-mineral sand operations in Australia [87]. For ^{232}Th and for ^{238}U, the corresponding activity concentration for the primary separation process is <0.12 Bq/g. The corresponding activity concentrations for the secondary separation process are significantly higher — in the order of 2 Bq/g for ^{232}Th and 0.4 Bq/g for ^{238}U.

6.2.6.4. *Airborne dust*

Activity concentrations in airborne dust generated by the dry separation process are provided in Table 106 of Appendix VIII. The activity concentrations in dust within dry separation plants are typically 1–20 Bq/g for ^{232}Th and 0.1–10 Bq/g for ^{238}U. However, in areas where monazite is separated, higher activity concentrations can be expected as a result of the greater monazite content of the airborne dust — data provided in Refs [93, 94] suggest values of about 10–200 Bq/g for ^{232}Th and 5–20 Bq/g for ^{238}U.

The average activity concentrations in airborne dust particles measured in one Western Australian dry separation plant were compared with concentrations measured in the heavy-mineral concentrate feedstock [95]. The ^{232}Th activity concentration in the dust (2.8 Bq/g) was found to be about twice that in the feedstock, while the ^{238}U activity concentration in the dust (1.2 Bq/g) was about half of the feedstock value. It was noted that the isotopic composition of the airborne dust appeared to correspond to a dilute form of monazite. The process involved in producing this airborne concentration of thorium rich compounds was not explained, but it has been noted elsewhere that monazite has a propensity to concentrate preferentially in airborne dust in separation plants because it is softer and finer than other heavy minerals [96, 97].

Typical activity concentrations in stack particulates generated by secondary separation processes in Australia are of the order of 2–20 Bq/g for ^{232}Th and 0.1–6 Bq/g for ^{238}U [87]. These are similar to concentrations measured in airborne dust within the plants.

Overall, the activity concentration of ^{232}Th is 2–10 times that of ^{238}U, depending mainly on the monazite content of the dust. When calculating overall dose coefficients for inhalation from gross alpha activity data (see Section 3.3.2.2), it would seem reasonable to assume a ^{232}Th: ^{238}U ratio of 2 for non-monazite dust, 10 for monazite dust and 4 for a mixture of the two.

6.3. OCCUPATIONAL EXPOSURE

6.3.1. Exposure pathways

6.3.1.1. Mining and primary separation

Mining operations pose few radiological concerns for workers because radionuclide concentrations in raw sand are usually very low (see Table 98 in Appendix VIII). External gamma dose rates depend on the concentration of monazite in the sand being mined and the degree to which the mining is mechanized. Internal exposure from airborne dust is rarely a cause for concern except in the case of raw sand containing a large proportion of fine particles or that with a high monazite content. Emanation of thoron gas from the mineral is extremely low and the very short half-life and open nature of such mining preclude the buildup of thoron and its decay progeny.

The radiological impact during primary separation is small because the bulk material has a low activity concentration and the moisture content of the heavy-mineral concentrate prevents dust generation.

6.3.1.2. Secondary separation: External exposure to gamma radiation

External exposure is an important exposure pathway in secondary separation plants handling monazite in significant concentrations, typically where monazite represents more than 0.5% of the total mineral content. Following the ilmenite removal stage (often the first stage of the process) the remaining non-magnetic mineral fractions become progressively enriched with monazite. Elevated gamma dose rates in the zircon processing stages arise from spillages and accumulation of monazite enriched sand fractions.

6.3.1.3. Secondary separation: Internal exposure to radionuclides in airborne dust

Inhalation of dust is the major exposure pathway in dry separation plants because the heavy-mineral concentration increases and the separation processes can generate significant concentrations of airborne dust due to the requirement for a free running material. Even so, the silica content of dust is likely to be of greater concern for health than radioactivity content. Each of the physical separation steps (involving electrostatic, magnetic, gravity and screening techniques) may produce copious amounts of airborne dust. Exposure due to inhalation of this dust is nearly always a concern, particularly in circuits where monazite rich mineral streams are being treated. Monazite tends to be softer and finer than the titanium and zirconium minerals, and may readily fragment during physical separation processes and become preferentially concentrated in airborne dust. In some sections of a dry separation plant, the monazite concentration in airborne dust has been found to be about 10–30 times that of the concentration in feedstock [96, 97].

A typical large dry separation plant contains several hundred units of separation equipment, each unit having several potential dust emission points, for instance at all mineral feed and discharge points. Plant operation is reliant on frequent visual inspection and manual adjustment of separation equipment, requiring close operator contact. The degree of radiation hazard depends on plant layout, the presence and location of significantly radioactive materials such as monazite, time spent on and tasks undertaken by workers in a plant and the existence, use and maintenance of control measures. The operation of some dry separation plants without any dust control measures has in the past led to extreme levels of airborne dust.

Significant inhalation exposures can occur during work tasks that involve manual cleaning methods and certain maintenance activities. Dust generation is aggravated by operating practices such as the use of compressed air and dry sweeping for housekeeping tasks, manual cleaning (pulling) of machine screens and feed slips, paddling air table decks to clean cloth substrates and overfilling of surge bins. Often it is the type of plant rather than its size that determines the radiation hazard. In plants that utilize labour intensive operations there may be close and lengthy contact with sources of airborne dust. Some small plants rely on separate pieces of separation equipment, with manual handling and mineral transport generally done using shovels, buckets and wheelbarrows.

Many design features of a dry separation facility, as well as the nature of the processing equipment and techniques, exacerbate the generation of dust, including:

(i) Material that is continually moving and cannot always be completely enclosed;
(ii) Dry sand that is extensively transported between floors and separation equipment using pipes, open belt conveyors or elevators;
(iii) Receiving chutes of conveyors, elevators and dryers, as well as certain separation techniques such as vibrating screens and air agitation (air tables), are particular sources of dust generation;
(iv) Falling sand may arise from blocked conveyors and elevators as a result of power failures;
(v) Use of gravity in the separation process means that material is likely to become suspended through air displacement;
(vi) Numerous exposed beams, pipes, cable trays and other equipment surfaces provide settling points for airborne dust which, along with dust on floors and window sills, may become resuspended by the movement of men and vehicles;
(vii) The hot and dry conditions that favour efficient separation allow particles to be carried on local thermal air currents;
(viii) Abrasion and attrition, as well as carryover of clay fines (slimes) from raw sand feed, introduce small particles that are more likely to become suspended;
(ix) Buildings are multi-storey, with grid mesh flooring, hindering effective spillage identification and control;
(x) Separation equipment is often designed to ‘spill’ material in the case of overload, for reasons of metallurgical convenience.

6.3.1.4. Secondary separation: Internal exposure to thoron, radon and their decay progeny

High concentrations of thoron, radon and their decay progeny are unlikely to be found in mineral separation plants owing to the low emanation of thoron and radon from process material, the short half-lives of the radionuclides involved and the good ventilation conditions in such processing plants. The inhalation of thoron and radon is therefore not usually an exposure pathway of concern. However, elevated levels of thoron and its decay progeny are likely to be observed in closed storage areas where large quantities of monazite or monazite rich tailings are stockpiled.

6.3.2. Gamma dose rates

Detailed information on absorbed dose rates associated with mining and beneficiation processes is presented in Appendix IX. The data can be summarized as follows:

(a) In areas where heavy-mineral sands are industrially exploited and where the monazite content of heavy-mineral sand deposits is high, the natural gamma background can be elevated well above normal levels. In India, for instance, natural background dose rates in such areas are in the range of 0.2–3 µGy/h;

(b) In mining operations, dose rates are highly variable, depending on the monazite content of the sand. In some cases, they are similar to local background levels, while for deposits with a high monazite content they can be as high as 35 µGy/h. Dose rates at backfilled mining areas are indistinguishable from local background levels;

(c) In the primary separation process, dose rates in the general plant area are not significantly above background levels, with a maximum of 3 µGy/h. The range of dose rates associated with exposure to heavy-mineral concentrate is significantly higher (1–20 µGy/h), while for tailings the range is lower (0.2–1.5 µGy/h);

(d) In the secondary separation process, exposure to gamma radiation depends on location within a facility and on the monazite content of the raw sand (and thus of the heavy-mineral concentrate). Dose rates associated with stages other than monazite separation are generally in the range 0.5–12 µGy/h, but in plants where the monazite content of the raw sand is high (typically more than 0.5%), values of up to 30 µGy/h have been reported. Monazite separation gives rise to dose rates of 2–30 µGy/h, while monazite bag filling gives rise to dose rates of 5–50 µGy/h. The highest dose rates are those associated with exposure to bulk amounts of monazite in stockpiles or stores, where the range is 30–250 µGy/h. Dose rates also depend on the type of facility. For instance, plants that utilize labour intensive operations are more likely to involve close and lengthy contact with sources of external radiation, resulting in higher dose rates.

6.3.3. Radionuclides in airborne dust

6.3.3.1. Mining and primary separation

Dust activity concentrations in mining and primary separation facilities are provided in Table 23. The concentrations are very low because of the wet nature of the process.

6.3.3.2. Secondary separation

Mass concentrations of airborne dust in dry separation plants are provided in Table 111 in Appendix X. Typically, average concentrations are in the range of 0.5–3 mg/m^3, but much higher concentrations, of the order of 10 mg/m^3, were reported in the 1970s and 1980s when airborne dust was not as well controlled. In four separation plants in Vietnam, it is reported that “primitive techniques without any safety measures were used for mineral separation”, resulting in exceptionally high airborne dust concentrations of 300–4200 mg/m^3 [100].

TABLE 23. AIRBORNE DUST ACTIVITY CONCENTRATIONS IN MINING AND PRIMARY SEPARATION

	Th-232 activity concentration (mBq/m^3)
Australia, mine site [98]	4–20 (average 10)
Australia [53][a]	
Mining	<2
Wet concentration	3
India [99][a]	
Dredge site	0.04
Beach washing plant	0.08
Beach sand site	0.07
Inland deposit site	0.11

[a] Derived from the gross alpha activity concentration assuming decay chain equilibrium in the airborne particles and 100% thoron retention on the sampling filter (see Section 3.3.2.2).

Various investigations have been conducted to determine the AMAD of airborne dust in dry separation plants. The results are shown in Table 24. For purposes of dose assessment, an AMAD of 5 µm is generally taken to be a conservative estimate. A higher value of, say 10 µm, might be considered in some situations, where supported by routine monitoring. For instance, there are indications that the AMAD is higher in the monazite section than in other sections of a plant.

Activity concentrations of ^{232}Th in airborne dust in dry separation plants are provided in Tables 112 and 113 of Appendix X. The data in Table 112, which have been gathered from literature dating back to 1984, show that in sections of a plant associated with the separation of heavy minerals other than monazite (mostly ilmenite, rutile and zircon), mean activity concentrations are in the range of 6–240 mBq/m^3, with a tendency for activity concentrations at later stages of the separation process to be higher than those at earlier stages. In sections where monazite is separated, activity concentrations are higher, with mean values in the

TABLE 24. PARTICLE SIZE OF AIRBORNE DUST IN DRY SEPARATION PLANTS

	AMAD (µm)	
	Mean	Range
Australia, 2 plants [98]	6	
Ilmenite separation section		3.6–7.5
Rutile separation section		4.8–6.5
Zircon separation section		5.3–8.5
Monazite separation section		5.6–7.0
Australia [101]	6	2–12
Australia [53]		
Stationary measurements		8–10
Personal measurements	>10	
Australia, 1 plant [95]	3.2	2.8–6.8
Australia [48]		10–15
Australia, monazite circuit [51]	14	10.3–17.9
India, 2 plants [94]	7	2.7–15[a]

[a] The AMADs at the monazite air tables were at the higher end of the range.

range of 20–700 mBq/m^3. In monazite bagging and storage areas, the range is 7–70 mBq/m^3. Mean activity concentration in any particular section of the plant can vary by an order of magnitude between plants, even within the same country. This variation appears to be due mainly to differences in the thorium content of feedstock and differences in workplace practices.

Airborne activity concentrations in dry separation plants have decreased substantially since the 1970s and 1980s. This is well illustrated by the situation in Australia, as shown by the data in Table 113. The average airborne ^{232}Th activity concentration in Australian plants, which stood at 350 mBq/m^3 in 1977, is currently less than 10 mBq/m^3. There has been a corresponding reduction measured in daily intake by plant workers — the average intake of alpha activity (estimated without accounting for the use of protective respiratory equipment) decreased from 8.5 Bq in 1986 to about 1.6 Bq in 1991 [48]. (These values correspond to ^{232}Th intakes of about 1 and 0.2 Bq, respectively.) This has been achieved through a combination of engineering controls and improved work and management practices.

The determination of average daily ^{232}Th intake by 241 workers employed for 1–40 years in Western Australian dry separation plants, based on records of airborne dust activity concentrations and employment periods, is described in Refs [34, 35, 48]. The employment periods generally covered the time in which airborne activity concentrations within the plants were being progressively and substantially reduced. Intake determination did not take into account the use of protective respiratory equipment, nor did it include contributions from alpha emitters in the ^{238}U decay chain. Refs [34, 35] describe, in addition, the determination of thorium lung burdens of 207 of these workers using the ‘thoron in breath’ technique, calibrated using direct in vivo counting conducted on 25 workers selected from the same group. Average daily ^{232}Th intake is summarized in Table 25, based on the results reported in Refs [34, 35, 48], with the following considerations:

(a) The ^{232}Th intake determined from airborne dust activity concentrations has been recalculated so as to include the contribution of alpha emitters in the ^{238}U decay chain (see Fig. 3), assuming a ^{232}Th:^{238}U activity concentration ratio of 2 for non-monazite sections, 10 for monazite sections and 4 for the plant overall. Detailed data on ^{232}Th intake derived from Ref. [48] in this way are supplied in Table 114 of Appendix X;

(b) Lung burdens, which overall were found to be in the range of <6–96 Bq with a mean value of 10 Bq, have been converted to long term average daily ^{232}Th intake by assuming that long term inhalation of ^{232}Th at a rate of 1 Bq/a results in an accumulated lung burden of 0.16 Bq (see Section 3.3.1).

TABLE 25. LONG TERM AVERAGE DAILY INTAKE OF AIRBORNE ^{232}Th BY WORKERS IN AUSTRALIAN DRY SEPARATION PLANTS

No. of workers	Employment period (a)	Job category	Method of determination	Average daily Th-232 intake (Bq)		Ref.
				All workers	Individual workers	
13	1–20	Primarily operation of heavy-mineral sections other than monazite sections	Air sampling	0.25	0.09–0.53	[48]
8	1–11	Primarily operation of monazite sections	Air sampling	0.49	0.23–-0.97	[48]
9	1–6	Operation of both monazite and non-monazite sections	Air sampling	0.46	0.25–1.04	[48]
4	3–9	Plant maintenance	Air sampling	0.30	0.23–0.37	[48]
62	2–34	Plant operation and maintenance	Air sampling	0.33	—	[34]
			Thoron in breath	0.17	<0.1–1.3	
145	1–40	Not specified	Air sampling	0.14	—	[35]
			Thoron in breath	0.17	<0.17–1.6	

6.3.4. Thoron and radon progeny

Emanation of thoron and radon from heavy minerals has been found to be very low, as shown in Table 26. Consequently, the concentrations of thoron and radon progeny in air are generally also low, as shown in Tables 27 and 28.

TABLE 26. THORON AND RADON EMANATION COEFFICIENTS FOR SOME HEAVY MINERALS

	Emanation coefficient		Ref.
	Thoron	Radon	
Monazite	0.0002	0.002	[92, 102]
Zircon	—	0.0013–0.048	[8, 102]
Xenotime	—	0.0009	[102]

TABLE 27. CONCENTRATIONS OF THORON AND THORON PROGENY IN WORKPLACES

	Thoron concentration (Bq/m^3)		Thoron progeny concentration (μJ/m^3)		Ref.
	Mean	Range	Mean	Range	
Mining					
Australia				0.02–0.2	[53]
India		64–119	0.35		[103]
Primary separation					
Australia, wet concentration				0.02–0.2	[53]
Secondary separation					
Australia, primary and secondary circuits				0.02–0.2	[53]
Australia, ‘latter stages’		Up to 1200			[101]
Australia, monazite circuits				0.02–0.2	[53]
Australia, monazite bagging and storage				0.02–0.4	[53]

TABLE 27. CONCENTRATIONS OF THORON AND THORON PROGENY IN WORKPLACES (cont.)

	Thoron concentration (Bq/m^3)		Thoron progeny concentration ($\mu J/m^3$)		Ref.
	Mean	Range	Mean	Range	
Australia, bulk monazite store		Up to 5000	0.3		[101]
India, Plant 1				0.06–0.6	[82]
India, Plant 2				0.02–0.4	[82]
India	200	100–300	0.06; 0.1[a]	0.01–0.6	[91]
India				0.02–0.1	[104]
Overall					
Australia				<0.002–0.1	[98]

[a] The lower figure is for plants where the monazite content of feedstock is less than 0.5%. The higher figure is for plants where the monazite content is more than 0.5%.

TABLE 28. CONCENTRATIONS OF RADON AND RADON PROGENY IN WORKPLACES

	Radon concentration (Bq/m^3)		Radon progeny concentration ($\mu J/m^3$)		Ref.
	Mean	Range	Mean	Range	
Mining, India		3.7–12.2			[103]
Secondary separation, India					[105]
Plant 1	206		0.33		
Plant 2	189		0.49		
Plant 3	295		0.50		
Plant 4	217		0.42		
Overall					
Australia, most locations		Up to 200		Up to 0.02	[101]

6.3.5. Effective dose

When establishing effective dose for the purposes of radiation protection, usually only exposures to external gamma radiation and airborne dust are taken into consideration. Exposures to thoron and radon progeny are generally ignored, as exposure levels are not significantly elevated. For an annual exposure period of 2000 h, the levels provided in Tables 27 and 28 correspond to annual effective doses of a few tenths of a millisievert with a maximum of the order of 1 mSv. Informal measurements made in Australian dry separation plants confirm this, indicating an annual effective dose from thoron progeny of 0.2–0.3 mSv [106].

Doses received by workers depend on the type of plant and its layout, the presence and location of significantly radioactive materials such as monazite, the time spent on and tasks undertaken by workers in a plant and the existence, use and maintenance of control measures. The data in Appendices IX and X show that there are very large variations in gamma dose rates and airborne dust activity concentrations. Consequently, it is not possible to translate these data directly into reliable estimates of annual effective dose. It is clear, however, that there is a potential for some workers to receive high doses from both gamma radiation and inhalation of airborne dust.

6.3.5.1. External exposure to gamma radiation

Effective doses received by workers from external gamma exposure in Brazilian dry separation plants were determined from workplace monitoring data. Annual doses were found to be 15.4 mSv for monazite milling, 35 mSv for magnetic separation and 21 mSv for 'light fraction deposition'. Annual effective doses received by workers in four Indian dry separation plants, as determined using TLDs deployed at fixed locations, were 3.06, 3.49, 4.42 and 3.29 mSv, respectively [105]. In Australia, gamma doses received by certain designated workers (essentially those working in the operation and maintenance of dry separation plants) are routinely determined through individual monitoring using TLDs [106, 107]. The mean effective doses from external exposure during the period 1986–2007 are shown in Table 29. In the 1970s, exposure of monazite plant operators and monazite product baggers to external radiation caused them to regularly receive annual effective doses exceeding 10 mSv [96]. Since then, there has been a significant reduction, with mean doses from external radiation now in the order of 1 mSv per year. The reduction in doses since 1994 can be explained by the fact that there was no monazite production after this date.

TABLE 29. DOSES RECEIVED BY WORKERS IN AUSTRALIAN DRY SEPARATION PLANTS *(from data provided in Refs [106, 107])*

Year	Mean annual effective dose (mSv)			Year	Mean annual effective dose (mSv)		
	Gamma radiation	Airborne dust	Total		Gamma radiation	Airborne dust	Total
1977		50.7		1993–1994	0.7	0.9	1.6
1978		32.6		1994–1995	0.7	0.8	1.5
1979		28.7		1995–1996	0.6	1.1	1.7
1980		27.8		1996–1997	0.9	1.1	2.0
1981		27.2		1997–1998	0.9	1.2	2.0
1982		25.9		1998–1999	1.0	1.0	2.0
1983		17.8		1999–2000	0.9	1.1	2.1
1984		18.1		2000–2001	0.8	1.3	2.2
1985		24.4		2001–2002	0.8	1.2	2.0
1986	2.4	15.1	17.5	2002–2003	0.8	2.3	3.1
1987	1.7	10.6	12.3	2003–2004	1.0	1.9	2.9
1988	2.1	9.1	11.2	2004–2005	0.7	1.1	1.8
1989	2.0	8.7	10.7	2005–2006	0.7	0.9	1.6
1990	1.4	4.8	6.2	2006–2007	0.9	1.3	2.2
1991	1.3	2.8	4.1	2007–2008	0.6	0.7	1.3
1992	1.5	3.2	4.7				

6.3.5.2. Internal exposure to airborne dust

Table 29 also shows mean committed effective doses received by workers in Australian dry separation plants through exposure to airborne dust for the years 1977–2007. There was apparently a fifty fold reduction in dose over this period. Part of this reduction may have been be due to changes in dust monitoring procedures over the years and the way in which the annual exposure period was determined.[11] Doses have been determined from the data reported in Refs [106–108] in the following way:

(a) For the period 1977–1992, doses were calculated from airborne gross alpha activity concentrations reported in Refs [107, 108], assuming an annual exposure period of 2000 h, a mean breathing rate of 1.2 m^3/h and a dose coefficient of 7.4 μSv per becquerel of gross alpha activity (corresponding to an AMAD of 5 μm and a mixture of monazite and non-monazite dust with a ^{232}Th:^{238}U activity ratio of 4, see Fig. 4);

(b) For the period 1993–2007, the doses reported in Ref. [106] were used, but with a small upward adjustment of 3.4% to maintain consistency with the dose coefficients provided in Fig. 4 — the assumed dose coefficient was 7.0 μSv per becquerel of gross alpha activity.[12] As in (a), the doses reported in Ref. [106] were based on a mean breathing rate of 1.2 m^3/h, but instead of assuming an annual exposure period of 2000 h, the actual time spent in different sections of the plant and performing various duties were taken into account.

Long term average values of the committed effective doses received per year from exposure to airborne dust in Australian dry separation plants were calculated from the average daily intake given in Table 25, assuming a ^{232}Th:^{238}U activity concentration ratio of 2 for non-monazite sections, 10 for monazite sections and 4 for the plant overall. The intake data cover selected groups of workers with employment periods of up to 40 years extending back from the early 1990s. During those employment periods, airborne activity concentrations within the plants were being progressively and substantially reduced. The results are shown in Table 30.

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[11] Dose assessements for the earlier part of the period were based on the results of stationary air sampling, typically carried out in the more dusty areas of the plant, assuming an annual exposure period of 2000 h. Dose assessments in later years were based on the results of personal air sampling, which expanded to include a wider range of workers, some of whom may not have necessarily spent much time in dusty areas. In addition, actual rather than assumed annual exposure periods were used.

[12] There was no monazite production during this period, so a ^{232}Th:^{238}U activity ratio lower than that for 1977–1992 would be expected. It is reported in Ref. [106] that the thorium to uranium mass ratio in the dust was between 7 and 9. Taking a mean mass ratio of 8, this corresponds to a ^{232}Th:^{238}U activity concentration ratio of 2.6 and, from Fig. 4, a dose coefficient of 7.03 μSv per becquerel of gross alpha activity (for low solubility particles with an AMAD of 5 μm). The dose coefficient assumed in Ref. [106] was 6.8 μSv per becquerel of gross alpha activity.

TABLE 30. LONG TERM AVERAGE DOSES FROM DUST INHALATION IN AUSTRALIAN DRY SEPARATION PLANTS

No. of workers	Employment period (a)	Job category	Method of determination	Committed annual effective dose (mSv)		Source of data on intakes
				Average for all workers	Averages for individual workers	
13	1–20	Primarily operation of heavy-mineral sections other than monazite sections	Air sampling	5	2–12	[48]
8	1–11	Primarily operation of monazite sections	Air sampling	11	5–21	[48]
9	1–6	Operation of both monazite and non-monazite sections	Air sampling	10	5–23	[48]
4	3–9	Plant maintenance	Air sampling	7	5–8	[48]
62	2–34	Plant operation and maintenance	Air sampling	7	—	[34]
			Thoron in breath	4	<2–28	
145	1–40	Not specified	Air sampling	3	—	[35]
			Thoron in breath	4	<4–35	

6.3.5.3. Total effective dose

Mean values of the total annual effective dose received from both gamma radiation and airborne dust over the period 1986–2007 by workers in Australian dry separation plants are given in Table 29 and shown graphically in Fig. 16. Data on effective dose received by dry separation plant workers in India and Vietnam are given in Table 31.

6.3.6. Measures to reduce doses

As pointed out in Section 6.3.1, the main area of radiological concern is the secondary separation process. Many opportunities for reducing doses have been identified, which, through a combination of engineering and administrative controls, have led to significant reductions in doses, particularly in doses received through the inhalation of airborne dust (see Section 6.3.5.2). Effective dust suppression requires careful attention to every potential source of air contamination. Even then, complete containment of dust is often not practical and ventilation equipment and adequate operational procedures are prerequisites for satisfactory control. Measures to reduce doses received through external exposure have also been identified, especially in areas where large quantities of

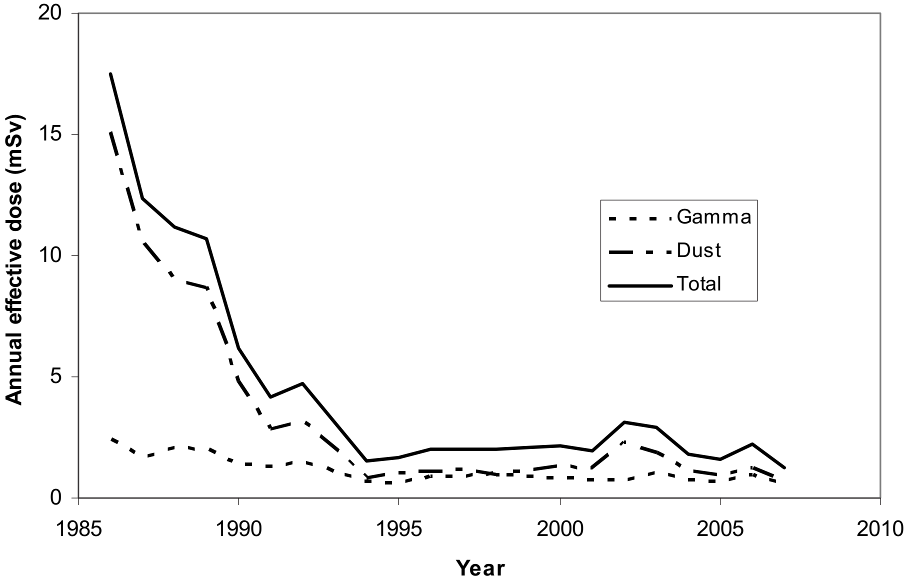

FIG. 16. Doses received by workers in Australian dry separation plants during the period 1986–2007.

TABLE 31. DOSES RECEIVED BY WORKERS IN DRY SEPARATION PLANTS IN INDIA AND VIETNAM

	Committed annual effective dose (mSv)
India, over a period of years up to 1996 [109]	3–10
India, 2 plants, 1997–2001 [91]	
Plant 1 (relatively low monazite content in sand)	1.1–3.2
Plant 2 (relatively high monazite content in sand)	4.3–5.1
India [110]	
Plant 1 (~0.5% monazite in raw sand)	1.98 (1.5–2.5)
Plant 2 (2% monazite in raw sand)	4.91 (4.2–7.0)
India [90]	
Plant 1	3
Plant 2	7
Vietnam [111]	6

process material are stored or handled. It is important to bear in mind that exposure to radiation is only one of several hazards at a mining or mineral processing site and needs to be treated as part of an overall OHS programme for a particular workplace.

6.3.6.1. Administrative measures

In most cases, adequate protection against external exposure can be achieved through the careful siting of materials and by discouraging workers from spending unnecessary time in these areas. Unnecessary exposures may be avoided if workers are reminded of the presence of radioactive minerals by demarcating areas of higher dose rates with appropriate floor markings and caution signs. For areas in which high gamma dose rates can occur, such as areas where significant quantities of monazite are stored or disposed of, it has been found necessary in some facilities to impose access controls and restrictions on occupancy periods. In extreme cases, consideration is given to job rotation. Good industrial hygiene, including restrictions on eating, drinking and smoking in certain areas, and maintaining an appropriate standard of general housekeeping, are widely regarded as being effective in contributing to the control of intake by workers.

Since there is always the possibility of high gamma dose rates and high airborne dust activity concentrations at certain locations and times, vigilance is needed in ensuring that individual and workplace monitoring programmes for external and internal exposure are implemented effectively. It has been found that widely varying levels of exposure for identical tasks can be avoided by providing in-house, standardized and practical work training programmes, including retraining. Other measures found to be important are supervision and methods to improve work discipline, promotion of a safety culture, work standardization, documented procedures at all levels and ISO certification.

6.3.6.2. Process considerations

Airborne dust is of particular concern because the secondary separation process is normally conducted under dry conditions. The introduction of wet process methods, especially in the initial stages, substantially reduces the opportunity for dust generation. Possibilities that have been investigated include wet magnetic separation and mineral froth flotation, particularly for minerals with fine particle size. However, the feasibility of these alternative processes is very mineral dependent and they may not yet be able to supersede contemporary dry separation processes on a commercial scale. Air table separation techniques are particular sources of airborne dust, and the use of alternative separation techniques is an option worth considering.

Coating larger particles of mineral sands with fine particles (slimes) can be a significant contributor to airborne dust generation in a dry separation plant. Two types of pre-treatment can be considered in order to reduce slimes coating: scrubbing of mineral particles in wet attritioning circuits (with the released coating being removed in desliming circuits) and wet classification (such as hydrosizing) directed towards fines removal. Improved removal of the mineral grain coating of heavy-mineral concentrate has the added advantage of increasing the efficiency of electrostatic separation, thus reducing the required number of dust generating mineral separation stages.

The periods of time spent by workers close to process materials can be reduced by the use of systems for remote plant monitoring and process control. Worker exposures can also be reduced through the use of automatic on-stream analysis for product quality control rather than relying on manual mineral sampling.

6.3.6.3. Plant layout and design

The design and layout of a dry separation plant can have significant implications for doses received by workers. One of the most important considerations in this regard is the segregation and separation of radioactive minerals from regularly occupied workplaces. This includes the establishment of a separate, dedicated monazite separation facility that is isolated from the main ilmenite and zircon separation areas, the location of monazite stores away from normally occupied areas and the direct pumping of crude monazite concentrate to silos or earthen trenches for storage. Other considerations in the design and layout of a plant include the following:

(a) Isolation of the process stream to reduce dust migration by convection currents — this can be achieved in new plants by design or in existing plants through suitable metallurgical or process stream modifications;
(b) For new plants, a low profile design rather than a multilevel construction (even though this results in a less compact plant layout), to enable replacement of downward gravitational movement and upward transfer of minerals by bucket elevators with other means (such as conveyor belts) that cause less mineral abrasion and thus less dust generation;
(c) Provision of rooms or cubicles with filtered air supplies and self-closing doors (preferably a simple airlock with two doors) and air conditioned cabs on machinery;
(d) Designing of operational areas in such a way that the effects of regular cleaning and careful product control are more readily visible to operators.

6.3.6.4. Engineering measures for controlling airborne dust levels

Transfer of airborne dust between various parts of a plant via natural and thermal air currents may be prevented by wall partitioning and solid flooring between various plant levels. Covering grid mesh floors can reduce dust migration, but is limited in effectiveness and may aggravate noise and heat problems.

Enclosures fitted around equipment limit the migration of dust and can reduce airborne dust activity concentrations by as much as an order of magnitude. For separation processes that require little manual operation, enclosures around large sections of a separation circuit (or the whole circuit where it is not too large) connected to dust filtration and collection systems via suitable ducting can be effective in reducing exposures provided that the control of access to these enclosures is practical. Such enclosures may be fitted to equipment such as electrostatic and magnetic separators, air tables and vibrating screens. Similar dust control methods have also been effective on transfer, feed and collection systems of processed minerals such as bucket elevators, bins, hoppers and conveyer feed and discharge points. These include the provision of hoods for chutes receiving sand from elevators and conveyors, covers for conveyors, enclosures at discharge points onto and from conveyor belts and from gravity transfer chutes, and seals on the housings of bucket elevators used for the vertical transport of minerals. The enclosure and insulation of equipment that handles hot minerals, such as feed elevators and driers, reduces thermal convection currents and allows for better ventilation without causing increased dust circulation.

Airborne dust levels can be reduced by avoiding large falling distances of minerals when they are transported via pipes and conveyors to destination equipment, particularly in multilevel plants. This can be achieved through the use of slide plates and dust proof chutes. The chutes connect discrete solid floor areas at each level with dust collection bins at the ground floor level and are vented to minimize dust emissions caused by batch discharges of dust.

More detailed information on good engineering design for mineral handling and conveying, electrostatic and magnetic separating equipment, screening equipment, air tables and sample cutters and containers is given in Ref. [112].

Local exhaust ventilation is effective in controlling airborne dust levels, especially for potentially high dust generating sources such as screens, feed and discharge points, electrostatic and magnetic separators, air tables, conveyors and elevator discharge and feed points, but can present a complex engineering problem and involves significant capital outlay. Dilution ventilation lowers overall dust levels by displacing dust laden air from the plant, but high velocities can be counterproductive because they might cause increased dust resuspension and keep larger particles in the air for longer. Suitable ventilation can be provided through the use of radial exhausts on roofs or by using roof designs that promote natural ventilation.

6.3.6.5. Engineering measures for controlling gamma radiation

The following measures can be taken to reduce external exposures from materials with high activity concentrations:

(a) The use of physical barriers to segregate radioactive minerals or, if space constraints do not permit an adequate segregation distance, the use of appropriate shielding techniques to lower dose rates in accessible areas (for example, the placement of bags or drums of low activity materials around the radioactive mineral);
(b) Shielding for bins containing monazite;
(c) The use of containers for shipment of monazite.

6.3.6.6. Surface contamination control

Strict measures to prevent the spread of contamination may be both impractical and unnecessary. Surface contamination is more easily controlled than in chemical processing operations. In general, prompt cleanup of monazite contamination will minimize exposure via the surface contamination pathway. When feasible, vacuum cleaning of spillage and dust deposited on surfaces is preferred over manual cleaning. Some plants are equipped with an integrated vacuum system serving strategic points at each section and operating level. Installation of a stand-by electrical power supply removes the possibility of material spillage in the event of a power failure.

6.3.6.7. Respiratory protection

A respiratory protection programme introduced in an Australian dry separation plant required the use of half mask cartridge type respirators in all plant areas where airborne gross alpha activity concentration exceeded 400 mBq/m^3 (equivalent to a dust concentration of about 3 mg/m^3). These areas were clearly delineated and signs were posted. This programme had been put in place while engineering control measures were being installed to reduce dust and radioactivity concentrations [113]. The filtration efficiency was specified as particulate matter penetration not exceeding 2% for particles with a size distribution of between 0.02 and 1 μm diameter and a mass median diameter of 0.6 μm. The mandatory use of respirators was accompanied by worker instruction on fitting and use of the masks, including basic maintenance, cleaning and storage requirements. Date of issue, respirator details and other relevant information were recorded for each worker. Workers were also instructed in simple fitting tests to determine if gross leakage was occurring. Follow-up training was provided. Evaluation of the workplace performance of the respirators revealed a mean protection factor of 5.1 for dust exposure (range 1.8–13) and 7.5 for radioactivity exposure (range 2.5–21). In a follow-up study [114], a mean radioactivity protection factor of 5.1 (range 1.5–21.8) was determined.

6.4. PUBLIC EXPOSURE

The main exposure pathways of interest during mining and beneficiation operations are those associated with atmospheric discharges, process water discharges and the management of solid process residues.

6.4.1. Atmospheric discharges

Atmospheric discharges originate primarily from the secondary (dry) separation process. Investigations of airborne dust, as well as of thoron and its progeny, were carried out in areas around various mining and beneficiation operations in India. The airborne concentrations at these locations were found to be comparable with natural levels in surrounding high natural background radiation areas, indicating that mining and beneficiation operations have no discernible radiological impact [82, 86, 91, 99, 115, 116]. Airborne ^{232}Th activity concentrations measured within 2 km of the boundaries of two dry separation plants were 1–6 mBq/m^3 for one plant (mean 3.8 mBq/m^3) and 1.9–3.6 mBq/m^3 for the other plant (mean 2.9 mBq/m^3). The thoron progeny concentrations at these locations were 0.01–0.1 µJ/m^3. A similar result was found for airborne dust in the vicinity of a heavy-mineral sand separation plant in Brazil. Characterization of the dust particles suggested that the heavy-mineral component originated not only from the plant but also from a nearby natural deposit of heavy-mineral sand. It was not possible to distinguish any clear, direct impact of atmospheric discharges from the plant [117].

6.4.2. Discharges of processing water

In India, the gross alpha activity concentration in the effluent water generated from the pre-concentration of raw sand and wet mineral separation was found, after settling, to be 0.007–0.009 Bq/L, which is similar to the gross alpha concentrations measured in the local high background area (0.002–0.023 Bq/g for groundwater and 0.022 Bq/g for river water) [82]. A similar result was found for recycled processing water [91]. Radioactivity levels in liquid effluent from dry separation plants in India are also very low, with a ^{228}Ra activity concentration of 0.03 Bq/L (range 0.01–0.04 Bq/L) [104]. At a primary separation plant in Brazil, where water used for wet separation was extracted from a coastal lagoon and discharged at the same point after use, the activity concentrations of ^{228}Ra and ^{226}Ra in the effluent were monitored and found to be similar to or less than those in the lagoon [118]. Activity concentrations upstream of the discharge point were higher than those downstream, as shown in Table 32.

6.4.3. Management of solid residues

Residues from mining and primary separation operations are of little radiological concern, because of their low activity concentrations (see Section 6.2.6). Activity concentrations in tailings from the secondary separation process are higher, but only moderately so because most of the radioactivity in

TABLE 32. ^{228}Ra AND ^{226}Ra IN LIQUID EFFLUENT FROM A BRAZILIAN WET SEPARATION PLANT COMPARED WITH NATURAL LEVELS IN THE RECEIVING WATER BODY [118]

Distance from head of water body (km)	Activity concentration in water (Bq/L)	
	Ra-228	Ra-226
0	1.52	0.63
0.8	1.52	0.36
1.9	0.49	0.25
3.7	0.20	0.15
4.5 (discharge point)		
Lagoon water	0.18	0.17
Effluent	0.19	0.07
4.9	0.10	0.14
5.4	0.12	0.08

the heavy-mineral concentrate tends to end up in the products rather than in the tailings. There is some off-grade residual material containing radionuclides at greater concentrations, but this material is readily diluted with non-radioactive material. Radioactive contaminants in the tailings are effectively still in their natural state, with no alteration of their chemical or physical form, and can be considered to be bound within the sand grains. As a consequence, since they are not prone to leaching into groundwater, the solubility or mobility of individual radionuclides is not a major consideration in the disposal of tailings [119]. Monitoring of drinking water from wells located within 5 km of a mining area refilled with tailings and other residues in Tamil Nadu, India did not show any significant impact of earlier mining and residue disposal activities [99]. Thoron and radon emissions from tailings are insignificant because of the very short half-lives (especially of thoron), the extremely low emanation rates and, in the case of radon, the relatively low activity concentration of ^{226}Ra.

6.4.4. Exposures from past operations

After termination of mining operations, there is a net reduction of radioactivity in the ground, even taking into account the backfilling of a mining void with tailings and other solid residues from the beneficiation process. Exposure of the public to gamma radiation from mined out areas can therefore be expected to be similar to or lower than pre-mining levels. Gamma doses rates at mine sites in India, before and after mining, are provided in Table 33. The data generally reflect measurements made at chest level and refer to two types of site: dredge mining areas, which are typically mined to a depth of 6 m and subsequently refilled with low activity material, and beach mining areas, which are not refilled after mining. The post-mining values measured at beach mining areas reflect levels immediately after mining, before heavy-mineral sand deposits were replenished by wave action in the following monsoon season. Clearly, mining operations significantly reduce gamma dose rates. In addition, concentrations of thoron progeny in air at the mined and refilled areas were found to be three times lower than the original levels [82, 99].

Dose assessments conducted at mining areas in India, based on post-operational data and environmental monitoring results, show that the annual effective doses received by members of the public in mined out areas after remediation are significantly lower than those received before mining started. This is due mainly to a substantial reduction in external gamma doses. The internal dose is very small by comparison and is not affected by mining

TABLE 33. GAMMA DOSE RATES AT MINING SITES

	Absorbed dose rate (μGy/h)		Ref.
	Before mining	After mining (and refilling in the case of dredge sites)	
Dredge mining areas			
Manavalakurichi	3–5	0.2–0.3	[116]
Tamil Nadu	1–3	0.4–0.8	[99]
South coast	1–3	0.2–0.8	[120]
Chavara and Manavalakurichi	—[a]	0.3–0.5	[91]
Beach mining areas			
Manavalakurichi	—[b]	—[b]	[116]
Tamil Nadu	0.5–20	0.5–3	[99]
South coast	1–20	0.5–3	[120]

[a] The dose rates before mining were reported to be an order of magnitude higher than those after mining.

[b] The dose rates before mining were reported to be three times higher than those after mining.

operations. The results of environmental radiological assessments carried out for three mining areas are shown in Table 34.

Some past operations were terminated without having been brought under adequate control. One such situation reported from the Russian Federation had implications for public exposure [122]. A heavy-mineral sand processing facility operating between 1949 and 1964 resulted in the contamination of 10 200 m^2 of ground (8000 m^2 of which was as a result of sand storage) and uncontrolled use of the sand as a material for the construction of buildings and roads. Ground samples were found to contain ^{232}Th at activity concentrations of up to 6 Bq/g. In the middle of the most highly contaminated area, the gamma exposure (which corresponded to a dose rate of approximately 10 µSv/h[13]) was 40–50 times the local outdoor mean value. Inside the contaminated buildings, the maximum gamma exposure (which corresponded to a dose rate of about 2 µSv/h) was nearly 7 times the indoor mean value for the village. Thoron concentrations in contaminated buildings were 2.5–15 Bq/m^3 compared with a mean value of 1.7 Bq/m^3 and maximum value of 2.5 Bq/m^3 in uncontaminated houses. No evidence was found of elevated radon concentrations or of contamination in air or groundwater samples by long lived alpha emitting radionuclides. The maximum annual effective dose in the contaminated buildings was assessed to be 16.2 mSv from gamma radiation and 3.8 mSv from thoron progeny.

TABLE 34. DOSES RECEIVED BY MEMBERS OF THE PUBLIC IN MINING AREAS IN INDIA

	Annual effective dose (mSv)					
	Area 1 [121]		Area 2 [121]		Area 3 [99]	
	Before operation	After operation	Before operation	After operation	Before operation	After operation
External gamma radiation	7.36	2.63	4.82	2.63	15.4	4.82
Dust inhalation	0.46	0.46	0.46	0.46	1.5	1.5
Thoron progeny inhalation	0.30	0.10	0.30	0.10	0.1	0.1
Water and food ingestion	0.03	0.03	0.03	0.03	<0.1	<0.1
Total	8.15	3.22	5.61	3.22	17.0	6.42

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[13] Dose rates are derived from exposure rates reported in röntgen per hour, assuming that an exposure rate of 1 µR/h roughly corresponds to a dose rate of 0.01 µSv/h.

6.5. TRANSPORT OF MATERIAL

The transport of large quantities of raw sand, concentrates and tailings can generate appreciable quantities of airborne dust. In India, this has led to the adoption of the following control measures: periodic cleanup of spillage, watering of dust prone areas on roads and within plants, wetting of tailings before transport and the use of spill proof trucks [82, 99]. The transport of monazite requires particular attention because of its high activity concentration. At a dry separation plant in India, monazite is filled from silos directly into 3 t steel transport containers loaded inside a truck. The containers are provided with appropriate transport labels [91].

6.6. REGULATORY CONSIDERATIONS

The ranges of radionuclide activity concentrations in the following process materials extend (or may extend) beyond 1 Bq/g:

(a) Heavy-mineral concentrate, monazite, xenotime, zircon, leucoxene;
(b) Tailings, oversize solids and clay fines from secondary separation process.

Activity concentrations in all other processing materials are generally less than 1 Bq/g. As a result, it is clear from Section 3.1.1 that the mining and beneficiation of heavy-mineral sands needs to be considered for regulation as a practice.

Doses received by workers are of regulatory concern only for those workers involved in secondary (dry) separation processes. In current operations, annual effective doses are generally about 1 mSv in well managed operations, but extending upward to a few millisieverts per year in some plants. Much higher doses were received in the past when measures to control exposures (particularly exposures involving airborne dust) were not well established. Because of the potential for doses to approach or even exceed the occupational dose limit, a comprehensive monitoring programme and a combination of engineering and administrative controls are required. Effective implementation of such measures should avoid the need to rely on the use of personal protective equipment in the form of respiratory protection.

Exposure to member of the public is minimal. Indeed, after a mining area is refilled, doses are lower than previous natural background levels.

As a result of this situation, mining and primary separation operations would appear not to require specific regulatory control measures, other than perhaps some general oversight. Secondary separation operations clearly need to be controlled in a more comprehensive manner, and should therefore be conducted under authorization in the form of a registration or licence. Examples of regulatory guidelines for mineral sand mining and beneficiation operations are provided in Refs [123, 124].

Radionuclide activity concentrations in monazite and xenotime are such that the transport of these materials would be subject to the requirements of the Transport Regulations.

7. RECOVERY OF HEAVY MINERALS FROM TIN MINING RESIDUES

Tin bearing alluvial deposits have been the major focus of mining in south-east Asia for more than a century. The ore contains about 0.01% tin in the form of cassiterite, together with various other heavy minerals, mainly ilmenite, zircon, monazite, xenotime, strüverite ($(\text{Ti,Ta,Fe}^{2+})\text{O}_2$), and columbite ($(\text{Fe,Mn})(\text{Nb,Ta})_2\text{O}_6$). In tin mining operations, after wet gravity separation of lower density gangue material consisting mainly of quartz sand and clay, the resulting rough concentrate of cassiterite is further upgraded to about 75–76% tin, leaving a sandy residue, known as ‘amang’, containing heavy minerals and sand tailings. Since the mid-1950s, this residue has been periodically processed to recover residual cassiterite as well as ilmenite and other heavy minerals, including the minerals monazite and xenotime, which have REO contents of 60% and 54%, respectively [10]. In a typical amang plant, the main minerals produced are ilmenite and cassiterite. Semi-concentrated forms of other mineral products are either sold to larger plants for purification into saleable grades of minerals such as monazite, zircon, xenotime and strüverite, or stored on-site for future processing, depending on demand.

7.1. PROCESS DESCRIPTION

Heavy-mineral separation processes use combinations of equipment such as wet shaking tables, rotary or vertical driers, magnetic separators, electrostatic separators and air tables. Examples of process flow charts can be found in Refs [11, 125]. The processes are similar to those used for heavy-mineral sands (see Section 6.1), but are generally simpler and on a smaller scale, with a typical plant throughput of only 50–100 t/d in smaller plants. Typical wet processing and dry processing operations are shown in Figs 17 and 18, respectively. Larger plants in Malaysia and Thailand are reported to have a monazite production of up to 240 t/d. Mineral handling and transportation within a plant is often done manually with the aid of shovels, buckets and wheelbarrows, and minerals are sometimes accumulated on the floor prior to bagging (see Fig. 18). Plant operation relies on frequent visual inspection and manual adjustment of separation equipment. In larger and more sophisticated plants, equipment and techniques are more complex, to enable production of export grade quantities of other heavy-mineral sand products such as zircon, monazite, strüverite and xenotime.

Reports from Malaysia claim that, due to inefficiencies in extraction processes, treated amang may be retreated several times, depending on the demand for tin and other mineral products [125]. Sometimes plants suspend operations, resulting in large variations in production quantities. In 2005, data from nine amang plants indicated that about 84 000 t of amang were processed.

In a 1991 study [126], more than 50% of the amang plants in Malaysia were reported to employ less than 10 workers and 24% less than 5 workers. Only one plant employed more than 50 workers. A more recent report puts the mean

FIG. 17. Wet processing of amang.

FIG. 18. Dry processing of amang.

number of workers at 12 per plant, with an average annual working period of 2500 h [125]. Plants in Thailand and Indonesia were generally reported to employ more than 20 workers.

The recycling of large quantities of water used in the wet separation process involves the use of recycling ponds. Water is first drawn from a river and then pumped into a natural pond. Water from this pond is used in a plant and then returned to the pond. The sediments accumulating in recycling ponds exhibit enhanced levels of radioactivity and this has created some concern with regard to reclamation of the ponds for future land use [127].

7.2. RADIONUCLIDE ACTIVITY CONCENTRATIONS

Activity concentrations in amang and various mineral concentrates derived from amang are provided in Table 35. The data given for monazite, xenotime, zircon and ilmenite are summaries of more detailed data provided in Appendix XI. The wide ranges of activity concentration are a reflection of differing levels of purity, especially with regard to monazite content and, in the case of ilmenite, the presence of zircon as an impurity [125]. Due to impurities, ranges of activity concentrations in ilmenite and zircon recovered from amang include values that are significantly higher than those in ilmenite and zircon recovered from heavy-mineral sands (see Section 6.2).

TABLE 35. ACTIVITY CONCENTRATIONS IN AMANG PROCESS MATERIALS

	Activity concentration (Bq/g)	
	Th-232 or Ra-228	U-238 or Ra-226
Amang [128, 129]	0.6–1	1–3.2
Monazite[a]	67–370	12–46
Xenotime[a]	13–200	37–247
Zircon[a]	1–88	4.8–62
Ilmenite[a]	0.06–10.5	0.07–8.2
Mixed rutile, tin oxide [125]	0.25	0.44
Tin oxide, 72% [125]	0.02	0.04
Strüverite [128]	0.24[b]; 7.7	1.06[b]; 29
Tourmaline, 80% [125]	2.3	0.99
Wolframite, 74% [125]	0.03	0.23

[a] More detailed data are given in Appendix XI.
[b] These values refer to a 10% concentrate.

At Malaysian amang plants, sediment samples were collected from water recycling ponds at points of discharge from the plants and at further sampling locations 25–100 m from discharge points [127]. The activity concentrations measured in these samples are shown in Table 36.

Activity concentrations of ^{228}Ra and ^{226}Ra in soil contaminated to a depth of 0.2 m from the storage of amang feedstock were found to be 1.036 and 0.855 Bq/g, respectively. Corresponding activity concentrations from the storage of ilmenite were 0.599 and 2.571 Bq/g [129].

7.3. OCCUPATIONAL EXPOSURE

7.3.1. Exposure pathways

Operating practices in amang processing plants (see Section 7.1), together with housekeeping tasks such as sweeping and machine cleaning, result in close

TABLE 36. ACTIVITY CONCENTRATIONS IN SEDIMENT IN PROCESSING WATER RECYCLING PONDS AT MALAYSIAN AMANG PLANTS [127]

	Activity concentration (Bq/g)					
	Th-232		U-238		Ra-226	
	Mean	Range	Mean	Range	Mean	Range
Plant 1						
Discharge point	0.52	0.43–0.64	0.41	0.36–0.47	0.19	0.16–0.23
25–100 m away (1)	0.17	0.11–0.26	0.10	0.10–0.11	0.05	0.02–0.11
25–100 m away (2)	0.18	0.11–0.34	0.31	0.08–0.86	0.05	0.03–0.08
25–100 m away (3)	0.21	0.16–0.30	0.10	0.10–0.13	0.10	0.04–0.20
Plant 2						
Discharge point	0.31	0.13–0.61	0.20	0.12–0.34	0.05	0.05–0.15
25–100 m away (1)	0.10	0.05–0.22	0.08	0.06–0.11	0.03	0.02–0.07
25–100 m away (2)	0.11	0.06–0.20	0.10	0.08–0.16	0.04	0.03–0.07

and lengthy worker contact with sources of external and internal exposure. Since workers spend most of their work shift in a plant, both types of exposure are likely to be significant. Many of the secondary separation processes are dry, resulting in the emission of significant amounts of dust into the workplace atmosphere. As explained in Section 6.3.1.3, this dust may have a higher monazite content than that of the feedstock. As amang plants usually have quite good natural ventilation, airborne concentrations of dust and of thoron and radon progeny are expected to be moderated to some extent by dilution [125].

7.3.2. Exposure to gamma radiation

Average absorbed dose rates measured in several amang plants in Malaysia are reported in Ref. [130]. Further measurements were made in eight plants in Malaysia and four plants in Thailand [126] and outside seven plants in Malaysia [131]. The results of these investigations are given in Table 37. The following general observations on measurements reported in Ref. [126] were made:

(a) Dose rates were about 0.5 μGy/h in most office areas;
(b) Dose rates were generally less than 2.5 μGy/h in wet concentration areas, above tailings piles and in other plant areas in which there were no accumulations of mineral concentrates;

TABLE 37. GAMMA DOSE RATES MEASURED IN AMANG PLANTS

	Absorbed dose rate at various distances (μGy/h)[a]			
	0.1 m	0.3 m	1 m	Unspecified
Malaysia, several plants [130]				
Monazite storage room	>100	91		
Xenotime storage room	40	25		
Zircon storage room	26	20		
Tin ore storage room	15	10		
Ilmenite pile in workplace	10	3		
Tin tailings outside plant	5	4		
Strüverite and rutile outside plant	3	3		
Pyrite pile outside plant	1	2		
Work area with monazite storage				Up to 100
Other work areas				1–3
Natural background				0.5
Malaysia, general level outside plant [133]				10
Malaysia, above access tracks around plants and stockpile areas [134]				0.5–10
Malaysia, 8 plants; Thailand, 4 plants [126][b]				
Office areas			<0.6 (0.3–3)	
Wet concentration area			1–2 (0.6–5)	
General plant			2–5 (0.5–20)	
Ilmenite separation area				1–2
Amang			3–4 (1.2–8)	
Ilmenite			2–3 (1.8–7)	
Zircon			5–7 (4–15)	
Monazite			90–100 (35–180)	
Cassiterite			4–8 (1.6–14)	
Strüverite			14–18 (8–35)	
Access tracks			3–5 (0.2–10)	
Site boundary			0.5–1 (0.2–4)	
Tailings			1–2 (0.3–2)	

TABLE 37. GAMMA DOSE RATES MEASURED IN AMANG PLANTS (cont.)

	Absorbed dose rate at various distances (μGy/h)[a]			
	0.1 m	0.3 m	1 m	Unspecified
Malaysia, working areas of 16 plants [125]				1.3 (0.42–3.5)
Malaysia, outside 7 plants [131]				
Plant 1				1.6 (0.1–10.7)
Plant 2				5.5 (0.7–18.7)
Plant 3				3.7 (0.4–16.5)
Plant 4				3.6 (1.2–10.2)
Plant 5				4.8 (0.6–56.0)
Plant 6				1.6 (0.4–3.8)
Plant 7				2.2 (0.5–6.8)

[a] Values denote mean or typical levels. Ranges are given in parentheses.

[b] Measurements were made 1 m from the ground and generally 1 m from equipment or material, although in some plants it was not possible to obtain typical levels away from stockpiles or bags of mineral as such accumulations were ubiquitous.

(c) Dose rates tended to be below 7.5 μGy/h in dry separation areas, if no minerals were stockpiled close to regularly occupied work areas;

(d) Dose rates were above 7.5 μGy/h where significant quantities of mineral concentrates of varying purity (for example impure or semi-concentrates of zircon) were stored close to regularly occupied work areas, as was the situation in many plants. In some cases, dose rates were above 25 μGy/h, as reported also in Refs [130, 132];

(e) Dose rates near mineral concentrates generally reflected radionuclide activity concentrations in such materials, with monazite having by far the highest dose rates and ilmenite the lowest. However, values varied markedly from plant to plant and, in some cases, minerals of supposedly moderate activity concentrations (for example strüverite, zircon, cassiterite and ilmenite) gave rise to a wide range of values, some of which were much higher than expected, indicating contamination by more radioactive minerals such as monazite;

(f) Dose rates above access tracks around plants and stockpile areas also varied widely, the higher values being indicative of contamination of soil by minerals rich in monazite and xenotime. Such contamination was also evident in other areas accessible by workers, including some areas outside offices, laboratories and plants. Dose rates near site boundaries also varied widely.

7.3.3. Radionuclides in airborne dust

Mass concentrations and activity concentrations of airborne dust in amang processing plants are given in Table 38. The gross alpha activity concentrations reported in Refs [126, 133, 135, 136] have been converted to ^{232}Th and ^{238}U activity concentrations assuming a ^{232}Th:^{238}U concentration ratio of 1.6, this being the mean for 16 plants in Malaysia (range 0.6–4.2) according to the data in Ref. [125]. The measurements reported in Ref. [126] were reported to be mostly associated with short term samples collected in fixed locations 1.5–2 m above the ground and in generally occupied work areas. The mean dust activity concentration, ignoring one anomalously high result, was 11 mBq/m^3 for ^{232}Th and 7 mBq/m^3 for ^{238}U. It was pointed out that actual dust concentrations experienced by workers could be higher or lower than reported values, but were more likely to be higher. On the basis of airborne dust measurements, the monazite content of dust was estimated in Ref. [126] to be about 10%, considerably higher than that in the feedstock (<0.5%). This finding is consistent with the observation that, in some sections of dry mineral sands separation plants, the concentration of monazite in airborne dust was about 30 times that of the mineral feedstock [137] (see Section 6.3.1). The AMAD of airborne dust measured in eight plants in Malaysia was in the range of 3.0–5.3, with a mean value of 3.9 [125].

7.3.4. Thoron progeny

Thoron progeny and radon progeny exposures have been determined in several amang plants in Malaysia. The results are shown in Table 39.

7.3.5. Effective dose

In Ref. [126], for purposes of calculating effective doses, rough estimates of the average gamma dose rate experienced by workers in eight amang plants in Malaysia and four in Thailand were derived from average gamma dose rates measured in different plant areas (see Table 37). In the absence of specific time and motion data, the following occupancy factors for different sections of the plant were assumed: 40% in dry processing sections, 30% in wet sections, 10% close to amang stockpiles, 10% near ilmenite stockpiles, 4% near zircon, cassiterite or strüverite stockpiles, 1% near monazite piles, and 5% outside the

TABLE 38. AIRBORNE DUST ACTIVITY CONCENTRATIONS IN AMANG PLANTS

	Mass concentration (mg/m^3)	Activity concentration (mBq/m^3)[a]	
		Th-232	U-238
Malaysia, respirable dust (<7 μm) [133]	—	22	14
Malaysia, air tables, magnetic separators, mean of 7 samples [135]	3.6	—	
Malaysia, 29 plants, ~90 samples [136][b]			
Range	—	0.7–1700	0.4–1060
Typical	—	5–14	3–9
Mean	—	12	7
Malaysia, 8 plants; Thailand, 4 plants [126]			
Dry separation plant	1.9	21	13
Dry separation plant, not operating	0.3	2	1
Dry separation plant and monazite plant	0.7	19	12
Monazite plant	—	12	7
Dry separation plant, partially operating	0.4	2	1.5
Dry separation plant	2.8	30	19
Dry separation plant, partially operating	0.4	11	7
Dry separation plant	84	610	380
Dry separation plant	—	11	7
Dry separation plant	<3	26	16
Malaysia, 16 plants [125][b]			
Range	2.2–9.2	0.3–7.3	0.4–3.7
Mean	5.9	1.9	1.2

[a] The values from Ref. [125] are as reported. All other values have been derived from measurements of gross alpha activity concentration assuming a Th-232:U-238 activity ratio of 1.6.

[b] The highest values were those measured near magnetic and electrostatic separators and vibrating screens.

TABLE 39. THORON PROGENY AND RADON PROGENY CONCENTRATIONS IN AMANG PLANTS IN MALAYSIA [138]

	Potential alpha energy concentration (μJ/m^3)						
	Thoron progeny						Radon progeny
16 plants [125]							
Range	0.04–0.17						0.01–0.07
Mean	0.08						0.04
6 plants [138][a]	Plant 1	Plant 2	Plant 3	Plant 4	Plant 5	Plant 6	
Amang mixture, magnetic separation	0.03	0.04	N[b]	0.02			
Monazite, magnetic separation		0.02					
Monazite, air table separation and nearby storage						0.10	
Monazite store room				0.26	0.16		
Zircon store room					N		
Strüverite, air table separation	0.07						
Strüverite, store room			N				
Xenotime, store room					0.30		

[a] Values are derived from airborne Pb-212 concentrations (see Section 3.4.1).

[b] N: not detectable.

plant. The result was an overall gamma dose rate of about 1–17 μGy/h, with a mean of 3.7 μGy/h. The largest contribution to the dose rate (about 25%) came from proximity to monazite stockpiles, even though the assumed occupancy factor was only 1%, illustrating the importance of controlling exposure to such material. The annual exposure period was taken to be 2400 h, giving annual effective doses ranging from <2 to 40 mSv with a mean value of about 9 mSv. Corresponding figures for dust inhalation (based on dust activity concentrations reported in Ref. [126], an annual exposure period of 2400 h, a ^{232}Th:^{238}U activity ratio of 1:6 and low solubility dust with an AMAD of 5 μm) gives annual effective doses ranging from 0.4 to 12 mSv with a mean value of 2 mSv. It is pointed out in Ref. [126] that the intermittent use of respiratory protection for dusty tasks would result in lower doses in practice. Combining the effective doses from external gamma exposure and dust inhalation gives a total annual effective dose of about 2–50 mSv with a mean of 11 mSv.

Annual effective doses assessed in 16 Malaysian amang plants [125] are shown in Tables 40 and 41. External gamma doses were assessed from TLD measurements at fixed locations (see Table 37). The assessed doses were expected to overestimate actual doses received because workers were not always working close to the sources of gamma exposure. Dust inhalation doses were assessed from personal air sampling measurements (see Table 38). Doses received from the inhalation of thoron progeny and radon progeny were assessed by continuous monitoring using active air sampling at fixed locations (see Table 39). The assessed doses were calculated using dose coefficients that were twice the value of those listed in Table 11. Consequently, for consistency with Table 11, the doses for thoron progeny and radon progeny reported in Tables 40 and 41 are 50% of those reported in Ref. [125].

TABLE 40. ANNUAL EFFECTIVE DOSES IN 16 MALAYSIAN AMANG PLANTS BY WORKPLACE *(derived from Ref. [125])*

	Annual effective dose (mSv)			
	External gamma	Airborne dust	Progeny of thoron and radon	Total
Air table (n=11)	7.2	0.4	0.3	7.9
Plate separator (n=3)	4.6	0.1	0.4	5.1
Dryer (n=9)	4.1	0.5	0.2	4.8
High tension separator (n=22)	2.9	0.5	0.3	3.7
Magnetic separator (n=36)	3.5	0.5	0.3	4.3
Intermediate separator (n=5)	3.2	0.3	0.3	3.8
Rapid magnetic separator (n=17)	2.7	0.2	0.2	3.1
Wet shaking tables (n=18)	2.0	0.1	0.3	2.4
Electrostatic separator (n=4)	1.3	0.1	0.3	1.7
Mean	3.5	0.3	0.3	4.1

Note: n denotes the number of measurements.

TABLE 41. ANNUAL EFFECTIVE DOSES IN 16 MALAYSIAN AMANG PLANTS BY PLANT *(derived from Ref. [125])*

Plant number	Annual effective dose (mSv)			
	External gamma	Airborne dust	Progeny of thoron and radon	Total
1	7.8	1.2	0.4	9.4
2	9.8	0.2	0.5	10.5
3	4.6	0.9	0.2	5.7
4	4.2	0.4	0.2	4.8
5	4.6	0.2	0.2	5.0
6	2.4	0.3	0.2	2.9
7	2.5	0.4	0.3	3.2
8	3.0	0.1	0.3	3.4
9	2.5	0.1	0.2	2.8
10	1.2	0.6	0.2	2.0
11	2.4	0.2	0.1	2.7
12	1.8	0.1	0.2	2.1
13	2.4	0.1	0.2	2.7
14	1.9	0.1	0.3	2.3
15	0.9	0.7	0.2	1.8
16	1.0	0.1	0.3	1.4
Mean	3.3	0.4	0.3	4.0

The annual effective dose received by a bulldozer driver transporting minerals to plants was estimated from the mean gamma dose rate and the annual exposure time (assumed to be 3 μGy/h and 500 h, respectively), giving an annual effective dose of 1.5 mSv [131].

7.3.6. Measures to reduce dose

As a result of investigations of exposure conditions in amang plants [126, 130, 132], the following dose reduction measures have been identified:

(a) Instruction of workers to increase their awareness of radiation hazards in amang processing plants and to familiarize them with work practices that reflect good radiation protection practice;

(b) Isolation of monazite and xenotime concentrates by:
  - (i) Storing them away from regularly occupied areas under good ventilation conditions;
  - (ii) Labelling of bags and drums;
  - (iii) Where necessary, shielding of concentrates (especially monazite) using bags or drums of low activity tailings or sand, or by incorporating shielding into the design of concentrate storage bins and feed hoppers;

(c) Improved design and layout of plants, including:
  - (i) Semi-automation of processes such as material conveying and transfer;
  - (ii) Introduction of slide plates or chutes to minimize airborne dust generated by large free fall distances;
  - (iii) Operation of feed pipes to separation equipment in the 'choke feed' mode to minimize entrainment and subsequent emission of dust as a result of a pipe not being filled with mineral;
  - (iv) Provision and proper use of ventilation systems that achieve frequent air changes without increasing or prolonging the suspension of dust particles in the atmosphere and avoiding the passage of dust laden air through occupied workplaces;
  - (v) Use of extraction hoods or enclosures (connected via ducting to an air cleaning system of suitable capacity) around dust generating equipment, especially screens, feed and discharge points to electrostatic and magnetic separators, air tables and pipes and chutes feeding conveyors and elevators;
  - (vi) Where necessary, removal of very fine particles from mineral before it enters the separation process by pre-washing or attritioning with clean water or other attritioning aids;

(d) Programmes for ensuring that equipment is maintained and operated as intended, for instance, ensuring that bucket elevators are not operated with missing side panels;

(e) Reduction of close worker contact with monazite by using larger capacity bags that have to be handled by machines rather than by hand, for example using 1–2 t bags instead of 50 kg sacks;

(f) Demarcation and signposting of areas with elevated gamma dose rates;

(g) Limiting the time spent by workers in areas where monazite and xenotime are separated and stored, for instance, by providing a room with a filtered or clean air supply so that workers do not have to spend unnecessary amounts of time in areas of high exposure;

(h) Enforcement of high standards of industrial hygiene in areas involving high activity material such as monazite processing sections, including measures to reduce spills and unnecessary accumulations of material;

(i) Emphasizing the need for individual monitoring of external exposure and airborne dust to allow for more accurate dose estimation;

(j) Where necessary, provision of suitable protective respiratory equipment conforming to international design standards and enforcement of their use.

7.4. PUBLIC EXPOSURE

In Malaysia, elevated gamma dose rates have been recorded in public areas near mineral processing plants with mineral stockpiles [139]. The highest dose rates were recorded in an area where large amounts of monazite, zircon and ilmenite had been deposited by the roadside — 1.56 μGy/h inside a car on the road, 1.98 μGy/h outside the car and 2.67 μGy/h at one of the mineral piles.

A study was carried out in a residential area neighbouring a typical amang plant in Malaysia to investigate the radiological impact on its residents [140]. Five single storey brick houses in a low cost housing estate next to a medium sized amang processing plant were selected for the study. One house was only about 20 m from an open amang stockpile. Indoor measurements of absorbed dose rate, airborne dust activity concentration, thoron progeny concentration and radon progeny concentration were carried out. For comparison purposes, corresponding measurements were carried out in an office in the amang plant and in a clean (air conditioned) building at a local university. Annual effective doses were assessed from these measurements and are shown in Table 42. The reliability of the results is compromised by the use of conservative assumptions and outdated dose coefficients. Nevertheless, the doses assessed for the five residential situations are comparable with average worldwide values and with values generally found in Malaysia. The dose in the amang plant office was significantly higher, owing to a higher contribution from external gamma radiation. The dose at the university clean area was also higher, in this case owing to a higher contribution from radon progeny. The results showed no evidence of any radiological impact on nearby residents.

TABLE 42. EFFECTIVE DOSES RECEIVED BY RESIDENTS NEAR AN AMANG PROCESSING PLANT *(from Ref. [140])*

	Annual effective dose (mSv) (rounded)				
	External gamma	Dust inhalation	Thoron progeny inhalation	Radon progeny inhalation	Total (rounded)
House 1	1.11	0.001	0.07	0.75	1.9
House 2	1.27	0.001	0.22	0.92	2.4
House 3	1.02	0.001	0.12	0.73	1.9
House 4	1.00	0.002	0.11	1.08	2.2
House 5	1.01	0.001	0.11	0.90	2.0
Office, amang plant	2.74	0.001	0.12	0.84	3.7
Clean area, university	1.26	0.003	0.22	1.92	3.4

Exposure modelling was conducted to predict the doses that could be received by members of the public arising from the use of contaminated land following the closure of an amang processing plant [129]. Based on activity concentrations measured in a 0.2 m layer of soil contaminated by the storage of amang feedstock and ilmenite product (see Section 7.2), two types of exposure scenarios were considered: an individual living in a house built on contaminated soil and an individual working in a factory built on contaminated soil. Four exposure pathways were included in the calculations: external radiation, inhalation of dust, inhalation of radon and ingestion of contaminated soil. Various ventilation rates and thicknesses of soil cover were considered. The results are summarized in Table 43. The air exchange rate and soil cover thickness were predicted to influence dose mostly at air exchange rates of up to 2 h^{-1} and soil cover thicknesses of up to 0.2 m. Above these values, any further reductions in dose would be small. In all cases, ^{226}Ra would initially be the major contributor to total dose, and this contribution would increase even more after the passage of a few years. Because of the relatively short half-life of ^{228}Ra (5.75 years), its contribution to total dose, which would already be relatively small from the outset, would decrease quite rapidly with time. For soil contaminated by amang, total dose would reduce by 90% after about 20 years, whereas for soil contaminated by ilmenite a similar reduction would be obtained only after about

TABLE 43. ESTIMATED DOSES FROM EXPOSURE TO CONTAMINATED LAND AFTER THE CLOSURE OF AN AMANG PROCESSING PLANT [129]

	Annual effective dose (mSv)			
	Air exchange rate 1 h^{-1}		Air exchange rate 10 h^{-1}	
	No soil cover	1 m soil cover	No soil cover	1 m soil cover
Residential scenario				
Amang contamination	10	1	3	0.1
Ilmenite contamination	35	7	30	0.6
Factory scenario				
Amang contamination	2	1	0.3	Negligible
Ilmenite contamination	6	2	3	0.1

100 years. It was also estimated that an increase in thickness of the contaminated soil layer (from 0.2 to 0.4 m) would increase total dose by 30%.

7.5. REGULATORY CONSIDERATIONS

Radionuclide activity concentrations in amang, as well as in the monazite, xenotime, zircon and ilmenite separated from it, are above 1 Bq/g. Consequently, in terms of Section 3.1.1, the recovery of heavy minerals from amang must be considered for regulatory control as a practice.

Annual effective doses received by workers are typically about 4 mSv, but were considerably higher in past operations. Doses received by members of the public are indistinguishable from natural background levels. The situation is therefore very similar to that for the recovery of heavy minerals from mineral sands, as described in Section 6.6, although the need for regulatory control measures in the workplace could be greater for amang processing because operations are typically less mechanized and less automated. As with mineral sand secondary separation operations, a range of administrative and engineering control measures (and, where necessary, respiratory protection) should be implemented through authorization in the form of a registration or licence.

The radionuclide activity concentrations in monazite and xenotime, as well as possibly some zircon, ilmenite and strüverite, are such that the transport of these materials would be subject to requirements of the Transport Regulations.

8. MINING AND BENEFICIATION OF OTHER DEPOSITS

8.1. RARE EARTH BEARING CLAYS: SOUTHERN CHINA

Commercially attractive quantities of rare earth bearing ion adsorption clays are found in the south-eastern Chinese provinces of Jiangxi, Guangdong and Fujian. This material has some desirable characteristics that are not possessed by any other type of ore, including:

(a) The low content of cerium in relation to lanthanum;
(b) The relatively high content of yttrium in some deposits and middle rare earths in others;
(c) The ease of mining and processing to recover rare earths, even though the actual rare earth content (a few tenths of a per cent [72]) is much less than that of other types of ore.

The ore bodies consist of loose layers, 3–10 m thick, of completely weathered granitic rocks that have been altered to mostly clays in which rare earths, rich in yttrium and heavy rare earths, are adsorbed. As a result of the effect of natural weathering processes, the REO composition varies according to mining location and depth, but does not exceed 1%. The low concentration of rare earths in the ore is made up for by the relative ease of mining and processing and the high value of the heavy rare earths. The clays are mined from open pits with power shovels. The clays are sent directly for chemical treatment without the need for any beneficiation.

Concentrations of ThO_2 and U_3O_8 in the deposits are very low (typically about 0.005%), and the corresponding thorium and uranium series radionuclide activity concentrations are therefore well below 1 Bq/g. No exposure data are available but, considering the low activity concentrations and the mining and beneficiation methods involved, exposures are unlikely to be of any significant concern and would thus not require regulatory control.

8.2. BASTNÄSITE: MOUNTAIN PASS, USA

This deposit was once the most important source of rare earths worldwide, but mining operations have been suspended since 2000. The ore, which comprises 10–15% bastnäsite (average 12%), contains rare earth elements in the form of a mixed rare earth fluorocarbonate. The rare earth concentration in the ore is 2–12% REO and the Th and U concentrations are 0.02–1 and 0.002%, respectively (see Table 89 in Appendix III). In past operations, the ore was extracted by drilling and blasting in an open pit approximately 150 m deep. The broken ore was loaded into large haul trucks using front end loaders and a hydraulic mining shovel and transported to the processing plant. Water spray trucks were used for dust control. In the plant, the ore was crushed in three stages to a maximum size of about 10 mm and stockpiled. Different grades of stockpiled material were blended to form a suitable feedstock having a uniform rare earth concentration of about 7–9% REO. After conveying to the flotation plant, the fine ore was ground in a ball mill to reduce particle size to 100% passing through a 150 mesh screen (approximately 100 μm). A series of conditioning steps followed by hot froth flotation steps produced a bastnäsite concentrate with a rare earth content of about 60–65% REO. Some of this concentrate was shipped to outside processing locations and the rest was used on-site for chemical processing. Ponds with an asphalt pad and liner were used to hold the bastnäsite concentrate. Tailings generated in the beneficiation process were deposited in a tailings pond.

Radionuclide activity concentrations in the various mining and beneficiation process materials are shown in Table 44. The ^{232}Th concentrations in the bastnäsite concentrate (3.1–4.6 Bq/g) are similar to those obtained from ores mined in China — bastnäsite concentrate from Bayan Obo has a ^{232}Th activity concentration of 5.3–7.8 Bq/g [11], while that from Sichuan Province has a ^{232}Th activity concentration of 3.9 Bq/g [74].

TABLE 44. RADIONUCLIDE ACTIVITY CONCENTRATIONS IN PROCESS MATERIALS IN THE MINING AND BENEFICIATION OF BASTNÄSITE ORE AT MOUNTAIN PASS MINE

	Activity concentration (Bq/g)		Ref.
	Th-232	U-238	
Ore	*0.8–4*	*0.25*	[141, 142]
Ore	0.93	0.3	[143]
Mineral concentrate	*3.9–4.6*		[10]
Mineral concentrate	3.1	0.15	[143]
Tailings	0.5	0.15	[143]

Note: Values in italics are derived from mass concentrations, assuming that 1 g of natural thorium contains 4057 Bq of Th-232, 1 g of ThO_2 contains 3566 Bq of Th-232 and 1 g of natural uranium contains 12350 Bq of U-238.

Personal monitoring using TLDs was carried out for three consecutive months during 1992 [141]. The TLDs were placed on individuals expected to receive the highest exposure. The results, converted to an hourly dose rate, are shown in Table 45. No further exposure data are available but, based on the TLD results given in Table 45 and dust inhalation calculations for similar types of material given in Appendix III of Ref. [7] which are based on the activity concentrations given in Table 44, it can be concluded that a worker exposed to bastnäsite concentrate could receive an annual effective dose of about 0.3–0.4 mSv from exposure to external gamma radiation and about 0.4 mSv from exposure to airborne dust. Such a situation would not require an occupational radiation protection programme to be implemented, even though radionuclide activity concentrations may exceed 1 Bq/g, as long as normal OHS measures for the control of dust inhalation (such as respiratory protection in dusty atmospheres) were in place. Consequently, operations involving the mining and beneficiation of bastnäsite, while being treated as practices, may be candidates for exemption.

8.3. LOPARITE: RUSSIAN FEDERATION

Commercially attractive quantities of loparite ore are found in the Kola Peninsula region of the Russian Federation. The ore contains 2–3% loparite, which in turn is composed of 37–40% titanium dioxide, 28–37% REO, 7–13% niobium oxide and 0.5–0.8% tantalum oxide [144], with the balance being mainly lime (CaO). Due to its high titanium content, the exploitation of loparite is associated with large titanium recovery operations. The ore is mined using underground and open pit methods and, after size reduction, is beneficiated by gravity and electromagnetic separation methods to yield a concentrate containing 95% loparite.

TABLE 45. GAMMA DOSE RATES FOR WORKERS AT MOUNTAIN PASS MINE *(derived from Ref. [141])*

	Average dose rate (μSv/h)	
	Driller in mine	Crusher operator
February 1992	0	0.07
March 1992	0.06	0
April 1992	0.4	0.6

Reported values of rare earths, thorium and uranium concentrations in the 95% loparite concentrate are given in Table 46. Assuming decay chain equilibrium and representative ThO_2 and U_3O_8 concentrations in the concentrate of 0.6 and 0.03%, respectively, the estimated activity concentrations in the concentrate are 21 Bq/g for ^{232}Th and 3 Bq/g for ^{238}U. Assuming that concentrations in the ore are 40 times lower, the corresponding activity concentrations in the ore are 0.5 Bq/g for ^{232}Th and 0.08 Bq/g for ^{238}U. No exposure data are available, but based on calculations for similar types of material given in Appendix III of Ref. [7] and the activity concentrations given above, a worker exposed to loparite concentrate could receive an annual effective dose of about 4 mSv from exposure to external gamma radiation and about 2 mSv from exposure to airborne dust. Such a situation would require an occupational radiation protection programme to be implemented. For a worker exposed only to ore, the corresponding annual effective doses would be about 0.1 and 0.05 mSv for gamma radiation and dust, respectively, and exposure control would most likely not be necessary.

8.4. VEIN TYPE MONAZITE DEPOSIT: STEENKAMPSKRAAL, SOUTH AFRICA

A small monazite mining and beneficiation operation was run at Steenkampskraal in South Africa from 1950 to 1965 and there are plans to resume operations at this facility. The mineral body forms an elongated deposit of high grade monazite (up to 75% [148]), dipping steeply to a depth of at least 400 m. The ore contains about 30% REO, 2.5–3% ThO_2 (originally up to 6%) and 0.1–1% U_3O_8. The monazite itself contains 57% REO (principally La, Ce, Pr, Nd, Sm, Gd and Dy), 3% Y_2O_3 and 8% ThO_2. The production of monazite

TABLE 46. CONCENTRATIONS OF RARE EARTHS, THORIUM AND URANIUM IN LOPARITE CONCENTRATE

Ref.	Mass concentration (%)		
	REO	ThO_2	U_3O_8
[10, 11, 145]	30	0.5–0.6	0.02–0.03
[144]	27.6–36.6	0.5–1	
[146]	28.4–34.3	0.58	0.03
[147]		0.65	

concentrate, containing 44% REO and 6% ThO_2, involves underground mining followed by sorting, crushing, screening, milling, gravity concentration, magnetic separation and flotation.

Based on the above mentioned mass concentrations of thorium and uranium, ^{232}Th activity concentration is about 100 Bq/g in the ore and 200 Bq/g in the concentrate, while ^{238}U activity concentration in the ore is 10–100 Bq/g.

During the period 1953–1963, gamma exposures were determined at various times using area monitoring with dose rate meters and personal monitoring with film badges. Airborne dust exposures were determined during the same period by gravimetric sampling. The results, reported in Ref. [149], are summarized in Tables 47 and 48. Measures to reduce exposures were progressively introduced, the effect of which can be clearly seen from monitoring results. These measures included:

(a) The use of protective clothing and improved facilities and arrangements for personal hygiene;
(b) Improved ventilation systems underground;
(c) Dust suppression with water and through the installation of dust exhaust systems in the plant, especially where concentrates were being handled;
(d) Arrangements for film badges to be worn by all workers when at work and for the keeping of dose records;
(e) The use of job rotation where necessary to avoid excessively high doses;
(f) Education of workers exposed to concentrates to enable them to minimize their doses, for instance by reducing time periods spent in contact with, or close to, the material.

Even after the introduction of these measures, there was still the possibility for a worker to receive an annual effective dose of some tens of millisieverts from external gamma radiation and in the order of 10 mSv from dust inhalation.

Thoron progeny concentrations in the underground air were determined in 1962 by alpha activity measurements conducted on air sampling filters. Thoron progeny concentrations were reported as being just below 1.3×10^5 MeV of alpha energy (1 working level) in the working stopes, as well as in the main return air of the mine. Taking this to be equivalent to a PAEC of about 20 $\mu J/m^3$, it implies an annual effective dose of about 19 mSv for an annual exposure period of 2000 h (see Section 3.4.2).

The abandoned site presents various legacy issues. Radionuclides in ore and tailings remaining on the site have migrated into the surrounding environment as a result of wind erosion and infrequent but heavy falls of rain. Contamination has been detected in local water courses over distances of several kilometres.

TABLE 47. GAMMA EXPOSURES DURING FORMER OPERATIONS AT STEENKAMPSKRAAL RARE EARTHS FACILITY [149]

	Area monitoring, dose rate (µSv/h)		Personal monitoring					
			Workers receiving >25 µSv/h		Workers receiving >60 µSv/h		Workers receiving >200 µSv/h	
	Mine	Plant	Super-visors	Manual workers	Supervisors	Manual workers	Supervisors	Manual workers
1953–1954	≤250	—	—	—	14%	25%	9%	9%
1955–1956	≤190	≤86	—	—	0.8%	5%	None	None
1958	30–100	0–85	—	—	—	—	—	—
1962–1963	≤50		6%	12%	None	None	None	None

Note: Dose rates have been derived from exposures, measured in units of milliröntgen, reported for specified time periods, assuming that an exposure of 1 mR gives rise to a dose of approximately 10 µSv.

TABLE 48. AIRBORNE DUST EXPOSURES DURING FORMER OPERATIONS AT STEENKAMPSKRAAL RARE EARTHS FACILITY [149]

	Th-232 activity concentration in airborne dust (mBq/m^3)		
	Mine workings	Return air from workings	Plant
1953	—	—	Maximum 50 000
1955	—	—	Maximum 32 000
1962	25–250	65	35–7000

Note: Activity concentrations have been derived from reported mass concentrations of thorium.

Numerous structures were erected on the site, including a clubhouse, school and employee housing. Most were constructed of waste rock from the mine, containing significant amounts of thorium and uranium.

Any future resumption of operations will necessitate comprehensive measures to control doses received by workers and members of the public, as well as impact on the environment, including measures to prevent surface water and groundwater pollution, airborne dust and erosion of areas denuded of vegetation. Resumption of operations will also be contingent on the implementation of remedial measures to improve the existing situation, including the processing of tailings and ore remaining on the surface from past operations, the demolition of derelict structures and remediation of the affected area. Such operations would therefore need to be authorized by way of a registration or licence.

9. CHEMICAL TREATMENT OF MINERAL CONCENTRATES

9.1. PROCESS DESCRIPTION

9.1.1. Treatment of mixed bastnäsite–monazite concentrate

The processes described below have been used in China to produce rare earth concentrate from the mixed mineral concentrate originating from Bayan Obo ore. The process of rare earths recovery is complicated by the fact that this concentrate contains a mixture of bastnäsite and monazite, with the monazite component tending to have a high phosphorus and iron content. For the separated bastnäsite and monazite concentrates, the chemical processes are similar to those used elsewhere for these types of minerals (see Sections 9.1.2 and 9.1.4).

9.1.1.1. Treatment with sulphuric acid

The mixed mineral concentrate is reacted with concentrated sulphuric acid in a furnace or kiln at an elevated temperature. The fluorocarbonate mineral matrix is destroyed, gaseous HF, CO_2 and SO_3 are emitted, and the rare earths are converted to sulphates. Gases emitted from the reaction are recovered by passing through milk of lime (a suspension of calcium hydroxide in water). The basic reactions are as follows:

Bastnäsite: $2REFCO_3 + 3H_2SO_4 \rightarrow RE_2(SO_4)_3 + 2HF + 2CO_2 + 2H_2O_2$

Monazite: $2REPO_4 + 3H_2SO_4 \rightarrow RE_2(SO_4)_3 + 2H_3PO_4$ and

$$Th_3(PO_4)_4 + 6H_2SO_4 \rightarrow 3Th(SO_4)_2 + 4H_3PO_4$$

A temperature of 200°C is sufficient for these reactions to proceed. However, through calcination of the reaction mass at temperatures of up to 600°C, or by conducting the reaction itself at such temperatures, the sulphates of thorium and iron are converted into insoluble products while the rare earth sulphates remain stable. Leaching of the reaction product in water followed by filtration produces a rare earth sulphate solution with minor amounts of impurities and a filter cake containing thorium, iron and a few other heavy metals, together with other insoluble matter such as barium sulphate, calcium sulphate and silica.

In the older version of the process, the addition of sodium sulphate to the solution causes the rare earths to precipitate as a double sulphate according to the following reaction:

$$RE_2(SO_4)_3 + 2NaSO_4 + 2H_2O \rightarrow RE_2(SO_4)_3 \cdot 2Na_2SO_4 \cdot 2H_2O$$

These double sulphates are recovered through filtration and converted to hydroxides by the addition of sodium hydroxide according to the following reaction:

$$RE_2(SO_4)_3 \cdot 2NaSO_4 + 6NaOH \rightarrow 2RE(OH)_3 + 5Na_2SO_4$$

Fractional dissolution using hydrochloric acid causes the remaining thorium, uranium and iron to precipitate, leaving rare earth chloride in solution. This solution serves as the starting point for producing commercial rare earth products. However, this version of the process leads to a low recovery of rare earths. A solvent extraction technique is now preferred for removing rare earths from the sulphate solution. The required rare earth chloride solution is obtained by stripping the loaded solvent with hydrochloric acid.

The sulphuric acid process generates a large amount of HF-containing waste gas. The slag-to-concentrate mass ratio is up to 0.66, resulting in the production of thousands of tonnes of slag per year. This slag contains 0.2% Th and has to be stored as a radioactive residue.

9.1.1.2. Treatment with hydrochloric acid and alkali

This process is similar to that used for treating bastnäsite concentrate (see Section 9.1.2). The mixed mineral concentrate, containing 40–60% REO, is first treated using a proprietary process to eliminate calcium fluoride. The concentrate is then leached with hydrochloric acid to dissolve part of the rare earth content, yielding rare earth chloride in solution:

$$RE_2(CO_3)_3 \cdot REF_3 + 6HCl \rightarrow 2RECl_3\ (soln) + REF_3\ (s) + 3H_2O + 3CO_2$$

Rare earth fluoride and monazite are carried in the cake. This is digested hot with 20–50% NaOH to convert the phosphate and fluoride into water soluble salts and the rare earths into hydroxides, which are separated and dissolved to obtain rare earth chlorides. The remaining cake is discarded or recycled to alkali digestion, depending on its composition, and the rare earth chloride solution is purified by precipitating and removing the impurities. The purified rare earth chloride can either be evaporated to obtain solid rare earth chloride or treated using solvent extraction to obtain a major part as light rare earth chloride with a minor concentrate of medium and heavy rare earths for further separation into individual rare earths.

9.1.1.3. Carbo-chlorination and chemical vapour transport

A stepwise process involving carbo-chlorination followed by chemical vapour transport has been developed and is known as the ‘SC-CVT’ process [150]. Using chlorine gas as a chlorination agent and carbon as a reductant, efficient rare earth chloride conversion can be achieved through carbo-chlorination of the mixed mineral concentrate under $SiCl_4$ gas as a defluorination agent at temperatures as low as 500°C. Complete removal of thorium from the rare earth chloride product is achieved with the subsequent chemical vapour transport reaction at 800°C in the presence of $AlCl_3$ as a vapour complex former. The SC-CVT process has, with some modifications, also been applied successfully to the extraction of rare earths from the bastnäsite concentrate obtained from Bayan Obo ore [151].

9.1.2. Treatment of bastnäsite concentrate with hydrochloric acid

This process has been used at Mountain Pass, USA, to produce rare earths concentrate from bastnäsite concentrate (60–65% REO) obtained from the physical beneficiation process. The bastnäsite concentrate is subjected to an initial hydrochloric acid leach to dissolve carbonate gangue. The resulting slurry is thickened, filtered and dried to yield a leached bastnäsite concentrate with a rare earth content of 68–72%. This is calcined at 600–800°C to produce an oxide mixture containing 85–90% REO. In the calcining step, any carbonate that has earlier resisted treatment is decomposed and about half the Ce^{3+} ions present are converted to Ce^{4+} ions. The calcined material is slurried with water and subjected to leaching in 30% hydrochloric acid.

The leach residue, containing insoluble Ce^{4+} chloride along with fluorides, is converted to a low grade cerium concentrate containing 65–70% REO and 55–60% CeO_2. It is sold as a product for glass polishing applications or is upgraded to a high grade (96%) cerium product through the digestion of fluorides with sodium hydroxide and further leaching with hydrochloric acid. The cerium concentrate upgrading process produces barium sulphates, a leach residue containing mixed rare earth fluorides, and a wastewater stream containing sodium chloride. Cerium concentrate is stored in a pond with an asphalt berm and lining, in readiness for drying when needed.

The leachate, containing chlorides of soluble Ce^{3+}, other rare earth elements, iron and lead, is purified by contacting with a solution of soda ash and tailings slurry to convert iron chloride to insoluble iron hydroxide, which is precipitated into the tailings. After recovering residual rare earths, the iron and tailings precipitate is thickened, washed, neutralized and sent to the tailings pond.

The rare earth chloride, now free of iron, is treated with sodium hydrosulphide solution to precipitate lead as lead sulphide, which is then processed into a sludge cake containing 50% water and 5–7% lead sulphide. This is shipped to a lead recycling facility or disposed of in a controlled hazardous waste landfill facility. The purified rare earth chloride solution, now free of both iron and lead, is then ready for further treatment to produce commercial rare earth products.

Before 1984, iron was not removed from the rare earth chloride solution in a separate processing step and a mixed iron and lead filter cake was produced. This was stored in three small surface impoundments at the mine and subsequently reintroduced into the process so that the iron and lead could be removed separately. After completing recycling of the mixed iron and lead filter cake, it was necessary to close the three surface impoundments in an approved manner.

Wastewater from throughout the plant is neutralized, following which rare earth oxides and lead are precipitated and the purified effluent is piped to an evaporation pond located in a dry lake bed underlain by low permeability clay. The precipitated solids are reintroduced into the processing streams to recover rare earth oxides.

9.1.3. Treatment of mineral concentrate from Mount Weld, Australia, using high temperature acid digestion

The mineral concentrate will be shipped to Malaysia for chemical treatment. Details currently available of the process are given in Ref. [79]. The plant, which will be constructed in the near future, will use high temperature acid digestion and various hydrometallurgical processes to process approximately 34 000 t of concentrate per year. The basic chemical treatment process will be followed by further processing to produce six different rare earth oxide products. Processing is expected to produce the following residues:

(a) 25 000 t/a of flue gas desulphurization residue;
(b) 50 000 t/a of neutralization underflow solids;
(c) 31 000 t/a of water leach purification solids.

There is also potential for the buildup of scale within various types of processing equipment. Regular surveys will be undertaken in the plant to monitor any such buildup.

9.1.4. Treatment of monazite concentrate with sodium hydroxide

9.1.4.1. Main process

In past operations, rare earths have been obtained from monazite concentrate through digestion with 98% sulphuric acid at 200–220°C. The mineral is decomposed exothermically, forming a pasty mixture of sulphates and acid sulphates suspended in phosphoric acid and excess sulphuric acid. Usually, sodium sulphate is added to the clear solution to cause the selective precipitation of a double sulphate $RE_2(SO_4)_3 \cdot Na_2SO_4 \cdot 3H_2O$ of essentially light rare earths. Thorium, some of the heavy rare earths and other impurities such as iron and uranium remain in solution. However, a significant part of the thorium precipitates with the rare earth double sulphate, requiring extensive purification of the rare earth fraction to meet market requirements. For this reason, the acid digestion process has now been largely replaced by digestion with sodium hydroxide. The sodium hydroxide method is currently the preferred method and is used in most parts of the world. It has the advantage of recovering phosphate in the monazite as trisodium phosphate, a marketable by-product, as well as yielding the intermediate product rare earth chloride with a minimum number of operations. A typical representation of the sodium hydroxide process is given in Fig. 19.

The monazite feedstock is first dry ground in a ball mill to a grain size of 90% less than about 50 μm and 50% less than about 10 μm. This reduction in grain size is necessary in order to prevent the monazite grains from becoming coated with sodium hydroxide and rendering the reaction incomplete. The ground material is digested at 140–160°C for 3–9 h in a 60–70% solution of sodium hydroxide.[14] About 0.75 kg of sodium hydroxide per kilogram of monazite is required to complete the reaction, corresponding to an excess of sodium hydroxide of about 50%. This excess can be recovered and reused. The main reactions involve conversion of rare earth and thorium phosphate into hydroxides:

$$RE(PO_4) + 3NaOH \rightarrow RE(OH)_3 + Na_3PO_4$$

$$Th_3(PO_4)_4 + 12\ NaOH \rightarrow 3Th(OH)_4 + 4Na_3PO_4$$

Treatment of the reaction mass with hot water dissolves the trisodium phosphate, which is then decanted as a solution, leaving the entire thorium and rare earths content in the form of a hydroxide cake. The trisodium phosphate solution contains 99.7% of the original phosphate content, together with sodium hydroxide at a concentration of 47%. The solution is clarified and fed to vacuum evaporators to crystallize the trisodium phosphate. The crystals are separated from the dilute sodium hydroxide solution by centrifuging and are then dried in a hot air pneumatic conveyor drier in readiness for bagging as a by-product that contains 17.5–19% P_2O_5. Since lead and, to a lesser extent, uranium are soluble in

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[14] In a variation of this process, known as the 'caustic fusion' process, monazite is added to fused sodium hydroxide at 400°C.

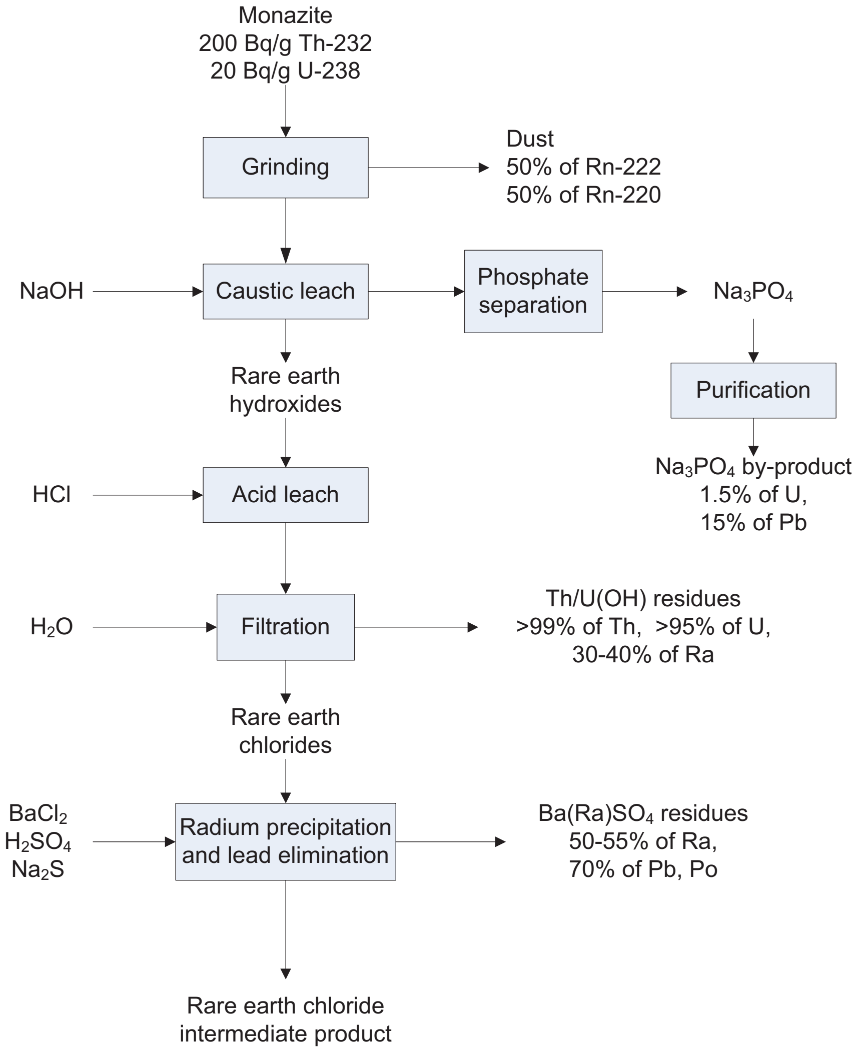

FIG. 19. The sodium hydroxide process for treatment of monazite concentrate (derived from Ref. [152]).

strong alkaline solutions, about 15% of radioactive lead isotopes (^{210}Pb and ^{212}Pb) and 1.5% of the uranium contained in the monazite end up in the trisodium phosphate. The mother liquor from the centrifuge, which is mainly unreacted caustic soda with a small percentage of P_2O_5, is evaporated to the required strength and reused in the monazite digestion process. In a variation of the process, lime may be added to the trisodium phosphate to produce a tricalcium phosphate by-product and sodium hydroxide, the latter being recycled to the monazite digestion process. The tricalcium phosphate by-product is reported in one instance to contain 1.15% thorium, 0.05% uranium, 53% tricalcium phosphate and 11.7% rare earth phosphate [153].

The hydroxide cake from the monazite digestion process contains mixed hydroxides of rare earths and thorium together with some uranium, iron, titanium and unreacted monazite. It is washed one or two more times and the leachate is decanted to render the cake free of sodium hydroxide and trisodium phosphate. After slurrying the cake in water and agitating, 30% hydrochloric acid is added while controlling the pH of the slurry at 3–4 and the temperature at 70–80°C. Owing to differences in basicity among the various consitutents, most of the rare earths (97.7%) become selectively dissolved as chlorides, leaving the other constituents (including most of the thorium) undissolved in a residue of solid hydroxides. The residual rare earths in this cake can only be made available if the cake is processed to recover thorium. In a variation of this process, leaching is carried out using nitric acid to produce rare earth nitrates. In another variation, residue from the monazite digestion process, instead of being selectively leached at a controlled pH, is dissolved completely in hydrochloric acid to form a mixed chloride solution. By gradual addition of sodium hydroxide to the chloride solution and controlling the digestion at a pH of 5.8, thorium, uranium and iron are precipitated, leaving rare earth chlorides in solution.

The rare earth chloride solution contains small amounts of impurities such as thorium, uranium, lead, iron and radium. These are removed through a ‘deactivation and lead elimination’ process. Barium chloride and sodium sulphate or sulphuric acid are added to co-precipitate lead and other heavy metals along with the barium–radium sulphate formed. Later, sodium sulphide is added in excess to precipitate lead as lead sulphide as well as thorium, uranium and iron. The combined precipitates are removed by filtration as a barium sulphate cake containing radium and lead. This residue, which is sometimes referred to as ‘mixed cake’ or ‘mesothorium cake’[15], is reported in one instance to contain 1.07% thorium, 0.076% uranium, 27% barium sulphate and 21% rare earth phosphate [153]. Modifications to the deactivation and lead elimination process, involving multiple deactivation steps and pH control, have resulted in lower radioactivity levels in the final rare earth chloride product. Multiple deactivation involves further additions of barium chloride and sodium sulphate, resulting in a larger volume of material and, consequently, lower concentrations of radionuclides in the residue as well as in the product.

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[15] Mesothorium is a name given in the past to ^{228}Ra.

The purified rare earth chloride solution is converted into an intermediate product that can be further processed to separate and purify individual rare earths. One option is for the rare earth chloride solution to be fed into a battery of steam jacketed glass lined vats where it is concentrated in stages through evaporation, flaked and packed into steel drums as the main product known as ‘mixed rare earth chloride’. This contains 46% REO and forms the starting material for the production of various rare earth products such as rare earth fluorides, carbonates and oxides, cerium oxide, and middle and heavy rare earth fractions. Another option is to subject the rare earth chloride solution to further processing in which the medium and heavy rare earths are separated from the lighter rare earths by solvent extraction or, less commonly, by ion exchange (see Section 10).

9.1.4.2. Management of residues

The hydroxide residue from the hydrochloric acid leaching process contains most of the thorium (99.3–99.99%) and uranium (95–99.7%) present in the monazite feedstock, together with other minerals such as iron, titanium and unreacted monazite. It also contains about 30–40% of the radium from the feedstock. The composition of this residue is given in Table 49. In India, the hydroxide residue is either put into storage or is treated with concentrated hydrochloric acid to produce thorium chloride solution for the subsequent extraction of thorium (together with uranium and residual rare earths). The unreacted monazite component, together with traces of rare earths, thorium, radium and uranium, is removed by filtration as an insoluble sludge. It represents

TABLE 49. COMPOSITION OF HYDROXIDE RESIDUE FROM THE HYDROCHLORIC ACID LEACHING PROCESS

	Composition (%)		
	Brazil [154]	India [90]	Malaysia [155]
Thorium (oxide or hydroxide)	22	25–28	15
Uranium (oxide or hydroxide)	0.9	0.6–0.8	0.45
Rare earths (REO)		10–12	
Chloride		4–5	
Insolubles		15–20	
Moisture		30–35	

about 8–10% of the original monazite mass, resulting in the generation of about 300 t per year from a typical plant. The sludge is neutralized and converted into a cake by mechanically operated filters, before being disposed of as a ‘monazite insolubles’ waste in engineered storage facilities in the form of concrete trenches. Trench design takes account of local conditions such as rainfall, acidity, water table, flooding and seismic activity and includes shielding to control gamma fields. After filling, the top of the trench is closed with a concrete slab. Monitoring wells are provided around the trenches to check groundwater for any migration of radioactivity.

The management of hydroxide residues in other countries, where thorium production is not (or is no longer) anticipated, is described in the following examples:

(a) In former operations in Brazil [156], the mass of hydroxide residue was typically about 2% of the original monazite mass, corresponding to an annual amount of 30 000 t during peak production in the 1950s and 1960s. Part of the residue was put into shallow ground silos in rubber drums at a controlled uranium mining and processing site and part was buried at a repository site in specially constructed concrete trenches 3 m deep;

(b) In Malaysia [155], the amount of hydroxide residue generated during the period 1982–1992 was estimated to be 12 000 t (including residue from the treatment of xenotime). The residue was kept in drums in long term storage at an approved above ground facility with concrete storage bays capable of holding more than 20 000 t of material. The future plan is to decommission this storage facility and dispose of the hydroxide residue as waste in an engineered cell at the site. The cell will have several layers, including a geochemical barrier, concrete capping and soil cover;

(c) Should operations be resumed at Steenkampskraal, South Africa to further exploit the vein type monazite deposit there, it is intended that the hydroxide residue will be initially stored at the site. At a later stage, it may be placed underground in mined-out parts of the workings or transported to a near surface low and intermediate level radioactive waste disposal facility 75 km from the mine.

Barium sulphate residue from the deactivation and lead elimination process contains a mixture of barium–radium sulphate and lead sulphide, together with traces of rare earths, thorium and uranium. The management of this residue is described in the following examples:

(a) In former operations in Brazil [156], this residue typically represented 6–7% of the original monazite mass, corresponding to an annual amount of 100 000 t during peak production in the 1950s and 1960s. It was stored in drums at a controlled uranium mining and processing site;

(b) In India [86, 91], the residue accounts for 6–10% of the original monazite mass, with an annual production of approximately 150 t. The residue is pumped into trenches lined with fibre reinforced plastic. These trenches are sealed with a concrete slab when full and surrounded by earthen shielding. Monitoring wells are provided around the trenches and groundwater samples are taken quarterly.

One proposal for a monazite processing facility in Western Australia involved the processing of 12 000 t of monazite annually [157]. A principal element of this proposal was to dispose of the residues at a low level radioactive waste repository at a remote arid site. The total amount of residue was estimated to be 6000 t per year, comprising both the hydroxide residue and the mixed cake residue. It was proposed that this would be disposed of in trenches where layers of bulk bags containing the residue would be separated by layers of clay. The filled trenches would be capped by a 5 m thick clay cover.

Solid residues are also generated during plant decommissioning. In Malaysia, 50 000 m^3 of contaminated equipment and soil had to be dealt with in the decommissioning of a monazite treatment plant [155]. The material was transported to an engineered cell at an approved disposal site. This cell included a geochemical barrier, concrete capping and a soil cover, similar to that proposed for the disposal of hydroxide residue.

It is reported from India [115] that liquid effluents from the chemical treatment of monazite comprise alkaline effluent from sodium hydroxide digestion, acidic effluent generated in subsequent steps in the process, and water and chemicals used for decontamination. About 15 m^3 of such effluents are generated per tonne of monazite processed. Settling of the alkaline effluent removes suspended particles and, with them, about 90% of the radium in the alkaline effluent stream. The acidic effluents contain suspended matter that is not easily settled out. About 50% of the radium in this effluent stream is contained in the suspended particles. The alkaline and acidic streams are then mixed thoroughly in a flash mixer, resulting in a precipitate carrying most of the remaining radionuclides. The effluent is partially neutralized with hydrochloric acid and then fed to another flash mixer to which calcium chloride is added, precipitating hydroxy apatite of calcium phosphate $Ca_{10}(PO_4)_6(OH)_2$ and calcium fluoride. The chemical reactions are:

$$6Na_3PO_4 + 2NaOH + 10CaCl_2 \rightarrow Ca_{10}(PO_4)_6(OH)_2 + 20NaCl$$

$$CaCl_2 + 2NaF \rightarrow CaF_2 + 2NaCl$$

Formation of these precipitates enables co-precipitation of residual radionuclides along with most of the phosphates and fluorides. The suspension is then fed to a further flash mixer to which a flocculating agent is added. The slurry from this mixer is fed to the flocculator compartment of a clariflocculator and allowed to settle for about 3 h. The overflow is taken to a post-treatment tank for monitoring, after which it is either discharged or sent for further treatment, depending on its quality. The annual discharge of ^{228}Ra is reported to be less than 1 GBq and is within established discharge limits [115]. The settled mass is pumped to an intermediate storage tank from where it is filtered in a pre-coat type rotary drum filter. The sludge from this, commonly referred to as effluent treatment cake, amounts to about 10% of the original monazite mass. In India, about 250 t (300 m^3) of this cake is generated annually. It is collected in HDPE laminated bags and disposed of in earthen trenches with suitable soil topping. The groundwater is sampled periodically for analysis.

During the various chemical processing stages, gases and airborne particulates, including chlorine, hydrogen sulphide and hydrochloric acid vapours, are discharged from stacks. A description of how these are controlled in Indian plants is given in Refs [86, 91, 115]. Reaction tanks and storage tanks are connected to the main ventilation system. Releases to the atmosphere are controlled using water and dilute sodium hydroxide scrubbers. The annual release of ^{232}Th varies between 1 and 125 MBq. Thoron and its progeny are also released. The mean annual release of thoron progeny is 198 GBq (range 70–360 GBq). Discharges are regularly monitored for radioactive and non-radioactive pollutants.

9.1.5. Treatment of xenotime

Xenotime is chemically processed in Malaysia mainly for the production of yttrium but is also used as a source of lanthanides, using either sulphuric acid digestion [158] or sodium hydroxide digestion [11].

In the sulphuric acid process, xenotime is first milled to the required grain size before roasting in a furnace. This is to ensure good yttrium recovery in the sulphuric acid digestion stage, in which the yttrium phosphate contained in the xenotime is converted into water soluble yttrium sulphate. Water is used as a leaching medium in the next stage to improve recovery. Yttrium is then precipitated as yttrium oxalate through the addition of oxalic acid. The final stage is the calcination of yttrium oxalate into an yttrium oxide concentrate containing 60% yttrium.

The sodium hydroxide process follows more or less the same steps as those used for the extraction of rare earths from monazite. Caustic fusion of fine ground xenotime at 400°C using sodium hydroxide is followed by the addition of water and filtration to extract trisodium phosphate and residual sodium hydroxide. The rare earth hydroxide cake is dissolved in hydrochloric acid and the insoluble residue is removed by filtration. The rare earth chloride solution is treated with oxalic acid and impurities are removed by filtration. The oxalate cake is dried and calcined to produce an yttrium oxide concentrate containing 40–60% yttrium. Management of the hydroxide residue is undertaken in the same way as the treatment of monazite (see Section 9.1.4.2).

The yttrium oxide concentrate is the starting material for further separation using solvent extraction to obtain purified yttrium and yttrium products.

9.1.6. Treatment of ion adsorption clays

A simple ore leaching with a dilute aqueous solution of sodium salts (sodium chloride, for instance) or ammonium chloride removes attached rare earths from the clays and leads to most of the rare earth content being solubilized by means of an ion exchange process [11]. The rare earths concentrate so produced contains a minimum of 90% REO, typically 95% [72]. Since leaching is quite selective, very few impurities are present in the solution, which is then used as a feed material for individual rare earth separation. A precipitation with, for instance, oxalic acid leads to the recovery of rare earths from the solution, at the same time affecting further purification of impurities. Calcination of the oxalate produces a 90% pure rare earth oxide, which then becomes the starting material for various rare earth products.

9.1.7. Treatment of loparite concentrate

Loparite concentrate produced in Kola, Russian Federation was used as a source of rare earths production in Estonia between 1970–1991 [146, 159]. Mixed light rare earths were produced as carbonates. The individual rare earths Ce, La and Nd were also separated and purified in extraction cascades and sold as fluorides. The chemical treatment of loparite concentrate is described in Refs [10, 11, 146]. It was generally carried out by dissolution in sulphuric acid or mixed acids, or by chlorination. Mention is made in Ref. [160] of various other processing routes, including those based on nitric acid.

In the sulphuric acid method, loparite concentrate is digested with 85% sulphuric acid at 150–200°C in the presence of ammonium sulphate. The treated material is leached with water and ammonium double sulphates of rare earths are filtered out to leave sulphates of titanium, niobium and tantalum in solution. The composite sulphates of rare earths and thorium are converted to carbonates and dissolved in nitric acid. The nitrate solution is then treated for separation of thorium, ^{228}Ra and other impurities by precipitation and the purified rare earth bearing solution is concentrated. The solution forms the starting material for separation of individual rare earths such as La, Nd, Pr, Sm and Eu through solvent extraction.

In the mixed acid method [146], concentrate is dissolved in a mixture of sulphuric and hydrofluoric acids (4 t of sulphuric acid and 0.55 t of 40% hydrofluoric acid per tonne of loparite concentrate). During this treatment, most of the niobium, tantalum and rare earths are dissolved, but thorium remains as insoluble fluoride in the moist thorium cake (2.5% ThO_2 equivalent) which also contains about 27% barium sulphate from ^{228}Ra co-precipitation, 12% REO, 6.8% fluorine, 3% titanium dioxide, 3.7% calcium oxide and 7.9% silica. This thorium cake was regarded as waste and disposed of as a minor component in a repository together with other waste from uranium and loparite processing and a large amount of oil shale ash from the local power plant. This moderately active waste forms the ‘upper grey layer’ of the repository profile, 5–10 m thick and containing 4 million t of material. It lies above the much more active uranium production waste containing at least a hundred times more ^{226}Ra (several kilograms), although in a very insoluble form (barium–radium sulphate). The repository has been recultivated and declared as not posing a radiological hazard. Long term storage or burial of this type of thorium residue can be regarded as an acceptable option, perhaps even for final disposal. This is supported by experimental data on leaching of thorium. The thorium content of the leachate from the repository is 5 µg/L, equivalent to a ^{232}Th concentration of 0.02 Bq/L.

In the chlorination process, finely ground loparite concentrate is treated with chlorine gas at 750–850°C using coke or coal as a reducing agent. The more volatile chlorides of niobium, tantalum and titanium are separated from the less volatile chlorides of calcium, rare earths and thorium by distillation. The ‘fusion cake’ containing chlorides of calcium, rare earths and thorium is left in the processing tank. The cake is later dissolved in water and impurities such as iron, uranium, thorium and ^{228}Ra are separated from the solution, leaving the rare earth chlorides in solution. In Ref. [148], it is reported that the fusion cake is dissolved in hot sulphuric acid in the presence of ammonium sulphate. The solution is diluted with water, precipitating double sulphates of rare earths and thorium that are converted to carbonates through the addition of sodium carbonate. The carbonates are dissolved in nitric acid and thorium is precipitated by raising solution alkalinity or through solvent extraction. The remaining rare earth nitrate solution is separated and purified via selective precipitation and solvent extraction. An analysis of the rare earth tailings reveals 1% La, 0.004% Y, 0.375% Th and 0.004% U [161].

9.2. RADIONUCLIDE ACTIVITY CONCENTRATIONS

The migration of radionuclides during the rare earths extraction process, and thus activity concentrations in the various process materials, vary according to the type of feedstock, the process used and particular process conditions. Since activity concentrations broadly reflect levels in the feedstocks, monazite feedstocks generate the most highly active residues, by-products and intermediate products. For the chemical treatment of monazite concentrate, the following general points can be made (see Ref. [152]):

(a) Practically all the thorium and most of the uranium remain as insoluble hydroxides throughout the acid leach stage and are removed by filtration. Since the hydroxide residue has a smaller mass than the monazite feedstock, activity concentrations of ^{232}Th and ^{238}U in the residue are approximately 2–3 times those of the feedstock. Some uranium may be dissolved during the caustic leach process (reported values vary from less than 1% to 19%, with an average of about 1.5%) and end up in the trisodium phosphate by-product;

(b) The amount of radium solubilized during hydrochloric acid leaching varies from 40% to 90%, with an average of 50–55%. For a plant utilizing nitric acid leaching, the dissolution of radium is more than 95%. The removal of radium in the deactivation and lead elimination process results in a residue with high activity concentrations of ^{228}Ra and ^{226}Ra;

(c) The potential for liberation of thoron and radon is significantly enhanced in the grinding and caustic leach processes and, on average, 50% is liberated along with fugitive dust;

(d) Most of the lead and polonium content (typically 70%) remains insoluble and is removed as part of the hydroxide residue. Since lead is soluble in strong alkaline solutions, some of the ^{210}Pb and ^{212}Pb (typically 15%) migrates to the trisodium phosphate by-product.

Radionuclide activity concentrations in solid process residues and by-products from the chemical treatment of various types of rare earth concentrate are given in Tables 50 and 51. Activity concentrations of ^{228}Ra in liquid effluent streams from the chemical treatment of monazite in India are shown in Table 52. The activity concentrations of gaseous releases from monazite plants in India (mostly thoron and thoron progeny) were found to be insignificant

TABLE 50. ACTIVITY CONCENTRATIONS IN RESIDUES AND BY-PRODUCTS FROM THE PROCESSING OF BASTNÄSITE, MOUNT WELD AND LOPARITE CONCENTRATES

	Activity concentration (Bq/g)[a]				Ref.
	Th-232	Ra-228	U-238	Ra-226	
Treatment of bastnäsite concentrate with hydrochloric acid					[143]
Stabilized lead–iron filter cake	1.2		42.6		
Pond lead–iron residues	1.6		57.0		
Lead sulphide concentrate	0.2		2.7		
Treatment of Mount Weld concentrate using high temperature acid digestion[b]					[79]
Flue gas desulphurization residue	0.04		0.01		
Neutralization underflow solids	0.20		0.05		
Water leach purification solids	6.8		0.3		
Treatment of loparite concentrate					
Loparite waste	15–26		1.5–2.2	1.4–1.8	[145]
Th–$BaSO_4$ residue	*89*				[146]
Th–$BaSO_4$ residue dispersed in a repository[c]	0.2–4		0.18–3.6	0.13–2.3	[145]
Th–$BaSO_4$ residue dispersed in a repository[c]	0.194–1.83 *Average 0.4*				[146]

[a] Figures in italics are derived from mass concentrations, assuming that 1 g of natural thorium contains 4057 Bq of Th-232 and 1 g of ThO_2 contains 3566 Bq of Th-232.

[b] Values are predicted from modelling, assuming that both the thorium and uranium decay chains remain in equilibrium during processing. This modelling will be confirmed after start-up by means of additional assessments, but the radiochemistry is well established and no significant changes are expected.

[c] The residue was unevenly dispersed within less active waste, mainly oil shale ash.

(less than 3.7 Bq/m^3) [86, 162]. Mixed rare earth chloride produced from the chemical treatment of monazite in India is reported to have had a ^{232}Th activity concentration of 37 Bq/g when using the original deactivation and lead elimination process. As a result of improvements to this process, the activity concentration was reduced to 0.5–1 Bq/g [163]. The ^{228}Ra concentration is reported to be 0.45 Bq/g [162].

TABLE 51. ACTIVITY CONCENTRATIONS IN RESIDUES AND BY-PRODUCTS FROM THE PROCESSING OF MONAZITE AND XENOTIME CONCENTRATES

	Activity concentration (Bq/g)[a]				Ref.
	Th-232	Ra-228	U-238	Ra-226	
Treatment of monazite concentrate with sodium hydroxide					
Tricalcium phosphate	47	23	6	3	[153]
Hydroxide residue	*785*		*94*		[154]
Hydroxide residue	*890–1000*		*60–80*		[90]
Hydroxide residue	*568*				[96]
Hydroxide residue	*487*				[164]
Hydroxide residue	*600*		*55*		[155]
Hydroxide residue, 40% water	*426*				[157]
Hydroxide residue, 10% water[b]	*637*				[157]
Monazite insolubles		500–2000			[165]
Monazite insolubles		400–1000			[115]
Barium sulphate residue, dry		~10 000			[91]
Barium sulphate residue, dry[c]		2000–5000			[91]
Barium sulphate residue	43	3150	9	450	[153]
Barium sulphate residue		1000–3000			[166]
Effluent treatment residue		25–100			[91]
Treatment of xenotime concentrate with sodium hydroxide					
Hydroxide residue		250		190	[155]
Hydroxide residue	34		130		[130]
Hydroxide residue	117.7			174.2	[167]
Hydroxide residue[d]		50		190	[128]

TABLE 51. ACTIVITY CONCENTRATIONS IN RESIDUES AND BY-PRODUCTS FROM THE PROCESSING OF MONAZITE AND XENOTIME CONCENTRATES (cont.)

	Activity concentration (Bq/g)[a]				Ref.
	Th-232	Ra-228	U-238	Ra-226	
Treatment of xenotime and monazite concentrates with sodium hydroxide					[128]
Tricalcium phosphate		0.6		0.1	
Hydroxide residue	*20*		*55*		
Barium sulphate residue	Negligible	350	Negligible	26	

[a] Figures in italics are derived from mass concentrations assuming that 1 g of natural thorium contains 4057 Bq of Th-232 and 1 g of ThO_2 contains 3566 Bq of Th-232.

[b] This lower water content was the predicted equilibrium level as a result of drying out after disposal.

[c] This particular residue is that obtained following changes in the process entailing repeated deactivation. The activity concentration is lower than the original value of 10 000 Bq/g because of the larger volume of residue generated.

[d] This residue is referred to as ‘xenotime waste’ and is assumed to be hydroxide residue.

TABLE 52. RADIUM-228 ACTIVITY CONCENTRATIONS IN LIQUID EFFLUENTS FROM THE PROCESSING OF MONAZITE

	Ra-228 activity concentration (Bq/L)	Ref.
Alkaline effluent		
Before treatment	300–400	[115]
Outflow from settling tank	0.4	[168]
Outflow from settling tank, improved settling efficiency	0.1	[168]
Acidic effluent		
Before treatment	150–200	[115]
After radium precipitation	0.0019	[168]
After radium precipitation, with deactivation process	0.0004	[168]
Combined effluent		
After precipitation	0.000044	[168]
After further treatment with lime	<0.000015	[168]

9.3. OCCUPATIONAL EXPOSURE

9.3.1. Exposure pathways

The chemical treatment of mineral concentrates gives rise to occupational exposure via external radiation and the inhalation of airborne dust and thoron progeny. Exposures are likely to be more significant when treating feedstocks with high activity concentrations, such as monazite.

In monazite processing, barium sulphate residue from the deactivation and lead elimination processes can exhibit very high radium concentrations (see Table 51), leading to the possibility of significant exposure to external gamma radiation. Air contamination occurs in the monazite grinding stage owing to the release of dust and thoron progeny. Some of the other operations in the process, such as filtration and drying of thorium concentrates and evaporation of rare earth chloride, also give rise to air contamination. Without adequate ventilation and confinement of process streams, significant thoron progeny concentrations may occur in areas of the plant involved in grinding and radium removal operations, and routine monitoring for thoron progeny may be necessary. Surface contamination results when spillages of monazite process streams settle on plant surfaces and equipment. The contamination may also be resuspended following drying and present an inhalation risk. Monazite processing presents a number of waste disposal issues, the principal ones being associated with the management of highly active waste streams and contaminated filters, piping and other processing equipment.

9.3.2. Exposure levels

9.3.2.1. Gamma

Gamma dose rates have been measured in and around plants for the chemical treatment of concentrate derived from Bayan Obo ore in China [76]. The results are provided in Table 53.

Average gamma dose rates, derived from TLD monitoring results in the chemical treatment of bastnäsite at Mountain Pass, USA, are given in Table 54. Gamma dose rates in the chemical treatment of bastnäsite are very low and similar to background levels. Gamma dose rates in the chemical treatment of monazite are given in Table 55 and are, as expected, considerably higher owing to the higher activity concentrations of the process materials. In the general plant areas, absorbed dose rates are generally less than 10 µGy/h. Higher dose rates, up to 2000 µGy/h, are observed near the radium removal circuit owing to the high activity concentration of radium in the residue and the formation of a radium rich

TABLE 53. GAMMA DOSE RATES IN THE PROCESSING OF BAYAN OBO CONCENTRATE [76]

	Absorbed dose rate (μGy/h)
Plant and adjacent storage areas	Generally 0.5–1, maximum 1.5
Central area of plants	Generally 0.2–0.6, occasionally >0.6
Slag stockpile	0.6–2
Plant and surrounding environment	0.065–1.2
Tailings pond water cover	Generally 0.65–1.2, maximum 1.3
Sorting plant	0.300
Sintering plant	0.328
Rare earth plant	0.257
Local natural background, for comparison	0.065

TABLE 54. GAMMA EXPOSURES IN THE PROCESSING OF BASTNÄSITE AT MOUNTAIN PASS, USA *(derived from Ref. [141])*

	Average dose rate (μSv/h)
Routine operations (January–April 1992)	
Worker, Pb–Fe carbonate operation	0.37
Pb sand filter area	0.42
Mill drum filter area	0.04
Roaster operator	0.21
Bastnäsite bagging operator	0.18
Bastnäsite bagging and warehouse area	0.60
Cerium dryer operator	0.31
Cerium bagging operator	0.40
Cerium bagging area	0.37
Plant superintendent	0.27

TABLE 54. GAMMA EXPOSURES IN THE PROCESSING OF BASTNÄSITE AT MOUNTAIN PASS, USA *(derived from Ref. [141])* (cont.)

	Average dose rate (μSv/h)
Stabilization and reintroduction of Pb–Fe filter cake (July 1995 to October 1996)	
Filter cake storage area	1.1
Filter cake reintroduction area	1.2
Individually monitored workers	0.06
Background, at guard house (July 1995 to October 1996)	1.0

TABLE 55. GAMMA EXPOSURES IN THE PROCESSING OF MONAZITE

	Absorbed dose rate (μGy/h)	Ref.
Monazite storage		
Monazite room	180	[169]
Monazite drying room, with thorium concentrate drums stored	180	[169]
Powdered monazite bin	40	[169]
Monazite storage area	>150	[152]
Sodium hydroxide storage		[169]
Caustic soda room	Background	
Caustic soda room, with used thorium concentrate drums stored	4	
Trisodium phosphate storage		[169]
Trisodium phosphate storage room	Background	
Trisodium phosphate storage room, rejected filter cloths stored	1	
General plant areas		[91]
Monazite grinding, NaOH treatment, Na_3PO_4 production	2 (0.5–50)	
Deactivation and lead elimination, rare earth chloride production[a]	5 (2–600)	
Filtration equipment		[169]
Moore filters	50	
Filter presses	30	

TABLE 55. GAMMA EXPOSURES IN THE PROCESSING OF MONAZITE (cont.)

	Absorbed dose rate (μGy/h)	Ref.
Deactivation and lead elimination		
Radium removal circuit	Up to 1000	[152]
Radium removal circuit	30–2000[b]	[152]
Barium sulphate residue	500–1000	[152]
Barium sulphate residue	400–600	[91]
Rare earth chloride production		[169]
Evaporators	25	
Evaporators, without charge	Background	
Drum of product	50	
Hydroxide residue and thorium concentrate		
$Th(OH)_4$ drying room, ~100 drums of wet cake stored	80	[169]
Thorium concentrate stores	250–350	[169]
Thorium–uranium residue, after ingrowth of Ra-228	300	[152]
Monazite insolubles		
Bag	60–100	[91]
Bag, in contact	100–150	[165]
Effluent treatment cake	<2–3	[91, 115]

[a] Also includes thorium extraction and thorium oxalates production.

[b] The upper value was registered close to the filtration unit where radium sludge is separated, reflecting a buildup of a radium rich crust inside the unit.

crust in the filtration unit. Dose rates of up to 350 μGy/h are found in storage areas of high activity process materials such as hydroxide residue.

9.3.2.2. Dust

Exposure to airborne dust in the chemical treatment of mineral concentrates is mainly of concern in the treatment of monazite, because of the relatively high activity concentrations in the process materials involved. However, the dust levels in monazite treatment plants are generally moderate, at least in more recent years. In monazite plants in India, mass concentrations of respirable dust are reported to be 0.03–2.5 mg/m^3, with an average of 0.3 mg/m^3 [91]. Information on particle size distribution in Indian plants is given in Table 56, giving an average AMAD of 5 µm. Data on ^{232}Th activity concentrations in air are given in Table 57. All the data are from Indian monazite plants except for the data for a Brazilian plant given in Ref. [170]. Some of the dust activity concentrations, particularly those reported several years ago, are very high. Opportunities for reducing airborne dust activity concentrations through engineering measures and better housekeeping were identified at that time [169], leading to generally lower concentrations in more recent years.

TABLE 56. PARTICLE SIZE DISTRIBUTION OF AIRBORNE DUST IN AN INDIAN MONAZITE PROCESSING PLANT [171]

	AMAD (µm)	GSD
Individual production areas		
Initial process area including ball mill	2.1–9.10	3.2–3.92
Extraction area	3.40–5.30	3.29–3.4
Deactivation area	3.05	6.44
Rare earth and $Th(OH)_4$ filters	3.20–4.06	3.33–6.20
Mean (rounded)	5	4

TABLE 57. AIRBORNE DUST ACTIVITY CONCENTRATIONS IN MONAZITE TREATMENT PLANTS

	Th-232 activity concentration (mBq/m^3)	Ref.
Initial process areas	64–74	[171]
Monazite store, 20 t stock	480	[169]
Monazite, 100 t stock	200	[169]
Monazite drying room	100	[169]
Monazite ball milling	20	[169]
Monazite milling	0.2–4	[170]
Monazite powder storage bin	300	[169]

TABLE 57. AIRBORNE DUST ACTIVITY CONCENTRATIONS IN MONAZITE TREATMENT PLANTS (cont.)

	Th-232 activity concentration (mBq/m^3)	Ref.
Automatic monazite balance	1	[169]
Automatic monazite balance, unfavourable wind conditions	18 600	[169]
Monazite grinding, NaOH treatment, Na_3PO_4 production	50 (2–220)	[91]
Extraction area	90	[171]
Moore filter	40	[169]
Deactivation area	100	[171]
Deactivation, lead elimination, rare earth chloride production[a]	60 (2–590)	[91]
Barium sulphate cake filter	103	[171]
Mixed hydroxide area	81	[171]
$Th(OH)_4$ filter press	3500	[169]
$Th(OH)_4$ drying room	9210	[169]
$Th(OH)_4$ drying room, dusty condition	25 090	[169]
Rare earth chloride evaporators	35	[171]
Non-production areas		[169]
Plant office	15	
General stores	26	
Chemistry laboratory	4	
Within plant, 15 year period	40 (1–2600)	[36]
Local population, for comparison	0.1 (<0.03–0.2)	[36]

[a] Also includes thorium extraction and thorium oxalate production.

There is little information on airborne dust exposures in plants treating mineral concentrates other than monazite. In the chemical treatment of bastnäsite at Mountain Pass, USA, airborne dust activity concentrations were measured as part of a project to stabilize the lead–iron filter cake and reintroduce it into the process stream instead of continuing to treat it as waste (see Section 9.1.2) [141]. Average airborne gross alpha activity concentrations associated with the storage of filter cake and its reintroduction into the process stream were 1.2 and

8.9 mBq/m^3, respectively. The background concentration (at the guard house) was 1.6 mBq/m^3. In support of dust monitoring, urine analysis was conducted during both periods and results were all below the lower limits of detection of 5 µg/L for uranium and 0.04 Bq/L for ^{226}Ra.

9.3.2.3. Thoron and thoron progeny

Thoron progeny concentrations have been measured in plants for the treatment of Bayan Obo ore in China [76]. The PAECs were found to be in the range of 25–500 MeV/L (0.020 µJ/m^3).

Airborne concentrations of thoron and thoron progeny measured in monazite treatment plants in India are given in Table 58. Values for individual contributions of ^{212}Pb and ^{212}Bi to the PAEC have been derived from activity concentrations reported for these radionuclides in Ref. [68], which were determined from air samples collected at a breathing zone height of 1.5 m. The conversion from activity concentration to PAEC as given in Section 3.4.1. Ref. [68] also provides data on equilibrium conditions between thoron and its progeny. Within the plant, the mean ^{212}Bi:^{212}Pb activity concentration ratio was 0.20 (range 0.11–0.37) and the mean equilibrium factor was 0.003 (range 0.002–0.007). In the open air outside the plant, the mean activity concentration ratio was 0.55 (range 0.24–0.84) and the mean equilibrium factor was 0.02 (range 0.006–0.054). Thus, while thoron activity inside the plant was much higher than outside, the degree of equilibrium between thoron and its progeny was significantly lower.

The data in Table 58 indicate that the highest thoron gas concentrations are found in areas where monazite and hydroxide residue are stored. A similar picture emerges for thoron progeny. The PAECs are typically 0.1–6 µJ/m^3 in general plant areas, somewhat higher in radium removal sections and much higher (20–60 µJ/m^3) in areas in which substantial amounts of high activity materials are kept and to which access would be controlled.

9.3.3. Effective dose

9.3.3.1. External radiation

Annual effective doses received by workers in the processing of concentrate derived from Bayan Obo ore, given in Table 59, are all less than 1 mSv, as would be expected from the low activity concentrations of the process materials [76].

As pointed out in Section 9.3.2.1, external exposure is only likely to be of concern during the chemical treatment of monazite, where absorbed dose rates in general plant areas can be up to 10 µGy/h. For a 2000 h annual exposure period,

TABLE 58. THORON AND THORON PROGENY CONCENTRATIONS IN INDIAN MONAZITE TREATMENT PLANTS

	Thoron gas concentration (Bq/m^3)	Thoron progeny PAEC (μJ/m^3)	Ref.
Monazite store, 20 t stock	7800		[169]
Monazite store, 100 t stock	52 500		[169]
Monazite drying room	4800		[169]
Monazite ball milling	5900		[169]
Monazite powder storage bin	5900		[169]
Automatic monazite balance	40		[169]
Monazite grinding, NaOH treatment, Na_3PO_4 production		4 (0.1–6)	[91]
Moore filter	3000		[169]
Deactivation, lead elimination, rare earth chloride production[a]		7 (0.5–37)	[91]
$Th(OH)_4$ filter press	11 000		[169]
$Th(OH)_4$ drying room	38 100		[169]
$Th(OH)_4$ drying room, dusty condition	34 400		[169]
Contaminated areas and bulk storage areas to which access is controlled		20–60	[110]
General plant areas		0.4–6	[65]
General plant areas	5000 (2000–50 000)		[91]
General plant areas		0.5–4	[110]
General plant areas, including thorium hydroxide storage	1200–43 500 000	Pb-212: 0.76–4700 Bi-212: 0.02–120	[68]
Non-production areas			[169]
Plant office	400		
General stores	70		
Chemistry laboratory	70		
Open air outside plant, for comparison	56.4–448	Pb-212: 2–20 Bi-212: 0.6–9	[68]

[a] Also includes thorium extraction and thorium oxalates production.

TABLE 59. DOSES FROM EXPOSURE TO EXTERNAL RADIATION RECEIVED BY WORKERS IN THE PROCESSING OF BAYAN OBO CONCENTRATE [76]

	Annual effective dose (mSv)
Sorting plant	0.36
Sintering plant	0.41
Rare earth plant	0.29
Ferrous slag dump	0.33
Slag brick manufacture	0.61
Tailings pond area	0.28

this would imply annual effective doses of up to 20 mSv. Although much higher dose rates are observed in mineral storage areas and near radium removal circuits, the annual occupancy periods in these areas are likely to be relatively short. A review of data on the annual effective doses received by workers in monazite processing plants prior to 1993 is provided in Ref. [152]. The results are summarized in Table 60. In plants in France and Malaysia, job rotation schemes appeared to be in place for tasks involving the highest exposures, implying that without such schemes the doses would have been considerably higher. A more recent review of the situation in India between 1995 and 2003 [110] suggests much lower doses. It is stated that, typically, external and internal exposures contribute equally to total dose. On this basis, the annual effective dose from external exposure was 1.5–4.5 mSv.

9.3.3.2. Airborne dust

As pointed out in Section 9.3.2.2, exposure to airborne dust is only likely to be of concern in the chemical treatment of monazite. In the treatment process, compounds other than oxides and hydroxides of thorium (lung absorption class S) are found in workplace air, and for this reason lung absorption class M is generally assumed for dose calculation purposes in the absence of more specific information [171]. Data on effective doses received by workers from exposure to airborne dust in monazite plants are given in Table 61.

TABLE 60. DOSES RECEIVED BY WORKERS FROM EXPOSURE TO EXTERNAL RADIATION IN MONAZITE PROCESSING PLANTS PRIOR TO 1993

	Annual effective dose (mSv)	Ref.
Brazil		[172]
Sodium hydroxide treatment	8	
Filter press	10.5	
Brazil, 1973–1984		[173]
Average in ‘hot’ section of plant	14	
Maximum	<50	
France		[152]
Mean	5–10	
Monazite handling and debagging, radium removal operations (<15% of plant workers)	15–30	
India, based on a 3 week film badge survey reported in 1963		[169]
Typical (57 of 87 workers surveyed)	<8	
Maximum (2 store workers handling Th concentrate and rare earth chloride drums)	>30	
India, survey reported in 1969		[174]
Typical (70% of workers)	<10	
Maximum	<30	
Malaysia, film badge monitoring in 1987		[152]
Typical (>75% of plant workers)	<5	
Transport of $Th(OH)_4$, average	5–7.5	
Transport of $Th(OH)_4$, maximum (3 out of 159 plant workers)	10–12.5	
USA, averages in ‘restricted areas’ in 2 plants	10–15	[152]

9.3.3.3. Thoron progeny

On the basis that thoron progeny PAECs in the production areas of monazite plants are typically 0.1–6 μJ/m^3 (see Section 9.3.2.3), an exposure period of 2000 h a year would imply an annual effective dose of a few

TABLE 61. DOSES RECEIVED BY WORKERS FROM EXPOSURE TO AIRBORNE DUST IN MONAZITE PROCESSING PLANTS

	Annual effective dose (mSv)
India, assuming no respiratory protection [103]	1–5
General review 1993 [152]	May exceed 5
India [171]	2–3
France, 127 plant workers [23]	
Average	1.5
Maximum	>20

millisieverts. This is consistent with the statement in Ref. [152] that the dose is not expected to exceed 5 mSv.

9.3.3.4. Total effective dose

The chemical treatment of monazite is, of course, the most likely source of significant doses and some dose assessments for this have been reported. The results of dose assessments in an Indian monazite treatment plant are reported in Ref. [91]. In the early 1960s, the annual average effective dose received by workers was assessed to be 15–17 mSv. This was followed by a significant overall reduction in doses over the next few years as a result of process modifications, decommissioning of old plants and commissioning of new plants. The downward trend was, however, interrupted by some temporary increases in dose as a result of the introduction of new process streams, the generation of additional quantities of monazite waste and the accumulation of relatively highly radioactive material on the premises owing to a lack of storage space [109]. Increases in internal exposures, which accounted for 50–70% of the total dose, had resulted from increases in airborne activity concentrations caused by a buildup of contamination in plants and on equipment, a reduction in available ventilation due to the introduction of newer processing equipment and the proximity of thorium storage silos. In more recent years (1995–2003), the average dose has remained within the range of 3–9 mSv per year [110] and after 1996 no worker received a dose of more than 30 mSv in a year [91].

It is reported in Ref. [79] that dose modelling calculations regarding the future chemical treatment of mineral concentrate from Mount Weld, Australia, predict that external gamma radiation will be the dominant exposure pathway, contributing 71% of the total dose, with exposure to thoron contributing the remaining 29%. The maximum annual effective doses received by process operators are expected to be: 4.1 mSv (concentrate handling), 2.8 mSv (kiln feed), 1.3 mSv (oversize handling) and 0.8 mSv (water leach purification solids filtration). A machine operator involved in waste solids handling is predicted to receive a maximum annual effective dose of 2.2 mSv. The assessment indicates that the highest annual doses, potentially up to 6.1 mSv, may be received by workers handling water leach purification solids. The actual doses received by workers are likely to be lower than those predicted, owing to the conservative approach adopted in dose modelling.

In the chemical treatment of bastnäsite concentrate at Mountain Pass, USA, reported gamma dose rates are 0.04–0.6 μSv/h in general plant areas and 0.06–1.2 μSv/h near lead–iron residues. If the respective annual exposure periods are assumed to be 400 and 100 h, the corresponding annual effective doses from external radiation are approximately 0.02–0.2 and 0.01–0.1 mSv. The bastnäsite concentrate contains predominantly ^{232}Th series radionuclides at an activity concentration of about 4 Bq/g, while the lead–iron residue contains predominantly ^{238}U series radionuclides at an activity concentration of about 50 Bq/g. In both cases, the dose coefficient for lung class M and an AMAD of 5 μm, expressed in terms of the activity concentration of the relevant head-of-chain radionuclide, is in of the order of 50 μSv/Bq (see Section 3.3.2.1). Assuming a dust concentration of 1 mg/m^3 and annual exposure periods of 2000 h for the feedstock and 100 h for the lead–iron residue, the annual effective dose from dust inhalation is in both cases approximately 0.5 mSv. Thus the total annual effective dose is expected to be approximately 0.6 mSv, irrespective of whether the source of exposure is feedstock or lead–iron residue.

For the chemical treatment of loparite concentrate, no dose assessments have been reported, but it is possible to make some rough calculations based on radionuclide activity concentrations reported in Section 9.2. The ^{232}Th activity concentration in loparite concentrate is approximately 20 Bq/g, five times that of the bastnäsite concentrate from Mountain Pass referred to above. Therefore the total annual effective dose received by workers exposed to the concentrate is also expected to be about five times higher, that is, about 3 mSv. Residues from the chemical treatment of loparite have ^{232}Th activity concentrations of 20–90 Bq/g, similar to that of lead–iron residue from the processing of Mountain Pass bastnäsite. This implies a similar effective dose, that is, about 0.6 mSv.

No information is available on the annual effective doses received by workers in the processing of mineral concentrates from bastnäsite–monazite ore from Bayan Obo, China or on rare earth clays from southern China, but the doses can be expected to be of little or no concern because of the low radioactivity content of the ores concerned.

9.3.4. Monitoring

In an Indian monazite treatment plant [91], monitoring for external exposure is carried out by means of area monitoring (using portable and installed gamma monitors) and quarterly personal monitoring using TLDs. Monitoring for internal exposure due to dust inhalation is carried out by means of ambient air dosimetry involving measurements of airborne activity concentration, particle size, particle solubility and occupancy periods. Personal air sampling and analysis is also used. Estimates of intakes are complemented and validated by whole body counting, urine analysis and thoron in breath measurements. Internal exposure due to thoron inhalation is monitored by personal air sampling and analysis for thoron and thoron progeny. Contamination on surfaces is monitored using portable survey meters, contamination monitors and/or swipes. Contamination of body and clothing is monitored by hand, foot and clothing monitors.

9.3.5. Measures to reduce doses

The adoption of suitable measures for reducing doses is particularly important in the chemical treatment of monazite, since this is where doses received by workers are highest and will need to be controlled. The measures generally applied in monazite treatment operations are described in Section 9.3.5.1 to 9.3.5.7 and are in some respects similar to those applied in the secondary stage of the beneficiation of heavy-mineral sand (see Section 6.3.6). The adoption of similar measures in the chemical treatment of other mineral concentrates may also be worth considering since some of them, especially those of an administrative nature, may provide significant benefits at a relatively low cost.

9.3.5.1. Adminstrative measures

As is the case for the dry separation of heavy-mineral concentrates, unnecessary exposures may be avoided if employees are reminded of the presence of radioactive minerals through demarcation of areas of higher dose rate with appropriate floor markings and caution signs. For monazite treatment plants, this is especially important because of the presence of a greater variety and amount of process materials with relatively high activity concentrations. For areas where high gamma dose rates can occur, such as areas where significant quantities of monazite concentrate and the more highly active residues (hydroxides, monazite insolubles, barium sulphate) are stored, access controls, special work permits and restrictions on occupancy periods should be applied. In some cases, consideration is given to job rotation between plants or locations within plants with relatively high radiation levels to those where levels are low. High standards of industrial hygiene, including restrictions on eating, drinking and smoking in certain areas, and maintaining an appropriate standard of general housekeeping, are essential for controlling intake by workers.

Since there is always the possibility of high gamma dose rates and high airborne dust activity concentrations at certain locations and times, vigilance is needed in ensuring that individual and workplace monitoring programmes for external and internal exposure are implemented effectively, that exposure records are maintained, and that periodic health surveillance is carried out. It has been found that widely varying levels of exposure for identical tasks can be avoided through the provision of in-house, standardized and practical work training programmes, including retraining. Job hazard evaluation, job planning and 'dry runs' are important, especially for work of a less routine nature such as maintenance and decommissioning. Other measures found to be important are supervision and other means of improving work practices (to ensure, in particular, the enforcement of administrative and engineering controls), promotion of a safety culture, work standardization, documented procedures at all levels and ISO certification.

9.3.5.2. Process considerations

Significant reductions in dose amounts have been achieved through the introduction of process modifications involving greater mechanization and automation, such as:

(a) The replacement of manual handling of monazite by a silo and conveyor system;
(b) The replacement of manually operated filter presses (plate and frame filters) by rotary drum filtration;
(c) The introduction of remote handling equipment for process material with high activity concentrations, for example:

    (i) The introduction of a conveyor system for filling large HDPE bags with insoluble monazite residue and the use of a fork lift to transport them to the disposal site;
    (ii) The direct pumping of barium sulphate residue to the disposal trenches.

As is the case for the dry separation of heavy-mineral concentrates, the periods of time spent by workers close to process materials can be reduced by the use of systems for remote plant monitoring and process control. Worker exposures can also be reduced through the use of automatic on-stream analysis for product quality control rather than relying on manual mineral sampling.

9.3.5.3. Plant layout and design

The variety and amounts of process materials with high activity concentrations within a plant can have significant implications for doses received by workers. The segregation and separation of these materials from regularly occupied workplaces is therefore especially important in the design and layout of a plant. This involves:

(a) The isolation of more highly active process streams;
(b) The segregation of processing equipment involving more highly active material;
(c) Designated storage for products and wastes;
(d) Storage of more highly active materials in leakproof trenches and silos.

9.3.5.4. Engineering measures to control airborne dust and thoron levels

Airborne dust concentrations need to be maintained at low levels (ideally below about 0.1 mg/m^3) in plant areas in which dry materials with high activity concentrations are handled, such as those where monazite grinding and radium removal operations are carried out. This is achieved through the confinement of process streams and the use of adequate ventilation. Reaction vessels are enclosed and connected to ventilation systems provided for each process building and gases and vapours pass through sodium hydroxide scrubbers before being discharged through stacks. The areas around dust generating equipment are enclosed and normally not occupied by workers. Enclosures are fitted with mechanical exhaust ventilation. Access, when needed, is obtained through double door airlocks. General ventilation in plants is augmented by wall mounted exhaust fans and installed air circulators. Air changes in plants vary from 1 to 8 per hour. Good general workplace ventilation is effective in reducing the accumulation of thoron gas.

The design and manufacture of material handling equipment to relevant standards, with due regard given to facilitating access, minimizes manual handling and prevents the liberation of dust at material transfer points.

9.3.5.5. Engineering measures for controlling gamma radiation

More highly active process streams and associated equipment are shielded by walls or enclosures where necessary.

9.3.5.6. Surface contamination control

Various control measures are applied to avoid the buildup and spread of surface contamination by materials with high activity concentrations. These measures include the following:

(a) The use of bunded areas to reduce the release of contamination;
(b) Regular decontamination of floor and equipment surfaces by cleaning and regular housekeeping campaigns to reduce the spread of contamination;
(c) The use of non-absorbent surface finishes, such as epoxy paint, and screeding of floors and other surfaces using resins to facilitate decontamination;
(d) The prevention of spills of highly active material;
(e) The prompt removal of spills and other accumulations of highly active material on surfaces within a plant;
(f) The use of suitable protective clothing, such as overalls, gloves and head and shoe covers to prevent skin contamination resulting from process spillages and the plate out of thoron progeny;
(g) The use of protective footwear barriers and decontamination facilities.

9.3.5.7. Respiratory protection

Respiratory protection is used in some plant areas and for some particular tasks when administrative and engineering controls are not sufficient to achieve acceptable conditions.

9.4. PUBLIC EXPOSURE

Some data on public exposure associated with the treatment of Bayan Obo ore are reported in Ref. [76]. In the vicinity of the treatment plant, the topsoil is contaminated by dust blown from the tailings pond. The ^{232}Th activity concentrations in the soil are generally 0.08–0.2 Bq/g, but are more than 0.4 Bq/g near the tailings pond. Gamma dose rates of 0.085–0.15 μGy/h are reported. In the nearby city area of Bayan Obo, thoron progeny PAECs were found to be 69–125.6 MeV/L (0.011–0.02 μJ/m^3), with an average value of 94 MeV/L (0.015 μJ/m^3). Incremental annual effective dose from gamma radiation near the tailings pond was reported to be 0.043 mSv, leading to the conclusion that the radiological impact on members of the public was not significant.

Exposure of members of the public to the chemical treatment of monazite has been identified as being more of an issue, because of the higher activity concentrations of the various process materials. The main exposure pathways to members of the public arise from the storage and disposal of solid residues and the discharge of liquid and gaseous effluents. Investigations of these exposure pathways have been conducted at a plant in India in which, in addition to the chemical processing of monazite, further processing is carried out to extract individual rare earth products. Concerns about public exposure have also arisen as a result of legacy sites.

9.4.1. Disposal and storage of solid residues

In a general review of exposures in various NORM industries [175], it was concluded that annual effective doses received by members of the public from the shallow land burial of waste from monazite processing were less than 0.25 mSv. This conclusion is supported by the results of investigations conducted at the monazite processing plant in India. Gamma dose rates measured over sealed shallow land burial sites (trenches and silos) are reported in Table 62. The dose rates are comparable to those in local natural high radiation backround areas. Annual effective doses from gamma radiation received by members of the public, as determined from environmental monitoring at the plant boundary, and in some of the inhabited areas adjacent to the plant boundary, were found to be 0.24–1.8 mSv [115, 165]. These doses are comparable with those from natural background radiation as determined by gamma radiation monitoring at greater distances from the plant, namely 0.08–1.48 mSv up to 15 km away [115] and 0.2–1.4 mSv up to 35 km away [165]. Groundwater monitoring around trenches and silos between 1991–2000 [165] gave ^{228}Ra concentrations of 0.01–1.60 Bq/L. Annual mean values were in the range 0.07–0.87 Bq/L, with no clear trend over time. It was concluded that groundwater samples did not show activity

TABLE 62. GAMMA EXPOSURES ABOVE STORAGE AND DISPOSAL SITES AT A MONAZITE TREATMENT PLANT

Solid residue	Type of containment	Absorbed dose rate (μGy/h)	Ref.
Barium sulphate cake	Contained in PVC lined concrete casks in concrete trenches, or pumped directly to FRP lined concrete trenches	3–5 2–5	[165] [115]
Monazite insolubles	Contained in HDPE bags in concrete trenches	2–4	[165]
Effluent treatment cake	Earthen trenches with soil topping	0.5–1.0	[115]
Thorium hydroxide	Storage in 30–90 cm thick concrete silos	Local natural background level	[165]

enhancement over the period, thus confirming the integrity of the trenches and silos. It was further stated in Ref. [165] that the progeny of thoron and radon emanated from thorium hydroxide residue decay within the storage silos, thus avoiding any airborne contamination.

9.4.2. Discharge of liquid effluent

Treated liquid effluent from a monazite processing plant in India is discharged into a nearby river. A series of investigations has been conducted over many years to determine the radiological impact of these discharges [91, 168, 176, 177]. Variations in ^{228}Ra activity concentrations in the river water over time and distance are shown in Table 63. Concentrations near the discharge point are significantly elevated, but less so in the two most recent periods (1980–1988 and 1996–2000) compared to concentrations in earlier years. This is attributed to improvements in effluent management practices — the effluent treatment plant at the monazite processing plant became operational in 1980. The ^{228}Ra measurements conducted in 1974–1975 and 1977–1978 were made during various times of the year and show a strong seasonal variation, although in all cases the increase in concentration in the vicinity of the discharge point remained significant. The impact of the release of ^{228}Ra is observed mainly downstream of the discharge point, but tidal movements in the pre- and non-monsoon seasons transport radioactivity in the upstream direction. The average thorium levels in the river water for the period 1980–1988 are reported in Ref. [177]. Activity concentrations of ^{232}Th (derived from the reported mass concentrations of thorium) were found to be at background levels (0.0001–0.0002 Bq/L) at all sampling locations except at the discharge point and 2 km downstream, where the levels were 0.004 and 0.0005 Bq/L, respectively.

Corresponding variations of ^{228}Ra levels in river sediment are shown in Table 64. Again, the impact of the release of ^{228}Ra is evident, including some impact at upstream locations during pre- and non-monsoon seasons. Because of the large variability of the data over time, it is not possible to identify any consistent reduction in ^{228}Ra levels as a result of the commissioning of the effluent treatment system in 1980. The average activity concentration of ^{232}Th in sediment at the discharge point (derived from the reported mass concentration of thorium) was 0.022 Bq/g, compared with a background level of 0.03 Bq/g measured 9 km upstream [177]. The ^{232}Th activity concentrations in sediment 2 km upstream and downstream were only slightly above background levels, at 0.004 and 0.005 Bq/g, respectively.

TABLE 63. CONTAMINATION OF RIVER WATER BY AQUEOUS DISCHARGES FROM A MONAZITE TREATMENT AND RARE EARTHS EXTRACTION PLANT [91, 177]

Zone	Distance from discharge (km)[a]	Ra-228 activity concentration (Bq/L)					
		1974–1975		1977–1978		1980–1988 Mean	1996–2000 Mean
		Pre-monsoon	Post-monsoon	Monsoon	Non-monsoon		
Background	–28	0.11	—	—	—	Average for zone 0.03	0.01
	–15	—	—	—	—		0.01
	–9	0.20	<0.01	0.06	0.11		0.025
	–5	0.17	<0.01	0.03	—		—
Industrial	–2	0.50	<0.01	0.01	0.32	Average for zone 0.05	—
	0	0.19	1.67	0.15	0.76		—
	+0.3	—	—	—	—		0.062
	+2	0.18	0.11	0.11	0.65		0.035
Intertidal estuarine	+5	0.17	<0.01	0.02	0.15	Average for zone 0.02	0.03
	+14	—	—	—	—		—
Coastal estuarine	+17	—	—	—	—	Average for zone 0.01	—
	+22	—	—	—	–		—

[a] Measured in the downstream direction.

TABLE 64. CONTAMINATION OF RIVER SEDIMENT BY AQUEOUS DISCHARGES FROM A MONAZITE TREATMENT AND RARE EARTHS EXTRACTION PLANT [91, 177]

Zone	Distance from discharge (km)[a]	Ra-228 activity concentration (Bq/g)					
		1974–5		1977–8		1980–1988 Mean	1996–2000 Mean
		Pre-monsoon	Post-monsoon	Monsoon	Non-monsoon		
Background	–28	0.02	—	—	—	Average for zone 0.07	0.162
	–15	—	—	—	—		0.141
	–9	0.06	0.07	0.05	0.11		0.165
	–5	—	—	0.12	0.04		—
Industrial	–2	0.04	0.03	0.06	0.06	Average for zone 0.13	—
	0	20.57	19.09	0.24	4.31		—
	+0.3	—	—	—	—		0.747
	+2	0.74	0.58	0.24	0.17		0.430
Intertidal estuarine	+5	0.04	0.04	0.14	0.06	Average for zone 0.10	0.216
	+14	—	—	—	—		—
Coastal estuarine	+17	—	—	—	—	Average for zone 0.05	—
	+22	—	—	—	—		—

[a] Measured in the downstream direction.

The results of biological uptake and food chain studies are reported in Ref. [178]. The levels of ^{228}Ra in fish caught from the river were found to be 0.01 Bq/g in flesh and 0.1 Bq/g in bone. On the basis of fish consumption and the use of the water for irrigation, the annual effective dose per capita was determined to be 0.5 μSv for the population residing within 3 km of the plant. It was concluded, therefore, that, as a result of improved effluent treatment (annual ^{228}Ra discharges were reduced from 200 GBq in 1971 to 0.3 GBq in 1996), the environmental impact of liquid discharges from monazite processing operations had fallen to insignificant levels.

9.4.3. Airborne discharges

Thoron and its progeny, as well as thorium containing particles, are discharged into the atmosphere from monazite treatment plants. From a public exposure point of view, only thoron progeny are of any significance. At a monazite treatment plant in India, thoron progeny concentrations were found to be 0.01–0.2 μJ/m^3 within 300 m of the plant boundary and at background levels beyond this point [91]. Only a small number of individuals reside in the region of elevated thoron progeny concentrations because the area is mainly an industrial zone. The annual effective dose received by these individuals was estimated to be 10–30 μSv [115].

9.5. DECOMMISSIONING OF MONAZITE TREATMENT PLANTS

9.5.1. Udyogamandal, India

Experience gained in the decommissioning of this plant is described in Ref. [179]. The plant was in continuous operation between 1952 and 1988. It was decommissioned over a nine month period during 1990–1991. Operations at the plant over the years had resulted in a buildup of activity on equipment surfaces, floors, pipes, walls and associated structures. The reaction tanks had accumulated elevated levels of activity as a result of impregnation of radionuclides into the rubber linings and pitting of surfaces due to chemical corrosion. This was observed particularly in the tanks used for deactivation of rare earth chloride, for which the absorbed dose rate on the internal surfaces of the tanks having risen from less than 200 μGy/h in 1970 to nearly 1200 μGy/h in 1989. The activity was mainly associated with ^{228}Ra and its progeny. The activity concentration of ^{228}Ra in the rubber linings was 55 500 Bq/g, considerably higher than the value of 13 000 for the Ba(Ra)SO$_4$ produced in the deactivation process. Gamma exposure levels at the time of decommissioning are shown in Table 65.

TABLE 65. GAMMA EXPOSURE LEVELS IN A RARE EARTH PLANT PRIOR TO DECOMMISSIONING

	Absorbed dose rate (μGy/h)	
	General working area	On equipment
Entrance	50	—
Deactivation tanks	800–1200	1000–2500 (outside) Up to 5000 (inside)
Extraction tanks	50	30–60
Weak chloride tanks	—	100
Wooden tanks	50–100	200–250
Central passage	300–500	—
Thorium slurry tanks	60	—
Extraction platform	30	—
Deactivation platform	800–1000	—
Thorium press	10–40 (floor)	20–50
Mixed cake ($PbS + Ba(Ra)SO_4$) press	30–200	100–400
Wooden tank platform	50–100	—
Washing machine	50	100
Slurry pipes	—	5–100
Switch panels	—	5–10
Motors and pumps	—	5–600
Moore filter	30–70	30–60
Thorium tank	—	150

The activity concentrations of airborne dust and thoron progeny prior to decommissioning are shown in Table 66. Average thoron progeny activity in the thorium filter press area increased from about 6 μJ/m^3 in 1983 to about 37 μJ/m^3 in 1987–1988.

TABLE 66. DUST AND THORON PROGENY EXPOSURE LEVELS IN A RARE EARTH PLANT PRIOR TO DECOMMISSIONING

	Th-232 activity concentration in airborne dust (mBq/m^3)	Thoron progeny PAEC (μJ/m^3)
Thorium concentrate press floor	34	36.7
Mixed cake press floor	12	26.7
Deactivation section	15	9.0
Thorium concentrate drying area	58	13.5
Mixed cake cask filling area	52	6.2
Extraction section	12	5.2
Wooden tanks area	24	24.8
Washing machine	8	58.3
Moore filters	23	4.2

The sequence of decommissioning operations was as follows:

(i) Sludge was removed from pipes, vessels, pits and drains by pumping or by manual recovery into polyethylene lined concrete casks in readiness for disposal;

(ii) The rubber linings of the extraction and deactivation tanks were not amenable to decontamination. Instead, the tanks were cut open from the outside with gas cutting torches and the linings were removed using scraping tools with long handles. This prevented workers having to enter the tanks, where gamma exposure levels were very high. The lining fragments were transferred to concrete casks for disposal. Sections of the tanks and other contaminated metallic items were decontaminated to the extent possible through garnet sand blasting in ventilated booths with exhaust air scrubbing. The workers engaged in these activities were equipped with full protective clothing and compressed air line respirators and were given suitable instructions and training;

(iii) Items with fixed contamination such as wooden tanks, plates and frames of filter presses, wooden platforms, Moore filter assemblies, agitators and supporting structures and pipes were, as necessary, dismantled, cut into small pieces and cleaned to remove any loose surface contamination prior to disposal;

(iv) Pumps, electric motors and other electrical equipment were decontaminated and salvaged for reuse after being checked for residual contamination;

(v) Contaminated floors and walls were scrubbed and washed to remove loose contamination. The remaining fixed contamination was then removed by chipping away the plaster layer. The contaminated chippings were collected in HDPE bags for disposal;

(vi) The buildings, after the removal of equipment and contaminated floor and wall surfaces, were demolished. Structural components and rubble were salvaged or disposed of as non-radioactive waste.

Disposal methods for the various wastes are summarized in Table 67. The experience gained in the decommissioning operations demonstrated that the amounts of contaminated material requiring disposal as waste could be significantly and cost-effectively reduced through decontamination of the building structures by removing the plaster layers from floors and walls and through decontamination of metal surfaces by sand blasting. In the case of metallic items, waste volume was reduced by a factor of 10.

Exposure levels for airborne dust and thoron progeny are shown in Table 68.

Total effective doses received by workers from external and internal exposures over the duration of decommissioning operations were assessed to be in the range of 0.20–8.94 mSv, with a mean value of 7.2 mSv. After the completion of decommissioning operations, underlying groundwater was monitored for a number of years to check for contamination from the waste disposal sites. The ^{228}Ra concentrations in the groundwater were very low (<0.001–0.004 Bq/L).

9.5.2. Santo Amaro, Brazil

Experience gained in the decommissioning of this plant, known as the USAM plant, is described in Refs [180, 181]. The plant, located in what is now an urban area within the city of São Paulo, operated between 1949 and 1992. During this period, the storage and transport of process residues had been carried out using no radiation protection measures, resulting in spillages, inappropriate on-site disposal and consequently significant soil contamination. Decommissioning of the site was carried out between 1994 and 1998. The site area was 16 500 m^2 and the area of the buildings was 13 000 m^2. Decommissioning was carried out in four stages:

TABLE 67. DISPOSAL OF DECOMMISSIONING WASTES

	Volume (m^3)	Absorbed dose rate (μGy/h)	Disposal method
Sludge in concrete casks	20	50–500	Concrete trenches
Liquid residues from tanks and decontamination operations	300	—	Treatment at effluent treatment plant followed by discharge
Rubber tank linings in concrete casks	10	50–5000[a]	Concrete casks in concrete trenches
Mild steel items not fully decontaminated	25	1–200	Concrete trenches
Wooden items	27	30–200	Concrete trenches
Plastic items not fully decontaminated	3–6	1–50	Concrete trenches
Wall and floor chippings in HDPE bags	15	1–10	Between casks in concrete trenches
Contaminated garnet from sand blasting	—	—	Concrete casks in concrete trenches

[a] Rubber linings with high dose rates were removed by ‘gas cutting’ the tanks and using scrapping tools with long handles. The total volume of the linings with high dose rates is low.

TABLE 68. EXPOSURE TO AIRBORNE RADIOACTIVITY DURING DECOMMISSIONING OPERATIONS

	Airborne dust		Thoron progeny PAEC (μJ/m^3)
	Mass concentration (mg/m^3)	Th-232 activity concentration (mBq/m^3)	
Sludge removal	0.10	10	6–17
Dismantling of services	0.10	10	6–17
Dismantling of tanks	0.19	Ra-228: 40–60 (mean 50)	6–8
Gas torch cutting of tanks and removal of rubber lining	0.33	Ra-228: 70–2720 (mean 900)	6–8
Scrubbing and washing of floors and walls	0.20	10	6
Chipping of floors and walls	0.54	30	2–4
Sand blasting of contaminated items[a]	120–180	230–650 (mean 440)	0.6–2
Demolition of buildings	0.2–0.5	10	0.6–2

[a] In practice, exposures to workers were much lower than levels quoted because of the use of air line respiratory equipment.

(i) Packaging and removal of residues remaining at the plant, comprising 532 t of thorium hydroxide residue (3250 drums) and 73 t of barium sulphate residue (745 drums);
(ii) Decontamination and dismantling of equipment;
(iii) Decontamination of floors and walls, followed by demolition of the buildings;
(iv) A radiation survey of the site and its cleanup.

The ultimate objective of the decommissioning operation was the release of the site for unrestricted use, since the site was to be sold for redevelopment as a residential area. The cleanup criterion was based on the annual dose received by members of the public. Factors such as the urban location, the size of the site and public concerns led to a rather restrictive value of 1 mSv being chosen. Exposure pathway analysis was then used to derive corresponding activity concentration criteria. The following criteria were adopted:

(a) Soil with a ^{228}Ra activity concentration exceeding 30 Bq/g was sent for storage to another site, pending the availability of a suitable disposal facility;
(b) Soil with a ^{228}Ra activity concentration of 0.65–30 Bq/g was disposed of at a municipal landfill site;
(c) Soil with a ^{228}Ra activity below 0.65 Bq/g remained at the site.

The application of these criteria led to the removal from the site of 60 m^3 of soil for storage and 2240 m^3 for disposal in a landfill facility. In addition to contaminated soil, surface contaminated material had to be dealt with. Surface contamination criteria of 0.25 and 2.5 Bq/cm^2 were adopted for alpha and beta radiation, respectively. Material with contamination levels exceeding these criteria were subjected to various decontamination processes. Materials remaining above the criteria were sent for storage at another site, pending the availability of a suitable disposal facility. This resulted in about 6900 m^3 of material, including metallic items, wood, clothing and paper, being removed from the site for storage. The total cost of decommissioning the facility was estimated to be about 20% of the resale value of the property.

9.6. REGULATORY CONSIDERATIONS

Radionuclide activity concentrations in mineral concentrate feedstocks exceed 1 Bq/g, regardless of their origin. Activity concentrations in residues and by-products range from <1 to 3000 Bq/g, with the highest values relating to the chemical treatment of monazite, as would be expected. Consequently, all chemical treatment operations need to be considered for regulatory control as practices.

For the processing of monazite, annual effective doses received by workers are typically about 3–9 mSv, although they were much higher in the past, when fewer control measures were in place. The maximum annual effective dose received by workers in the treatment of Mount Weld concentrate could reach 6 mSv, although much lower doses are expected more generally. Annual effective doses associated with the chemical treatment of bastnäsite and loparite concentrates are expected to be <1 mSv.

Annual effective doses received by members of the public appear to be well below 1 mSv but, in the case of monazite treatment operations, this is contingent on the implementation of a variety of control measures for the storage and disposal of residues and for the discharge of effluents.

On the basis of this information, the chemical treatment of monazite needs to be comprehensively controlled through authorization. In many regulatory jurisdictions, this would take the form of a licence. An example of regulatory guidelines for monazite processing operations is given in Ref. [123]. The chemical treatment of other types of rare earth concentrate would require significantly less regulatory control, although the management of residues and discharges would, in most cases, require some regulatory attention because of the activity concentrations involved.

In terms of Section 3.1.3, the Transport Regulations are unlikely to apply to process feedstocks in the form of bastnäsite concentrates, concentrates derived from Mount Weld ore and concentrates from ion adsorption clays, because of their moderate activity concentrations. Other process feedstocks are likely to fall within the scope of the Transport Regulations, namely:

(a) Monazite concentrates derived from mineral sands, tin mining residues (amang), vein type deposits and, in some cases, Bayan Obo ore;
(b) Xenotime concentrates;
(c) Loparite concentrates.

The Transport Regulations would also apply to almost all residues from the chemical processing of monazite, xenotime, bastnäsite and loparite. They are unlikely to apply to residues from the processing of concentrates from ion adsorption clays.

10. EXTRACTION AND PURIFICATION OF INDIVIDUAL RARE EARTHS

10.1. PROCESS DESCRIPTION

This section provides a short summary of the main processes used for the commercial extraction and purification of rare earths, including some information on the preparation of rare earth compounds. A more detailed description of the technology behind separation and purification processes can be found in Ref. [13].

10.1.1. Separation

Rare earths show unusual chemical resemblance. Consequently, individual rare earths are difficult to separate and purify. Development of the processes of selective oxidation, selective reduction, fractional crystallization and fractional precipitation stretches back over many years; these processes have largely been superseded by the more modern techniques of ion exchange and, more recently, solvent extraction. Selective oxidation and selective reduction are based on differences in chemical behaviour of certain rare earth elements in their divalent or tetravalent states compared with that of their normal trivalent state. All other separation methods for individual trivalent rare earths are based on very small differences in basicity, resulting from equally small differences in the ionic radii of adjacent rare earths and their ionization potentials (electron binding energy).

10.1.1.1. Selective oxidation and selective reduction

Selective oxidation and selective reduction can be used for the separation of rare earths that exist in stable form in the tetravalent state (Ce, Pr and Tb) or in the divalent state (Sm, Eu and Yb), because the behaviour exhibited by these oxidized (RE^{4+}) and reduced (RE^{2+}) states is markedly different from that exhibited by the trivalent (RE^{3+}) state:

(a) Selective oxidation has been used primarily for the separation of Ce, the most abundant rare earth element, early in the separation sequence, in order to produce cerium concentrates suitable for further upgrading or for sale as a product with glass polishing applications. The valency change from Ce^{3+} to Ce^{4+} occurs, for example, when bastnäsite concentrate is heated in air (see Section 9.1.2) or when rare earth hydroxides are dried in air at

120–130°C. Once oxidized, insoluble Ce^{4+} can be readily separated from the soluble trivalent components of the mixture;

(b) Reduction of Sm, Eu and Yb to the divalent state allows them to be easily separated from other rare earths that remain in the trivalent state. Since these elements are much less abundant than Ce, their separation using selective reduction has generally been carried out only after having been enriched using other methods.

Selective oxidation and selective reduction continue to be used, particularly for separating Ce and Eu, as part of the overall technique of separation either preceding or following ion exchange or solvent extraction.

10.1.1.2. Fractional crystallization and fractional precipitation

The processes of fractional crystallization and fractional precipitation can be used to achieve the separation of rare earths by taking advantage of slight differences in the solubility of their salts:

(a) During fractional crystallization, a part of the salt in solution is precipitated by a change in temperature or by evaporation of a saturated solution. During repeated crystallization, the crystals become enriched in the less soluble component while, at the same time, the liquor becomes enriched in the more soluble components. This technique is particularly suited to separation of the light rare earths, for which the differences in ionic radius are greatest, and is known for its use in the separation of lanthanum at high levels of purity. However, it is slow and tedious;

(b) Fractional precipitation entails the removal of part of the rare earths from solution by the addition of a chemical reagent to form a new, less soluble compound. The rare earths remaining in solution can be recovered via further precipitation. The double sulphates $RE_2(SO_4)_3\ Na_2SO_4\ nH_2O$ are usually precipitated by the addition of Na_2SO_4. The light rare earths La, Ce, Pr, Nd and Sm form sparingly soluble double sulphates whereas the heavy rare earths Ho, Er, Tm, Yb, Lu and Y form soluble double sulphates. The rare earths Eu, Gd and Dy form double sulphates of intermediate solubility. Generally, use of this method is confined to the crude separation of rare earth mixtures into these three groups.

10.1.1.3. Ion exchange

The ion exchange technique for the separation of rare earths is based on the differential absorption and differential elution of ionic species, both of which are affected by the basicity of the rare earths. To increase differences in the basicity of adjacent rare earths, various complexes are formed by individual rare earths in the mixture through the use of chelating agents (complexing agents) such as ethylene di-amine tetra-acetic acid (EDTA), 2-hydroxy ethylene diamine triacetic acid (HEDTA) and nitrilo triacetic acid (NTA). The technique is noted for its simplicity and its ability to produce high purity rare earths, but the process is very slow and is not continuous. Consequently, the ion exchange technique, like its predecessors, has become largely outdated in favour of solvent extraction, even though it is still regarded as superior for the production of extremely pure materials.

10.1.1.4. Solvent extraction

Solvent extraction is now the most widely used technique for separating groups of rare earths and separating (and purifying) them individually. It has the following advantages over other methods:

(a) The process is fast, continuous and works on more concentrated solutions;
(b) It is economical for handling large quantities of materials;
(c) The cost of raw materials is low;
(d) The extraction plants are compact and require little manual control.

According to Ref. [13], solvent extraction technology is now used for commercial scale separation (and purification) of at least 11 of the rare earths that occur in bastnäsite, monazite and xenotime ores, although it would not be the method of choice when a product purity of more than 99.9% is required or when the lesser abundant rare earths such as Tm, Yb and Lu are to be separated. Solvent extraction is the main technique used for the separation and extraction of yttrium.

The solvent extraction technique depends on the preferential distribution of individual rare earths between two different immiscible solvents, usually water and an organic solvent, such as kerosene, containing an extractant which has the ability to remove rare earth in the form of a complex. The types of extractant used include a cation exchanger such as di-2-ethylhexyl phosphoric acid, an anion exchanger such as alamine-336, or a solvating type of extractant such as tri-n-butyl phosphate. Since differences in the basicity of adjacent rare earths are small, the separation factors between adjacent rare earths are also small. Hence, tens or hundreds of extraction stages may be needed to reach the desired level of purity and separation. Counter-current, multi-stage mixer settlers are usually used for effecting repeated contact between aqueous and organic phases and then separating one from the other. After separation, each organic stream is then scrubbed with an aqueous stream to transfer the rare earth element into an aqueous phase. A typical mixer settler installation in a rare earths separation plant is shown in Fig. 20.

Solvent extraction has been used for many years in India for the initial separation of groups of rare earths from rare earth chloride solution obtained from the chemical treatment of monazite (see Section 9.1.4.1). The rare earths in the solvent are stripped with hydrochloric acid and medium and heavy rare earths are recovered as carbonates. The chlorides of La, Ce, Pr and Nd are concentrated via evaporation in glass lined evaporating vessels and then poured into drums. They are allowed to cool to obtain a 'chloride crystal lump of rare earths'. The use of solvent extraction provides the option of allowing radium to remain in the rare earth chloride solution instead of having to remove it in the deactivation and lead elimination processes since, during solvent extraction, radium will be rejected into the raffinate. In recent times, it has become general practice to use solvent extraction (using various solvents) to produce La- and Ce-rich fractions, a Ce–Nd–Pr concentrate, a Sm–Gd–Eu concentrate and an yttrium concentrate. Subsequent extraction steps using an organophosphonic acid solvent separate the individual rare earth elements Sm, Eu and Gd, leaving residual light rare earths in the aqueous phase. Further extractions with varying molarity preferentially extract Nd, leaving residual La and Ce in the raffinate. High purity cerium compounds are then produced by repeated fractional precipitations and extractions of the Ce-rich component.

FIG. 20. Mixer settlers used for the separation of rare earths via solvent extraction.

Solvent extraction was used in former operations in the USA to separate rare earths from rare earth chloride solution obtained from bastnäsite (see Section 9.1.2). An extraction plant, which was set up with special characteristics for the recovery of Eu oxide, could separate 1% Eu, Sm and Gd from 99% light rare earths. The elements Eu, Sm and Gd were extracted in the organic phase, leaving the light rare earths La, Pr, Nd, and Ce in the aqueous solution. Further processing of the organic and aqueous phases were undertaken as follows:

(a) The elements Eu, Sm and Gd were stripped from the solvent with 4N HCl and the stripping solution was neutralized with soda ash to precipitate $Fe(OH)_3$ at 3–3.5 pH. Separation from the iron free stripping solution and further purification using selective reduction of trivalent Eu to the divalent state (see Section 10.1.1.1), followed by calcination, produced Eu_2O_3 at 99.99% purity;

(b) By adding ammonia to the aqueous solution, some of the light rare earths were precipitated as hydroxides, thickened, filtered, dried, screened and packed as a crude lanthanum hydroxide concentrate. The rest of the solution was subjected to further solvent extraction to separate high purity Nd, which loaded preferentially on the solvent. Further processing produced high purity Nd oxide.

Solvent extraction has also been used for the removal of individual rare earths from rare earth nitrate or chloride solutions obtained from the chemical processing of loparite (see Section 9.1.7).

10.1.1.5. Other methods

For the purposes of separating lanthanum and converting rare earth elements into nitrates, as well as separating uranium and thorium, studies have been conducted on the use of monocarboxylic acids in the processing of rare earth chloride solutions that have been obtained from loparite by chlorination and acid dissolution (see Section 9.1.7). Further details, including information on the distribution of radioactivity, are given in Ref. [182].

10.1.2. Production of rare earth compounds

Rare earth purification plants produce a variety of rare earth compounds such as chlorides, fluorides, oxides, carbonates and nitrates. These are used as intermediates for the manufacture of final products containing rare earths at various levels of purity, depending on the application. Some examples of the processes used are given below:

(a) Mixed rare earth carbonate is produced by treating rare earth chloride solution with sodium carbonate:

$$2RECl_3 + 3Na_2CO_3 \rightarrow RE_2(CO_3)_3 + 6NaCl$$

Didymium (Nd, Pr) carbonate, containing nearly 40% La, 40% Nd and 10–12% Pr, is produced in the same way, except that cerium is first removed from the other rare earths (as a cerium hydrate precipitate) by oxidation and hydroxide precipitation using sodium hypochlorite:

$$2CeCl_3 + NaOCl + H_2O \rightarrow 2Ce(OH)_4 + NaCl$$

The didymium solution may be used to prepare other compounds such as fluorides and nitrates via precipitation methods;

(b) Mixed rare earth oxide is produced through the calcination of mixed rare earth carbonate:

$$RE_2(CO_3)_3 \rightarrow RE_2O_3 + 3CO_{2;}$$

(c) Cerium hydrate from the chloride oxidation process in (a) is used, at 92–95% purity, in glass decolorization. It may also be calcined at 1000°C to produce cerium oxide, which after grinding and sieving is used as a polishing powder for optical and ophthalmic glass (see Section 11.1.1). Cerium hydrate may also be dissolved in nitric acid and the solution slowly added to boiling water. In the presence of sulphate ions, the Ce^{4+} becomes hydrolysed and precipitates as basic sulphate. This helps in improving purity to more than 99.9%. This basic sulphate can be converted to pure cerous nitrate or oxide, or other salts such as ceric ammonium nitrate and ceric ammonium sulphate;

(d) Mixed rare earth fluoride is precipitated when dilute rare earth chloride solution is treated with sodium silico-fluoride:

$$2RECl_3 + 3H_2O + Na_2SiF_6 \rightarrow 2REF_3 + Na_2SiO_3 + 6HCl$$

10.1.3. Production of rare earth metals by reduction

The end products of rare earth separation processes are generally rare earth oxides. These are the starting point for the production of rare earth elements in metallic form, although reduction to metal is difficult because the oxides are extremely stable. In many cases, the oxide is first converted to an intermediate in the form of a rare earth chloride or fluoride, which is then reduced by lithium or calcium to the metal. Direct reduction from the oxide with lanthanum is, however, used for the production of Sm, Eu and Yb, since these elements cannot be produced using halide reduction. Other methods include metallothermic reduction in molten salt and electrolysis of molten rare earth chloride or oxide–chloride mixture.

10.1.3.1. Halide reduction

In the halide reduction process, an anhydrous rare earth trihalide is first prepared from rare earth oxide. In applying the so-called wet route to chloride reduction, rare earth chloride hexahydrate is crystallized from an HCl solution of rare earth oxide and is then dehydrated by heating with a dry HCl flow under reduced pressure. Alternatively, dry methods such as chlorination of oxide by heating with ammonium chloride or chlorination of oxide or even rare earth ore concentrate via heating with carbon under chlorine flow have also been used. Preparation of the rare earth metal is then achieved through lithium reduction of the trichloride. Chloride reduction is an involved procedure on account of the hygroscopicity and volatility of chlorides. This problem can be circumvented by the application of wet or dry methods to fluorides instead of chlorides. Fluoride reduction is achieved using calcium. Ten of the rare earths (La, Ce, Pr, Nd, Gd, Tb, Dy, Ho, Er and Lu) as well as Sc and Y can be produced through calcium reduction, and up to 99.99% purity can be achieved.

10.1.3.2. Oxide reduction

The oxide reduction process is used for preparing metallic Sm, Eu and Yb. The reduction process exploits the pronounced volatility of these metals. A mixture of lanthanum (or another metal with similar relevant characteristics such as cerium, mischmetal, zirconium or thorium) and rare earth oxide is heated in a tantalum crucible fitted with a long tantalum condenser. The oxide is reduced and the rare earth metal, being volatile, escapes and collects in the condenser.

10.1.3.3. Metallothermic reduction in molten salt

This process is used for the production of Nd metal. Neodymium oxide is reduced to metal through reacting with calcium in a molten salt bath ($CaCl_2$–NaCl) and simultaneously the Nd metal is extracted into a molten Nd–Zn alloy. The Nd–Zn alloy product is treated in a vacuum to remove zinc and produce Nd metal. In variations of this process, reduction may be carried out using sodium instead of calcium and the reduced metal may be extracted into a molten alloy of Nd–Fe alloy instead of Nd–Zn.

10.1.3.4. Electrolysis of molten rare earth salts

Electrolytic methods account for the largest quantity of rare earth metals production worldwide. A solution of rare earth chloride or rare earth oxide–fluoride in another salt or mixture of salts is kept molten in an inert container fitted with an anode and cathode. When an electric current is passed through the circuit, the salt is reduced and the rare earth metal is deposited and recovered as a liquid (if the electrolyte temperature is high enough) or as an agglomeration of solid particles.

In some cases, it is necessary to recover the rare earth metal as a binary alloy that is molten at electrolysis temperature, allowing it to be easily separated from the electrolyte. The rare earth is then recovered from the alloy by pyrovacuum distillation.

Molten salt electolysis of rare earth chloride is used to produce lanthanum, cerium and didymium (Nd + Pr) at purity levels of 99% or more. Electrolytic methods involving molten rare earth oxide–fluoride are used for producing all rare earth metals except for samarium, europium and ytterbium. One widely used cell design for the electrolysis of molten rare earth oxide–fluoride is capable of producing cerium at a purity level of at least 99.8%. Another type of cell, specially designed to operate at a higher temperature (1370–1700°C), is capable of producing high melting point rare earth metals gadolinium, dysprosium and yttrium in liquid form.

10.1.4. Refining of rare earth metals

Rare earth metals produced by reduction techniques are generally 98–99% pure, although higher levels of purity are possible. Additional purity is gained through the use of various refining processes, the choice of which is influenced by the nature and concentration of the impurity and the final purity level required.

Generally, a sequence of refining processes is applied to effect the removal of various impurities. The conventional purification steps applied initially involve the use of pyrovacuum techniques such as vacuum melting, vacuum distillation and vacuum sublimation, although electrorefining in an inert atmosphere has been used for the purification of Gd and Y, these being the two rare earth metals that are the most difficult to purify using only pyrovacuum techniques. The various pyrovacuum techniques all involve heating the metal up to or above its melting point in a tantalum crucible and are conducted under high vacuum or ultra-high vacuum conditions in order to prevent contamination from atmospheric impurities and to effect removal of the impurities. In vacuum melting, the impurities vaporize, while in vacuum distillation and vacuum sublimation the metal itself is vaporized off to condense as a purer product, leaving the impurities in the residue. The particular techniques used depend on the volatility and melting point of the rare earth metal concerned.

In the final ‘ultrapurification’ steps, involving the techniques of zone refining and solid state electrotransport, the remaining impurities are redistributed within the metal so that one physical portion of the metal becomes purer at the expense of the remainder. Zone refining involves generating a thin molten zone of metal by localized melting and passing the zone along the length of the metal in one direction several times. Purification takes place at the travelling liquid–solid interface. Impurities more soluble in the molten phase will move in the direction in which the molten zone moves and therefore concentrate and solidify at the finishing end of the metal. In solid state electrotransport, a direct current is passed through a rod or strip of metal held between two electrodes. The impurities move under the influence of the electric field and eventually accumulate at one end.

10.1.5. Management of residues

Solid residue types and amounts resulting from rare earth separation processes depend on the exact processes used, but may include solvent extraction residue, filter cake or sludge, and scrapped filter cloths and equipment (see, for instance, Ref. [183]). The solvent extraction residue is an insoluble organic complex comprising non-halogenated organics and organic phosphates and chlorides; it is generally disposed of in controlled industrial waste facilities, although solvent extraction sludge can be recycled. The amount of solvent extraction residue generated depends on plant conditions, plant size, and the degree to which spent solvent is recycled. In former operations in the USA, it was generated in quantities of about 100 t/a per plant, whereas in current operations in India the amounts are generally considerably smaller.

The various process steps in the separation and processing of rare earths result in some airborne discharge. In rare earths production in India, gaseous effluents include hydrochloric acid vapour, chlorine and hydrogen sulphide and are assessed as part of the operational radiation protection programme. These releases are controlled by alkaline (sodium hydroxide) scrubbers installed in the ventilation system to ensure compliance with local limits [165]. It is reported from past operations in the USA [183] that off-gases from the electrolytic reduction step include chlorine, carbon monoxide and carbon dioxide from the graphite anodes, and hydrochloric acid fumes. These gases were treated with water or alkaline scrubbers to cool them and to absorb particulates and hydrochloric acid vapours. The partially cleaned gases were then contacted with sodium hydroxide solution, resulting in the formation of sodium hypochlorite solution, which could be sold as a by-product.

Liquid waste streams are described in Refs [183, 178] and include rare earth carbonate decant and washings, cerium hydrate decant and leachings, rare earth fluoride decant and leachings, leachings from both heavy and light rare earth separation facilities, spent electrolyte cell quench water and various other washings and coolants. Quantities from a typical separation plant are reported to be in the range 10–80 m^3/d, while the pH varies from 2 to 8 [178, 184]. These streams are treated in an effluent treatment plant along with other liquid waste streams from chemical processing steps to render them suitable for discharge to the environment (see Section 9.1.4.2). Spent solvents from the solvent extraction process leave the process as residual aqueous streams, but can be recycled. Residual process water from cerium production can also be recycled.

10.2. RADIONUCLIDE ACTIVITY CONCENTRATIONS

In earlier operations in the USA, first documented in 1961 and reported subsequently in Ref. [142], rare earth sodium sulphate with a rare earth content of 42% REO was used as a feedstock for the production of various rare earth compounds. Purified rare earths were produced using ion exchange technology. The thorium concentrations in these materials were reported, from which it is possible to derive ^{232}Th activity concentrations. In current operations in India, the mixed rare earth chloride (derived from monazite) used as the starting point for separation and purification processes contains ^{228}Ra at an activity concentration of 0.45 Bq/g [162]. The major contaminant in the derived rare earth compounds is expected to be ^{228}Ra. A wide range of ^{228}Ra activity concentrations is observed, and values depend upon process conditions and the chemical nature of the compound concerned. A compilation of average activity concentrations from these two reference sources is provided in Table 69.

In addition to radionuclides in the ^{232}Th and ^{238}U decay series associated with rare earth compounds, the rare earth element samarium is itself radioactive as a result of the presence of radioactive isotopes ^{147}Sm, ^{148}Sm and ^{149}Sm. The natural abundances and half-lives of these isotopes are shown in Table 70. Using these values, the specific activities of pure natural samarium and samarium oxide (Sm_2O_3) are calculated to be 127.3 and 109.5 Bq/g, respectively. Almost the entire activity content (99.994%) is attributable to ^{147}Sm; the contributions of ^{148}Sm and ^{149}Sm can therefore be neglected.

Solvent extraction residue is reported to have a ^{232}Th activity concentration of 0.6 Bq/g and a ^{238}U activity concentration of 28.1 Bq/g [143].

The liquid effluent from two rare earths separation plants in India (a heavy rare earth oxide plant and a neodymium recovery plant) is reported to contain ^{228}Ra at activity concentrations of 3–10 Bq/L [178]. In a more recent publication

TABLE 69. ACTIVITY CONCENTRATIONS IN RARE EARTH COMPOUNDS

	Activity concentration (Bq/g)	
	^{232}Th [142][a]	^{228}Ra [162][b]
Cerium hydrate	6.4	
Rare earth oxide	7.8	
Purified rare earths	<2	
Rare earth sodium sulphate	3.9	
Rare earth chloride	<2	
Rare earth fluoride	7.8	
Mixed rare earth chloride		0.45
Mixed rare earth fluoride		1.23
Mixed rare earth carbonate		3.80
Mixed rare earth oxide		1.90
Cerium oxide, A grade, >85% purity		5.20
Cerium oxide, B grade, 90% purity		2.30
Cerium oxide, C grade, 99% purity		0.43
Cerium oxide, D grade, 99.5% purity		<0.15
Cerium hydrate		1.40
Cerium nitrate		<0.15
Neodymium–prometheum carbonate		2.30
Neodymium oxide		0.20
Yttrium oxide		0.53
Praseodymium oxide, 99%		0.54
Neodymium oxide, 99%		<0.15
Samarium oxide, 99%		0.83

TABLE 69. ACTIVITY CONCENTRATIONS IN RARE EARTH COMPOUNDS (cont.)

	Activity concentration (Bq/g)	
	^{232}Th [142][a]	^{228}Ra [162][b]
Gadolinium oxide, 99%		0.48
Yttrium oxide, 99.9%		0.33
Lanthanum oxide		7.80
Terbium oxide, 90%		0.63
Samarium–gadolinium concentrate (carbonate)		0.18

[a] Derived from the reported values of thorium concentration.

[b] Each value is the mean of three samples (or 15 samples in the case of mixed rare earth chloride).

TABLE 70. NATURAL ABUNDANCES AND HALF-LIVES OF SAMARIUM ISOTOPES [185]

	Natural abundance (%)	Half-life (a)
Sm-147	14.99	1.06×10^{11}
Sm-148	11.24	7×10^{15}
Sm-149	13.82	$>2 \times 10^{15}$

[162], the activity concentration of ^{228}Ra in liquid effluent is reported to be in the range of 5–46 Bq/L with a mean value of 24 Bq/L. These values refer to the effluent before treatment. The effluent treatment plant, which also treats liquid effluent from the chemical treatment of monazite (see Section 9.1.4.2), removes almost all radioactivity and allows treated effluent to be discharged into a river in compliance with local discharge limits [178].

Gaseous emissions from rare earth separation and purification plants normally only contain low concentrations of thoron progeny. Measurements conducted around plants in India have indicated no significant environmental concentration of thoron progeny attributable to plant operation.

10.3. OCCUPATIONAL EXPOSURE

10.3.1. Exposure pathways

Large scale handling and processing of rare earth compounds, particularly those sourced from monazite, result in occupational radiation exposures. The major routes of exposure are:

(i) External gamma exposure: The rubber linings of processing tanks containing composite rare earth chloride and other compounds become contaminated over long periods of operation and may result in significant gamma exposure of workers;
(ii) Inhalation of long-lived alpha activity in dust;
(iii) Inhalation of thoron progeny: The emanation of thoron from different compounds due to contamination of ^{228}Ra and ^{224}Ra may enhance background thoron and progeny levels in a workplace.

10.3.2. Exposure levels

10.3.2.1. Gamma dose rates

Dose rates measured in rare earth separation plants [91, 141, 162] are given in Table 71. The most recent measurements [162] were carried out 1 m above the ground in a plant in India on a monthly basis over a period of two years — they show that the mean dose rates were generally below 1 μGy/h except for areas near storage tanks with accumulated sludge and scale. These areas were generally unoccupied. The average value in areas occupied by workers was 0.5 μGy/h. Natural backround levels outside plant premises was 0.15 μGy/h, suggesting an average incremental exposure of 0.35 μGy/h. Earlier measurements of gamma dose rates (for plants in India and the USA) suggested higher mean values, in the range of 1–6 μGy/h.

10.3.2.2. Dust

In rare earth separation plants in India, the dust is reported to have the characteristics shown in Table 72. Airborne dust activity concentrations measured in rare earth extraction plants are given in Table 73. The most recent measurements, carried out in India [162], indicate a mean ^{232}Th activity concentration of 5 mBq/m^3 over a five year period.

TABLE 71. EXTERNAL GAMMA EXPOSURES IN RARE EARTH SEPARATION PLANTS

	Absorbed dose rate (μGy/h)	
	Mean	Range
USA, 1992 [141]		
Solvent extraction, raffinate cell	5.60	—
Solvent extraction, sump pump	1.32	—
Solvent extraction, feed tanks	4.82	—
India, 2003 [91]		
Production of diversified rare earth compounds	3	2–120
Heavy rare earth oxide production	2.5	2–100
Ce, Pr, Nd, Y compound production	1	1–5
India, 2008 [162]		
Rare earth chloride storage tanks	1.0	0.6–1.2
Rare earth chloride storage tank, sludge and scale:		
General	15	10–40
Prolonged scale accumulation	—	Up to 100
Cerium hydrate tanks	0.4	0.3–0.7
Rare earth carbonate tanks	0.8	0.7–1.0
Mixer settlers, solvent extraction	0.8	0.6–0.9
Rare earth fluoride process tanks	2.0	1.0–3.0
Product drier	0.7	0.5–1.0
Mixer settlers, cerium purification	0.3	0.2–0.4
Filter presses	0.5	0.4–0.6
Calciner, kiln, drier	0.7	0.5–1.0
Average general background in worker occupied areas	0.5	0.3–0.8
Natural background outside plant premises	0.15	—

TABLE 72. AIRBORNE DUST CHARACTERISTICS IN RARE EARTH SEPARATION PLANTS IN INDIA

	Value	Unit	Ref.
Mass concentration in air	0.03–2.5 (average 0.3)	mg/m^3	[91]
AMAD			
Solvent extraction area	4.57	µm	[171]
Rare earth compound process area	2.21–3.47	µm	[171]
Geometric standard deviation			
Solvent extraction area	3.3	—	[171]
Rare earth compound process area	3.44–6.2	—	[171]
Resuspension factor	1.7×10^{-8}	cm^{-1}	[186]

TABLE 73. DUST ACTIVITY CONCENTRATIONS IN AIR

	^{232}Th activity concentration in air (mBq/m^3)		Ref.
	Mean	Range	
Cerium hydrate production (drying in manually loaded and unloaded trays and wet separation of cerium from rare earth mixture)	1.5		[142]
Rare earth oxide production (closed room with continuous rotary calciners where material is screened and packed)	3.7		[142]]
Production of purified rare earths (ion exchange)	2.2		[142]
Production of rare earth sodium sulphate (rotary drying of filter cake and bulk handling)	0		[142]
Production of rare earth chloride	0.7		[142]
Production of rare earth fluoride (preparation, drying, pulverizing and packing)	2.2		[142]
Production of diversified rare earth compounds	20	10–90	[91]
Heavy rare earth oxide production	6	4–30	[91]

TABLE 73. DUST ACTIVITY CONCENTRATIONS IN AIR (cont.)

	^{232}Th activity concentration in air (mBq/m^3)		Ref.
	Mean	Range	
Pr, Y, Nd, Ce compounds production	8	2–20	[91]
Production of rare earth compounds, plant averages			[162]
2003	8	1–12	
2004	6	2–14	
2005	7	1–23	
2006	5	1–18	
2007	3	1–11	
Overall (2003–2007)	5	1–23	

10.3.2.3. Thoron progeny concentrations

PAECs of thoron progeny measured in rare earth extraction plants in India are given in Table 74. The most recent measurements [162] indicate a mean thoron progeny concentration of 0.62 μJ/m^3 over a five year period. Earlier measurements of thoron concentrations suggested similar or higher mean values.

10.3.3. Effective dose

In a plant in the USA, annual effective doses from external exposure received between 1959–1960 by individuals working with ion exchange columns for the purification of rare earths were determined using film badge measurements. The results, which were first documented in 1961, are reported in Ref. [142]. Estimated annual doses ranged from 0.6 to 11.55 mSv, with a mean value of 5.0 mSv.

A more up-to-date estimate of effective dose from all relevant exposure pathways can be determined from recently measured average exposure levels. The most recent sets of measurements reported in Section 10.3.2 suggest that the average gamma exposure level is 0.35 μGy/h above background, the average dust activity concentration for ^{232}Th is 5 mBq/m^3 and the average thoron concentration is 0.62 μJ/m^3. Assuming that 1 Gy of absorbed dose gives rise to approximately 1 Sv of effective dose, and assuming that the dust is of lung

TABLE 74. THORON PROGENY CONCENTRATIONS IN AIR

	Thoron progeny PAEC (μJ/m^3)		Ref.
	Mean	Range	
Production of diversified rare earth compounds	10	0.9–40	[91]
Heavy rare earth oxide production	0.5	0.2–2	[91]
Pr, Y, Nd, Ce compound production	0.3	0.1–3	[91]
Plant averages			[162]
2003	0.31	0.10–1.9	
2004	0.62	0.15–3.2	
2005	0.87	0.10–3.1	
2006	0.73	0.06–3.4	
2007	0.85	0.04–2.9	
Overall (2003–2007)	0.62	0.04–3.4	

TABLE 75. ANNUAL EFFECTIVE DOSE ESTIMATED FROM EXPOSURE MEASUREMENTS

	Annual effective dose (mSv)
External gamma exposure	0.7
Inhalation of radionuclides in dust	0.6
Inhalation of thoron progeny	0.6
Total	1.9

absorption class M and has an AMAD of 5 μm, the effective dose received in a 2000 h annual exposure period (excluding the dose from inhalation of ^{147}Sm, ^{148}Sm and ^{149}Sm) can be calculated using the relevant dose coefficients given in Sections 3.3.2 and 3.4.2. The results are shown in Table 75, indicating a total annual effective dose of 1.9 mSv. This is also the value reported in Ref. [162].

The effective dose received from the inhalation of ^{147}Sm, ^{148}Sm and ^{149}Sm in airborne dust can be calculated from the specific activity of samarium oxide

(127.3 Bq/g, see Section 10.2). Assuming that the dust contains 10% samarium oxide (lung absorption class M) with an AMAD of 5 μm and a mass concentration in air of 0.3 mg/m^3 (see Table 72) and that the annual exposure time is 2000 h, the annual activity intake is 79 Bq. The dose coefficient for ^{147}Sm is 6.1 × 10^{-6} Sv/Bq [2], giving an annual effective dose of 0.05 mSv. Given the conservative assumptions in this calculation, it can be concluded that the inhalation of radioactive isotopes of samarium in airborne dust does not contribute significantly to the effective dose received by a worker in a rare earths separation plant.

10.3.4. Measures to reduce doses

Doses received by workers in the separation of rare earths arise from exposure to radionuclides introdued via the rare earth chloride solution used as a starting material. Thus, the lower the radionuclide concentrations in the rare earth chloride, the lower will be the doses received by workers. Experience gained in Indian rare earth plants has demonstrated that it is possible to reduce the radium content of rare earth chloride by about 50 times, through modifications to the deactivation process conducted as part of the chemical treatment of monazite (see Section 9.1.4.1).

Doses from exposure to external gamma radiation can be reduced by ensuring that scale and sludge within process tanks is not allowed to accumulate. Doses from inhalation of airborne dust and thoron progeny can be reduced by attention to the ventilation system.

10.4. PUBLIC EXPOSURE

10.4.1. Exposure from operating facilities

As reported in Section 10.2, gaseous and liquid discharges from rare earth separation and purification plants in India contain very low concentrations of radionuclides. Consequently, no significant exposure to the public is apparent. Monitoring of environmental radioactivity levels around a plant processing rare earth hydroxides in Freeport, Texas, showed no evidence of off-site contamination [187]. Elevated gamma dose rates were detected at the boundary of the facility, but public occupancy at these locations was reported to be low.

10.4.2. Exposure to residual radioactive material from past operations

In Brazil, the separation of light rare earths from heavy rare earths was formerly carried out at Usina de Interlagos (USIN), a 60 000 m^2 site in an industrial district of São Paulo. The facility was closed in 1990. Management of the process residues and decommissioning and demolition of the facility was carried out without adequate radiation protection measures, resulting in significant residual contamination of 6500 m^2 of soil [188]. The contamination, which extended to a depth of 1 m, included some residues from the chemical processing of monazite carried out at another site (see Section 9.5.2). Activity concentrations in the soil were found to be 0.15–33 Bq/g for ^{228}Ra and 0.05–6.7 Bq/g for ^{226}Ra, respectively. The volume of contaminated soil that needed to be removed to a suitable waste repository depended on the future use of the land and the choice of clean-up criterion (the level of residual dose). For a future residential exposure scenario and to reach a residual dose criterion in the range of 0.3–10 mSv/a, the most recent estimate of the amount of soil to be removed varies from 500 to 1600 m^3 [181]. A dose criterion of between 1 and 5 mSv/a has been proposed for the remediation process.

10.5. REGULATORY CONSIDERATIONS

Some rare earth compounds have radionuclide activity concentrations greater than 1 Bq/g, depending on purity level. None have activity concentrations exceeding 10 Bq/g. The ^{238}U activity concentration in a solvent extraction residue was reported to be almost 30 Bq/g. On the basis of this information, it can be concluded that some operations involving the extraction and purification of individual rare earths should be considered for possible regulatory control.

Current estimates of annual effective doses received by workers in plants using rare earth feedstock derived from monazite suggest a value of about 2 mSv, although much higher doses, up to about 10 mSv, have been reported from past operations. This indicates that monitoring and radiation protection measures to control occupational exposures are needed, at least in plants handling rare earths compounds derived from monazite in large quantities. Annual effective doses received by members of the public from the operation of rare earth separation plants are insignificant, although the possibility of doses higher than 1 mSv has been demonstrated in a situation where land had become contaminated by former operations that were not subject to adequate radiation protection measures. Some activities involving the extraction and purification of rare earths, and at least those for which rare earth feedstock is derived from monazite, would therefore need to be authorized. Depending on the circumstances and local regulatory preferences, authorization in the form of registration might be sufficient.

With the possible exception of the transport of solvent extraction residue, the transport of process materials would not be subject to requirements of the Transport Regulations.

11. MANUFACTURE AND USE OF RARE EARTH PRODUCTS

11.1. PROCESS DESCRIPTION

11.1.1. Products used in the glass industry

Rare earth products with applications in the glass industry are discussed in Ref. [189]. Production processes can be summarized as follows:

(a) The production of cerium oxide glass polishing powders involves the calcination of cerium concentrate (typically at 1000°C) followed by grinding and sieving to obtain required particle size, and finally packing of the product into bags. The cerium concentrate feedstock may be diluted with other material by a factor of up to four. The process generates minimal amounts of residue. Stack emissions are controlled using efficient filter systems. In a typical plant producing 100 t/a of glass polishing powder, it has been estimated that up to 20 kg/a of product are washed away with water effluent and, at most, 5 kg/a are discharged to the atmosphere. Spills of material may give rise to discharge via other routes. Used polishing powder is mixed with non-radioactive material and disposed of in landfill facilities.

(b) In the manufacture of special glass, rare earth concentrate is mixed with silica before melting. A typical plant throughput is about 50 t/a. Feedstock is diluted in the final product by a factor of about 100. A small liquid discharge from floor washings and some discharge to the atmosphere from local ventilation is expected to occur, with the amounts being similar to those for a plant manufacturing cerium oxide glass polishing powder. The airborne discharge of rare earth concentrate from a furnace has been assessed to be about 60 kg/a.

11.1.2. Metal alloys

The use of rare earths in alloys and alloy products such as permanent magnets is discussed in Ref. [12]. Production processes can be summarized as follows:

(a) Mischmetal with a purity level suitable for most commercial applications is produced by molten salt electrolysis of anhydrous rare earth chlorides that have been prepared from wet rare earth chlorides or hydrated rare earth compounds in dehydration furnaces. Excess slag is removed and may be sold for its rare earth content [183]. Following electrolytic reduction, the mischmetal is cast into bars or ingots. In the process described in Ref. [183], the caustic wet air pollution control system following the water quench or water scrubber in mischmetal production is designed to recover chlorine from the gas stream. Sodium hydroxide is circulated through the scrubber and the reaction with chlorine forms sodium hypochlorite which is sold as a by-product. Wet air pollution control residues in the form of spent scrubber liquor and electrolytic cell caustic sludge may also be recycled. Mischmetal at higher purity levels can be gained through metallothermic reduction of mixed rare earth fluoride;

(b) Alloys of rare earths, iron and silicon are produced by the carbothermic reduction of a mixture of rare earth compounds (or ore concentrate), iron (or iron ore) and silica in a submerged electric arc furnace. Electric arc furnace reductions have also been used to produce alloys of rare earths, magnesium and silicon using blast furnace slag containing rare earths, calcined dolomite–ferrosilicon, calcium and silicon;

(c) Alloys of rare earths, aluminum and zinc and of yttrium and aluminium are produced by molten salt electrolytic processes;

(d) Alloys for permanent magnets are made by directly melting alloy components, by reduction of a mixture of oxides (co-reduction or reduction diffusion) or, in the case of Nd–Fe–B magnets, by a thermit type reduction[16]. In the production of Sm–Co magnets, the alloy is ground to a powder, mixed with a chemical binder and pressed under a magnetic field to produce a plastic bonded (sintered) magnet. Soft metal may also be used as a binder. Magnets based on Nd–Fe–B alloys are also produced through the traditional pressing and sintering route. Alternatively, they are produced by

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[16] Thermit (or thermite) reaction is the name given to a series of displacement reactions between powdered aluminium and various oxides or sulphides of metals that are below aluminium in the reactivity series of metals.

'melt spinning', a process in which molten alloy is subjected to rapid solidification by ejection through an orifice in a crucible onto the surface of a spinning copper disc. The ribbon-like material so formed is then consolidated into a dense form to produce either a bonded magnet or a hot pressed magnet. Permanent magnets are now also produced from Tb–Fe–Co and Sm–Fe–N alloys using an injection moulding process;

(e) The magnetostrictive material Terfenol-D (a Tb–Dy–Fe alloy) is produced either through the conventional method of direct melting of constituents in an arc furnace or by a thermit type reduction of the fluorides of Tb, Dy and Fe with Ca in a crucible lined with CaF_2;

(f) In the manufacture of magneto-optic storage media, an amorphous alloy such as Tb–Fe–Co or a Gd–Tb–Fe composition is deposited as a thin film onto a transparent plastic substrate using a sputtering process;

(g) Intermetallic $LaNi_5$, which is used as a hydrogen sponge, is prepared by melting constituent metals in an arc furnace. The alloy ingot is then granulated in a hardened steel mortar and pestle and the granules are plated with copper.

11.1.3. Superconductors

Various techniques for making high temperature ('high Tc') superconductors are discussed in Ref. [12]. Constituents may be deposited as an epitaxal film onto a single crystal structure via evaporation, laser ablation or sputtering. Thick films can be made by mixing the powdered constituents with polymers and solvents and depositing the mixture onto a polycrystalline ceramic or metallic substrate. A superconducting 1–2–3 yttrium barium copper compound can be prepared as a film through oxidation of the metallic precursors in a process involving electron beam bombardment of three separate metal targets followed by oxidation during deposition on a substrate. A similar compound can be incorporated along with zirconium oxide into a three layer nickel alloy tape using pulsed laser deposition of films 6–10 μm thick. Such a compound can also be made in bulk by sintering micron size particles of high purity oxides of yttrium, barium and copper.

11.1.4. Products made directly from rare earth minerals

The direct use of rare earth minerals for the production of domestic and industrial products is reported in Ref. [190]. Pulverized monazite has been used in health products, paint and spa bathing elements. Bastnäsite, after wet crushing, filtration, drying, roasting and size classification, is used for the production of abrasives.

11.2. RADIONUCLIDE ACTIVITY CONCENTRATIONS

Radionuclide activity concentrations in feedstocks and products involving relatively low purity rare earth materials are given in Table 76. Activity concentrations in feedstocks and products involving the use of high purity rare earth materials are likely to be much lower (see, for instance, Table 69). It is

TABLE 76. ACTIVITY CONCENTRATIONS ASSOCIATED WITH THE MANUFACTURE OF PRODUCTS INVOLVING LOW PURITY RARE EARTH MATERIALS

	Activity concentration (Bq/g)		Ref.
	^{232}Th	^{238}U	
Manufacture of glass industry products from rare earth concentrate			
Feedstock for polishing powder and special glass	5[a]	0.1	[192]
Feedstock for polishing powder and special glass	10	0	[193]
Feedstock for polishing powder and special glass	6	1	[194]
Feedstock for polishing powder	6	1	[189]
Feedstock for special glass	10	1	[189]
Polishing powder product	1.8–7.1	—	[142]
Polishing powder product[b]	1.25–6	0.025–1	[189, 192]
Special glass product[c]	0.05–0.1	0.001–0.01	[189]
Used products disposed of in landfill[d]	9		[142]
Manufacture of health products, paint, spa bathing elements from monazite			
Feedstock	~300	~40	[190]
Production of abrasives from bastnäsite			
Raw material yard	5.8	1.1	[190]
Raw material input hopper	4.9	1.0	[190]
Abrasive products yard	7.1	1.4	[190]
Pink tinted eyeglasses containing cerium oxide	1.8[d]		[191]

[a] One bag of material was found to have a ^{232}Th activity concentration of 25 Bq/g.

[b] Derived from reported dilution factors of up to four times.

[c] Derived from a reported dilution factor of about 100 times.

[d] Derived from the reported thorium concentration.

reported in Ref. [191] that during the production of cerium additives for use in certain metals and alloys, ^{228}Ra and ^{226}Ra appear to concentrate in the slag.

11.3. OCCUPATIONAL EXPOSURE

11.3.1. Exposure pathways

Depending on the activity concentration of the process material involved, there may be a potential for workers to receive significant exposures from external gamma radiation near large quantities of material, although exposure periods may be quite short.

Several of the processes could result in the generation of airborne dust with significant thorium activity concentrations. In the production of glass polishing compounds and special glass, inhalation of airborne dust is said to be the main pathway of exposure [195]. The process for manufacturing glass polishing compound is reasonably continuous and dust exposure can occur over long periods. Even where a plant has been designed to minimize dust through automatic transfer systems, airborne dust levels have been of concern [192]. It is reported in Refs [189, 193] that the furnacing of rare earth concentrate for the production of glass polishing powder generates loose surface dust on floors and other horizontal surfaces, although airborne dust appeared to be restricted to a small number of areas such as those associated with introduction of the raw material entry and bagging of the product.

The grinding and sintering of Sm–Co alloys in the manufacture of permanent magnets is reported to give rise to airborne dust [190].

11.3.2. Exposure levels

11.3.2.1. Gamma radiation

Gamma exposures have been measured in workplaces used for the manufacture of products using low purity rare earth materials. Such workplaces include those for the production of glass polishing powder, special glass and cerium containing additives for metals and alloys. Data are also available for workplaces involving the use of pulverized monazite for the production of health products, paint and spa bathing elements and the use of bastnäsite for the production of abrasives. The results are given in Table 77, from which it can be concluded that workers are generally exposed to dose rates of the order of 1 μSv/h or less. Gamma exposures in workplaces involving the use of high purity rare earth materials are likely to be much lower.

TABLE 77. GAMMA EXPOSURES ASSOCIATED WITH THE MANUFACTURE OF PRODUCTS INVOLVING LOW PURITY RARE EARTH MATERIALS

	Dose rate (μSv/h)	Ref.
Manufacture of glass polishing powders and special glass	Maximum 4 (near bulk material) 'Significantly lower' in general areas	[189]
Manufacture of glass polishing powders and special glass	1.8	[194]
Manufacture of cerium containing additives for metals and alloys, slag storage room	1.7–1.9	[191]
Manufacture of health products, paint and spa bathing elements from monazite		[190]
Monazite warehouse, on contact	100	
Product fabrication area, at 1 m	0.8 (0.75 excluding background)	
Manufacture of abrasives from bastnäsite		[190]
Raw material yard	1.9 on contact, 0.6 at 1 m	
Raw material input hopper	2.0 on contact, 0.10 at 1 m	
Abrasive products yard	3.6 on contact, 0.88 at 1 m (0.84 excluding background)	

11.3.2.2. Dust

Airborne dust characteristics have been determined in workplaces in the USA and the United Kingdom for the manufacture of products for the glass industry. The results are provided in Table 78.

11.3.3. Effective dose

11.3.3.1. External gamma radiation

Estimates of annual effective doses received by workers involved in the manufacture of rare earth products and their eventual disposal in landfill facilities are given in Table 79. For workers in manufacturing facilities, annual doses range from 0.05 to 0.8 mSv. For workers engaged in the disposal of products, annual doses are 0.0006–0.005 mSv.

TABLE 78. AIRBORNE DUST CHARACTERISTICS IN THE MANUFACTURE OF RARE EARTH PRODUCTS

	AMAD (μm)	Concentration in air		Ref.
		Mass (mg/m^3)	^{232}Th activity (mBq/m^3)	
Glass industry				
Bastnäsite and cerium oxide glass polishing powders		0.4–1.4[a]	2.6	[142]
Glass polishing powders	3	7[a]	70	[193]
Glass polishing powders	3	1	6[b]	[189]
Glass polishing powders	5	1	6[b]	[194]
Special glass	3	5	50[b]	[189]
Special glass	5	5	30[b]	[194]
Samarium–cobalt magnets				
Production of alloy			3.1	[190]
Manufacture of magnets			17	[190]

[a] Derived from the ^{232}Th activity concentration of the material (Table 76) and the activity concentration in air.

[b] Derived from the ^{232}Th activity concentration of the material (Table 76) and the mass concentration in air.

11.3.3.2. Airborne dust

Estimates of committed effected dose from the inhalation of airborne dust, using various dose conversion factors, are reported in Refs [189, 193, 194]. In all cases, a breathing rate of 1.2 m^3/h was assumed and the values of dust activity concentration and AMAD are those given in Table 78. If doses are recalculated using the most recent dose conversion factors (see Table 9), the results given in Table 80 are obtained. Also shown in Table 80 are the doses reported in Ref. [190] for the production of Sm–Co alloy and the use of this alloy in the production of permanent magnets, as well as the doses received by workers involved in the landfill disposal of used rare earth products [142].

TABLE 79. DOSES FROM EXTERNAL GAMMA RADIATION RECEIVED BY WORKERS IN THE PRODUCTION AND DISPOSAL OF RARE EARTH PRODUCTS

Product	Work activity or area	Annual exposure period (h)	Annual effective dose (mSv)	Ref.
Glass polishing powders	Production facility	—	0.2	[193]
		200	0.8	[189]
		200	0.36	[194]
Special glass	Production facility	—	0.8	[189]
		200	0.36	[194]
Metal and alloy additives	Slag storage room	24	0.05	[142]
Health products, paint, spa bathing elements	Product fabrication area	360	0.3	[190]
Abrasives	Product storage yard	480	0.4	[190]
Cerium containing products	Collection of used products	—	0.005	[142]
	Landfill disposal	—	0.0006	[142]

TABLE 80. DOSES FROM AIRBORNE DUST INHALATION RECEIVED BY WORKERS IN THE PRODUCTION AND DISPOSAL OF RARE EARTH PRODUCTS

	Assumed annual exposure period (h)	Assumed respiratory protection factor	Annual committed effective dose (mSv)	Ref.
Glass industry				
Glass polishing powders[a]	1400	50	0.16	[193]
	1600	10	0.09	[189]
	400	10	0.02	[194]
Special glass[a]	1400	10	0.60	[189]
	600	10	0.12	[194]
Samarium–cobalt magnets				
Production of alloy	2000	10	0.001–0.067[b]	[190]
Manufacture of magnets	2000	10	0.006–0.038[b]	[190]

TABLE 80. DOSES FROM AIRBORNE DUST INHALATION RECEIVED BY WORKERS IN THE PRODUCTION AND DISPOSAL OF RARE EARTH PRODUCTS (cont.)

	Assumed annual exposure period (h)	Assumed respiratory protection factor	Annual committed effective dose (mSv)	Ref.
Disposal of used rare earth products in a landfill facility				
Waste collector			0.001	[142]
On-site worker			0.0002	[142]

[a] The dose has been recalculated using the dose coefficients given in Table 9.

[b] The range corresponds to assumed particle sizes of 1–50 µm.

11.3.3.3. Total effective dose

Estimates of the total effective doses received by workers in the manufacture and disposal of rare earth products can be obtained by summing the doses received from external gamma exposure and from inhalation of airborne dust reported in Tables 79 and 80, respectively. The results, given in Table 81, indicate that the total effective doses received by workers in the manufacture and disposal of rare earth products are generally less than 1 mSv.

11.3.4. Measures to reduce doses

Airborne dust concentrations can be significant in workplaces for the production of glass polishing powders, special glass and permanent magnets. In such workplaces, it would appear that workers are provided with respiratory protection. However, it is reported in Ref. [192] that the eagerness of the workers to get on with the job has sometimes led to them not wearing the protective respiratory equipment that they have been provided with and trained to use.

11.4. PUBLIC EXPOSURE

Gamma dose rates at the site boundaries of facilities for the manufacture of rare earth products from monazite and bastnäsite have been measured [190]. Dose rates were found to be similar to natural background levels.

Discharges from facilities involved in the use of cerium concentrate to produce glass polishing compounds and as an additive to special glass represent a

TABLE 81. TOTAL EFFECTIVE DOSES RECEIVED BY WORKERS IN THE PRODUCTION AND DISPOSAL OF RARE EARTH PRODUCTS

	Annual committed effective dose (mSv)			Ref.
	Gamma radiation	Dust inhalation	Total	
Glass industry				
Glass polishing powder	0.2	0.16	0.36	[193]
	0.8	0.09	0.89	[189]
	0.36	0.02	0.38	[194]
Special glass	0.8	0.60	1.4	[189]
	0.36	0.12	0.48	[194]
Metal and alloy additives	0.05	—	0.05	[142]
Health products, paint, spa bathing elements	0.3	—	0.3	[190]
Abrasives	0.4	—	0.4	[190]
Samarium–cobalt magnets				
Production of alloy	—	0.001–0.067	0.001–0.067	[190]
Manufacture of magnets	—	0.006–0.038	0.006–0.038	[190]
Disposal of used rare earth products in a landfill facility				
Waste collector	0.005	0,001	0.006	[142]
On-site worker	0.0006	0.0002	0.0008	[142]

potential exposure pathway to members of the public. Doses from such discharges have been estimated using a conservative modelling approach [189]. The results are shown in Table 82. Calculated annual doses are very low indeed — less than 0.002% of the public dose limit of 1 mSv.

Estimates of the doses received by members of the public through the use of glass products containing rare earths are reported in Ref. [142]:

(a) An analysis of the radiological impacts on the public from the wearing of pink tinted eyeglasses containing cerium oxide with a 0.05% thorium content took into consideration exposure to the eye, primarily from irradiation by alpha particles, and whole body exposure from photons. Annual doses, estimated on the assumption that an individual wears eyeglasses for 16 h/d, were determined to be an equivalent dose to the cornea from alpha particles of about 40 mSv, an effective dose from

TABLE 82. DOSES RECEIVED BY MEMBERS OF THE PUBLIC FROM DISCHARGES ASSOCIATED WITH THE MANUFACTURE OF RARE EARTH PRODUCTS FOR THE GLASS INDUSTRY [189]

Dominant exposure pathway	Annual effective dose (μSv)	
	Glass polishing compound	Special glass
Inhalation of dust discharged to the atmosphere	0.0035	0.00067
Ingestion of water discharged to a river	0.036	0.0023
Ingestion of molluscs contaminated by water discharged to the sea	0.016	0.0012

Note: The values of annual effective dose are those received in the 50th year of continuous discharge.

irradiation of the cornea with alpha particles of ≤4 μSv (assuming a tissue weighting factor of $\leq 10^{-4}$) and an effective whole body dose from photons of about 0.2 μSv;

(b) Television faceplates contain about 0.2–0.3% cerium oxide to suppress discoloration of the glass over time. The annual effective dose received by a viewer as a result of gamma exposure has been conservatively estimated to be less than 0.01 μSv. For computer screens, the annual effective dose was estimated to be 0.03 μSv for an average person and up to twice this value for those individuals who use a computer at work;

(c) An estimate has been made of the dose received by a motorist from the use of cerium in specialty automotive glass to attenuate UV radiation and reduce solar heating. The annual effective dose was estimated to be 2 μSv for an average motorist and 7 μSv for a commercial driver.

Doses received by members of the public from the landfill disposal of rare earth products have been estimated. In one study [142], it was assumed that the annual rate of disposal corresponded to 270 kg of cerium concentrate containing 0.25% thorium. In another study [189], the landfill disposal of 2000 kg/a of used glass polishing powders and 1000 kg/a of used special glass came under consideration. The results are provided in Table 83. For individuals associated with future intrusion scenarios, calculated doses varied from <0.01 to 20 μSv/a. For off-site individuals, the dose was not expected to exceed 0.01 μSv/a.

TABLE 83. DOSES RECEIVED BY MEMBERS OF THE PUBLIC FROM THE LANDFILL DISPOSAL OF RARE EARTH PRODUCTS

	Primary exposure pathway	Annual effective dose (μSv)
Disposal of 270 kg/a of Ce concentrate [142]		
Off-site individual	Inhalation of dust	<0.01
Off-site individual, 30 years after closure	Ingestion of well water	<0.01
Intrusion 30 years after closure, residential	External gamma, inhalation of dust and radon, ingestion of soil	<0.01
Disposal of 2000 kg/a of used glass polishing powder [189]		
Off-site individual after closure[a]	Inhalation of dust	0.0058
Future intrusion, drilling[b]	Inhalation of dust	20
Future intrusion, excavation[b]	External gamma	5.6
Future intrusion, residential[b]	External gamma	2.9
Disposal of 1000 kg/a of special glass [189]		
Off-site individual after closure[a]	Inhalation of dust	0.000029
Future intrusion, drilling[b]	Inhalation of dust	0.32
Future intrusion, excavation[b]	External gamma	0.045
Future intrusion, residential[b]	External gamma	0.022

[a] Doses are those received 100 000 years after the site is closed.

[b] Intrusion occurs 300 years after closure of the landfill site, assuming all site records have been lost.

11.5. REGULATORY CONSIDERATIONS

Some rare earth products, such as those used in the glass industry, are manufactured using low purity rare earth materials. These materials may have radionuclide activity concentrations exceeding 1 Bq/g. One case is reported of products being manufactured directly from monazite with a ^{232}Th activity concentration of about 300 Bq/g. Consequently, manufacturing operations involving low purity rare earth feedstocks may need to be considered for regulatory control.

Annual effective doses received by workers are reported to be generally less than 1 mSv, even in situations where the thorium content of the process material is high. However, it would appear that measures to reduce dust inhalation, including the use of respiratory protective equipment where necessary, are often adopted as a matter of course. Doses received by members of the public, including doses received through the use and eventual disposal of products, are insignificant.

Based on this information, it can be concluded that, where low purity process materials are used, some form of control over dust inhalation is likely to be needed. In some workplaces this might already be required under general OHS regulations and no further regulatory control might be necessary. Where this is not the case, the appropriate regulatory option might be to require authorization in the form of a registration.

With the obvious exception of monazite used as a feedstock, the activity concentrations of all the process materials are less than 10 Bq/g and the transport of such material would be outside the scope of the Transport Regulations.

Appendix I

THORIUM AND URANIUM DECAY SERIES

The ^{232}Th and ^{238}U series are the main decay series of interest. The ^{235}U decay series is less important for radiation protection purposes, except for the radionuclide ^{227}Ac, which can contribute significantly to inhalation exposure. If necessary, the presence of ^{235}U (and, by implication, its decay progeny) can be taken into account on the basis of the abundances of ^{235}U and ^{238}U in natural uranium (0.711 and 99.284%, respectively) — the corresponding ^{235}U:^{238}U activity ratio is 0.046. Details of the ^{232}Th and ^{238}U decay series, as reported in Ref. [185], are given in Tables 84 and 85.

TABLE 84. THORIUM-232 DECAY SERIES

	Half-life	Mode of decay[a]	Gamma energy (keV)[b]
^{232}Th	1.405×10^{10} a	Alpha	
^{228}Ra	5.75 a	Beta	
^{228}Ac	6.15 h	Beta	911.204 (25.8%), 968.971 (15.8%)
^{228}Th	1.912 a	Alpha	
^{224}Ra	3.66 d	Alpha	240.986 (4.10%)
^{220}Rn	55.6 s	Alpha	
^{216}Po	0.145 s	Alpha	
^{212}Pb	10.64 h	Beta	238.632 (43.6%)
^{212}Bi	60.55 min	Beta 64.06% Alpha 35.94%	727.330 (6.67%)
^{212}Po	0.299 μs	Alpha	
^{208}Tl	3.053 min	Beta	583.191 (84.5%), 2614.533 (99.16%)
^{208}Pb	Stable	—	

[a] Only major modes of decay are shown.

[b] Only major gamma emissions of interest are shown.

TABLE 85. URANIUM-238 DECAY SERIES

	Half-life	Mode of decay[a]	Gamma energy (keV)[b]
^{238}U	4.468×10^9 a	Alpha	
^{234}Th	24.10 d	Beta	63.29 (4.8%), 92.38–92.8 (5.6%)
^{234m}Pa	1.17 min	Beta	1001.03 (0.837%)
^{234}U	245 700 a	Alpha	
^{230}Th	75 380 a	Alpha	
^{226}Ra	1600 a	Alpha	186.211 (3.59%)
^{222}Rn	3.8235 d	Alpha	
^{218}Po	3.10 min	Alpha	
^{214}Pb	26.8 min	Beta	351.932 (37.6%)
^{214}Bi	19.9 min	Beta	609.312 (46.1%), 1764.491 (15.30%)
^{214}Po	164.3 µs	Alpha	
^{210}Pb	22.20 a	Beta	46.539 (4.25%)
^{210}Bi	5.012 d	Beta	
^{210}Po	138.376 d	Alpha	
^{206}Pb	Stable	—	

[a] Only major modes of decay are shown.

[b] Only major gamma emissions of interest are shown.

Appendix II

RARE EARTH ELEMENTS

Table 86 provides details of the 15 lanthanides and related elements scandium and yttrium, which occur with the lanthanides in natural minerals and have similar chemical properties.

TABLE 86. RARE EARTH ELEMENTS

	Symbol	Atomic number	Atomic weight	Abundance	
				Value (ppm)	Rank
Lanthanum	La	57	138.9	30	28th
Cerium	Ce	58	140.12	60	26th
Praseodymium	Pr	59	140.98	7	37th
Neodymium	Nd	60	144.24	25	27th
Promethium	Pm	61	145	4.5×10^{-20}	—
Samarium	Sm	62	150.4	5	40th
Europium	Eu	63	151.96	1	50th
Gadolinium	Gd	64	157.25	4	41st
Terbium	Tb	65	158.93	0.7	58th
Dysprosium	Dy	66	162.5	3.5	42nd
Holmium	Ho	67	194.93	0.8	55th
Erbium	Er	68	167.26	2.3	43rd
Thulium	Tm	69	168.93	0.32	61st
Ytterbium	Yb	70	173.04	2.2	44th
Lutetium	Lu	71	174.97	0.4	59th
Scandium	Sc	21	44.96	8	36th
Yttrium	Y	39	88.91	30	29th

Appendix III

RARE EARTH, THORIUM AND URANIUM IN RARE EARTH ORES

TABLE 87. MIXED BASTNÄSITE–MONAZITE ORE, BAYAN OBO, CHINA

Ref.	Concentration (%)		
	REO	ThO_2	U_3O_8
[75]	5		
[74]		0.07[a]	0.0002[a]
[11]	1–7.85; typically 2.9		
[26]		0.04	
[148, 196, 197]	Average 6		
[198]	3–6		
[148]	5.2–6; locally >10		

[a] Derived from Th-232 activity concentration assuming that 1 g of ThO_2 contains 3566 Bq of Th-232.

TABLE 88. HEAVY-MINERAL SAND

Ref.	Location of deposit	Concentration (%)		
		REO	ThO_2	U_3O_8
[87]	Australia		0.0006–0.008[a]	0.0004–0.001[a]
[199]	Brazil	2.2[b]	0.18[b]	0.009[b]
[11]	China	0.5–1		
[103]	India	0.66	0.098	0.0045
[100]	India		0.004–0.20[c]	
[86]	India		0.008–0.16	0.0003–0.006
[84]	India, Manavalakurichi		0.27–0.4[b]	
[200]	Vietnam		0.011–0.015[a]	0.002–0.003[a]
[201]	General	0.1–5		

[a] Derived from the elemental concentration.

[b] Derived from the monazite content of the raw sand and the composition of the monazite.

[c] Derived from Th-232 activity concentrations assuming that 1 g of ThO_2 contains 3566 Bq of Th-232.

TABLE 89. BASTNÄSITE ORE

Ref.	Location of deposit	Concentration (%)		
		REO	ThO_2	U_3O_8
[10]	USA, Mountain Pass	5–10		
[202]	USA, Mountain Pass	~9		
[143]	USA, Mountain Pass	5–8		
[141]	USA, Mountain Pass		0.02	0.002
[197]	USA, Mountain Pass	7.67		
[142]	USA, Mountain Pass		0.02–0.1	0.002
[11]	USA, Mountain Pass	3–10 Average 7		
[198]	USA, Mountain Pass	8–12 Average 9.3		
[148]	USA, Mountain Pass	8.9		
[11]	China, Weishan (Shandong Province) and Maoniuping (Sichuan Province)	0.5–5		
[148]	China, Weishan (Shangdong Province)	Average 1.6		
[148]	China, Maoniuping (Sichuan Province	Average 2		
[74]	China, Maoniuping (Sichuan Province)		0.04[a]	0.004[a]

[a] Derived from Th-232 and U-238 activity concentrations assuming that 1 g of ThO_2 contains 3566 Bq of Th-232 and 1 g of U_3O_8 contains 10470 Bq of U-238.

TABLE 90. MOUNT WELD ORE, AUSTRALIA

Ref.	Concentration (%)		
	REO	ThO_2	U_3O_8
[79]	18	0.075	0.003
[75]	9.9–15.7, average 14.3		
[11]	Average ~10	<0.3	
[197]	Typical 10–25, maximum 42%		
[203]	9.7 (4 in southern zone)		
[148]	11.2		

TABLE 91. RARE EARTH BEARING CLAYS FROM CHINA

Ref.	Concentration (%)		
	REO	ThO_2	U_3O_8
[72]	A few tenths		
[75, 197]	1		
[10]	0.15 and 0.3–0.4		
[11, 75, 148]	0.05–0.2		
[204]		0.005	0.005
[205]	~0.1		

Appendix IV

INDIVIDUAL RARE EARTHS IN RARE EARTH MINERALS

TABLE 92. BASTNÄSITE AND ION ADSORPTION CLAYS

	Proportion of total REO content (%)								
	Bastnäsite				Ion adsorption clays, China				
	China			Mountain Pass, USA [197]	LongNan, Jiangxi			Xunwu, Jianxi [10]	Xin Fen [75]
	Bayan Obo [75]	Shandong [206]	Sichuan [206]		[197]	[75]	[10]		
La	26.5	35	27.5	33.2	43.37	7.8	2.18	29.84	26.2
Ce	50.8	50.7	50	49.1	2.38	2.4	<1.09	7.18	1.9
Pr	4.96	3.7	4.5	4.34	9.02	2.4	1.08	7.14	6
Nd	15.4	9.5	14	12	31.65	9	3.47	30.18	21.1
Sm	1.1	0.45	1.25	0.79	3.9	3	2.34	6.32	4.5
Eu	0.21	0.05	0.25	0.12	0.5	0.03	<0.1	0.51	0.71
Gd	0.6	0.22	0.58	0.17	3	4.4	5.69	4.21	4.8
Tb	0.03	0.03	0.04	0.02	Trace	0.9	1.13	0.46	0.77

TABLE 92. BASTNÄSITE AND ION ADSORPTION CLAYS (cont.)

	Proportion of total REO content (%)								
	Bastnäsite				Ion adsorption clays, China				
	China			Mountain Pass, USA [197]	LongNan, Jiangxi			Xunwu, Jianxi [10]	Xin Fen [75]
	Bayan Obo [75]	Shandong [206]	Sichuan [206]		[197]	[75]	[10]		
Dy	0.1	—	0.11	0.03	Trace	5.3	7.48	1.77	4.1
Ho	Trace	0.02	0.05	0.01	Trace	1.4	1.6	0.27	0.8
Er	Trace	0.01	0.07	0.00	Trace	3.6	4.26	0.8	2
Tm	Trace	—	—	0.00	Trace	Trace	0.6	0.13	Trace
Yb	Trace	0.11	0.032	0.00	0.26	2.7	3.34	0.62	1.6
Lu	Trace	—	—	0.00	0.1	0.3	0.47	0.13	0.2
Y	0.2	—	0.76	0.09	8	56.2	64.1	10.07	25.1

TABLE 93. MONAZITE, XENOTIME, MOUNT WELD ORE AND LOPARITE

	Proportion of total REO content (%)									
	Monazite				Xenotime				Mount Weld ore, Australia [75]	Loparite, Russian Federation [197]
	Heavy-mineral sand			Rock, South Africa [207]	Australia		Malaysia [10]	China [10]		
	Australia [197]	India [11]	China [10]		[10]	[206]				
La	23.89	22.0	23.35	21.63	3.5	11.4	0.5	1.2	25.5	25
Ce	46.02	46.0	45.69	46.55	7.2	22.4	5	8	46.74	53
Pr	5.04	5.5	4.16	5.00	1	2.58	0.7	0.6	5.32	6
Nd	17.38	20.0	15.74	16.66	3.5	7.85	2.2	3.5	18.5	14
Sm	2.53	2.5	3.05	2.50	1.9	1.86	1.9	2.15	2.27	0.9
Eu	0.05	0.016	0.1	0.08	0.1	0.07	0.2	<0.2	0.44	0.08
Gd	1.49	1.2	2.03	1.55	0.3	2.18	4	5	0.75	0.56
Tb	0.04	0.06	0.1	0.08	0.1	0.47	1	1.2	0.05	0.37
Dy	0.69	0.18	1.03	0.67	7.9	4.46	8.7	9.1	0.12	0.12
Ho	0.05	0.02	0.1	0.05	1.8	1.01	2.1	2.6	Trace	0.08
Er	0.21	0.01	0.51	0.08	11	3.86	5.4	5.6	Trace	0.016
Tm	0.02	Trace	0.51	0.07	1	0.61	0.9	1.3	Trace	0.003

TABLE 93. MONAZITE, XENOTIME, MOUNT WELD ORE AND LOPARITE (cont.)

	Proportion of total REO content (%)									
	Monazite				Xenotime				Mount Weld ore, Australia [75]	Loparite, Russian Federation [197]
	Heavy-mineral sand			Rock, South Africa [207]	Australia		Malaysia [10]	China [10]		
	Australia [197]	India [11]	China [10]		[10]	[206]				
Yb	0.12	Trace	0.51	0.07	5.5	4.04	6.2	6	Trace	0.008
Lu	0.04	Trace	0.1	0.01	0.2	0.59	0.4	1.8	Trace	0.002
Y	2.41	0.45	3.05	5.00	55	36.6	60.8	59.3	0.25	0.008

APPENDIX V

LOWER LIMITS OF DETECTION OF INHALED THORIUM

TABLE 94. LOWER LIMITS OF DETECTION FOR MEASUREMENT TECHNIQUES BASED ON THE ANALYSIS OF EXCRETA AND BLOOD

Method	Measurement technique	Lower limit of detection of Th-232	Ref.
Urine analysis	Alpha spectrometry	0.3–1.5 mBq/d	[208]
	Alpha spectrometry	46 ng/d	[209]
	Alpha spectrometry	1 mBq/L	[210]
	Alpha spectrometry	0.1 mBq/L	[211]
	Alpha spectrometry	1–2 mBq/L	[50]
	Alpha spectrometry	10 mBq/L	[212]
	NAA	0.06 mBq/d	[208]
	NAA	0.04 mBq	[213]
	NAA	0.025 ng/d	[46, 55, 209]
	Fluorimetry	10 ng/d	[209]
	Spectrophotometry	100 ng/d	[209]
	ICP–MS[a]	0.003 ng/d	[209]
	ICP–MS	7.4×10^{-5} ppm (0.3 mBq/L)	[210]
	ICP–MS	0.0004 mBq/L	[211]
	ICP–MS	~1 ng per 50 mL (~0.08 mBq/L)	[50]
	SSNTD[b] (alpha tracks)	0.1 mBq/cm^2	[209]
Faecal analysis	Alpha spectrometry	20 mBq/d	[208]
	Alpha spectrometry	1 mBq/g ash	[214]
	Alpha spectrometry	10 mBq	[212]
	NAA	20 mBq/d	[208]
Blood analysis	NAA	0.025 ng	[46, 55]

[a] ICP–MS: Inductively coupled plasma mass spectroscopy.

[b] SSNTD: Solid state nuclear track detection.

TABLE 95. LOWER LIMITS OF DETECTION FOR MEASUREMENT TECHNIQUES BASED ON AIR SAMPLING, DIRECT IN VIVO COUNTING AND THORON IN BREATH

Method	Measurement technique	Lower limit of detection of Th-232	Ref.
Stationary air sampling	Large area proportional counter[a]	0.4 mBq/m^3	[208]
Personal air sampling	Large area proportional counter[b]	2 mBq/m^3	[208]
Lung counting	Gamma spectrometry	10–30 Bq	[208]
		10–25 Bq	[41]
		15 Bq class S	[53]
Whole body counting	Gamma spectrometry	30–100 Bq	[208]
		40 Bq (lung 15 Bq)	[34]
		Lung: 45 Bq	[40]
		52 Bq (lung 12 Bq)	[27]
		40 Bq class S	[53]
Thoron in breath	Double filter system	14 Bq	[31, 40]
		10 Bq	[215]
		6 Bq[c]	[34]
	Electrostatic collection, alpha spectrometry	1–3 Bq	[208]
		10 Bq	[41]
		1.84 Bq	[26]
		1.2 Bq	[33]
		1.4 Bq[d]	[32]

[a] Flow rate ≥20 m^3/h, sample volume ≥750 m^3.

[b] Flow rate 0.2 m^3/h, sample volume 10 m^3.

[c] Derived from a lower limit of detection of 0.27 Bq of emanating equivalent activity of ^{224}Ra at the mouth and assuming an exhalation rate of 4.7% (see Section 3.3.1.2).

[d] Derived from a lower limit of detection of 0.068 Bq of emanating equivalent activity of ^{224}Ra at the mouth and assuming an exhalation rate of 5% (see Section 3.3.1.2).

Appendix VI

COMPOSITION OF HEAVY-MINERAL SAND

Data on the composition of heavy-mineral sand deposits in various countries are given in Table 96. The concentrations of individual heavy minerals (including, in particular, the rare earth bearing mineral monazite) are expressed as mass concentrations in raw sand. An appropriate correction factor has been applied in cases where literature values are expressed as concentrations in the heavy-mineral component.

TABLE 96. MINERAL COMPOSITION OF HEAVY-MINERAL SAND DEPOSITS

Location of deposit	Type of deposit	Total heavy-mineral content (%)	Individual heavy-mineral content (%)						Ref.
			Monazite	Ilmenite	Rutile	Zircon	Garnet	Sillimanite	
Australia									
East coast	Coastal	8–10		5.6	1.9	1.5			[11]
West coast	Coastal	5–8	1.0	4.2	0.6	1.6			[11]
West coast	Coastal		0.1						[216]
West coast	Coastal	Typical 7–8 Range 2–20							[88]
West coast (Eneabba)	Coastal	6	0.03–0.4						[148]
West coast (Capel)	Coastal	>6							[148]
Brazil	Coastal (beach)		3	≤96	0.5	2.5			[199]
India (east coast)									
Overall	Coastal	8–20							[84]
Orissa (Chatrapur)	Coastal	20–20.5	0.27–0.4	8.8	0.38	0.31	6.7	3.4	[11]
Orissa (Chatrapur)	Coastal (dune)	20	0.5						[84]
Andhra Pradesh	Coastal	25–30	0.2						[84]
Andhra Pradesh (Bhimilipatnam)	Coastal	20	0.1						[11]
Andhra Pradesh (Narasapur)	Coastal (beach)	14.7[a]							[217]

TABLE 96. MINERAL COMPOSITION OF HEAVY-MINERAL SAND DEPOSITS (cont.)

Location of deposit	Type of deposit	Total heavy-mineral content (%)	Individual heavy-mineral content (%)						Ref.
			Monazite	Ilmenite	Rutile	Zircon	Garnet	Sillimanite	
India (south and west coasts)									
Tamil Nadu	Coastal	25–30	0.2						[84]
Tamil Nadu	Coastal (beach)		0.2–1.5						[99]
Tamil Nadu (Manavalakurichi)	Coastal (beach)	64	3–4	45–50	2–3	4–6	7–14	2–3	[84]
Tamil Nadu (Manavalakurichi)	Coastal (dune)	50	3.3	35	1.35				[84]
Tamil Nadu (Manavalakurichi)	Coastal	40–60[b]	2[c]						[11]
Tamil Nadu	Inland	10 (ave.)	“Low”						[11]
Tamil Nadu	Inland	Up to 10	0.06–0.35	3–4					[84]
Kerala (Chavara)	Coastal (beach)	73	0.5–1	60–70		5–8	4–7	1.15	[84]
Kerala (Chavara)	Coastal (dune)	39 (25–55)[b,d]	0.7–1.0	24	1.8	2.0	5.5	3.5	[11]
North of Chavara to Ratnagiri	Coastal (beach)	20	0.06						[84]
Neendakara–Kayankulam bar	Coastal (sea bed)	5	0.05						[84]
Sri Lanka	Coastal		0.4	62	18				[84]
Vietnam	Coastal (beach)	1.1–7.8[e]	0.1	3–4		0.2–0.4			[100]

[a] The heavy-mineral content includes the rare earth bearing mineral xenotime at a concentration of 0.09–1.32%.

[b] When mining started in the beach areas, the heavy-mineral content was 80–90%.

[c] When mining started in the beach areas, the monazite content was 6%.

[d] The heavy-mineral content includes, in addition to the minerals indicated, leucoxene at a concentration of 0.9%.

[e] In Ref. [100], the total heavy-mineral content is quoted in units of kg/m^3. The values have been converted to% assuming a density of 4500 kg/m^3.

Appendix VII

RARE EARTH CONTENT OF MONAZITE

TABLE 97. RARE EARTH CONTENT OF MONAZITE, BY COUNTRY

Origin	Total rare earth content (% REO)	Ref.
Australia	61.33	[10]
	62.5	[218]
	57–60	[11]
Brazil	59.9	[218]
	≤74	[199]
China	50–60	[72]
	55[a]	[11]
	48–62[b]	[11]
	45.3[b]	[219]
Democratic People's Republic of Korea	42.65	[10]
India	59.5	[218]
	60	[115, 220]
	59.37	[169]
	58	[11]
	59.68	[10]
Malaysia[c]	59.65	[10]
Republic of Korea	60.20	[10]
Thailand[c]	60.20	[10]
United States of America	39.5	[218]
Vietnam	35–40	[111]

TABLE 97. RARE EARTH CONTENT OF MONAZITE, BY COUNTRY (cont.)

Origin	Total rare earth content (% REO)	Ref.
Unspecified	50–62.5	[206]
	60	[10, 184]
	70	[75]
	48	[153]
	60–65	[109]

[a] Monazite from Guangdong Province.
[b] Black monazite from Taiwan, China.
[c] Monazite recovered from tin bearing alluvial deposits (see Section 7).

Appendix VIII

ACTIVITY CONCENTRATIONS IN PROCESSING MATERIALS ASSOCIATED WITH THE MINING AND BENEFICIATION OF HEAVY-MINERAL SAND

Radionuclide activity concentrations in heavy-mineral sand, heavy-mineral concentrate, monazite, other heavy minerals (ilmenite, zircon, rutile, garnet, sillimanite, leucoxene and xenotime), tailings and airborne dust are provided in Tables 98–106. Where indicated in these tables, activity concentrations are derived from mass concentrations of natural thorium and natural uranium or of ThO_2 and U_3O_8. For the purposes of such derivations, it is assumed that 1 g of natural thorium contains 4057 Bq of ^{232}Th, 1 g of natural uranium contains 12 350 Bq of ^{238}U, 1 g of ThO_2 contains 3566 Bq of ^{232}Th and 1 g of U_3O_8 contains 10 470 Bq of ^{238}U.

TABLE 98. HEAVY-MINERAL SAND

Origin	Activity concentration (Bq/g)[a]		Ref.
	Th-232	U-238	
Australia	0.02–0.28	0.03–0.12	[93]
Brazil	0.520	0.099	[221]
India	0.15–7.2		[100]
	0.3–6.0	0.04–0.70	[86]
(Manavalakurichi)	9.6–14.3[b]		[84]
Vietnam	*0.39–0.55*	*0.21–0.35*	[200]

[a] Values in italics are derived from mass concentrations.

[b] Derived from the monazite content of the raw sand (3–4%) and the ThO_2 content of the monazite (9–10%).

TABLE 99. HEAVY-MINERAL CONCENTRATE

Origin	Activity concentration (Bq/g)[a]		Ref.
	Th-232	U-238	
Australia	1.36–1.60	2.00–2.23	[95]
	0.3–3	<0.1–0.8	[93]
Bangladesh	4.68	2.58 (Ra-226)	[222]
Brazil	7.58	2.1	[221]
Vietnam	*0.68–1.0*	*10.2–13.7*	[200]

[a] Values in italics are derived from mass concentrations.

TABLE 100. MONAZITE

Origin	Activity concentration (Bq/g)[a]		Ref.	Remarks
	Th-232	U-238		
Australia	251.6	30.34	[223]	90% concentrate
	240	*30*	[92]	
	230	*19*	[92]	
	24	6.4	[95]	65% concentrate
	100	17	[95]	85% concentrate
	146.5(144–149)	21(20–22)	[95]	95% concentrate
	210	—	[218]	
	200–280	*12–60*	[96]	
	200–280	*12–40*	[87]	
	40–220	*6–30*	[87]	Impure concentrate
	230	*40*	[10, 11]	
	210–240	—	[11]	
	180	—	[11]	
	40–250	6–30	[85]	
Brazil	*240*	*22*	[218]	
	210	*40*	[180]	
	214	*31*	[221]	

TABLE 100. MONAZITE (cont.)

Origin	Activity concentration (Bq/g)[a]		Ref.	Remarks
	Th-232	U-238		
China	*180–250*	*40–70*	[72]	
	140		[11]	Guangdong Province
	7–15		[11]	Black monazite, Taiwan
	28.4–31.6	5.06–5.85	[219]	Black monazite, Taiwan
Democratic People’s Republic of Korea	*163*	*19*	[10]	
India	*317 (267–320)*	*37*	[169]	West coast
	320–360		[84]	Beach and dune sand, Manavalakurichi
	320		[84]	Beach sand, Chevara
	340		[84]	Sea bed, Neendakara–Kayankulam bar
	290	*37*	[10, 220]	
	290–360		[224]	Manavalakurichi
	350	*28*	[218]	
	320	*37*	[11, 179]	
	370	38	[128]	
	340	*39*	[10, 11]	

TABLE 100. MONAZITE (cont.)

Origin	Activity concentration (Bq/g)[a]		Ref.	Remarks
	Th-232	U-238		
India	*230*	*40*	[11]	
	300–340		[82]	
	290–320	*37*	[115]	South coast
	350–400	35	[86]	
Republic of Korea	*205*	*47*	[10]	
United States of America	*~140*		[142]	
	120	*16*	[218]	
Unspecified	*340*	*30–40*	[184]	
	8–800	6–37	[225]	
	40–800	6–37	[226]	
	210	*26*	[153]	
	60–700	0–175	[227]	
	295	—	[228]	
	110–360	*<50*	[109]	
	180–250	*10–30*	[222]	

TABLE 100. MONAZITE (cont.)

Origin	Activity concentration (Bq/g)[a]		Ref.	Remarks
	Th-232	U-238		
Unspecified	*220–1000*		[201]	
	180–360		[10]	
	8–300	6–40	[15]	
	290–360	37	[206]	

[a] Values in italics are derived from mass concentrations.

TABLE 101. ILMENITE

Origin	Activity concentration (Bq/g)[a]		Ref.
	Th-232	U-238	
Australia	0.064 (0.035–0.090)	0.050 (0.031–0.071)	[95]
	0.2–2.0	*<0.1–0.4*	[87]
	0.73–1.8	*0.09–0.25*	[119]
Bangladesh[b]	0.388	0.348 (Ra-226)	[222]
Brazil	0.62	0.13	[221]
India	0.23–0.61	—	[82]
	0.57 (0.23–0.86)	—	[104]

[a] Values in italics are derived from mass concentrations.
[b] Incomplete separation of monazite.

TABLE 102. ZIRCON

Origin	Activity concentration (Bq/g)[a]		Ref.
	Th-232	U-238	
Australia	0.62 (0.57–0.66)	3.90 (3.60–4.27)	[95]
	0.6–1.0	*1.85–3.7*	[96]
	0.6–1.2	*1.85–3.7*	[87]
	0.7–0.8	*2.5–3.0*	[119]
	0.6–1.2	1–4	[93]
Bangladesh[b]	1.3	6.4 (Ra-226)	[222]
Brazil	0.5	2.3	[221]
India	0.56 (0.3–0.7)	2.5 (1.6–3.4)	[103]

[a] Values in italics are derived from mass concentrations.
[b] Incomplete separation of monazite.

TABLE 103. RUTILE

Origin	Activity concentration (Bq/g)[a]		Ref.
	Th-232	U-238	
Australia	0.07 (0.06–0.09)	0.56 (0.53–0.64)	[95]
	0.20–0.41	*0.12–0.37*	[96]
	0.20–1.4	*<0.12–0.25*	[87]
	0.12–0.26	*0.25–0.62*	[119]
Bangladesh[b]	9.3–14.0	5.9–7.4 (Ra-226)	[222]
Brazil	1.6	1.24	[221]
India	0.10 (0.08–0.15)	—	[104]

[a] Values in italics are derived from mass concentrations.

[b] Incomplete separation of monazite.

TABLE 104. OTHER HEAVY MINERALS

Mineral and origin	Activity concentration (Bq/g)[a]		Ref.
	Th-232	U-238	
Garnet			
India	0.25 (0.24–0.35)		[103]
Bangladesh[b]	7.9	4.0	[222]
Sillimanite, India	0.10 (0.06–0.15)		[103]
Leucoxene, Australia	*0.32–2.8*	*0.25–0.6*	[87]
	0.85–1.1	*0.12–0.5*	[119]
	0.3–3	0.2–0.6	[93]
	0.4–5	*0.3–1*	[88]
Xenotime			
Australia	*~60*	*~50*	[87]
Malaysia	*30*	*85*	[10]

[a] Values in italics are derived from mass concentrations.

[b] Incomplete separation of monazite.

TABLE 105. TAILINGS

Origin	Activity concentration (Bq/g)[a]		Ref.
	Th-232	U-238	
Primary separation process			
Australia	*<0.2*	*<0.2*	[88]
Australia	*<0.2*	*<0.12*	[87]
Brazil	0.044	0.010	[221]
India, dredge mining and wet concentration	0.036 (0.027–0.045)[b]	0.018 (0.013–0.022)[b]	[82]
India, wet concentrate upgrading plant and beach washings plant	0.052 (0.030–0.076)[b]	0.026 (0.015–0.038)[b]	[82]
India, mining	0.05[b]	0.02[b]	[86]
India, wet concentration	0.08[b]	0.04[b]	[86]
Secondary separation process			
Australia	*0.8–8*	*0.1–1.6*	[88]
Australia, monazite circuit	*24*	*2–10*	[88]
Australia, pre-1989 tailings stored on-site	*4.9 (0.4–122)*	*1.1 (0.1–12)*	[96]
Brazil	2.09	1.35	[221]
India	8.4 (6.9–10.2)[b]	2.1 (1.7–2.5)[b]	[82]
India, Tuticorin district, plant 1	2.253	0.337	[105]
India, Tuticorin district, plant 2	3.540	0.227	[105]
India, Tuticorin district, plant 3	0.505	0.055	[105]
India, Tuticorin district, plant 4	2.298	0.737	[105]
India	8[b]	2[b]	[86]
India	3.9 (2.3–5.4) (Ra-228)	—	[104]

[a] Values in italics are derived from mass concentrations.

[b] Derived from the gross alpha activity concentration assuming that the uranium and thorium decay chains are in equilibrium and that the activity concentration of Th-232 was twice that of U-238 for primary separation tailings and four times that of U-238 for secondary separation tailings.

TABLE 106. AIRBORNE DUST GENERATED VIA THE DRY SEPARATION PROCESS

Origin	Activity concentration (Bq/g)[a]		Ref.
	Th-232	U-238	
Australia			
Dust within plants	2.8	1.2	[95]
Dust within plant, ilmenite section[b]	2	1	[108]
Pure monazite dust within plant[b]	200	20	[108]
Dust within plant, typical[b]	20	5	[108]
Dust within plants	*1–4*	*0.1–1*	[88]
Dust within plants	*~1–10*	*~0.1–3*	[87]
Stack particulates	*~2–20*	*~0.1–6*	[87]
India			[94]
Dust within Plant 1[b]	0.4–1.1	0.1–0.3	
Dust within Plant 2, monazite section[b]	6.2–10.2	0.6–1.0	
Dust within Plant 2, other sections[b]	1.4–8.0	0.3–2.0	
Vietnam			[100]
Dust within Plant 1	*0.96 (0.53–1.42)*	*0.41 (0.33–0.80)*	
Dust within Plant 2	*0.97 (0.57–1.50)*	*0.57 (0.31–0.93)*	
Dust within Plant 3	*0.91 (0.39–1.83)*	*0.33 (0.12–0.74)*	
Dust within Plant 4	*0.89 (0.45–1.79)*	*0.44 (0.22–1.05)*	

[a] Values in italics are derived from mass concentrations.

[b] Derived from the gross alpha activity concentration using the relationship provided in Fig. 3 and assuming that the activity concentration of Th-232 was twice that of U-238 for non--monazite dust, ten times that of U-238 for monazite dust and four times that of U-238 for a combination of the two.

Appendix IX

GAMMA DOSE RATES IN THE MINING AND BENEFICIATION OF HEAVY-MINERAL SAND

Gamma dose rates provided in Tables 107–110 were generally measured at a distance of 1 m from the source, unless otherwise stated.

TABLE 107. LOCAL NATURAL BACKGROUND IN THE VICINITY OF MINING AND BENEFICIATION OPERATIONS IN INDIA

	Absorbed dose rate (μGy/h)		
General background in plant (office) [169]		1	
Areas of heavy-mineral sand deposits [229]		1–5	
Averages, high natural background areas [81]			
Ayiramthengu, 64 locations		0.2–1.4	
Neendakara, 64 locations		0.2–3	
Kudiraimozhi, 43 locations		0.2–0.9	
Bhimilipatnam, 40 locations		0.2–3	
Background at 3 plant sites [82]	Plant 1	Plant 2	Plant 3
Residential area around plant	0.7–3	0.6–1.5	0.6–1.5
General backround in plant	1–3	0.7–1.5	1.5–3
General background [110]		1–3	
General background in plant [104]		0.5–1	

TABLE 108. MINING OPERATIONS

	Absorbed dose rate (μGy/h)		
Australia, ore body [98]		0.1	
Australia, mining operations [53]		0.1–0.3	
India, 3 sites [82, 115]	Plant 1	Plant 2	Plant 3
Beaches	2–20	1.1–3	2–3
Mining area	2–10	1–5	—
Beaches after mining	0.5–3	0.5–2	—
Refilled area	0.6–0.8	0.2–0.7	0.2–0.7
Refilled and remediated area	0.3–0.8	0.2–0.7	—
India, raw beach sand [86]		0.5–35	
India [103]			
Dune sand deposits		0.4–0.9	
Inland sand deposits		0.25–0.3	
Beach sand deposits, <0.5% monazite		0.3–1.4	
Beach sand deposits, >0.5% monazite		1.1–20	
Beach sand dredge sites		1–2	
Dredge mining areas		2–5	
Dredge		0.8–1	
India [110]			
Mining areas		1–4	
Raw sand		1–4	
Raw sand		0.5–5	

TABLE 109. PRIMARY CONCENTRATION OPERATIONS

	Absorbed dose rate (µGy/h)		
Australia, wet concentration [53]		0.2–1	
Australia [216]			
Primary separation tailings		0.2	
Heavy-mineral concentrate stockpiles		1–2	
India, 3 sites [82]	Plant 1	Plant 2	Plant 3
Raw sand concentrate storage area	3–15	1	3
Sand tailings	0.6	1–1.5	0.9–1
India [86]			
Mine tailings		0.5	
Pre-concentration tailings		0.4	
India [103]			
Pre-concentration spirals		1–2	
Trommels		0.8–1	
Pre-concentrated sand		2–8	
Concentrate yards, chest level		5–15	
Concentrate storage sheds		5–20	
Pre-concentration tailings		0.4	
Heavy-mineral upgrading plant		1–3	
Upgraded sand concentrate		4–20	
India [104]			
Mineral concentrate storage area		2–5	
Mineral concentrate upgradation plant		0.6–1	
India [110]			
Concentration plant		1–1.5	
Sand concentrate (dump yard)		4–15	
Sand concentrate in storage		5–20	

TABLE 110. SECONDARY CONCENTRATION OPERATIONS

	Absorbed dose rate (μGy/h)
Australia [101]	
Primary circuits	1 (0.5–2)
Monazite air tables	2–10
Monazite air tables and bagging station	10 or more
Some locations inside monazite bag store	100 or more
Australia [53]	
Primary circuits	0.5–2
Secondary circuits	2–5
Monazite circuits	5–30
Monazite bagging and storage	5–150
Australia [95][a]	0.08–1.4 (average 0.4)
Australia [216]	
Tailings and stockpiled mineral products	‘Several’
Stockpiled monazite	>200
Australia, tailings stockpile > 10t [88]	
Th-232 activity concentration 0.8 Bq/g	<0.5
Th-232 activity concentration 4–8 Bq/g (typical)	2.5–5
Th-232 activity concentration 120 Bq/g	>50
India [169][b]	
Vibrating screen (sand)	5
Vibrating screen (after initial ilmenite removal)	18
Magnetic separation	5
Magnetic separation (catwalk between magnets)	3
Magnetic separation (non-magnetic fraction)	8
Magnetic separation (magnetic fraction)	3
Magnetic separation (ilmenite fraction)	3

TABLE 110. SECONDARY CONCENTRATION OPERATIONS (cont.)

	Absorbed dose rate (μGy/h)		
India [169] (cont.)[b]			
Air table (zircon separation)	2		
Air table (monazite concentrator)	4		
Bag filling station (at the hands)	50		
Bag filling station (chest level)	25		
Store containing about 20 t of monazite	160		
Entrance to monazite store	30		
India [230][b]			
Dry separation area	3–6		
Monazite storage and bag filling areas	100–200		
India, 3 sites [82]	Plant 1	Plant 2	Plant 3
Ilmenite store	1–2	0.5–1.5	0.7–1
Monazite store	50–100	40–80	40–70
Residue storage and disposal area[c]	30–40	10–20	15–17
India, dry separation area [91]			
<0.5% monazite content	1–12 (average 2)		
>0.5% monazite content	1–30 (average 3)		
India [86]			
Monazite	180–250		
Tailings	50		
India [110]			
Screens	2		
Primary high tension separators	4		
Monazite high tension separators	6		
Magnetic separators	2–4		
Monazite air tables	8–20		
Monazite wet circuit	5–30		
Monazite bags	200		
Zircon	2–4		
Ilmenite, rutile, garnet	2		

TABLE 110. SECONDARY CONCENTRATION OPERATIONS (cont.)

	Absorbed dose rate (µGy/h)			
India [104]				
High tension separators	2.0–2.5			
Magnetic separators	1.1–1.8			
Ilmenite, initial separation	1.7–2.8			
Rutile section	0.8–1.5			
Ilmenite final product section	0.5–1.5			
Vietnam, plants in 4 regions [100]	Plant 1	Plant 2	Plant 3	Plant 4
Dry separation areas	1.04	1.22	0.025	1.35
Storage areas	1.56	1.31	0.95	1.15

---

[a] The values refer to dose rates reported in units of microsieverts per hour. As an approximation, it can be assumed that these correspond numerically to absorbed dose rate in units of microgray per hour.

[b] The values are derived from exposure rates reported in units of milliröntgen per hour, assuming that 1 mR/h corresponds to an absorbed dose rate of 10 µGy/h.

[c] The residue in Plants 1 and 2 was subsequently recycled.

Appendix X

AIRBORNE DUST CHARACTERISTICS IN HEAVY-MINERAL DRY SEPARATION PLANTS

TABLE 111. DUST CONCENTRATIONS IN AIR

		Mass concentration (mg/m^3)	
Australia	(n=33) [95][a]	3.3	(0.49–9.09)
Australia	(n=26) [113]	3.8	(1.1–31)
Australia [108]			
1977	(n=165)	16.8	(0.1–453)
1978	(n=743)	10.8	(0.1–220)
1979	(n=804)	9.5	(0.1–262)
1980	(n=484)	9.2	(0.1–41)
1981	(n=516)	9.0	(0.1–52)
1982	(n=437)	8.6	(0.1–147)
1983	(n=348)	5.9	(0.1–36)
1984	(n=391)	6.0	(0.1–47)
1985	(n=458)	8.1	(0.1–593)
1986	(n=582)	6.4	(0.1–162)
1987	(n=1055)	5.1	(0.1–139)
1988	(n=1877)	4.7	(0.1–54)
1989	(n=2581)	3.6	(0.1–65)
1990	(n=2027)	2.7	(0.1–186)
1991	(n=1442)	1.4	(0.1–19)
Brazil [172]			
Magnetic separation		0.56	
Monazite section		0.25	
India [229]			
1986–1987		4–9	
1996		<1	

TABLE 111. DUST CONCENTRATIONS IN AIR (cont.)

	Mass concentration (mg/m^3)			
India, 2 plants [109][b]	Plant 1		Plant 2	
	0.6	(0.03–4.3)	0.5	(0.05–1.1)
India, 2 plants [231]	Plant 1		Plant 2	
Sand screening and storage	0.76	(0.10–2.10)	1.85	(0.13–3.86)
High tension separators	0.96	(0.12–3.33)	1.60	(0.80–3.03)
Magnetic separators	—		1.21	(0.33–2.80)
Monazite air tables	0.86	(0.60–1.10)	0.71	(0.20–2.53)
Ilmenite section	1.45	(0.27–7.51)	1.71	(0.40–2.67)
Zircon section	1.01	(1.13–2.53)	1.40	(0.20–1.87)
Garnet section	—		1.28	(0.50–2.06)
Rutile section	1.90	(0.50–4.67)	1.80	(0.86–3.86)
Leucoxene section	1.85	(0.31–4.60)	—	
General areas	1.00	(0.30–2.60)	—	
Overall	1.22	(0.10–7.51)	1.45	(0.13–3.86)
Vietnam, 4 plants [100]	Plant 1 (n=5)	Plant 2 (n=15)	Plant 3 (n=10)	Plant 4 (n=7)
Screening, gravity separation	2510	4200	1800	2300
Magnetic separation	350	410	300	350
Average	—	500	—	—

---

[a] n denotes the number of samples.

[b] Values are for the respirable fraction. Nearly 75% of the airborne dust was in the non-respirable region.

TABLE 112. THORIUM-232 ACTIVITY CONCENTRATIONS IN AIR

	Th-232 activity concentration in air (mBq/m^3)			
Australia, 2 plants [98]	Plant 1		Plant 2	
Ilmenite section	240	(180–360)	20	(10–50)
Rutile section	130	(50–260)	10	(3–10)
Zircon section	110	(60–170)	30	(5–30)
Monazite section	260	(70–470)	20	(10–30)
Bag store	7	(0–10)		—
Australia [101][a]				
Ilmenite section	10–200			
Rutile and leucoxene section	10–100			
Monazite air tables	70–700			
Mean, west coast operations	~100	(max. 300 in one shift)		
Mean, east coast operations	~10			
Australia [53][a]				
Primary circuits	10–100			
Secondary circuits	20–200			
Monazite section	70–700			
Monazite bagging and store	30–70			
Australia [95]				
Personal sampling (n=33)[a]	9	(2–20)		
High volume sampling (n=5)	12	(7.6–17)		
Australia, prior to implementation of engineering controls) [113][a]				
Engineering (n=359)	20	(1–400)		
Day crew (n=134)	60	(1–500)		
Ilmenite section (n=98)	90	(2–400)		
Zircon section (n=78)	70	(3–300)		
Monazite section (n=159)	150	(1–600)		
Shift foreman (n=242)	40	(1–500)		
Routine tasks (n=27)	60	(30–260)		
Australia, monazite section [51][a]	80	(50–150)		
India, 15 year average [36]	130	(1–4810)		

TABLE 112. THORIUM-232 ACTIVITY CONCENTRATIONS IN AIR (cont.)

	Th-232 activity concentration in air (mBq/m^3)			
India [82]				
Plant 1	8–70			
Plant 2	3–50			
India [91]				
Plant 1	15	(<0.1–50)		
Plant 2	20	(1–110)		
India [104]				
Ilmenite separation	6	(3–29)		
Rutile separation	10	(3–41)		
India, 2 plants [231]	Plant 1		Plant 2	
Sand screening and storage	12	(1–29)	13	(6–24)
High tension separators	16	(2–34)	21	(8–39)
Magnetic separators	—		39	(8–66)
Monazite air tables	—		35	(8–58)
Ilmenite section	23	(1–59)	16	(11–18)
Zircon section	24	(6–40)	9	(4–19)
Garnet section	—		8	(4–18)
Rutile section	16	(10–22)	15	(6–20)
Leucoxene section	20	(9–40)	—	
Monazite bagging, container filling	—		15	(13–18)
General areas	13	(2–40)	—	
Overall	16	(1–59)	19	(4–66)

[a] Derived from the gross alpha activity concentration using the relationship provided in Fig. 3 and assuming that the activity concentration of Th-232 was twice that of U-238 for non-monazite dust, ten times that of U-238 for monazite dust and four times that of U-238 for a combination of the two.

TABLE 113. AIRBORNE DUST ACTIVITY CONCENTRATIONS IN AUSTRALIAN DRY SEPARATION PLANTS BY YEAR

Year	Th-232 activity concentration in air (mBq/m^3)					
1977–1985 [108][a]	Mean					
1977	353					
1978	227					
1979	200					
1980	194					
1981	189					
1982	181					
1983	124					
1984	126					
1985	170					
1986–1992 [107][b]	Mean	Plant A	Plant B	Plant C	Plant D	Plant E
1986	105	12	167	136	26	63
1987	74	15	95	77	25	104
1988	63	11	71	62	24	90
1989	61	33	71	61	26	87
1990	33	27	52	26	28	22
1991	20	5	24	22	30	11
1992	22	4	25	32	19	11
1993–2008 [106][c]	Mean	Maximum				
1993–1994	7.5	520				
1994–1995	4.6	492				
1995–1996	5.3	262				
1996–1997	6.4	409				
1997–1998	7.4	275				
1998–1999	9.2	272				
1999–2000	10.1	193				
2000–2001	10.1	367				
2001–2002	8.5	486				
2002–2003	6.9	370				
2003–2004	8.5	233				

TABLE 113. AIRBORNE DUST ACTIVITY CONCENTRATIONS IN AUSTRALIAN DRY SEPARATION PLANTS BY YEAR (cont.)

Year	Th-232 activity concentration in air (mBq/m^3)	
2004–2005	8.3	176
2005–2006	6.8	323
2006–2007	5.3	337
2007–2008	6.0	206

[a] Derived from mass concentrations assuming a typical gross alpha activity concentration of 170 Bq/g (corresponding to a ^{232}Th activity concentration of about 20 Bq/g).

[b] Derived from gross alpha activity concentrations assuming a Th-232:U-238 activity concentration ratio of 4 (representative of a combination of monazite dust and non-monazite dust).

[c] Derived from gross alpha activity concentrations assuming a Th:U mass concentration ratio of 8 (corresponding to a Th-232:U-238 activity concentration ratio of 2.6), typical of processed heavy-mineral concentrate during this period, when there was no monazite production.

TABLE 114. INTAKE OF ^{232}Th IN AIRBORNE DUST BY WORKERS IN AUSTRALIAN DRY SEPARATION PLANTS *(derived from Ref. [48])*

Worker code	Employment period (d)	Mean daily Th-232 intake, by job category (Bq/d)[a]			
		Plant maintenance	Plant operation		
			Monazite sections	Other sections	Monazite and other sections
1	2381		0.25		
2	2087		0.23		
3	744			0.09	
4	1513			0.14	
5	1095		0.32		
6	1603				0.36
7	367				0.50
8	1570			0.47	
9	1071				0.37
10	1276	0.37			
11	737			0.25	
12	1290	0.35			
13	1094				0.49
14	5682			0.40	
15	4137		0.78		
16	444				0.26
17	2350				0.54
18	2722		0.47		
19	3352	0.24			
20	2953	0.23			
21	664				0.25
22	758			0.15	
23	570			0.19	
24	7387			0.19	

TABLE 114. INTAKE OF ^{232}Th IN AIRBORNE DUST BY WORKERS IN AUSTRALIAN DRY SEPARATION PLANTS *(derived from Ref. [48])* (cont.)

Worker code	Employment period (d)	Mean daily Th-232 intake, by job category (Bq/d)[a]			
		Plant maintenance	Plant operation		
			Monazite sections	Other sections	Monazite and other sections
25	1717			0.09	
26	1325			0.14	
27	1456				1.04
28	3219			0.26	
29	1400			0.53	
30	381		0.53		
31	1093		0.97		
32	528			0.30	
33	892				0.35
34	1848		0.35		
Average	1383	0.30	0.49	0.25	0.46

[a] The data in Ref. [48] were based on the gross alpha activity detected on air sampling filters, assuming Th-232 decay chain equilibrium but ignoring alpha emitters in the U-238 decay chain. The values provided in this table take account of U-238 series contributions by assuming a Th-232:U-238 ratio of 10 for workers in monazite sections, 2 for workers in non-monazite sections and 4 for all other workers (see Fig. 3).

Appendix XI

ACTIVITY CONCENTRATIONS IN HEAVY MINERALS RECOVERED FROM TIN MINING RESIDUES

Radionuclide activity concentrations in monazite, xenotime, zircon and ilmenite recovered from tin mining residues (amang) are provided in Tables 115–118. Where indicated in these tables, activity concentrations are derived from mass concentrations of natural thorium and natural uranium, assuming that 1 g of natural thorium contains 4057 Bq of ^{232}Th and 1 g of natural uranium contains 12 350 Bq of ^{238}U. Some values (as indicated) refer to the activity concentrations of ^{228}Ra or ^{226}Ra rather than ^{232}Th or ^{238}U.

TABLE 115. MONAZITE

Ref.	Activity concentration (Bq/g)		Comment
	Th-232	U-238	
[200]	67	20–21 (Ra-226)	
[130]	265	43.2	
[232]	90–200	20–32	
[167]	330	31	
[233]	83–370	12–37	
[126]	290	25	
[10]	210	25	
[10]	305	46	
[158]	243	37	From mass concentrations
[128]	178	20	From mass concentrations
[167]	327	30.6 (Ra-226)	
[125]	82 (Ra-228)	21 (Ra-226)	30% concentrate

TABLE 116. XENOTIME

Ref.	Activity concentration (Bq/g)		Comment
	Th-232	U-238	
[232]	16–24	68–110	
[167]	37	120	
[233]	41–200	37–87	
[126]	20	62	
[10]	30	85	
[130]	26	110	
[158]	28	247	From mass concentrations
[128]	14 (Ra-228)	71 (Ra-226)	
[167]	37	119 (Ra-226)	
[125]	41 (Ra-228)	93 (Ra-226)	30% concentrate
[200]	13–19	42–45 (Ra-226)	

TABLE 117. ZIRCON

Ref.	Activity concentration (Bq/g)		Comment
	Th-232	U-238	
[232]	2–88	16–21	
[167]	45	16	
[233]	41–57	25–50	
[126]	10	62	
[130]	33	18	
[128]	2.2 (Ra-228)	13 (Ra-226)	
[167]	43	16 (Ra-226)	
[125]	2 (Ra-228)	11.3 (Ra-226)	65% concentrate
[200]	2–3	4.8–5.4	
[200]	9.0–11.5	6.2–6.3 (Ra-226)	Initial concentrate
[200]	1.0–1.65	4.8–5.1 (Ra-226)	Final concentrate, for export

TABLE 118. ILMENITE

Ref.	Activity concentration (Bq/g)	
	Th-232	U-238
[130]	10.5	8.2
[126]	0.8	1
[128]	0.5 (Ra-228)	0.7 (Ra-226)
[125]	1.4 (Ra-228)	3.5 (Ra-226)
[200]	0.056–0.085	0.074–0.078 (Ra-226)

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