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Management of Problematic Waste and Material Generated During the Decommissioning of Nuclear Facilities



IAEA

International Atomic Energy Agency

MANAGEMENT OF PROBLEMATIC
WASTE AND MATERIAL GENERATED
DURING THE DECOMMISSIONING
OF NUCLEAR FACILITIES

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INTERNATIONAL ATOMIC ENERGY AGENCY
VIENNA, 2006

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FOREWORD

Problematic waste and material from the decommissioning of nuclear facilities contain radioactive and non-radioactive constituents that may adversely affect humans and the environment. Therefore the management of such waste and material should take into account both the radioactive and non-radioactive components and their associated hazards. This report provides an overview of the management options for some types of decommissioning waste and material with regard to their toxicity and relevant hazard. In particular, it identifies and discusses the following issues associated with chemically toxic and hazardous waste and material: (a) their origin and characteristics; (b) typical hazards; (c) recovery and reuse options; and (d) predisposal management options.

The information provided in this report is intended to contribute to a timely and rational development of processes and procedures for Member States' decommissioning programmes, taking into account not only radioactive contamination but also the other hazardous effects of some types of decommissioning material. In addition, it stresses the necessity to take into account the needs of dismantling and waste management during an early stage of the design of new facilities.

This report was prepared by a series of IAEA Consultants Meetings and a Technical Meeting. The IAEA officer responsible for this report was V. Efremkov of the Division of Nuclear Fuel Cycle and Waste Technology.

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1. INTRODUCTION

1.1. BACKGROUND

Decommissioning activities related to any nuclear facility present several problems in the management of the generated waste and obsolete redundant material. According to the estimates available, the volume of decommissioning waste and material can exceed the volume of operational waste accumulated over the life cycle of the facility by 10 to 200 times. The cost of waste management can reach 50% of the total decommissioning budget [1–5].

The waste arising from decommissioning is often different from the waste generated during normal operations or routine maintenance of the facility. These differences may include its chemical, physical and radiological characteristics, the physical form and the general amounts or volumes. Owing to these specific characteristics, some of the waste could be considered as being problematic, for example waste for which application of routine methods of handling, treatment and conditioning is not appropriate and therefore requires special considerations for the selection of specific management options. For such decommissioning waste and material proper planning and selection of appropriate waste management and material management options are of particular importance from the organizational, safety and economic points of view.

Some examples of the problematic nature of specific decommissioning waste are as follows:

- (a) High volume–low activity material may give rise to economic concerns over the disposal of the waste (e.g. contaminated concrete and contaminated soil). The volume of waste in this category is dependent on the national clearance levels.
- (b) Low volume–high activity material may require special remote handling techniques and additional shielding above the shielding required for other waste items (e.g. activated stellite rubbing pads and Nimonic components that contain high levels of ^{60}Co).
- (c) Some waste may be considered problematic because of the inventory of radionuclides that it contains (e.g. waste containing radionuclides of high radiotoxicity and mobile radionuclides such as ^{14}C and tritium).
- (d) Some waste may be considered problematic because it is difficult to encapsulate in cementitious matrices (e.g. aluminium, beryllium, Magnox and uranium metal). Corrosion of the material can lead to the generation of high levels of hydrogen, which can disrupt the encapsulation matrix

and can introduce a risk of explosion. Also, expansion of the waste form can occur, due to the formation of corrosion products.

- (e) Additional problems can occur in the encapsulation of waste material in a cementitious grout, in which the waste material can affect the product properties of the grout (e.g. zinc on galvanized metal surfaces, or contaminated zinc bromide from cell windows, can act as a cement setting retarder); this is also true for the immobilization of high nitrate, fluoride and borate bearing liquid waste. The immobilization of phosphate, such as tributyl phosphate, or high levels of sodium hydroxide in some waste streams, can cause accelerated cement setting, leading to 'flash' setting of the waste form.
- (f) Other waste can be problematic because of a combination of different factors; for example, irradiated reactor graphite can be of concern because of problems associated with stored Wigner energy, flotation of graphite during the encapsulation process and the immobilization of graphite dust, which tends to be hydrophobic and not easily wetted.
- (g) Galvanic interaction between different metals in contact can enhance the rate of corrosion, which can disrupt the waste form. There is also a risk of explosion from the hydrogen produced within the corrosion process.
- (h) Some types of waste can be problematic because of their physical nature (e.g. non-aqueous phase liquids such as oils, organic complexants and the degradation products of organic polymers). These components of waste may enhance the mobility of radionuclides in the disposal environment. They are difficult to immobilize because they are often just absorbed and not chemically bound within the immobilization matrix.
- (i) Waste may also be considered problematic because it is hazardous due to either its physicochemical properties or its inherent toxicity. These types of material represent a potential hazard to human health or the environment when improperly treated, stored or disposed of, or otherwise mismanaged.

Among these types of waste the main concerns are on material that is hazardous and/or toxic by its chemical or physical nature. An analysis of the specific characteristics of such waste, and of its possible management options, is important not only for ensuring the safety of decommissioning activities but also for the planning of maintenance activities for facilities still in operation and for identifying possibilities to diminish such problems in the future at the facility design stage. As a result, new material and procedures could be defined for improving the safety and efficiency of the decommissioning of nuclear facilities in the future.

Several Member States have experience in the decommissioning of nuclear facilities and in the management of the associated material and waste streams. A review of the relevant available information is of particular interest for Member States planning such activities or planning modifications of nuclear facilities and the design of new facilities. There is a range of publications devoted to the analysis of different potential problems associated with the decommissioning of nuclear facilities, in particular with the hazards of some decommissioning waste and possible options for dealing with associated problems. IAEA Technical Reports Series No. 401 [6] provides an overview of the general options and opportunities for the minimization of decommissioning waste. The specific problem of the management of operational low and intermediate level waste with regard to its potential chemical toxicity and the management of waste containing ^{14}C and tritium are discussed in two other IAEA technical reports [7, 8]. Many problems are associated with the management of the large amounts of graphite and sodium generated during the decommissioning of some boiling water reactors, gas cooled reactors and fast neutron breeder reactors [9, 10].

Analysis of the experience and information available on the management of the different types of problematic waste generated during decommissioning is important for identification of the most efficient and appropriate options for solving the problems associated with the management of such waste and material. Even the identification and outline of the problem itself is of particular importance, since it brings the attention of organizations and Member States to this specific problem. Therefore this report concentrates on those types of waste and material considered as toxic and hazardous that could arise from the decommissioning of a nuclear facility.

1.2. OBJECTIVE

The objective of this report is to review the available information and to outline the management options for problematic decommissioning waste and material, which are different from those for waste generated during the normal operation of nuclear facilities. Specifically, waste and material that have been identified as being conventionally hazardous require special attention because of their specific toxic properties or other hazardous characteristics. In this context 'toxicity' means poisonous to life and 'hazard' means a special danger or risk during the handling, processing and conditioning of these types of material and waste. In this report problematic waste and material are identified as those that require special handling and treatment because of their unique combination of radioactivity, toxicity or chemical and physical hazards. This

report reviews the origins of these types of waste and their characteristics, potential hazards and management options.

An integrated approach to the consideration of organizational principles, the regulatory background and the technical options for dealing with these types of waste and material is important in order to ensure the efficiency of the selected options, the safety of workers and the general public, and the protection of the environment. Information already exists on the management of some problematic types of waste and material and on particular technologies and their application for handling, storage and processing. A review of the available information on this subject, analysis of related data and experience, and discussion of related problems would be of particular benefit for Member States planning decommissioning activities.

The information summarized in this report should facilitate the selection of adequate technologies to solve particular waste management problems during decommissioning. The intention of this report is to identify and review the existing information on dealing with different types of problematic waste and material. It is intended that this information will assist responsible organizations in Member States in the selection of particular processes and technologies for the processing of some types of problematic waste and material generated during nuclear facility decommissioning.

1.3. SCOPE

The scope of this report is to review the available experience and information concerning the management of hazardous and toxic waste and material generated during the decommissioning of nuclear facilities. For that reason, discussion of radioactive material having no extraordinary hazardous or toxic concern is excluded from this report. Taking into account that the subject of the management of problematic waste from decommissioning may be rather broad, this report concentrates only on the most commonly occurring types of problematic waste and material. Organic waste is covered more generally, since management of this waste type is discussed in detail in another IAEA report [11]. The management of waste containing concrete, steel, cast iron, graphite and aluminium is also excluded from the scope of this report because there is only a minor conventional hazard or toxic concern associated with it.

This report is organized into the following topics:

- (a) Following this introductory section, Section 2 briefly describes the different options for decommissioning, the steps in the decommissioning process and the selection of the appropriate decommissioning strategy.
- (b) Section 3 considers the particular types of waste generated during decommissioning and the sources and characteristics of these problematic types of waste and material. This section describes the typical hazards associated with these types of material and the possibilities for their recovery for potential reuse or recycling. The available options for processing, storage and disposal are discussed.
- (c) Section 4 provides a summary and concluding remarks.

2. DECOMMISSIONING OPTIONS AND STEPS

2.1. OPTIONS FOR NUCLEAR FACILITY DECOMMISSIONING

References [12–14] refer to three primary decommissioning options that should be considered in planning a decommissioning project:

- (a) Immediate dismantling; that is, prompt removal of all radioactive material from the facility to a predesignated location.
- (b) Delayed dismantling, to take advantage of the natural decay of radionuclides.
- (c) Entombment; that is, a nuclear facility is wholly or partly disposed of at its existing location.

Other options may be considered but are generally combinations or variations of these three primary options. Each decommissioning option will, among other aspects, define the timing and the sequence of decommissioning activities.

In the context of the specific legal framework of the Member State and its social and economic conditions, a decommissioning option, including dismantling and waste management, follows some basic principles, considering some basic components and general steps. These main decommissioning steps are listed in Table 1. An extensive list of the key issues in the selection of a decommissioning option can be found in Refs [14, 15].

These principal steps are not influenced by the requirements for the management of toxic and hazardous waste. However, the presence of these

TABLE 1. PRINCIPAL STEPS IN FACILITY DECOMMISSIONING

Principal step	Action
Description of the facility	Physical, chemical and radiological characterization of the facility components: periodically updated
Decision to decommission a nuclear facility	Political, economic and technical considerations
Preparation of decommissioning plan, including safety documents and environmental impact assessment	Consequent steps, time schedule, methods, radiation protection, waste inventory, waste minimization, waste forms, interim storage, transport, disposal option, environmental impact study, provision of adequate financial resources
Analysis and approval of decommissioning documentation by the authorities/regulators	Feedback on and improvement of the decommissioning and material management plan
Allocation of required funds	Reporting, periodic assessment of the adequacy of financial resources and expenses
Dismantling	Practical dismantling, reporting, possibly based on stepwise approval
Waste management	Practical waste minimization with respect to clearance levels, decay storage and optimized waste treatment and conditioning for disposal
Final release procedures for the facility or site	Release of the land or facility for future reuse

material types needs to be fully considered. In contrast with radioactive waste, which decays with time, delayed dismantling would not lead to a decrease in the associated hazards and toxicity of such waste; in fact the opposite is the case, in that delay may lead to decreasing integrity of the material and components, which may cause additional problems with the dismantling and management of the associated waste. This important factor should be taken into consideration when defining the decommissioning strategy and making the selection of appropriate decommissioning options and associated techniques.

2.2. SELECTION OF A DECOMMISSIONING OPTION

The choice of a decommissioning option will mainly be based on technical, safety, economic and regulatory considerations. These considerations will enable the operator to select the most appropriate decommissioning option. Although radiological hazards predominate in decommissioning activities, toxic and other conventional hazards must be taken into account during the decision making process.

The definition of a decommissioning and waste management strategy needs to fully consider the technical problems associated with the management and processing of all radioactive and hazardous waste. Experience of decommissioning has shown that while the use of and requirements for personal protective equipment for radiological purposes during cleanup of these facilities may decline with time because of radioactive decay, the use of personal protective equipment for toxic and hazardous waste may remain constant or increase with time as material degrades.

For each option it is necessary to consider the volume and physicochemical form of the toxic and hazardous material generated. ‘Cradle to grave’ processes should be available for the handling and treatment of all material (including waste) arising from any decommissioning activity before these activities are undertaken. It should be kept in mind that in most Member States no waste repository is available and that therefore safe interim storage facilities should be provided for the material until a suitable disposal option becomes available.

To determine a suitable decommissioning strategy, information about the facility’s design and operational history is required. A full inventory of the material of construction of a facility and the material used in the facility operation and maintenance needs to be produced at the beginning of a project in order to support the development of a decommissioning strategy. It has to be stressed therefore that record keeping during the operational life of a facility and careful radiological and physicochemical characterization of waste and material are crucial.

The chosen decommissioning and waste management strategy has to take into account the technical, regulatory, economic and social considerations listed in Table 2.

The priority of the above listed parameters may vary from country to country and may also vary with time. This varying priority demands flexible management of the decommissioning process.

TABLE 2. FACTORS TO BE CONSIDERED WHEN SELECTING A DECOMMISSIONING STRATEGY

Area for consideration	Main objective
Facility components' condition after final shutdown	Status of equipment, structures and containments
Radiological characterization	Activation and contamination status
Physicochemical characterization	Presence of hazardous and toxic material and waste
Safety features	Radiological and conventional/industrial safety
Waste management	Waste treatment and conditioning on the site or at a centralized facility, local or centralized interim storage, availability of centralized disposal
Transport	If centralized facilities are to be used, refer to Ref. [16]
Regulations	With respect to radiation protection, toxicity and additional hazards as well as release and disposal of waste
Available services	Operation, maintenance, instrumentation and surveillance
Further use of site and buildings	The nature of the reuse of the buildings, either restricted or unrestricted use, influences the decontamination practices, waste volume and cost [17]
Available resources	Experienced personnel, knowledge of the history of the facility, knowledge of decommissioning techniques, characterization techniques (including modelling) and analytical laboratories
Finances	Balancing present versus future costs (depending on maintenance of existing equipment, available technology, interim storage, final disposal and waste volume)
Political and social considerations	Public perception, information policy and involvement of stakeholders

3. PROBLEMATIC WASTE AND MATERIAL ARISING FROM THE DECOMMISSIONING OF NUCLEAR FACILITIES

3.1. INTRODUCTION

As indicated in Section 1, this report considers the management of hazardous or toxic waste and material generated during the decommissioning of different types of nuclear facility. For the purposes of this report the following definitions of hazardous and toxic waste and material are used:

- (a) Hazardous: Waste and material that because of their quantity, concentration and/or physical, chemical or infectious characteristics may pose a substantial potential threat to human health or the environment when improperly handled, treated, stored or disposed of, or otherwise mismanaged.
- (b) Toxic: Waste and material that contain certain substances determined to be harmful to human health in very small concentrations.

To distinguish between the two definitions, it is helpful to consider that all toxic waste is hazardous but not all hazardous waste is toxic.

There are some general considerations that are common to toxic and hazardous waste. The disposal of toxic waste in either shallow land burial or in deep geological facilities needs to consider the long term behaviour of the waste and has to respect the national regulations for its disposal. Special requirements may also be defined by regulatory authorities for the long term storage of hazardous waste if a disposal option is at present not defined or not available. As was indicated in Section 2, it should be considered that unlike the hazards related to radioactivity, the hazard from toxic waste will not reduce with time. However, some unstable toxic waste, mainly of an organic nature, could degrade while in storage or disposal, resulting in the generation of non-toxic products.

There are various national regulations concerning the limits for emission of toxic compounds, their concentration in drinking water, etc. These particular limits should be respected when preparing the safety analysis for toxic waste treatment, conditioning and disposal.

One of the possible options for the management of decommissioning waste, including some hazardous components, is to consider recycling and reuse of components of the waste [18]. Another option is the processing of this waste for storage and final disposal. These options are discussed in general in the

following sections of this report in relation to particular types of hazardous material. Further specific details of some waste processing options are provided in the Annex.

All types of material arising during decommissioning activities, including chemically toxic and other hazardous material, could be activated or radioactively contaminated, depending on the nature of the nuclear facility in which the material originated and/or the purpose for which the material was employed. Therefore their treatment, conditioning and disposal consider both the radiological and non-radiological hazards associated with these types of material and waste. Table 3 summarizes the commonly occurring radiological hazards associated with the problematic waste and material generated during decommissioning that are considered and discussed in the following sections of this report.

The discussion of these types of waste and material is mainly concentrated on their non-radiological hazards; the radiological hazard of these material types is considered only briefly.

3.2. BERYLLIUM

3.2.1. Form of the waste

Beryllium metal or beryllium oxide (BeO) are used as moderators and as reflectors to enhance the thermal neutron flux densities in the cores of many research reactors. The design and geometry of the beryllium components are specific to the needs of each individual reactor. Depending on the design, different types of cladding of beryllium elements can be used, such as aluminium and zircaloy [19]. In some countries beryllium has been an important metal component in nuclear weapon production and has been used in the fuel rods of special reactors.

3.2.2. Typical hazards

Soluble beryllium compounds are toxic if incorporated or absorbed through the skin. Beryllium is accumulated permanently in tissues, with no known self-regulating excretory mechanism. The principal concern is the toxic and carcinogenic effects caused by the inhalation of airborne particulates of beryllium or beryllium compounds, which include inflammation of the respiratory system and lung disease. There is sufficient evidence to classify beryllium and its compounds as carcinogens [20–24].

TABLE 3. COMMONLY OCCURRING RADIOLOGICAL HAZARDS ASSOCIATED WITH PROBLEMATIC WASTE AND MATERIAL

	Probability of commonly occurring radiological hazard		Comments
	Activation	Contamination	
Beryllium	High	Medium	The degree of contamination of the beryllium depends on whether it is clad
Sodium and sodium-potassium alloys	High	Medium	Contamination in secondary circuit sodium is low and consists mainly of tritium
Cadmium	High	Low	When cadmium is used in fuel storage flasks it may be only slightly activated
Mercury	Low	High	Activated mercury may be used as shielding in research reactors or as target material in accelerators
Lead	Low	High	Where lead is activated it can be difficult to demonstrate compliance with clearance levels because of self-shielding effects
Cyanide	None	High	Cyanide is used for caesium removal purposes and hence is not activated
Decontamination chemicals	None	High	Some spent decontamination solutions may contain activation products
Asbestos	Low	Medium	Asbestos may be used as insulation material on reactor pressure vessels, but commonly the radiological hazard occurs from contamination on the surface
Polychlorinated biphenyls	None	Medium	Polychlorinated biphenyls are commonly found in oils, paints and other organic based material

Beryllium is not radioactive in its commonly found form. However, after use in a reactor core it may contain significant quantities of tritium. Activation of impurities within beryllium material can also lead to significant levels of other radionuclides, the most important of which is ⁶⁰Co [25]. This needs to be taken into account in the handling and processing of all beryllium components during facility decommissioning.

3.2.3. Possibilities for recovery and reuse

Beryllium oxide may swell when irradiated. Irradiated beryllium metal will become brittle and crack due to the production of helium by neutron irradiation. Owing to these features, beryllium components from reactor decommissioning are not commonly reused.

3.2.4. Waste management

Current practice for the management of the beryllium metal and beryllium oxide components generated during decommissioning is their interim storage awaiting final disposal. These components are placed in cans and the void between the component and the can is filled with quartz sand, which ensures the mechanical stability of the packaged waste and minimizes voids around the component. The cans are placed in an interim storage container.

Natural convection by ventilation will allow cooling of the containers and prevent any tritium buildup within the containers (in Germany studies have been performed with the aim of holding the tritium in the waste package [25]). After intermediate storage, to allow the decay of ^{60}Co to a suitable level and the consequential diffusion of the tritium, the void between this canister and the primary package can be filled with cement and the primary package then closed [22], as described in the Annex, and stored in an interim storage facility to await final disposal [26]. The behaviour of beryllium and its components in the repository conditions should be considered in the safety analysis of the repository.

It is not recommended to directly immobilize beryllium in cement, as done with many other waste types, since it reacts with water in the basic cementitious matrices, leading to high levels of hydrogen gas generation and volume expansion. Owing to its chemical properties, beryllium requires extensive research and development (R&D) work to investigate its compatibility with the encapsulation material and to identify the optimal conditions for storage and disposal.

3.3. SODIUM AND SODIUM–POTASSIUM ALLOY

3.3.1. Form of the waste

3.3.1.1. Physical and chemical information

Arisings of sodium or sodium–potassium alloy (NaK) waste in the nuclear industry, including decommissioning activities, are closely associated with the development of the liquid metal fast breeder reactor (LMFBR), in which they are used in the liquid form as a low moderating effect coolant to extract thermal energy from the core. NaK alloy is sometimes preferred to sodium, because it is liquid at ambient temperature.

Owing to their unique chemical and physical properties, sodium or NaK alloys are also used in other nuclear fields as a high performance heat transfer medium and oxygen trap. These applications will not be considered here specifically because in terms of the management of the associated sodium waste they are the same as for LMFBR activities.

(a) Bulk sodium and sodium–potassium from main circuits

After draining from the main circuits of the reactor or the facility to be decommissioned, bulk sodium or NaK is considered as a waste. The quantities of such alkali metal waste are generally large, for example 1600–5500 t for commercial sized liquid metal reactor power plants. This waste constitutes a significant hazard, especially in the course of decommissioning–dismantling activities. A summary of the main LMFBRs worldwide is presented in Table 4.

Several sodium cooled reactors also have a relatively small inventory of NaK used in auxiliary circuits, for example about 13 t in the Prototype Fast Reactor (PFR) in the UK and 7 t in the Superphénix commercial power plant in France.

Smaller quantities of bulk sodium are associated with critical or non-critical R&D facilities; these quantities range from a few kilograms to a few tonnes (5 t for Commissariat à l'énergie atomique's Cabri/Sura facility in France).

(b) Residual coolant in reactors

After draining of the main circuits in the course of decommissioning operations, some coolant can remain trapped inside. The capability of reactor circuits to be completely drained depends on the general concept (loop or pool type) and on the particular design features of the reactor. Depending on the construction of the reactor, the total volume remaining after draining will vary significantly.

TABLE 4. MAIN LMFBR FACILITIES WORLDWIDE

Name	Purpose	Country	Coolant circuit principle	Power (MW(th))/(e)	Coolant primary circuit/total (t)	Operating period	Status ^a
Rapsodie	R&D	France	Loop	40	Na 37/57	1967–1982	SS
KNK II	R&D	Germany	Loop	58/21	Na 36/88	1977–1990	DIP
DFR	R&D	UK	Loop	60/14	NaK 57/167	1959–1977	DIP
EBR I	R&D	USA	Loop	1.4/0.2	NaK 15/18.5	1951–1963	D
SEFOR	R&D	USA	Loop	20	Na	1969–1972	SS
EBR II	R&D	USA	Pool	62/20	Na 286/327	1964–1994	SS
Fermi I	R&D	USA	Loop	300/61	Na 290/421	1963–1972	DIP
FFTF	R&D	USA	Loop	400	Na 500/900	1980–1992	SD
Joyo	R&D	Japan	Loop	140	Na 126/199	1977	OP
BR 5/10	R&D	Russian Federation	Loop	10	Na 2/14	1959	OP until end of 2002
SRE	R&D	USA	Loop	20	Na 27	1957–1964	D
Hallam	R&D	USA	Loop	254	Na 260/370	1963–1964	D
BOR 60	R&D	Russian Federation	Loop	60/10	Na 22/42	1969	OP
FBTR	R&D	India	Loop	40/13	Na 27/150	1985	OP
Phenix	Prototype	France	Pool	563/250	Na 800/1281	1973	OP
PFR	Prototype	UK	Pool	670/250	Na 915/1450	1974–1994	DIP
BN350	Prototype	Kazakhstan	Loop	1000/350	NaK 550/1040 + 20	1972–1999	DIP

TABLE 4. MAIN LMFBR FACILITIES WORLDWIDE (cont.)

Name	Purpose	Country	Coolant circuit principle	Power (MW(th))/(e)	Coolant primary circuit/total (t)	Operating period	Status ^a
Monju	Prototype	Japan	Loop	714/280	Na 912/1675	1995	OP
Superphénix	Commercial	France	Pool	3000/1250	Na 3300/5520	1986	DIP
BN600	Commercial	Russian Federation	Pool	1470/600	Na 770/1600	1980	OP

^a OP: in operation. SD: shut down. SS: safe stored. DIP: decommissioning in progress. D: decommissioned. Status as of May 2003.

Additionally, other reactor components need to be considered separately because they are not designed to minimize residual coolant holdup and their geometry and dimensions are frequently not adapted for complete removal of sodium and NaK. One area of significant concern is the presence of stainless steel insulation material, which consists of thin layers of corrugated stainless steel; coolant can easily become trapped in the void spaces in this material.

(c) Cold traps

Among the various types of sodium and NaK waste arising from LMFBR operation and decommissioning, cold traps are probably the most difficult to dispose of. There are several reasons for this:

- (i) Design complexity, as internal structures are generally composed of curved deflectors to force the coolant flow through metallic mesh sections or other devices able to increase the specific surface on which the oxides and hydrides will deposit.
- (ii) The presence of internal or external devices to cool down the incoming liquid metal.
- (iii) The presence of significant quantities of impurities, mainly oxides and hydrides, trapped inside a solid form, making the cold trap very difficult to drain.
- (iv) The cold traps are, in most cases, not designed to be self-draining.
- (v) Hydrides are more reactive than alkali metal; moreover, the presence of hydride leads to the possible release of hydrogen.
- (vi) Owing to its trapping function, the cold trap tends to concentrate radioactive products contained in the coolant; thus the radioactive inventory is generally very substantial and leads to high dose rates as well as the danger of tritium release during the processing.

After final shutdown of the reactor, the impurity levels in the coolant may increase, especially the oxide and hydride content, depending on the storage conditions, the use of purification systems and the time since shutdown.

Chemical impurities in the primary circuit, and therefore also in the cold traps, may also result from corrosion/erosion of primary components and material as well as from the influx of seal alloy material and lubricants from the primary pumps.

3.3.1.2. Radiological characterization of sodium waste

(a) Reactor primary circuits

The radioactivity of bulk sodium or NaK from primary circuits has several causes:

- (i) Activation by neutron flux from the core and formation of radioelements, mainly ^{22}Na and ^{40}K (^{24}Na is also produced by activation, but it rapidly decays due to its short half-life).
- (ii) Contamination by fission products from the fuel due to initial external contamination, fuel cladding failures during operation or use of leaking fuel pins: typically radioisotopes of actinides (plutonium, americium, curium and uranium), caesium and tritium.
- (iii) Contamination by activated corrosion products from fuel cladding and primary circuit structures, such as ^{54}Mn and ^{60}Co .

The specific activity of radioactive products in the primary coolant is highly dependent on the operational history of the reactor and may vary from 2 to 15 kBq/g after the reactor has been shut down for two to five years. Major contributors to the radiological inventory, as indicated above, are generally ^{137}Cs , ^{22}Na (^{40}K in NaK) and tritium.

(b) Reactor secondary circuits

In normal operation, the coolant of secondary circuits is physically separated from the primary sodium or NaK by the intermediate heat exchangers. However, some of the tritium produced in the primary circuit diffuses through the walls of heat exchanger tubes and contaminates the secondary coolant. In the secondary circuit, tritium may diffuse through the walls of the steam generators or collect in the cold traps.

In the event of a failure of an intermediate heat exchanger the secondary coolant may be slightly contaminated by the primary sodium or NaK (with associated radioactive contaminants), although secondary circuits are designed generally to maintain a small overpressure margin relative to the primary circuit.

For pool type reactors the secondary coolant could be activated by the neutron flux from the core, but the main activation product is ^{24}Na , which decays rapidly after reactor shutdown, due to its short half-life (15 h).

Also for pool type reactors, activated corrosion products on the secondary side of the heat exchanger walls may be swept along by the

secondary coolant. However, given the operating temperatures of the secondary circuits, corrosion is negligible and the resulting radioactivity of the secondary coolant is in general below detection limits.

The main radiocontaminant of the secondary coolant is therefore tritium. The order of magnitude of the activity ranges in general from hundreds to thousands of becquerel per gram one to two years after shutdown. For the EBR II in the USA, the tritium activity in the secondary circuit was found to be 750 Bq/g. At the BOR 60 fast reactor in the Russian Federation, sampling analysis has indicated 2–4 kBq/g of tritium in the secondary sodium circuit.

(c) Other bulk alkali metal

Facilities used for fuel development, fuel post-irradiation examination or core safety research may contain significantly radioactive sodium with the presence of actinides and other alpha emitters. The radiological inventory of bulk alkali metal drained from R&D and test facilities obviously depends on their utilization.

An example is sodium from the irradiated fuel caves of the PFR reactor in the UK. PFR fuel caves are fuel post-irradiation examination cells containing sodium pools in which the fuel is stored. Owing to the various operations that have taken place in these cells, the chemical quality of the sodium has been degraded and fuel debris may also be present.

Another example of sodium coming from other than the main reactor circuits of a facility is that of the fuel storage of the Superphénix reactor in France. This sodium has been contaminated by the primary sodium of the main vessel during fuel assembly transfer. The radioactivity level is very low because the fuel storage has not been operated for long.

3.3.2. Typical hazards

Besides the risk associated with the manipulation of radioactive products, nuclear alkali metal waste generates specific risks due to the chemical properties of alkali metals. Sodium and potassium are very reactive. Reactions with water, air and oxygen are generally violent and produce hazardous by-products such as hydrogen and caustic products. Sodium can burn in air at temperatures above 115–130°C, but also at lower temperatures, depending on the amount and physical condition of the surface exposed and other factors such as humidity. When finely divided into particles, for example aerosol deposits, sodium can burn at room temperature in humid air. It is therefore recommended to store alkali metal waste under an inert atmosphere and to avoid transfer and manipulation in the open air.

NaK waste should be considered with special attention, due to the high chemical reactivity of NaK, especially when the presence of sodium or potassium superoxides is suspected. Specific solutions are applied on a case by case basis for the treatment of this type of waste.

Serious accidents with NaK have been reported from NaK cooled LMFBR operations in the UK and the USA. In 1999 an accident occurred at Oak Ridge, USA, due to a NaK spillage during the replacement of a NaK cooled furnace crucible. Eleven workers were injured [27].

3.3.3. Possibilities for recovery and reuse

The potential for recovery and reuse of sodium and NaK is limited. The most likely potential use would be as reactor coolant, but there are currently no LMFBRs being constructed that would use these alkali metals.

3.3.4. Waste management

There are two types of bulk alkali metal stream requiring treatment:

- (a) Alkali metal from reactor main circuits, for which the difficulties encountered are in general associated with the large quantities involved. As there is a minimal possibility of recycling, it must be transferred into a chemically stable form, with the resulting question of how to handle the large quantities of the resulting reaction products (release to the environment, in accordance with existing authorizations, or conditioning for final disposal).
- (b) Alkali metal from R&D and test facilities and specific reactor auxiliary fuel storage, which is generally present in smaller quantities compared with reactor circuits but is sometimes much more radioactive and contains other contaminants. The configuration of these sodium quantities may also present an issue as to the methods of removal from the containers and introduction into the treatment process.

It may be beneficial to reduce the radioactivity level of the sodium by pretreatment processes, in order to facilitate subsequent treatments and to simplify the required equipment. This could also have a bearing on the final effluent. Tritium and ^{137}Cs are the elements that are generally considered in sodium decontamination processes. In general, ^{137}Cs is the main gamma contaminant and tritium is the main contaminant of the gaseous effluent resulting from sodium treatment.

3.3.4.1. *Bulk sodium and sodium–potassium destruction/treatment*

Bulk sodium oxidation processes to obtain a stable product at ambient conditions are described in detail in Refs [10, 28, 29]. The two continuous processes for bulk alkali metal treatment are the NOAH process and the Argonne process. The NOAH process is based on the injection of small amounts of liquid sodium into a large flow of aqueous sodium hydroxide. The Argonne process is based on a two stage process involving a sodium hydroxide forming process step and a carbonate forming process step.

As indicated in Section 3.3.1, there is a relatively small inventory of NaK in sodium cooled reactors. The most common practice is to dilute this NaK into the bulk sodium before treatment. In this case the small proportion of potassium does not affect the physical and chemical properties of the sodium. At the EBR II reactor the NaK was mixed with the sodium inside the primary vessel, while at the PFR it is mixed in a buffer tank during the processing of the sodium. Mixing is also planned at the BN350 and Superphénix reactors.

3.3.4.2. *Treatment of residual coolant in reactors*

As previously described, following drainage of the primary vessel there will be some residual alkali metal that cannot be removed without additional engineering work. The treatment of residual sodium or NaK in LMFBR main circuits in most cases must be performed in situ (and sometimes reaction products must be eliminated). This is necessary in order to eliminate the potential hazard due to alkali metal chemical reactivity and associated surveillance requirements, or to allow in-air operations without fire and caustic hazards. This may be also the case when the decommissioning strategy is to keep the circuits or the vessels under a care and maintenance regime for a long period, in order to benefit from radioactivity decay or to defer the dismantling effort.

When NaK systems are drained, the empty system must be maintained under an inert atmosphere in order to prevent the ingress of air, which can potentially form unstable potassium superoxide.

The main treatment process for the residual coolant in reactors is the water vapour nitrogen (WVN) process. The WVN process is based on the circulation of a nitrogen carrier gas containing a small proportion of water vapour (1–6 vol. %). Care has to be taken with hydrogen production; hydrogen must be safely vented from the reactor circuit. Details of this process, along with other applicable processes, are provided in Ref. [30].

3.3.4.3. *Treatment of limited quantities of sodium and sodium–potassium waste*

For oxidation of limited quantities of radioactive alkali metal, several treatment methods have been adopted. These include:

- (a) Treatment with water in an autoclave reactor;
- (b) Treatment with water under reduced pressure;
- (c) Conversion into carbonates in a fluidized bed;
- (d) Burning in a vessel;
- (e) Incorporation in a 'geocement';
- (f) Solid phase oxidation;
- (g) Gas phase oxidation;
- (h) Burning in air;
- (i) Reaction with alcohol.

3.3.4.4. *Treatment of the oxidation reaction products*

Treatment of alkali metals with water generates radioactive liquid effluents. These effluents are caustic and could be considered as an intermediate product in the treatment process. Further treatment would be required for discharge or conditioning for admission into a final repository. Several conditioning processes have been developed for the high alkali solutions generated during the treatment of sodium waste:

- (a) Reduction of pH and conditioning in cement;
- (b) Reduction of pH, ion exchange treatment and sea discharge;
- (c) Conditioning in a ceramic matrix;
- (d) Conversion into dry carbonates.

3.4. CADMIUM

3.4.1. **Form of the waste**

Owing to its high thermal neutron cross-section, cadmium is used in reactor control rods to control criticality. There are three functional classes of control rod: regulating rods, used for fine adjustments; shim rods, used for coarse adjustments; and safety or scram rods, used for rapid or emergency reactor shutdown. Cadmium was mainly used in research reactors in the early years because of its low melting point and soft nature. Pure cadmium is usually clad in stainless steel or aluminium [31].

In power reactor applications cadmium is alloyed with elements such as silver and indium, which are more effective neutron absorbers in the intermediate and higher neutron energy ranges.

Cadmium is also used in spent fuel racks to increase spent fuel storage capacity; a cadmium plate is sandwiched between two stainless steel tubes surrounding the fuel assembly.

3.4.2. Typical hazards

The major acute hazard is inhalation of cadmium metal or oxides as fumes or dust. If ingested, only a small quantity of cadmium salt enters the bloodstream, because it causes vomiting and is poorly absorbed by blood from the digestive tract. Deposits of cadmium oxide in the lungs slowly pass into the bloodstream (50% reduction in five days); cadmium is distributed throughout the body, but is mainly retained in the liver and kidneys. Excretion of cadmium compounds from the body occurs, but extremely slowly. Absorption of cadmium salt by the skin is negligible [20]. Cadmium and its compounds are considered to be carcinogenic.

Owing to neutron irradiation, cadmium may contain several radionuclides. These can be produced by nuclear reactions with the bulk material; such products are ^{109}Cd , $^{109\text{m}}\text{Ag}$, ^{109}Ag , ^{113}Cd and ^{113}In . Also, impurities in the cadmium can contribute to the activity inventory, due to neutron bombardment. The main radionuclide resulting from activation of impurities is ^{60}Co , but other nuclides, including actinides, may also be produced [25, 32].

3.4.3. Possibilities for recovery and reuse

Owing to the nature of the use of cadmium in reactor control rods, and to the comparatively small volume of cadmium components, there is no economic need and little opportunity for recovery or reuse of this material. The cadmium and associated cladding will be highly activated and in most cases will require disposal as intermediate level radioactive waste.

The cadmium used in spent fuel racks may be, depending on its specific activity, suitable for reuse or recycling.

3.4.4. Waste management

During decommissioning of cadmium-containing equipment, attempts should be made to avoid exposing workers to the cadmium. Therefore, contrary to how work is typically performed, cadmium-containing equipment

should not be cut or sheared unless this is unavoidably necessary. Leaving the cadmium contained within its stainless steel or aluminium cladding is preferable. If control rods are broken, special attention must be paid to avoiding contact of the workers with the cadmium contamination.

The cadmium control rods or cadmium-containing component can be directly immobilized in a cementitious matrix and stored pending final disposal. The behaviour of cadmium and its components in the repository disposal conditions is considered in the safety analysis of the repository.

3.5. MERCURY

3.5.1. Form of the waste

Liquid mercury was used as a coolant for early experimental fast reactors. The first liquid metal cooled reactor, known as Clementine, at the Los Alamos Scientific Laboratory, was mercury cooled. The advantages of mercury, relative to other liquid metal reactor coolants, are its low melting point (-38.8°C) and the fact that it is chemically inert to air and water. The disadvantages of using liquid mercury are its toxicity and high density (hence heavy pumping load).

Mercury has also been used:

- (a) As a liquid seal for rotating shields above the core of fast reactors (e.g. almost 2 t of mercury in the seals of the PFR and the Dounreay Fast Reactor in the UK);
- (b) For radiological shielding, as in the case of the heat transfer reactor experiments in the USA;
- (c) As a catalyst in the dissolution of uranium alloy fuels;
- (d) In lithium isotope separation [33].

3.5.2. Typical hazards

Metallic mercury can enter the body by ingestion, through the skin or by inhalation, although it is not absorbed significantly by the gastrointestinal tract. Spilled mercury constitutes a potential hazard because of its high volatility. The proportion of inhaled vapour absorbed in the upper respiratory tract may approach 100%, but on average may be nearer to 70% [20].

The LD_{50}^1 values for mercury compounds are in the range of 10–40 mg/kg body weight. When given in massive doses, mercury in a wide variety of chemical compounds will denature proteins, inactivate enzymes and cause

severe disruption of any tissue with which it comes into contact in sufficient concentration.

The relative degree of toxicity of inorganic mercury compounds is dependent on the solubility — the greater the solubility, the more harmful if ingested. Mercury ions are partially absorbed (up to 15% absorption of inorganic mercury salts) in the gastrointestinal tract, but small quantities are readily excreted. Phenyl mercury compounds may be absorbed through the intact skin or mucous membranes, and can cause second degree chemical burns. Alkyl mercury compounds are readily absorbed from the intestinal tract and through the skin; they show affinity for lipid tissues and when taken up are excreted slowly compared with other forms of mercury. Chronic mercury poisoning can have serious effects on the nervous system and kidneys.

Suitable personal protective equipment should be used when dealing with mercury waste [34].

3.5.3. Possibilities for recovery and reuse

Contaminated mercury can be recovered and reused through a process of distillation. The principle of operation is to distil the mercury under vacuum, thereby reducing the boiling temperature from 356°C to about 200°C. This allows the distillation rig to operate with lower power consumption and reduces the need for thermal lagging. The rig is designed to allow the vacuum pump to be removed once the rig is fully primed. The rig is effectively a single piece of glass, constructed from about ten elements, to avoid the need for greased joints, since the operating temperature of the rig would render such joints useless for maintenance of a vacuum.

The operating principles of the distillation rig require that the mercury be free from high levels of volatile material, since this would degas from the metal in the first boiling chamber and pressurize the chamber. Distillation will remove the contamination from the mercury and leave clean mercury for reuse or clearance.

Activated mercury cannot be treated in this way due to its radioactive mercury isotopes, which cannot be separated during distillation. It is treated and disposed of as radioactive waste.

¹ LD stands for 'lethal dose'. LD₅₀ is the amount of a material, given all at once, that causes the death of 50% (one half) of a group of test animals. The LD₅₀ is one way to measure the short term poisoning potential (acute toxicity) of a material. It is usually expressed as the amount of chemical administered (e.g. milligrams) per 100 grams (or kilogram) of the body weight of the test animal.

3.5.4. Waste management

If distillation is not applicable, mercury has to be immobilized. Most of the information available on the immobilization/stabilization of mercury arises from studies and trials carried out in the USA. The methods employed are varied and deal with all types of mercury waste, from the treatment of metallic mercury to the immobilization of aqueous solutions of mercury compounds.

Some technologies for the immobilization of metallic mercury and mercury solutions are described below (these have not been demonstrated on the industrial scale).

3.5.4.1. Amalgamation

An amalgam is defined as an alloy of mercury and at least one other metal, such as copper, zinc, nickel or silver. In general, an amalgam is formed when metal powder is mixed with liquid mercury. The process is fairly simple, and involves the mixing of elemental mercury with an amalgamating material. The mixture is then agitated/stirred in batches for a prescribed period of time.

The most common metal used for amalgamation for the purposes of waste management is copper, principally because of its relatively low cost. Zinc is also used to form an amalgam, although this amalgam can be attacked by water, forming hydrogen and zinc hydroxide [35]. Investigations have been undertaken on the direct amalgamation of mercury with copper in the form of a fine powder. It has been found that a copper/mercury ratio of 3:1 (by weight) is required to produce a satisfactory amalgam at room temperature, with a mixing time of 40 min. Other trials have investigated precleaning of the copper to remove the non-reactive oxide layer prior to mixing with the mercury [35]. The mixing times and mercury waste loading of the amalgam using precleaned copper have been investigated. Two methods of cleaning have been identified:

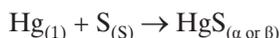
- (a) Cleaning with a solution of ammonium chloride and hydrochloric acid;
- (b) Heated hydrogen gas at temperatures higher than 600°C.

It is claimed that mixing times can be reduced to three to five minutes and the copper/mercury ratio reduced to 1:1 (by weight). Waste loadings of up to 85 wt% have been achieved.

Amalgamation has the additional benefit of producing a solid waste product requiring no further stabilization. Amalgamation is the recommended technology in the USA for the immobilization of radioactive mercury waste.

3.5.4.2. *Mercury sulphide*

Mercury sulphide is exceedingly insoluble in water and unreactive chemically, being attacked only by concentrated HBr, HI or 'aqua regia'² [36]. Therefore, mercury sulphide formation by the reaction



could be used for mercury immobilization. This is a relatively simple process involving the mechanical blending of mercury with sulphur. Although sulphur is not a metal, this process is categorized in the USA as amalgamation. Mercury (II) sulphide exists in two forms, a red hexagonal α -HgS (cinnabar) form and a black β -HgS (metacinnabar) form.

Some investigators have evaluated a high shear blending technique for the production of both cinnabar and metacinnabar mercury sulphide. Zinc amalgam has also been produced as a means of comparison [37]. It has been found that metacinnabar (black) is produced at low mixing speeds (<1000 rpm) and cinnabar (red) at high shear (mixing speed of 19 000 rpm) and high temperature (270–300°C). It has therefore been concluded that the production of HgS via a low shear process (metacinnabar) is preferred to a high shear process (cinnabar), as it is safer and easier.

3.5.4.3. *Sulphur polymer stabilization*

This method is used for the stabilization of mercury contaminated mixed waste and uses heated sulphur polymer cement and small amounts of unspecified additives to convert metallic mercury into HgS. The thermoplastic mixture is melted and poured into a mould with the mixed waste to be immobilized, where it cools and solidifies. The process is claimed to be very effective and is used on a commercial scale in the USA [38]. The process has a further stage, in which the solidified waste is subsequently encapsulated within a casing of clean sulphur polymer cement (i.e. a form of macroencapsulation). The process has not been proven for the immobilization of elemental mercury. The process is costly and substantially increases the volume of the final waste form.

² One part volume of concentrated nitric acid and three parts volume of concentrated hydrochloric acid.

3.5.4.4. *Immobilization in Portland cement*

Trials have been undertaken in which Portland cement was doped with 10 wt% aqueous mercuric nitrate — $\text{Hg}(\text{NO}_3)_2$ [39]. It is believed that the final chemical state of the dopant is mercuric oxide (HgO).

The mercury was added to the grout mix in the form of 10 wt% aqueous $\text{Hg}(\text{NO}_3)_2$. The grout samples were left to set over a 28 day period at room temperature and a relative humidity of 85%. The waste product was then analysed via X ray photoelectric spectroscopy to identify the oxidation state and chemical form of the mercury in the Portland cement, which showed it to be mercury oxide. The study was concerned solely with the form of the mercury in the final waste form. However, no data are available on the viability of the final waste form. As the mercury is immobilized in an aqueous form the potential for metallic mercury to be present within the waste form is greatly reduced.

The study concluded that:

- (a) HgO was the final state of the mercury ion contaminant;
- (b) The oxidation state of the mercury was 2+;
- (c) There was no chemical bonding of the mercury to the cement components.

3.6. LEAD

3.6.1. **Form of the waste**

Lead is widely used in nuclear facilities as a shielding material in the form of bricks, sheets, wool or shot. The physical form of the lead shielding material is dependent on the nature of its use. In addition, lead based paints and primers were routinely used during the construction of many facilities. Initially the toxic properties of lead were not fully understood, and as a result lead became widely used and the various sources of lead are generally not enclosed, encapsulated or labelled. In nuclear facilities lead is predominantly contaminated rather than activated.

3.6.2. **Typical hazards**

The effects on human health of exposure to lead vary according to the nature of the substance and its path of incorporation into the body. The principal toxic effects are on the central nervous system, on the blood and on the kidneys. The most serious cases of lead poisoning are usually the result of

cumulative exposure. The ingestion of quantities of elemental or alloyed lead sufficient to cause death from a single exposure is most unlikely [20].

Acute lead poisoning is often due to the accidental ingestion of a significant quantity of inorganic lead, particularly in the form of soluble lead compounds such as acetate and nitrate. Insoluble and stable inorganic compounds of lead, for example chromate, sulphate and sulphide, are less acutely toxic because their rate of absorption from the gastrointestinal tract is low. Such compounds can still be toxic, however, by the accumulation of insoluble ingested particles in tissues, since it is then possible for slow dissolution to result in the release of lead into body fluids.

The principal concern in the decommissioning of nuclear facilities is the inhalation of lead dust. This can result initially in accumulation in the lungs, and subsequently in gradual dispersion into bones, teeth and tissues. Therefore medical surveillance of employees exposed to lead above predefined trigger levels is normally undertaken, in the form of blood samples.

During all lead removal operations the decommissioning manager must ensure that engineering and work practices are controlled in order to reduce the exposure of employees to the lowest feasible level, whether respiratory protection is worn or not. Some of the measures that may be put in place to control lead exposure during dismantling operations include [40]:

- (a) Abatement of lead based paint using vacuum tools or chemical removal techniques;
- (b) Taping cut lines prior to mechanical cutting;
- (c) Using wet methods to control dust release;
- (d) Local exhaust ventilation;
- (e) Routine housekeeping;
- (f) Containment of the process by tents or enclosures;
- (g) Barriers to prohibit unnecessary personnel from entering the work area.

From the radiological point of view it can be difficult to confirm whether the lead has been activated or whether contamination has been incorporated into the matrix, due to the self-shielding effect of the lead.

3.6.3. Possibilities for recovery and reuse

There are many possibilities for the recovery and reuse of lead inside and outside of the nuclear industry. For reuse outside of the nuclear industry decontamination of the lead is generally required. There are many different methods that can be employed for this purpose. The method used is dependent on the form of the lead (i.e. mechanical methods are preferred for lead blocks)

and the form of the contamination. The methods include mechanical and non-mechanical means such as:

- (a) Shaving;
- (b) Sand blasting;
- (c) Grinding;
- (d) Chemical methods;
- (e) Lead melting.

Mechanical treatment methods involve the physical removal of the lead surface and the associated contamination. This can be done by shaving or planing to remove the surface of the lead block, by sand blasting or by grinding. However, sand blasting can have the effect of pushing the contamination further into the lead block.

Experience in the UK on the decommissioning of lead shielded enclosures is based on the use of planing (shaving of lead blocks). The process utilizes a purpose built partitioned containment tent, ventilated with a high efficiency particulate air (HEPA) filtration system, so that the receipt, decontamination and radiological monitoring of individual items can be segregated in order to minimize any cross-contamination. This process is summarized in Fig. 1. The contaminated lead blocks are sent for assessment and decontamination treatment as necessary [41]. Further details of the process are provided in the Annex.

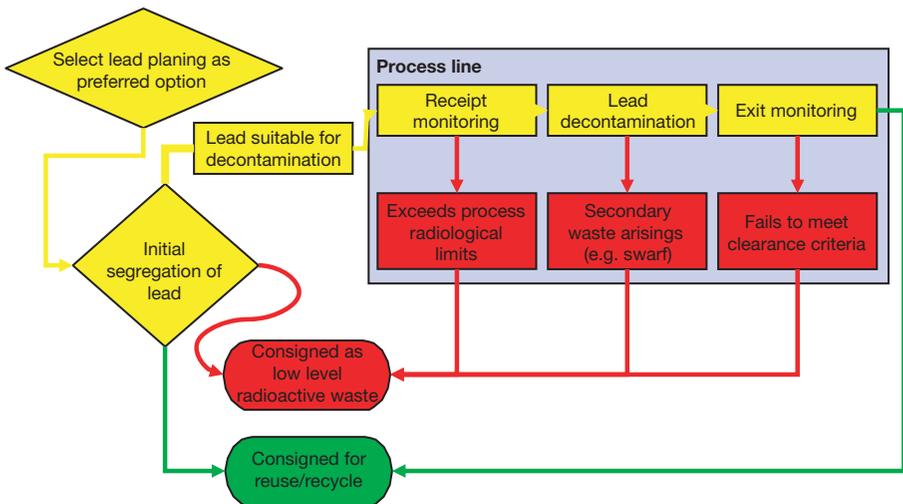


FIG. 1. Lead decontamination process flowchart.

Chemical methods are also applicable for the decontamination of lead; for example, the DECOFOR process, based on a patented formic acid process, can be used for chemically decontaminating lead to clearance levels [42].

The DECOFOR process involves immersing the contaminated metal samples in dilute formic acid for a short period of time, or the samples can be sprayed with acid solution. After reaction has been completed, the surface is rinsed with water to remove the contaminated surface layer. The acid depleted solution can be regenerated with H_2O_2 , as an oxidizing agent, which precipitates the insoluble compounds. Formic acid is added to the depleted solution to achieve the initial concentration, and the acid solution can be reused. The solution containing the dissolved metals is electrochemically processed to recover the metals. The acid solution from the electrolysis is returned to the decontamination tank for reuse (Fig. 2). This process will only be economic for large quantities of contaminated lead.

This process is operated as a closed cycle for water and the decontamination chemicals. As a result, there are no liquids requiring further treatment and disposal, and the amount of secondary solid waste for disposal is minimal: 0.01–0.1% of the original metal, as metal and metal oxide.

Melting of lead had also been used as a means of purifying contaminated lead. The melting of lead allows radionuclides to be concentrated in the slag,

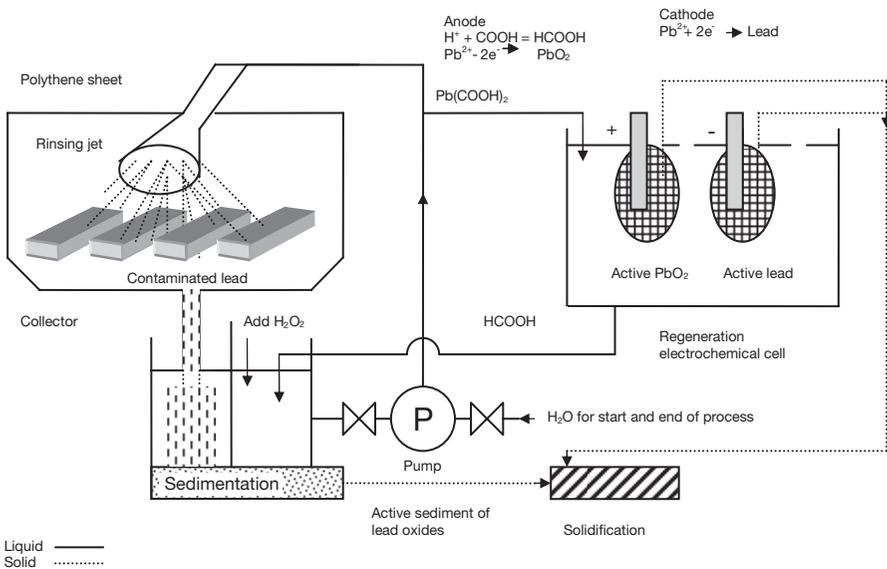


FIG. 2. General scheme of lead decontamination technology.

which has to be treated and disposed of as radioactive waste. Melting of the lead will allow a sample of the molten material to be taken and confirmation provided of the levels of contamination/activation, in order to demonstrate that it is within the clearance levels [6, 43]. Melting will also allow the conversion of finely divided lead into a massive form for disposal. Further details on treatment processes are provided in the Annex.

The reuse of lead in nuclear facilities is encouraged, especially if the waste is activated and the clearance of this material is difficult to demonstrate. As indicated above, it is difficult to confirm whether contamination has been incorporated into the matrix, because of the density of the lead. The history of the previous use of the lead must be known in order to ensure that the contamination is only a surface effect, otherwise samples of the lead need to be taken to ensure that it meets the clearance levels.

3.6.4. Waste management

If lead cannot be decontaminated for recovery and reuse, it is disposed of as radioactive waste. As a solid it can easily be incorporated into a cement matrix for disposal or used as shielding in a waste container. The total quantities of lead in the repository are considered as part of the repository post-closure safety case.

3.7. CYANIDE

3.7.1. Form of the waste

Cyanide is a carbon–nitrogen chemical unit that combines with many organic and inorganic compounds. Different types of cyanide-containing material are quite often used in the nuclear industry, in particular in waste management practices. Cyanides are important components of inorganic absorbents used for the selective removal of caesium from various liquid waste streams.

In the nuclear industry cyanides are mainly found in stored sludges or liquids arising at R&D facilities. In some cases cyanide-containing liquids and sludges have been cemented by the waste producer and have been stored on-site for a long time. Reprocessing tests, metal plating operations, etc., have generated waste contaminated with cyanide. Generally this type of waste is mixed with other chemicals in complex waste that requires careful characterization prior to treatment for storage and disposal.

Cyanides can also be found in ion exchange resin media, in which they occur as insoluble transition metal hexacyanoferrates. These have been used for decades for the removal of radioactive caesium and other radionuclides from solution. These types of ion exchange media can be produced during decommissioning activities. The issues associated with the management of spent ion exchange resins and sorbents are discussed in Ref. [44]. The cyanide complexes are stable during interim storage and processing in appropriate conditions, for example non-oxidizing media, but degradation of these complexes is possible in a deep geological disposal environment, with the liberation of free cyanide.

3.7.2. Typical hazards

The short term health effects of cyanide are that cyanide potentially causes rapid breathing, tremors and other neurological effects when people are exposed to it at levels above the maximum contaminant limit (MCL) for relatively short periods of time.

The long term health effects of cyanide are that cyanide has the potential to cause weight loss, thyroid effects and nerve damage from a lifetime exposure at levels above the MCL.

In the USA, for example, the MCL has been set at 0.2 ppm because, given present technology and resources, this is the lowest level to which water systems can reasonably be required to remove this contaminant should it occur in drinking water [45].

Releases to soil appear to be primarily from the disposal of cyanide waste in landfills and the use of cyanide-containing road salts. Chlorination treatment of some waste waters can produce cyanides as a by-product. Cyanides are generally not persistent when released to water or soil, and are not likely to accumulate in aquatic life, since they are broken down by microbes. They do not bind to soils and may leach to groundwater.

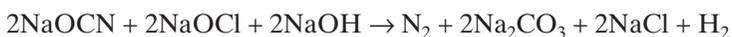
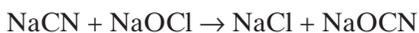
3.7.3. Possibilities for recovery and reuse

Owing to the chemical complexity of cyanide-containing waste, the possibility for recovery and reuse is excluded.

3.7.4. Waste management

Alkaline chlorination is the most widely used method for the destruction of cyanides and other chemical waste. This process, which has been in commercial use for over 35 years, is suitable for destroying free dissolved

hydrogen cyanide and for oxidizing all simple and some complex inorganic cyanides in aqueous media. The cyanide in very stable complexes such as ferrocyanides or ferricyanides is basically unaffected by chlorination. Cyanide that is complexed with copper, nickel and precious metals is amenable to chlorination but reacts more slowly than free cyanide and therefore requires excess chlorine for efficient cyanide destruction. If properly designed, maintained and operated (good pH and oxidation–reduction potential control), the process will oxidize cyanides that are amenable to chlorination. Traditionally this process involves successive oxidation of cyanides to cyanates, and then to nitrogen and non-toxic chlorides and carbonates by chlorine gas or sodium hypochlorite solution under high pH conditions (to avoid the formation of toxic CNCl or HCN gas):



Excess hypochlorite is neutralized by adding dry sodium thiosulphate in stoichiometric amounts. The final pH is adjusted to neutral (pH6–7).

Further chemical treatment is not necessary before the waste is discharged (assuming other waste types are not present). Any solids recovered are immobilized in a drum using an appropriate solidification process. The treated waste solution may be solidified in cement and landfilled or sent for storage or disposal as radioactive waste.

The advantages of direct alkaline chlorination for treating process effluents are its relative simplicity, high efficiency (approaching 100%), operation at ambient temperature (except for complexed cyanides), suitability for automatic control and low cost.

An obvious disadvantage of the above described method is that it requires the purchase and storage of large quantities of hazardous chlorine gas or hypochlorite solutions. However, the risks associated with these chlorine sources can be essentially reduced or fully excluded by the employment of innovative approaches and technologies. Other hazards associated with the process are summarized below.

3.7.4.1. *Chemical hazards*

Cyanogen chloride (CNCl), hydrogen cyanide (HCN), chlorine gas (Cl₂) and the caustic solutions in the reaction vessel and caustic scrubber represent

the principal chemical hazards. The process has to be designed to maintain a pH higher than pH10 in the reaction vessel and a pH higher than pH12 in the caustic scrubber in order to prevent the evolution of these hazardous gases. The ventilation blower maintains airflow through the reaction vessel and to the caustic scrubber. This airflow is the primary safety system. If the process gets out of tolerance and evolves any cyanogen chloride or hydrogen cyanide gas, the airflow carries all gases from the reaction container to the caustic scrubber container. High pH liquids quickly capture any cyanogen chloride or hydrogen cyanide gases. In addition to the airflow, the system includes a hydrogen cyanide detector near the caustic scrubber, set to produce an audible alarm before there is a health hazard.

3.7.4.2. *Explosive hazards*

The scenario of highest risk is the potential of a fire or explosion resulting from hydrogen gas buildup (see processing options below). The explosive concentration range of hydrogen gas in air is 4–76%, so it is necessary to control potential accumulation. Control is accomplished by proper airflow and ventilation. A hydrogen detector at the reaction vessel is normally set to produce an audible alarm before there is a fire hazard.

3.7.4.3. *Additional hazards*

Spills, releases and/or chemical burns are additional hazards of the system. These hazards have to be controlled by limiting the maximum pressures and flow rates used in the system, using containment trays under the reaction vessel and caustic scrubber system, using appropriate industry standard personal protective equipment and ensuring proper training of operational and management personnel.

In specific cases calcium hypochlorite can be used instead of sodium hypochlorite [46], or direct alkaline chlorination can be replaced by various kinds of electrochemical technology [47–54]. The advantage of using the calcium form [46] is that introduction of water into the reaction vessel (12% sodium hypochlorite solution) is eliminated with the solid granular calcium hypochlorite feed. Calcium hypochlorite is metered into the reaction vessel through the dry solid feeder, as required, to maintain a sufficient oxidation–reduction potential and to control the temperature in the reaction vessel. After the conversion to cyanates, conditions are adjusted to oxidize cyanate to carbon dioxide and nitrogen.

The advantage of this process is that there is no need for on-site storage of hazardous substances in gaseous or dissolved forms. The temperature of the

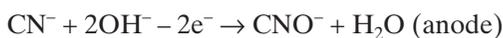
reaction is controlled by a combination of circulating water through the reaction vessel jacket and controlling the reagent addition rate. By limiting the dry hypochlorite addition, the pH rate and the oxidation–reduction potential will be more easily monitored and controlled [46].

The electrochemical chlorination method (in contrast to direct chlorination) enables chlorine gas to be generated in situ [47]. Sodium chloride (NaCl) is added to the cyanide solution and the solution is circulated through an electrolysing cell. Under high current, the salt solution breaks down, with release of chlorine gas. The chlorine, in turn, oxidizes the cyanide species into carbon dioxide and nitrogen. The results of a treatability study demonstrate that the electrochemical chlorination technique is a simple, effective process capable of achieving the destruction of soluble cyanide waste in a batch mode. The destruction time depends on the type of cyanide and its concentration and on interfering contaminants (e.g. ammonia). No toxic hydrogen cyanide gas has been detected in the off-gas from the scrubber–vent system [48].

Electrochemical oxidation of cyanides is a well investigated process [49–54]. It can be realized in standard, commercially available electrolytic cells or in a specially designed apparatus. Single cell modules are simply combined, much like building bricks, to deliver any desired output. The risk of scale-up problems and possible failures, quite often experienced by the chemical industry, are thus alleviated. In most instances the electrochemical processing does not require the addition of chemical reagents, and in many cases the reaction can be instantaneously terminated by simply turning off the power supply.

In practice, unused chemicals are mixed with spent plating bath solution (cyanides of potassium, sodium, copper or silver) and electrolysed in a cell with so called dimensionally stable anodes (titanium grids covered with a mixture of titanium oxide and rare earth oxides) and various cathodes (the material of the cathode depends on the composition of the waste to be treated). Usually during the electrolysis an addition of NaOH is required to keep the pH at a high level in order to avoid HCN gas evolution.

Typical reactions in an electrochemical cell are the following:



The post-electrolysis solution is acidified with concentrated H_2SO_4 (resulting in the evolution of a large quantity of non-toxic carbon dioxide) and

evaporated, so that the final products of the electrochemical treatment are pure metal and a mixture of sulphates.

The efficiency of the electrochemical process depends on the concentration of the metal and cyanide in solution and the applied current density. However, essential improvements in efficiency can be made through the use of packed bed or porous flow-through electrodes; for example, with a packed bed graphite electrode a solution with an initial cyanide concentration of ~600 ppm can be reduced to a concentration of less than 10 ppm at a lower cost than the conventional alkaline chlorination treatment [54]. Pulsed current techniques may also improve the overall current efficiency.

3.8. DECONTAMINATION CHEMICALS

3.8.1. Form of the waste

Chemical decontamination techniques are mainly applicable to metallic items. These processes use reagents that are able to erode the surface by chemical attack and chemical complexants that hold the contamination within the decontamination liquor. Aggressive chemicals such as strong mineral acids, for example nitric or fluoroboric acid, are usually found to be effective decontamination reagents. Some typical compositions can be found in Refs [6, 55].

Sequences of chemical treatments may be more effective for decontamination in some cases, for example the use of proprietary processes such as LOMI, CACITOX³, CORD or CITROX for the decontamination of water reactor circuits [56], and they also tend to be less aggressive. An example of this was the UK Atomic Energy Authority (UKAEA) using two different reagents in the past for the decontamination of the primary circuit in the Steam Generating Heavy Water Reactor (SGHWR) on the Winfrith site. The reagents were Turco reagent (80% ammonium oxalate, 15% oxalic acid and 5% citric acid) and LOMI reagent [57].

There are several different methods of application for chemical reagents:

- (a) Immersion: this technique may be enhanced by the use of heating, agitation or ultrasonic transducers.

³ Multicomponent leaching process including low concentrations of carbonate, oxidants and complexing agents, which convert insoluble or absorbed contaminants into soluble complexes.

- (b) Swabbing.
- (c) Recirculation, which may be used for large self-contained components or systems such as transport flasks and reactor circuits [58].
- (d) The use of foams or gels.
- (e) A combination of any of these techniques.

Whichever chemicals are used for decontamination, further treatment, conditioning and disposal of the spent waste reagent needs to be considered. There may be disposal limits on the concentrations of metal ions such as lead, chromium, etc. The use of organic reagents such as citric acid–EDTA (ethane diamino tetra acetic acid or its sodium salt) may also be limited, as these can cause problems with the complexing of heavy metals and actinides, thus preventing their removal in the secondary effluent treatment processes.

3.8.2. Typical hazards

The major hazard with the use of acids is their corrosivity. When diluted below corrosive levels mammalian toxicity is low, with certain exceptions (e.g. monofluoroacetic acid and oxalic acid) [20].

In general, severe aggressive effects on tissues are found only in the lower molecular weight saturated and unsaturated aliphatic carboxylic acids. Halogen substituted lower carboxylic acids and hydroxy and mercapto substituted acids are all severely aggressive to tissues in concentrated forms. Special care needs to be taken when hydrofluoric acid is used, because of the health effects on bone and tissue.

Complexing agents such as EDTA are of low acute oral toxicity, but some may be aggressive, causing severe damage to human tissue.

3.8.3. Possibilities for recovery and reuse

Once contaminated these types of material are rarely recycled or reused, since in most cases this could not be economically justified.

3.8.4. Waste management

Simple processes involving neutralization and filtration of reagents are often sufficient to remove the heavy metals and radionuclides that precipitate out of solution; the liquid waste stream can then be discharged. The corresponding sludge waste stream can be immobilized in a cementitious matrix and stored pending disposal.

Sometimes additional processes may be necessary to treat these chemicals to make them acceptable for disposal, such as destruction of the organic material present. A number of technologies have been investigated for this purpose; for example, the process described in Ref. [59] involves the following operations:

- (a) The solution containing residual chelate agent is placed in a precipitation vessel in which sodium hydroxide is added to precipitate heavy metallic ions as hydroxides. This results in the separation of heavy metallic ions from the chelate solution. The discharge from the precipitation vessel passes through a filtration process, where the precipitation filtrate is separated from the remaining chelate solution, which is directed to an electrolytic cell for organics destruction.
- (b) In the electrolytic cell the chelate solution is decomposed to carbon dioxide and water. Sodium hydroxide is added to support and enhance the electrolysis process. The carbon dioxide is exhausted to the atmosphere and the water is circulated within the process.
- (c) The remaining solution may be transferred to an ultraviolet photo-oxidation reactor or to a reverse osmosis membrane. In the ultraviolet reactor, air is ozonized when it passes near mercury lamps, bubbles into the solution and oxidizes the remaining organic matter and dissolves it.
- (d) The water solution with sodium hydroxide is then transferred to a reverse osmosis membrane, where the sodium hydroxide is recovered and is available for recycling.

To increase the efficiency of organics destruction a number of special methods can be employed, including, for example, ozonation [60] and various kinds of improved electrochemical oxidation [61, 62], or a combination of several methods may be used [63].

As an alternative to the separate destruction of the organic components, the decontamination chemicals can also be sent directly for incineration without pretreatment.

3.9. ASBESTOS

3.9.1. Form of the waste

Asbestos is the name of a family of naturally occurring minerals that consist of silicates (chemical fragments of silicon and oxygen) — a very common component of many minerals — and varying amounts of aluminium, calcium,

iron, magnesium, manganese, potassium and sodium. Each asbestos mineral forms long, thin needle-like fibres. The most common minerals are chrysotile, crocidolite and amosite [64]. Chrysotile is the type of asbestos most widely used within industry. Amosite is the major type of asbestos used in construction material within buildings. Further details on asbestos minerals and properties are provided in Ref. [64].

Nuclear laboratories and other nuclear facilities have long used asbestos cement board, since its strength and chemical and heat resistance made it a popular choice for the interior walls and backs of older style fume hoods. It is usually easy to decide if a fume hood contains asbestos cement board. The board has a very hard and smooth, grey, low lustre finish. The board is normally a medium grey colour. Newer fume hoods use plastic or stainless steel panels, and it is easy to distinguish these types of material from asbestos cement board. The same asbestos board was sometimes used as a counter top in laboratories or workshops to protect the cabinet from chemicals or heat.

Asbestos cement tiles were used until the late 1960s on building exteriors. The tiles come in various sizes but, again, the tiles are very hard and they sometimes have a textured surface. Since they are usually painted, colour is not usually helpful in identifying asbestos cement tiles. Occasionally, asbestos mixed with cement to form asbestos mortar was used as an exterior decorative finish on buildings. The mixture was trowelled or sprayed to form a continuous textured surface.

Heating pipes are usually covered with insulation to prevent heat loss. In buildings, glassfibre insulation normally insulates straight runs of heating pipe. However, asbestos is often present at elbows and around joints and valves. Both fibreglass and asbestos pipe insulation were invariably covered with a paper or fabric covering. In turn, the covering is often painted. It is fairly easy to assess whether pipe insulation is fibreglass or asbestos, since fibreglass pipe insulation is fairly pliable and 'gives' slightly when squeezed. In contrast, asbestos insulation is very firm.

Reactor pressure vessels and reactor containment buildings were commonly insulated with material containing asbestos. At the Windscale advanced gas cooled reactor in the UK the insulation material comprises several layers of a substance called Metadextramite. This is a mixture of magnesium carbonate and asbestos. This insulation was tied to the pressure vessel by an outer covering of wire mesh and cement, which was fixed to tangs on the pressure vessel exterior.

3.9.2. Typical hazards

Asbestos is only a concern when the fibres become airborne, because it is only when the fibres are present in the air that people can inhale them. Although there have been concerns raised about taking in asbestos fibres in food or water, there is no evidence that ingestion exposure to asbestos poses any risk. Neither is skin contact a concern. Some types of asbestos can cause skin irritation, but skin contact is not a direct cause of serious illness. Only inhalation of asbestos fibres presents a health hazard. If asbestos fibre is loose or can easily be crushed, the risk is high that the fibre can get into the air. In such cases a serious health risk might be present, particularly if a person were to be exposed to a lot of dust or to be exposed over a period of years. Asbestos fibres are extremely small and can remain airborne for an extended period of time. Of the asbestos fibres that can be inhaled, the most hazardous fibres are about 5–8 µm in length and about 1.5 µm in diameter.

Asbestos that can be easily broken or crushed is called friable asbestos. When asbestos is present in products in which the fibres are effectively bound, there is normally no hazard. However, drilling or cutting can free fibres even from these bound products. Under such situations, even tightly bonded asbestos-containing material can present a hazard.

The human respiratory system has evolved ways to deal with inhaled dust. The body can remove the large dust particles that become trapped in the upper parts of the respiratory system, and the body has other ways to capture and remove the very finest dust particles that can reach well down into the lungs. Unfortunately, the body's systems that clear dust from deep within the lungs do not work very well with asbestos. This poor performance of the deep lung dust clearing systems may be partially responsible for the illnesses that asbestos fibres cause.

The most significant medical problems that can be caused by the inhalation of asbestos fibres are asbestosis, lung cancer, mesothelioma and other cancers; these are discussed in more detail in Ref. [64].

3.9.3. Possibilities for recovery and reuse

Owing to its hazardous nature and its specific physicochemical properties there are no possibilities for recovery or reuse of asbestos. In many Member States the use of asbestos is now forbidden.

3.9.4. Waste management

The removal of contaminated asbestos from piping, vessels, walls, etc., has to be carried out by an authorized specialized firm. Preference should be given to companies that have experience of working in nuclear installations. For most other staff, observing special rules will minimize the chances of a significant asbestos exposure. Workers carrying out asbestos removal observe many rules to ensure their own safety and that of other building occupants. Normally such work is carried out by trained and authorized personnel with personal protective equipment and special precautions to prevent release of airborne fibres [65].

The safety precautions to be observed during asbestos removal are more stringent than for many other decommissioning works or activities at a nuclear facility. The removal of the contaminated insulation greatly improves the work environment for further decommissioning work. After removal, the contractor places the asbestos and any contaminated material into sealed heavy duty, labelled plastic bags.

‘Wrap and cut’ refers to a method of asbestos abatement. This method is used when a building or facility component, such as a length of piping with asbestos-containing material on it, is first wrapped in plastic sheeting. The entire wrapped component is then removed from the building. Sometimes asbestos-containing material is removed from part of the component to free it from the items around it. This method can also be used to wrap components with asbestos-containing material inside them. This method can be used instead of full containment procedures only when the asbestos-containing material in or on the component is in good condition. Other restrictions for this process also apply [66].

Although vitrification of asbestos is safer than incorporation in concrete, the latter is industrially used for asbestos originating in non-nuclear applications. Generally, asbestos originating from the decommissioning of nuclear facilities is only slightly contaminated and, depending upon the national policy, can be incorporated in a cementitious matrix after supercompaction with other inorganic material in a sacrificial container. However, if necessary, high temperature technologies are available for vitrification of highly contaminated asbestos [67–69]. The choice between solidification with cement and vitrification is dependent on the amount of asbestos to be solidified.

Acidic decomposition of asbestos to an amorphous silica suspension has also been considered as a method for treating asbestos prior to disposal [70]. This allows for the complete destruction of the asbestos fibres. The amorphous silica solution is solidified by the addition of lime, sodium silicate and possibly other reagents in preparation for disposal.

A mineral conversion process has been developed that changes asbestos to stable, non-hazardous minerals at temperatures significantly below those required for melting [71]. The asbestos minerals chrysotile and the amphiboles are known to convert to other mineral phases, such as pyroxines and olivines, at high temperatures. In a patented conversion process known as the Asbestos Conversion Process, proprietary chemical conversion agents are added to the asbestos-containing material. These additives greatly increase the rate and completeness of conversion. Sintering agents cause the fine material particles to combine into hard, durable masses that are free of fibres.

This conversion process has been tested on a variety of types of asbestos-containing material from actual abatement sites. A commercially viable transportable production system is currently in operation that is capable of successfully processing tonnes of asbestos per day. A volume reduction by a factor of ten can be achieved.

In addition to the Asbestos Conversion Process, a number of high temperature technologies are available for the transformation of radioactively contaminated asbestos to the form most appropriate for safe storage and subsequent disposal. Further details are provided in the Annex.

3.10. POLYCHLORINATED BIPHENYLS

3.10.1. Form of the waste

A commonly realized problem during decommissioning is the occurrence of polychlorinated biphenyls (PCBs). Due to their superior technical properties (e.g. water insolubility, fire resistance, long life, chemical inertness, high thermal conductivity, high electrical resistance) and their low cost, PCBs were widely used in technical installations, including nuclear facilities, as components in many types of organic material [72]. When such types of material are used in controlled areas they can become radioactively contaminated.

The following list [72] gives a summary of the types of material in which PCBs have to be expected:

- (a) Epoxy paints;
- (b) Oil impregnated electrical cable insulation;
- (c) Electrical transformer oils;
- (d) Hydraulic oils;
- (e) Machine cutting oils;
- (f) Lubricants;
- (g) Impregnation material;

- (h) Plasticizer for organic polymers;
- (i) Dielectric in capacitors.

When characterizing a facility for decommissioning, the possible PCB content has to be considered carefully and chemical analysis of the above mentioned sources is recommended.

3.10.2. Typical hazards

PCBs are a group of very stable organic compounds, and because of this they are persistent in the environment, resulting in an enrichment in the food chain. PCBs are soluble in lipids and therefore are enriched in lipid tissue when incorporated. PCBs are suspected to be carcinogenic. Incorporation cannot only take place by inhalation or ingestion but also by uptake through the unprotected skin. Although the acute toxicity is rather low, PCBs are enriched in lipid tissue due to their chemical stability. Therefore the chronic toxicity is of special concern. There is a long list of health problems related to the intake of PCBs [72–74].

3.10.3. Possibilities for recovery and reuse

The inherent hazards associated with this class of compounds do not allow the future reuse of PCB-containing material. In many Member States the production and use of PCBs is now forbidden.

3.10.4. Waste management

During decommissioning the initial removal of PCB-containing material and its processing are the biggest problems with respect to the personal protection of workers from exposure to these types of material. Once PCB-containing waste is removed it is recommended to incinerate it in an appropriate incinerator at temperatures exceeding 1200°C [74]. Destruction by incineration is the preferred treatment method; however, if incinerated improperly, another toxic component, dioxins, can be formed.

An advanced process for PCB treatment has been developed in Canada. This powerful process, based on a low temperature, atmospheric pressure reaction between a chlorinated contaminant and finely dispersed metallic sodium, is equally efficient in treating low level PCB contaminated oils as it is in treating decontamination liquids and solid waste containing high levels of PCBs. The toxic contaminant is converted to NaCl, NaOH and an organic by-product (polyphenols) [75].

If an appropriate incinerator is not available, PCB-containing waste must be embedded in an inert matrix such as cement and isolated from the environment. The way to achieve this depends on the technical and physical properties of the waste.

4. CONCLUDING REMARKS

The management of decommissioning waste and material can be site specific, facility specific or waste and material specific. When planning and implementing decommissioning options careful analysis should be performed in terms of the availability and applicability of special or conventional technologies for the safe and efficient management of all waste and material.

There are a number of types of waste and material that can be generated during the decommissioning of a nuclear facility that are problematic due to a combination of their conventional hazardous or toxic nature and their radiological hazard.

This report describes several specific forms of material and waste generated during the decommissioning of some nuclear facilities; they include beryllium, sodium, cadmium, mercury, lead, cyanide, decontamination chemicals, asbestos and PCBs. With these examples the scale of particular decommissioning waste management problems has been highlighted. The possibilities for reuse of some of these types of material have been considered; however, it should be emphasized that in many cases their recycle or reuse is not possible or feasible. Potential methods have been described for the processing and treatment of these problematic types of waste and material.

This report has considered the most commonly occurring problematic waste and material types. Other types of problematic material may arise, especially in research facilities in which complex tasks and a variety of experiments have been undertaken over decades. These, often small, quantities of problematic waste and material are challenging and are considered on a case by case basis.

Poor record keeping on the use of potentially hazardous material at the facility can result in unexpected combinations of problematic waste during decommissioning. To compensate for poor record keeping the decommissioning plan and strategy should include the careful characterization of all problematic material present in the facility in order to avoid difficulties at a later stage of the decommissioning process and in the management of the generated waste and material. To minimize these problems in the future it is

recommended to consider this issue at the design stage of the facility, as well as during operation and maintenance.

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Annex

EXAMPLES OF PROCESSING PROBLEMATIC WASTE AND MATERIAL

A-1. PROCESSING OF BERYLLIUM WASTE AT THE PAUL SCHERRER INSTITUTE, SWITZERLAND

During the decommissioning of the SAPHIR research reactor at the Paul Scherrer Institute, beryllium reflectors from the reactor core had to be conditioned for interim storage to wait for the availability of a disposal facility. The waste consisted of ten beryllium metal reflectors and ten BeO reflectors.

The ten beryllium metal reflectors each contained 6.1 kg beryllium metal with impurities of 1500 ppm aluminium, 1500 ppm carbon, 1800 ppm iron and 10 ppm cobalt. The beryllium metal was covered by lateral aluminium plates and fitted with aluminium end fittings, an aluminium holder and steel and aluminium screws, resulting in 3.4 kg aluminium and 0.1 kg steel screws each. The nuclide inventory at the time of conditioning was determined by means of an ORIGEN-2 calculation. The main radionuclides were tritium (7.05 PBq) and ^{60}Co (6.13 TBq).

The ten BeO reflectors each contained 6.5 kg BeO with impurities of <1 ppm cobalt. Each BeO reflector was totally covered by 2.2 kg aluminium. The nuclide inventory was determined by means of an ORIGEN-2 calculation. The main radionuclides were tritium (1.88 PBq) and ^{60}Co (450 GBq).

The total dimensions of the reflectors were $873 \times 83.5 \times 76.1$ mm (beryllium) and $873 \times 80.5 \times 76.1$ mm (BeO).

For conditioning of the beryllium reflectors for long term storage they were encapsulated in special containers for the purpose of:

- (a) Encapsulation of the tritium content;
- (b) Shielding of the gamma radiation caused by the ^{60}Co ;
- (c) Protection of employees, equipment and the environment with respect to the chemical toxicity of the beryllium and BeO;
- (d) Flexibility, due to a long interim storage period and unknown final repository.

A-1.1. Encapsulation process

The reflectors were not dismantled, to prevent any contamination of the equipment with beryllium or BeO. The reflectors were transferred by means of

a shielded transport flask to the hot cell facility. The flask was loaded under water and afterwards dried.

The reflectors were loaded into steel cans (Fig. A-1), which had dimensions of $950 \times 100 \times 100$ mm and a wall thickness of 4 mm. The cans consisted of Type 1.4301 stainless steel. The cans were gas tight welded to encapsulate the tritium. The 4 mm wall thickness was chosen on the basis of a corrosion rate of $1 \mu\text{m/a}$ in the environment of a potential final repository. The gap between the beryllium/BeO reflector and the can was filled with quartz sand (1.5–2.2 mm) to eliminate the voidage. On average, 5.2 kg of sand was used per can.

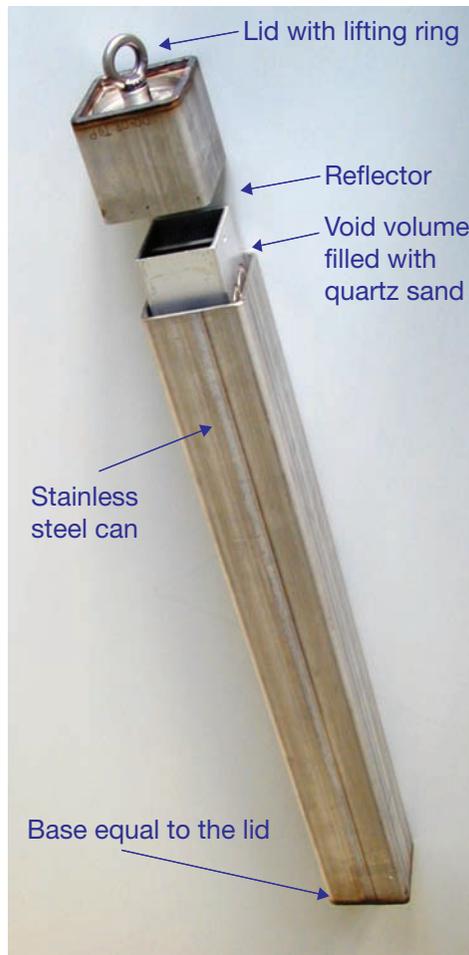


FIG. A-1. Beryllium reflector in a can designed for storage and disposal.

All 20 cans containing the reflectors were placed in one MOSAIK container (type II-15 U) and kept in their positions by means of a grid (Fig. A-2). The MOSAIK container provides shielding for the ^{60}Co gamma radiation.

The container itself is gas tight closed and allows gas sampling with respect to tritium contamination. In addition, the single cans are retrievable and can be re-evaluated at the time of final disposal with respect to their future dose rate after decay of the ^{60}Co . The MOSAIK container is accepted as a transport package and as a container for final disposal. The MOSAIK container with the canned reflectors has been placed at the federal interim storage facility at the Paul Scherrer Institute.

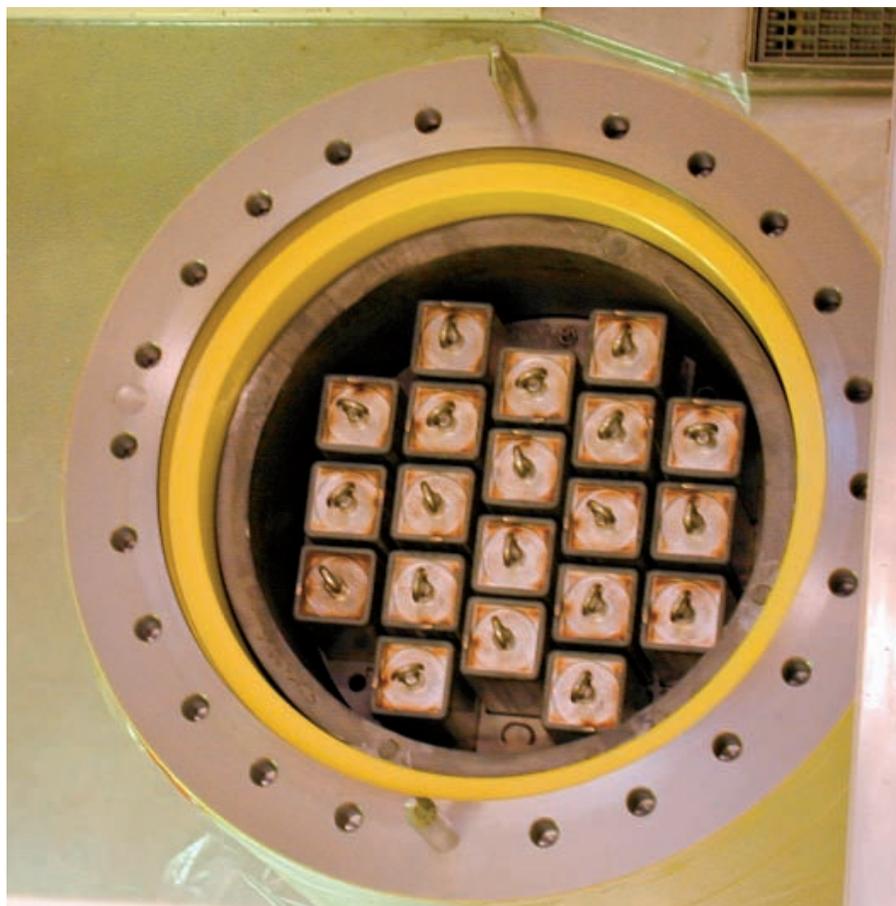


FIG. A-2. MOSAIK container loaded with canned beryllium reflectors.

A-2. PROCESSING OF BERYLLIUM WASTE AT THE BR2 REACTOR, SCK•CEN, BELGIUM

The BR2 reactor in Belgium is an MTR using high enriched uranium fuel and beryllium as the moderator (Fig. A-3). During the reactor decommissioning, the beryllium moderator and the stainless steel extension pieces needed to be unloaded and transferred to the decay pool. After 17 years of decay, the radioactivity (mainly in the stainless steel parts) decreased in such a way that treatment and transport of the beryllium moderators to the storage facility were feasible.

Each moderator contains 79 beryllium channels. There are three different channel types with different diameters and different geometries. Only the central part of the rod is beryllium (900 mm); the rest is stainless steel. The waste that originated from the first moderator contains 535 kg of beryllium. When packing the beryllium, the actual dose rates of different parts of different channels of the first moderator were measured; a large range was observed, due to the differences in irradiation history and geometry of the different parts. The dose rates of the heavy steel extension pieces ranged from 450 to 55 000 mSv/h, and for the beryllium from 350 to 1300 mSv/h. Table A-1 gives an overview of the radionuclide inventory of the moderator at the date of packing.

The management of the beryllium of the BR2 reactor can be summarized as follows:

- (a) The beryllium tubes and the stainless steel extension pieces were transferred from the storage pool to the hot cell of BR2.
- (b) The tubes were cut under water (to avoid beryllium contamination inside the hot cell) using hydraulic reciprocating saws.

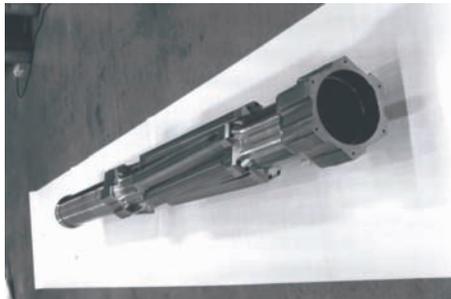


FIG. A-3. Beryllium channel with its two stainless steel extension pieces.

TABLE A-1. MEASURED, CALCULATED OR ESTIMATED SPECIFIC ACTIVITY OF THE NUCLIDES CONTRIBUTING TO THE TOTAL SPECIFIC ACTIVITY OF THE BERYLLIUM OF THE FIRST MODERATOR OF BR2 (AFTER 15 YEARS OF OPERATION AND 17 YEARS OF DECAY)

	Specific activity (Bq/g)	Methods for evaluation
Tritium	$0.3-2.0 \times 10^{10}$	Liquid scintillation counting
	$0.8-1.2 \times 10^{11}$	Correlation calculation through helium estimation
Beryllium-10	$0.9-1.7 \times 10^6$	Activation calculation
Carbon-14	$0.9-1.4 \times 10^7$	Calculation of $^{14}\text{N}(n, p)^{14}\text{C}$
Cobalt-60	$0.7-1.2 \times 10^7$	Gamma spectrometry
	$0.4-1.7 \times 10^7$	Activation calculation
Nickel-63	$0.1-2 \times 10^9$	Activation calculation
Nickel-59	$0.1-2 \times 10^7$	Activation calculation
Strontium-90	2.2×10^6	Estimation through contamination correlation with ^{137}Cs
Caesium-137	$1.5-2.5 \times 10^6$	Gamma spectrometry
Caesium-134	5.4×10^3	Gamma spectrometry
	4.2×10^3	Estimation through contamination correlation with ^{137}Cs
Niobium-94	$0.2-2.6 \times 10^3$	Activation calculation

- (c) The beryllium parts with the clamed top and the bottom ends of the extension pieces were loaded into canisters (Figs A-4, A-5).
- (d) After the drying process, the canisters were filled with quartz sand and closed with a screwed cover.
- (e) The closed canisters were then transferred back to the storage pool, where they were loaded by four into the transport container.
- (f) After transport to the treatment facility, the canisters were unloaded in a hot cell and placed inside a bigger canister.
- (g) The 4 cm void between the two concentric canisters was filled with cement.
- (h) Covers equipped with a carbon filter were welded on to the outer canister.
- (i) The canisters were then transported to the storage facility to wait for final disposal.



FIG. A-4. Four empty canisters for holding the beryllium.

A-3. PROCESSING OF LEAD BY MECHANICAL DECONTAMINATION AT UKAEA HARWELL

The UKAEA and its contractor (NNC) have decontaminated lead blocks arising from the decommissioning of a metallurgical suite that comprised three concrete shielded remote handling cells and 36 lead shielded enclosures. The primary decommissioning and dismantling work entailed the dismantling of the 36 lead enclosures, which were expected to yield over 1000 t of lead shielding bricks as waste [A-1].



FIG. A-5. Four empty canisters for holding the beryllium and the empty transport container arriving at the BR2 MTR.

During the initial dismantling of the lead shielded enclosures, all the lead bricks were monitored for radioactive contamination; clean items were segregated and set aside for detailed clearance and assurance checks. The contaminated blocks were sent for assessment and decontamination treatment, as necessary.

The decontamination process utilized a purpose built partitioned containment tent, ventilated with a HEPA filtration system, so that the receipt, decontamination and radiological monitoring of individual items could be segregated in order to minimize any cross-contamination. The dismantled lead blocks comprised a range of standard thicknesses (2, 4, 9 and 10 in, or 3, 8, 13 and

15 cm) and incorporated a variety of chevron, concave and convex shapes, which are utilized to avoid weaknesses within the assembled shielding.

The primary technical issues for the mechanical processing of the contaminated lead blocks were consideration of the individual lead brick shapes (i.e. the bricks were contoured) and the individual weight of the bricks, which had a range of 10–75 kg. The preferred option was a manual dry cutting technique using a handheld rotary industrial planer (the selected planer is normally associated with the joinery trade).

The dry cutting option considered the malleability of the lead, which under certain circumstances during dry cutting could give rise to localized heating effects, leading to melted lead over the cutting surface, resulting in limited effectiveness in the removal of the contaminated layer. To mitigate this effect the planer was set to take cuts typically 0.5–2 mm deep.

In addition, several handheld abrasive tools were utilized to remove contamination from areas locally inaccessible to the mechanical planer.

A-3.1. Lead decontamination process line

The containment tent was set up as a mini ‘production line’ with three distinct areas (Fig. A-6).

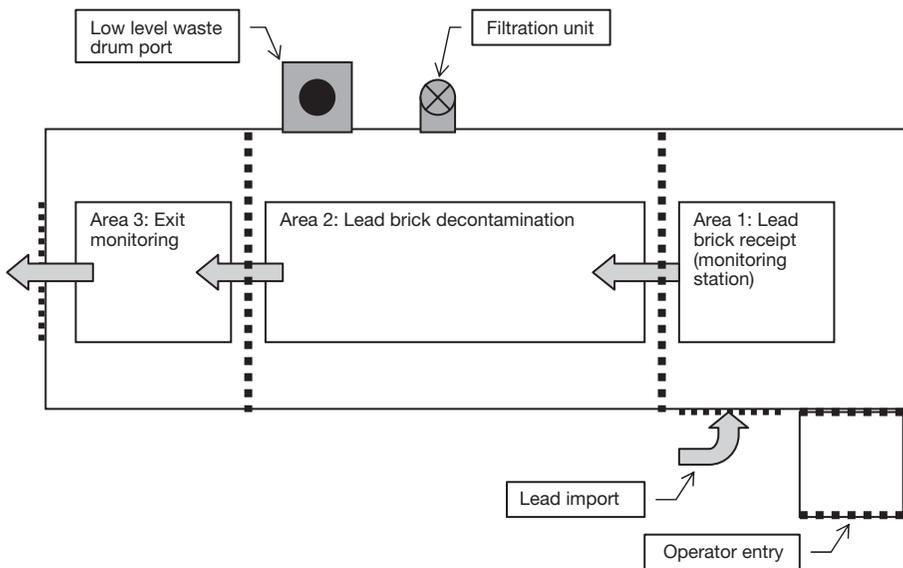


FIG. A-6. Lead decontamination process line.

Area 1: Lead brick receipt. Within the entry area the lead bricks (positioned on wood pallets and wrapped in polythene sheeting) were brought into the containment using a forklift truck. An access flap was opened and the palletted lead bricks were introduced into the containment tent and placed on a steel table. The polythene sheeting was removed and local receipt monitoring was undertaken at this point in order to ensure that the received items did not exceed the agreed level of contamination and radiation dose rate.

Area 2: Lead brick decontamination. Individual bricks were transferred to area 2. Bricks weighing less than 15 kg were transferred by hand, and bricks in excess of 15 kg were transferred by chain blocks and runner beam. The lead brick was placed on to a steel table and the exposed face cleaned using a degreasing agent and was radiologically monitored. If the radioactive contamination was recorded at levels higher than the clearance criteria, the exposed face was mechanically planed and remonitored. This process was repeated until the clearance criteria were met. The work surfaces were cleaned to remove any swarf and the brick was turned to expose the next face. The process was continued until the lead block was considered below the established clearance criteria (Figs A-7, A-8).

Area 3: Exit monitoring. The individual bricks were transferred to the exit area. They were radiologically monitored to ensure that they were below the clearance criteria set by the UKAEA and, if considered acceptable, an external exit flap was opened and they were transferred to a designated area for final monitoring and unrestricted release. The UKAEA then undertook routine audit monitoring of the bricks following processing, to provide assurance that the released bricks were within the agreed clearance criteria.

The primary factor in the waste management arrangements was to achieve as much unrestricted release of the potentially contaminated lead as possible. The process has been successful, in the free release after monitoring (for the year 2004) of approximately 400 t of lead as clean, 250 t of decontaminated lead for reuse in nuclear related applications and some secondary waste arising from contaminated lead bricks, lead swarf, protective clothing and contaminated polythene sheeting.

Throughout this process, reassurance monitoring of the lead was carried out to ensure correct classification of the decontaminated blocks. Final collection of the blocks and transfer from the site were carried out following detailed method statements and under the supervision of the UKAEA Project Supervisor to ensure safe carriage and compliance with the statutory requirements for lead handling.



FIG. A-7. Lead block planing.

A-3.2. Acknowledgement

Both NNC and the UKAEA acknowledge the support given by the UK Department of Trade and Industry to the decommissioning project at Harwell from which the lead brick decontamination process arose.

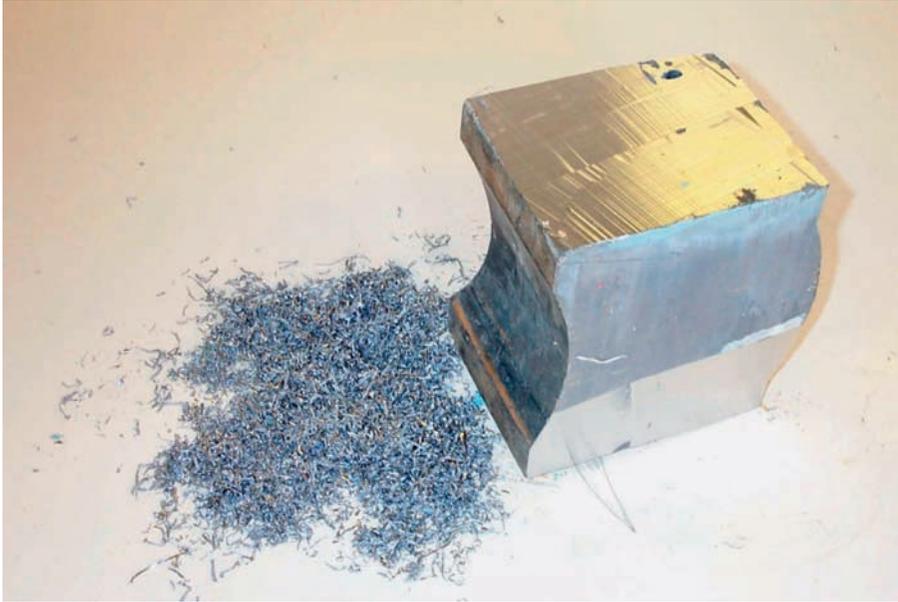


FIG. A-8. Lead brick and shavings.

A-4. PROCESSING OF LEAD BY MELTING: SCK•CEN, BELGIUM

An example of melting for the separation of clean lead from contaminated casing was demonstrated during the decommissioning of the BR3 reactor in Belgium [A-2]. A wall constructed in the BR3 refuelling pool shielding the first set of reactor internals had to be dismantled. This wall, made of seven shielding elements, each weighing about 5 t, contained lead cast in a stainless steel casing (Fig. A-9). The casing was contaminated and slightly activated (close to 100 Bq/g ^{60}Co for the external part of the shielding) and needed to be treated as radioactive waste. However, the activation calculation on the lead that was inside the casing showed, owing to the high purity of the lead (99.94%), that the activity should be lower than the clearance level. The risk of contaminating the lead during cutting of the elements was high, therefore extraction of the lead before dismantling was proposed.

The shielding elements were transferred into a containment tent and then heated batchwise up to 450°C by means of thermal resistance placed around the casing (Figs A-10–A-12). Applying electric power of about 127 kW, the melting temperature of the lead was reached after 5 h. The liquid lead was then



FIG. A-9. Removal of the last element of the shielding wall.

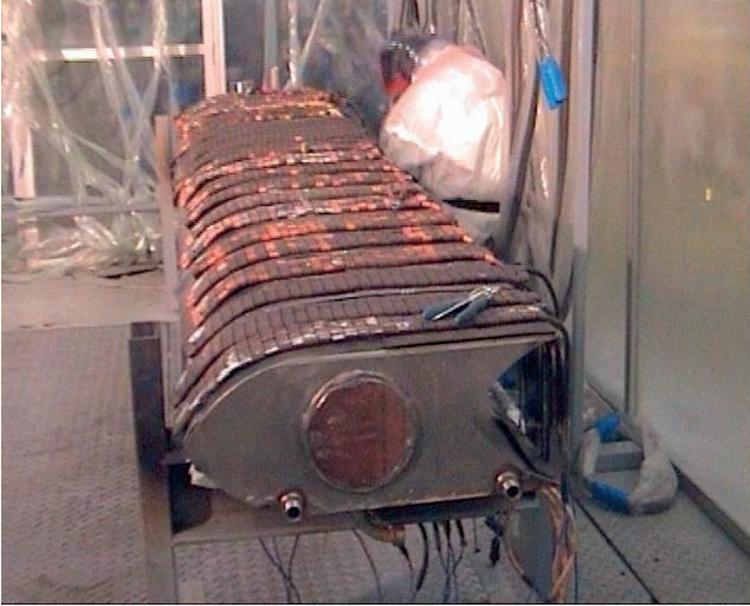


FIG. A-10. Placement of the heaters.



FIG. A-11. Placement of the insulation material.



FIG. A-12. Melting process.

drained from the casing into 300 kg casts. For the treatment of all elements, 120 ingots were produced for a total mass of 34 t.

The SCK•CEN has developed a specific procedure for the clearance of such material based on an activation calculation and a statistical measurement of samples taken during the casting of the molten lead. Activation and contamination products likely to be present in the lead moderator are ^{108m}Ag , ^{125}Sb , ^{60}Co and ^{137}Cs . This methodology allowed the project to clear all the lead ingots (Figs A-13, A-14). They have been sent to a scrap dealer for recycling in either nuclear or non-nuclear industries.

A-5. HIGH TEMPERATURE PROCESSING OF ASBESTOS WASTE

In direct arc heating melters the relatively high temperature of the process ($\sim 1400^\circ\text{C}$) leads to accelerated corrosion of the furnace lining and electrodes and, as a consequence, to a reduction of the inter-repair time of such facilities. Application of induction crucible melting furnaces is limited by the crucible campaign: about 30 fuses. Owing to the finite lifetime of the melters, both technologies are characterized by the large volumes of difficult to treat



FIG. A-13. Removal of the lead ingots after cooling.



FIG. A-14. Thirty-four tonnes of lead has been extracted and unconditionally cleared.

secondary waste in the form of radioactively contaminated cap moulds and ceramic material.

In order to prolong the operating time of the melters and to diminish the volumes of secondary waste, it is necessary to decrease the temperature of the process. This can be done, for example, by mixing the initial material with FeO (10–25% mass) or with the products of pressurized water reactor liquid radioactive waste calcinations, which contain oxides of potassium and boron (20–40% mass).

This approach enables the operator to decrease the temperature of the melting process to about 200°C. However, it complicates the technological scheme of the process and leads to an increase in the volume of the final waste form.

It is considered that some disadvantages of the methods described above can be avoided by employing an induction heated, water cooled skull melter made of stainless steel ('cold crucible' [A–3]) or an electroslag skull furnace with direct resistance heating [A–4].

A–5.1. High temperature processes: Cold crucible

The features of the cold crucible melter (in particular the loss of direct melt–wall contact) enable the furnace to melt various types of material at a broad range of temperatures (up to 3000°C) without problems with regard to corrosion stability of the structural material. Therefore asbestos can be melted without fluxing additives decreasing the temperature of the process or an increase in the volume of the end product.

Heating by a high frequency circuit enables the operator to intensify the process of the melting and thereby to increase considerably the relative productivity of the cold crucible in comparison with ceramic melters utilizing direct arc or resistance heating methods. In turn, at equal productivity the volumes of the secondary dismantling waste of a cold crucible are, as a rule, 40–50 times less than those of direct heating furnaces (see Table A–2).

Although the cold crucible melter system is non-repairable, it can be replaced remotely, and with essentially less complexity than the replacement of ceramic melter equipment.

A basic technological scheme of heat insulating material (HIM) melting in a cold crucible is shown in Fig. A–15 [A–5].

At present, equipment commercially produced in the Russian Federation provides an opportunity for manufacturing cold crucibles with an output of up to 70 kg(melt)/h or 1 m³(HIM)/h (Table A–3). However, the theoretical efficiency of such melters is up to 500 kg(melt)/h [A–5].

TABLE A-2. COMPARATIVE CHARACTERISTICS OF CERAMIC MELTERS WITH RESISTANCE HEATING AND COLD CRUCIBLE MELTERS FOR BOROSILICATE GLASS MELTING [A-5]

	CMRH ^a	CC ^b
Structural material	Refractory ceramics	Stainless steel, copper
Melt-structural material contact	Yes	No
Melt-electrode contact	Yes	No
Surface area of the melt (m ²)	0.4	0.15
Specific melt capacity (kg(melt)/(m ² h))	50	180
Temperature range (°C)	900-1200	900-3000
Power consumption (kW·h/kg)	2.5-3.0	5-6
Radionuclide loss (%)		
Caesium-137	≤3	≤3
Strontium-90	≤0.5	≤0.5
Alpha emitters	≤0.2	≤0.2
Efficiency (kg(melt)/h)	25	25
External dimensions (m)	3.0 × 1.5 × 2.0	0.59 × 0.30 × 0.65
Volume of secondary dismantling waste (m ³)	15	0.3

^a CMRH: ceramic melters with resistance heating.

^b CC: cold crucible.

A simplified version of an electrothermal facility for HIM melting has been developed and tested by the All-Russia Design and Research Institute for Integrated Power Technology (VNIPIET) [A-4]. This is an electroslag skull furnace with direct resistance heating with submersible electrodes (Fig. A-16). A standard metallic drum is used as a water cooled crucible and, concurrently, as the waste package. A thin layer of slag skull, generated on the interior surface of the crucible, guards it against burning and diminishes the heat loss.

This experimental facility used a 160 kW-50 Hz heater, a 200 L drum as the crucible container and various types of electrode — graphite and metallic tubes with various fillers. It has been shown that the paint on the interior surface of the crucible under the skull layer is preserved in its initial state, which indicates the normal temperature regime of the furnace.

As a result of remelting, the leachability of the strontium and caesium from the end product does not exceed 3.2×10^5 Bq/cm²/d in the case of ¹³⁷Cs and 5.3×10^5 Bq/cm²/d in the case of ⁹⁰Sr.

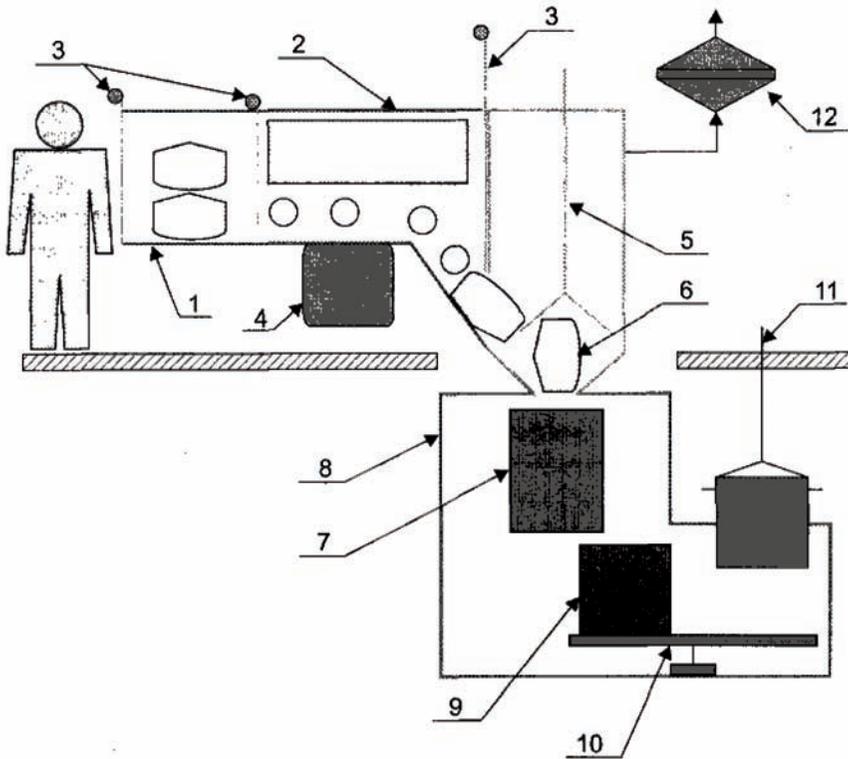


FIG. A-15. Basic technological scheme of HIM melting [A-2]. 1: receiving compartment of separator. 2: separator. 3: gate valves. 4: bags and wire bin. 5: pusher. 6: HIM. 7: melter. 8: technological chamber. 9: receiver. 10: anchor system mooring turntable. 11: elevating system. 12: bulk filter.

TABLE A-3. CHARACTERISTICS OF COMMERCIALY PRODUCED EQUIPMENT FOR INDUSTRIAL PROTOTYPES OF COLD CRUCIBLES

Type of generator	Power (kW)	Frequency (MHz)	Diameter of crucible (mm)	Height of crucible (mm)	Output (kg(melt)/h)	Output (m ³ (HIM)/h)
HFS-60/1.76	60	1.76	200	500	8.0	0.1
HFS-160/1.76	160	1.76	400	500	30.0	0.5
HFS-250/0.44	250	0.44	600	800	70.0	1.0
HFS-250/0.88	250	0.88	600	800	70.0	1.0

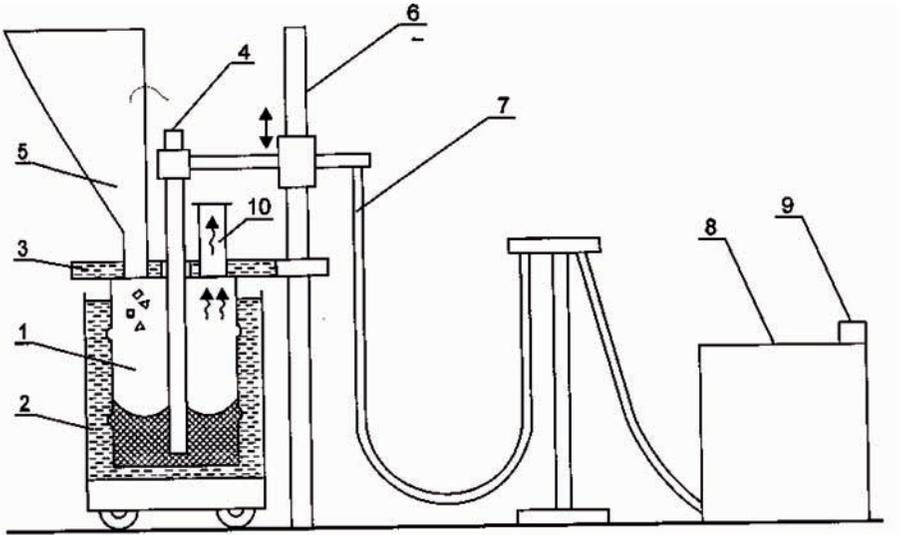


FIG. A-16. Scheme of electrothermal facility. 1: crucible/container (barrel). 2: cooling water tank. 3: water cooled cap. 4: electrode. 5: loading hopper. 6: leg. 7: power cable. 8: transformer. 9: gas flue.

The obvious advantages of such melters are their simplicity, their low cost, their small dimensions, their high specific output, the small volume of the secondary waste and the broad range of permissible temperatures. The latter advantage enables, in principle, the operator to employ the facility for conditioning (along with HIM) radioactively contaminated concrete, asbestos, brick, building debris, ash, furnace black and metals generated in decommissioning activities. At present, pilot facilities with three melters of 500 kW total power are under construction both in fixed and mobile variants [A-4].

REFERENCES TO THE ANNEX

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Decommissioning of nuclear facilities may present several problems in the management of some types of material and waste having not only a radioactive nature but also chemical toxicity, other dangerous properties or problematic characteristics. Material such as asbestos, beryllium, cadmium, mercury, lead and polychlorinated biphenyls, which were widely used in the past in the construction of facilities, are considered. This report outlines the management options for such waste and material, which are different from those for waste generated during the facility's normal operation.