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A Preliminary Inventory and Assessment of Uranium Resources in Mine Wastes

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A PRELIMINARY INVENTORY AND ASSESSMENT OF URANIUM RESOURCES IN MINE WASTES

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INTERNATIONAL ATOMIC ENERGY AGENCY
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FOREWORD

The IAEA provides support to its Member States through several mechanisms, including publications and databases related to uranium and thorium resources and production cycles. The IAEA's World Distribution of Uranium Deposits (UDEPO) database contains original geological resources for uranium deposits, with few records for remaining resources in the deposit and even fewer for remaining resources in mine waste. To increase the range of applications for assessing resource material flows, where original resources are converted to produced and remaining resources, this publication commences the collection of data for uranium inventories in mine waste products. The main focus is the extraction of uranium in waste from uranium mines, with a secondary focus being the extraction of uranium from deposits that are not solely uranium (so-called unconventional uranium deposits). Additional metals in waste from uranium mining are also briefly discussed.

Traditionally, discussions of the uranium production cycle focus on either the economic extraction of resources or on the environmental and remediation aspects as a linear cradle-to-grave process. Herein, the emphasis on resource evaluation is expanded to the full life of the mine, including evaluation of resources in residual waste products, in the context of potential reuse or remining of so-called anthropogenic resources in a circular economy. This publication is intended not only to provide a preliminary uranium inventory basis for the long-standing goal of comprehensive extraction in the (uranium) mining industry, but also to provide a basis for integrating this aim with the aims of environmental and remediation considerations in achieving zero waste.

The IAEA officers responsible for this publication were B. Gerstmann and M. Fairclough of the Division of Nuclear Fuel Cycle and Waste Technology.

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1. INTRODUCTION

1.1. BACKGROUND

Traditional linear economies follow a '*take-make-dispose*' plan, whereby raw materials are extracted and transformed into goods that are used until they are finally discarded as waste. Similarly, traditional uranium mining pursues a linear economic value chain by arranging value-adding activities in a sequence to fulfill the requirements of customers (Fig. 1). Such traditional economic activities generate a great diversity of wastes, and these wastes are commonly discarded near their production sites as mining, mineral processing or metallurgical wastes (e.g. uranium mill tailings are placed into tailings storage facilities).

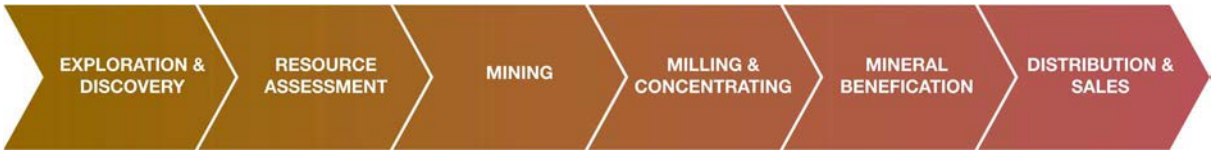

FIG. 1. Uranium mining value chain.

Within the linear uranium mining value chain, much focus is on the minimum grade required in order for uranium to be economically mined (i.e. cut-off grade) and on the quantities of uranium ores in the ground (i.e. in situ tonnages). In fact, in uranium resource evaluation, geological measurements of ore grades and tonnages are perceived as indicators of resource, and mining project and company value.

There are many other important factors that control the successful development of uranium resource projects, including the social licence to operate, knowledge of mine closure costs, environmental monitoring and remediation and finally the production, management and disposal of mine wastes. In particular, the traditional or historic uranium mining value chain does not only generate uranium ores and concentrates, it also commonly separates those valuable materials from wastes that are perceived to be worthless and may require safe disposal as per local regulations. An opportunity exists to mitigate short-term linear economic thinking, which is nearly exclusively focused on uranium ore extraction. In addition, the historic lack of local regulations and poor management of wastes in some earlier uranium mining facilities compared to modern uranium mines may have contributed to the extraordinary mine closure liabilities of some uranium mine sites.

As a result, many countries around the world are now facing challenges in relation to uranium mine legacy sites [1]. Historical mining methods were in part less efficient than today, environmental legislation did not exist, and contaminated mine wastes were often used for construction purposes. Consequently, there are several examples of historical uranium mine sites posing risks to the environment caused by the presence of uranium and other environmentally significant trace elements (e.g. As, Co, Ni) as well as radiological hazards [2–4]. These sites require ongoing long-term monitoring and in some cases remediation or treatment of effluent or groundwater. For example, effluent from some historic uranium mines and mills requires active or passive water treatment [5].

In a linear economy, mine waste production and waste repositories are an inevitable part of every mine. Therefore, past and present practices have led to variable mine waste quantities at individual mine sites, with an accumulation of significant very large amounts of mine wastes.

Today, an estimated 20 000 to 25 000 Mt of solid mine waste (waste rock and tailings) are produced annually worldwide [4]. Waste production varies greatly from mine to mine, nation to nation, continent to continent, and from year to year. For example, the mining industry of the European Union Member States (EU-28) represents the second greatest industrial sector in terms of generated wastes (25% or 642 Mt) [6], with at least 1200 Mt of historic tailings stored in the European Union (EU) [7]. Contemporary mine tailings production is significant, particularly in countries with a strong mining industry, with studies estimating between 5 000 and 14 000 Mt of tailings material produced per year worldwide [8–10].

Global mine waste production is forecasted to increase [4] because most high-grade ores have been previously mined and as a result contemporary mining tends to focus on the extraction of lower-grade ores [11, 12]. This includes most of the new uranium deposits as they are typically lower in grade [13], but there are regional differences, as evidenced by uranium deposits in Canada [14]. Even when richer deposits are discovered, they will be most likely deeper and accessing them will generate relatively more wastes. As a result, future surface and underground uranium mining operations will be associated with higher volumes of waste than previous operations.

At many locations, uranium mines and their waste repositories have been well managed, but there are opportunities for continual improvement with regard to waste management, improved uranium recoveries and recovery of other metals of economic interest. Linear economic mining practices and production of large mine waste quantities, coupled with long-term environmental impacts and loss of mineral resources to waste streams, have less than optimal sustainability for mining companies and statutory regulators that pursue the highest standards of environmental protection and responsibility [15]. Today and into the future, there is an obvious opportunity to minimize mine waste production, to reduce any uranium losses to waste streams and to integrate the uranium mining industry into a circular economy.

The circular economy concept looks beyond the traditional mantra of '*take-make-dispose*' and instead creates industrial circular systems with the environment in mind, following the '*make-use-return*' goal. In circular economies, mining is still vital to support the new industrial circular systems (Fig. 2) with the extraction, processing and use of raw materials including mineral resources optimized to generate little waste. In addition, historical or contemporary mine wastes are reprocessed, recycled, used or reused as raw material, and the remaining minerals or metals are extracted [16]. An increase in the recovery of minerals and metals including uranium from wastes, has the potential to result in reduced long-term environmental impacts (and related ongoing costs) from waste disposal.

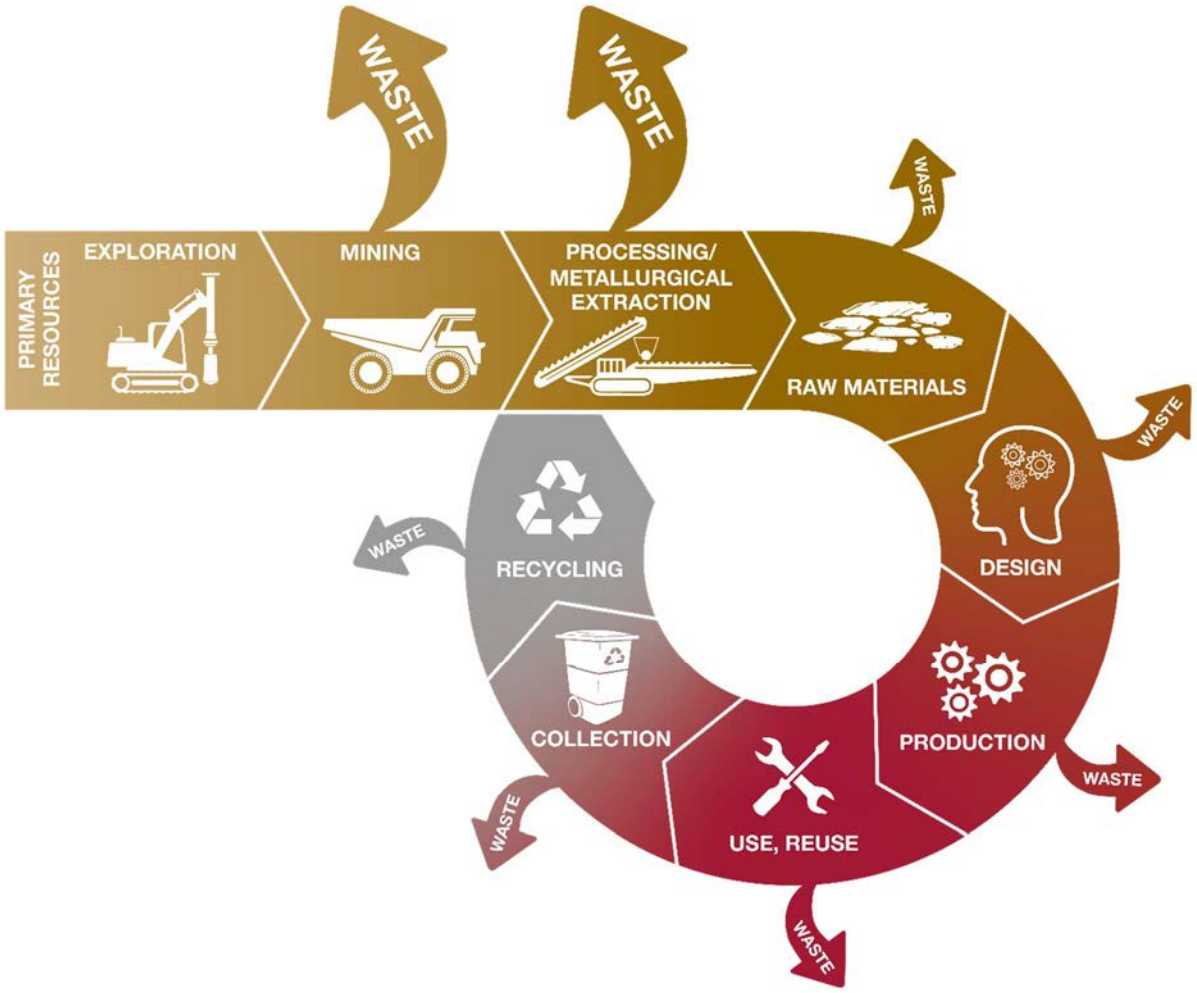

FIG. 2. The circular economy from a raw materials perspective (after [17]), illustrating the magnitude and challenge of significant waste production during mining, mineral processing and metallurgical extraction.

The knowledge that additional raw materials can be extracted from mine wastes is not new to the modern world. Recycling and reprocessing of mine wastes have been practiced in central Europe for at least hundreds, if not thousands, of years [18]. The aim of such pursuits has always been to recover marketable products from solid and liquid wastes of mining, mineral processing and metallurgical activities.

In the case of uranium mine wastes, the reprocessing of mine wastes subsequent to original uranium mining or site abandonment, the extraction of uranium as a by- or co-product from other metal ore deposits, and the treatment of mine waters to extract uranium have attracted interest since the beginning of uranium mining. There are plenty of examples to demonstrate this:

(1) The recovery of uranium during the wet acid processing of phosphorite ores has been pursued since the 1950s [19];
(2) Uranium has been recovered from copper leach solutions since the 1960s [20, 21];
(3) Sorption and ion exchange technologies have been used to extract uranium from mine waters since the 1970s [22, 23];
(4) Reprocessing of uranium mill tailings has been pursued since the 1980s [24, 25].

Should the political or market conditions for uranium improve, uranium wastes may represent a commercial value for a project proponent, e.g. [26].

To date, there have been numerous individual and targeted activities to extract uranium and other mineral resources from both uranium mine and other mining-type wastes. However, a systematic review of the global mine waste literature has demonstrated that research activities on mine wastes have concentrated on environmental impacts as well as the remediation of mine wastes and evaluating the rehabilitated mined areas for alternative uses [27]. Only a limited number of studies have analyzed uranium-bearing mine wastes from an economic point of view (e.g. Witwatersrand gold tailings), and there are no publications that document how uranium mine waste reprocessing may contribute to the goals of a circular economy. Thus, there remain significant gaps in our knowledge of how the extraction of uranium and other elements of economic interest from mine wastes may contribute to economic benefits and environmental stewardship in a circular economy. The properties of uranium mine wastes and the potential extraction of uranium and other elements of economic interest from solid and liquid mine wastes need to be documented for all waste types including former uranium mines. Such a wide-ranging assessment, which summarizes the opportunities related to uranium mine wastes, is lacking and filling this gap is one of the objectives of this document.

1.2. OBJECTIVE

This report presents information on major mine waste types from the extraction of conventional and unconventional uranium resources. The work focusses on the following aims:

(1) Provide a critical review of relevant readily available information (e.g. mineralogy, geochemistry, location, tonnages) in regard to uranium-bearing mine wastes;
(2) Provide a description of the scope of work required to gain a solid understanding of the potential of uranium supply from mine wastes;
(3) Incorporate uranium resources in mine wastes into the UDEPO database.

The aims of this study were met by: (a) reviewing existing company reports, publications and databases as well as in-house IAEA reports; and (b) comparing available data on mine wastes with best practice protocols and methodologies of established mineral resource classifications.

1.3. SCOPE

This desktop study reports on the occurrence and concentration of uranium in diverse mine waste types. The report specifically aims to:

— Provide information on the different mine waste types originating from mining, mineral processing and metallurgical extraction of U from uranium and other deposits (Chapter 3);
— Elaborate on the different wastes that originate from conventional and unconventional uranium resources (Chapter 4);
— Provide details on the distribution of uranium in mine wastes for selected major ore deposit types (Chapter 5);
— Document properties of the Witwatersrand gold tailings of South Africa that are relevant for uranium extraction from these wastes (Chapter 6);
— Document properties that are relevant for uranium extraction from wastes in legacy uranium mines in the former Soviet Union area of Central Asia (Chapter 7);

— Quantify the amount of uranium that may be available from selected uranium mill tailings and tailings storage facilities using available reports, publications and databases (Chapter 8);
— Describe any gaps of knowledge that need to be addressed to ensure responsible and successful extraction of uranium from mine waste sources in future (Chapter 9);
— Comment on the future of the uranium mining industry in a circular economy world (Chapter 10).

1.4. STRUCTURE

The focus of this study is to demonstrate that different wastes of conventional (e.g. polymetallic iron-oxide breccia complex deposits, palaeo-quartz-pebble conglomerate deposits) where uranium was not considered as an economic commodity and unconventional uranium deposits (e.g. phosphate rock deposits, black shale deposits) represent potential sources of uranium. If mine wastes are to be considered as a uranium resource, then any statements about possible resources, reserves and the extraction of uranium from mine wastes will require an increased knowledge and confidence on mine waste properties. Akin to geological resources, data and evidence on uranium waste sources would need to be documented and communicated using best practices, agreed assessment methods, classification frameworks and reporting standards. Therefore, a gap analysis was performed to identify any operative requirements for the successful extraction of uranium from mine wastes and related public classification and reporting standards.

This report is organised in a series of chapters that document the different properties of uranium mine wastes. Contents of Chapters 3–5 are inherent to mine sites extracting conventional and unconventional uranium resources. The content of Chapter 6 is primarily of importance to the mining industry of South Africa, but the principles of reprocessing tailings within the Witwatersrand area have been, and still are, relevant to tailings reprocessing operations elsewhere. Chapter 7 provides similar insights to Central Asia. Contents of Chapters 8–9 document the likely quantities of uranium stocks in tailings and present gaps in our understanding that need to be addressed to set minimum standards for public reporting of uranium extraction from wastes. Chapter 10 outlines a vision for future uranium mining, where innovation and entrepreneurship could potentially reduce waste and deliver the circular economy in uranium mining.

2. METHODOLOGY

2.1. BIBLIOMETRIC METHODS

A bibliometric analysis was performed to identify, organize, and analyze the main components within the topic of 'uranium in mine wastes'. Online databases including Google Scholar, Web of Science and OneMine were explored. Web of Science is considered a large repository of peer–reviewed literature, Google Scholar includes a large number of non–peer reviewed articles, and OneMine is a collective online digital library of mining and minerals technical papers, periodicals, books, and publications from professional societies and government records. Consequently, information from peer-reviewed and non-peer reviewed, as well as academic and mining industry related sources was obtained. To study the various topics of uranium in mine wastes, a descending search was performed. Such a search involved searching the database for a general topic and subsequently, more restricted searches of the topic were conducted until a specific source material was identified. Initially, a search was performed using parameters like 'uranium mine waste', with the aim of identifying all of the public information related to uranium in all waste types. The searches were limited to the years ranging from 1960 to 2020, and the results of these searches are given in Chapters 3 to 9.

2.2. DATABASES FOR QUANTITATIVE APPROACHES

Records of national and international agencies document uranium resources, deposits and production data based on statutory reporting codes. These records provide facts and figures for individual uranium mines or entire countries, such as uranium resources and uranium production. For example, IAEA TECDOC-1843 [28] and Hall and Coleman [29] document resource and grade ranges and resources and grades for individual deposits, respectively. Also, the OECD-NEA and IAEA [30] provides data on identified resources, which consists of resources and production of both conventional and unconventional resources that provides a country-level basis for comparing resource to production material flows to mine waste inventories, as well as information on South African resources from gold mine waste.

Mineral waste registries exist in some countries and, these databases are the best known information sources for assessing the material recovery potential from mine waste, tailings and metallurgical waste [31]. The databases from France, Hungary, Italy, Portugal, Slovenia, Spain, and the United Kingdom of Great Britain and Northern Ireland document a range of metals and metalloids, however, exclude uranium. Thus, these national mineral waste databases are not suitable as a source for data for potential uranium recovery projects and, they could not be used in this study for an assessment of uranium stocks in mine wastes.

This problem is not unique to uranium and the European Commission (EC) Directorate-General for Environment (DG ENV) has recently commissioned a study that aims to better understand the national reporting of mines wastes in EU Member States inter alia also in the context of enhancing a circular economy. The currently available aggregated data for mine wastes by Eurostat, the statistical office of the European Union, proved unsuitable for the purposes of this report. The EC (DG ENV) study is expected to be available in late 2020. Practical experience during the data collection exercise for this study has also shown that data pertaining to uranium are still considered 'state secrets' in some former Eastern Block countries although active mining may have ceased decades ago [32].

A new online global tailings dam portal that was launched in January 2020, 'Global Tailings Portal', provides details that mining companies have made about their tailings storage facilities [33]. The portal provides communities, investors, regulators and the media access to information about mine waste. However, the current version does not document various past and present uranium mine sites, and therefore the portal has been of little value to the current study.

With few exceptions, until recent years, the mining industry itself publishes very limited, if any, information on wastes generated at uranium mine and mill sites, in particular the details on the quantities and properties of waste rocks, tailings or water treatment sludges. Waste relevant data are generally missing from annual company reports, or the published data have not had the necessary resolution to deduct meaningful and quantitative assessments. In fact, there are only limited data freely available on waste production in uranium mining and milling (i.e. volume and mass). This lack of data and data density probably reflects the fact that mine wastes are still perceived worthless by some. Such data may also be considered sensitive, as they allow insights and conclusions to the drawn on the profitability and locations of individual uranium operations. Moreover, mine wastes were not systematically monitored, measured, analysed and documented for their chemistry and mineralogy in many legacy mines. Instead, most of the industry focuses on traditional profitability parameters and the physical and chemical integrity of modern waste impoundments.

The lack of publicly available uranium mine waste data and the deficiencies of national mineral waste databases makes direct deductions on quantitative uranium inventories in mine wastes difficult. Nevertheless, limited annual and cumulative uranium processing and production data were available for some individual uranium mines and mills. Mass balance equations and metallurgical accounting, commonly used to quantify mineral process operations [34], were applied to these data, which in turn allowed some quantification of the uranium present in uranium mill tailings and tailings repositories. The results of these assessments are given in Chapter 7. Calculated data in text and tables are given in triuranium octoxide (U_3O_8) values, whereas data from the literature are given as elemental (U) or oxide uranium (UO) values as stated in the bibliographic source.

2.3. LIMITATIONS AND CONSTRAINTS OF THE STUDY

A number of limitations of the study influence the interpretation of the findings in this work. These limitations constrain generalizability, applications to practice and the utility of the findings:

(1) Prior to this study, there have been limited review studies on the topic of uranium in mine wastes and uranium recovery from mine wastes and waste impoundments;
(2) Due to the lack of data, the study included self-reported data of mine waste production and wastes properties that could not be independently verified;
(3) There is a lack of available and/or reliable data on waste production and properties for particular mine sites and uranium mills that extract and process conventional and unconventional uranium resources;
(4) There is very limited information available on the global production data (mass, volume) of uranium and other mine wastes (e.g. tailings, waste rocks, mine waters, drainage treatment sludges);

(5) The precise amount of uranium contained in mine wastes locally, regionally and globally is difficult to quantify due to the lack of reliable data that document the production and properties of mine wastes.

2.4. DEFINING WASTE TERMS

In the following chapters, the properties and resource recovery options for uranium from liquid and solid mine wastes are presented and discussed, concentrating on wastes that accumulate at uranium mine sites. Considering that diverse words beginning with 'Re' are inconsistently used in the scientific literature and specifically when used to cover aspects of **re**mining, **re**processing, **re**cycling and resource **re**covery, these terms need to be defined in the context of uranium mine wastes. In this document, **remining** refers to the physical extraction of waste from waste repositories such as tailings impoundments. **Reuse** of mine wastes is the practice that finds a new use or application of the waste in its original form for a clear purpose without any processing or treatment. **Recycling** of mine wastes is defined as the activity that either extracts valuable resource ingredients using physical, thermal, biological or chemical methods, or uses the waste as feedstock for other purposes and converts the entire mine waste into a new valuable product. **Reprocessing** is defined as the targeted activity that uses mine wastes as feedstock to produce only certain valuable products such as uranium. By contrast, **treatment** of mine wastes aims to lessen the waste's toxicity or reduce its volume or mass. If commodities of interest are extracted from waste, this is termed resource **recovery**.

3. CLASSIFICATION OF WASTE TYPES

3.1. THE NOTION OF WASTE

For the purpose of this document, mine wastes are defined as solid or liquid by-products of mining, mineral processing, and metallurgical extraction [4]. At the time of production such wastes have no economic value based on company evaluations, are deemed unwanted or unusable, and accrue mostly at mine and mineral processing sites. Uranium-bearing mine waste refers to waste from a uranium mine or to any mining waste containing uranium. Therefore, uranium-bearing mine wastes are unwanted or unusable, have elevated concentrations of uranium and other by-products (e.g. Au), and consist of diverse materials (e.g. rock, sediment, tailings, metallurgical wastes). They are typically found at or near uranium mine sites.

Generally, the classification of mine wastes considers their origin and source, and such a classification scheme is applied in this document. The uranium mining industry extracts uranium ore from the ground (i.e. mining), processes uranium ore in mineral processing plants, and extracts uranium from uranium minerals (i.e. hydrometallurgical extraction). Each activity generates its own unique waste and therefore, uranium mine wastes can be categorized as mining, mineral processing and metallurgical wastes, and mine waters (Fig. 3).

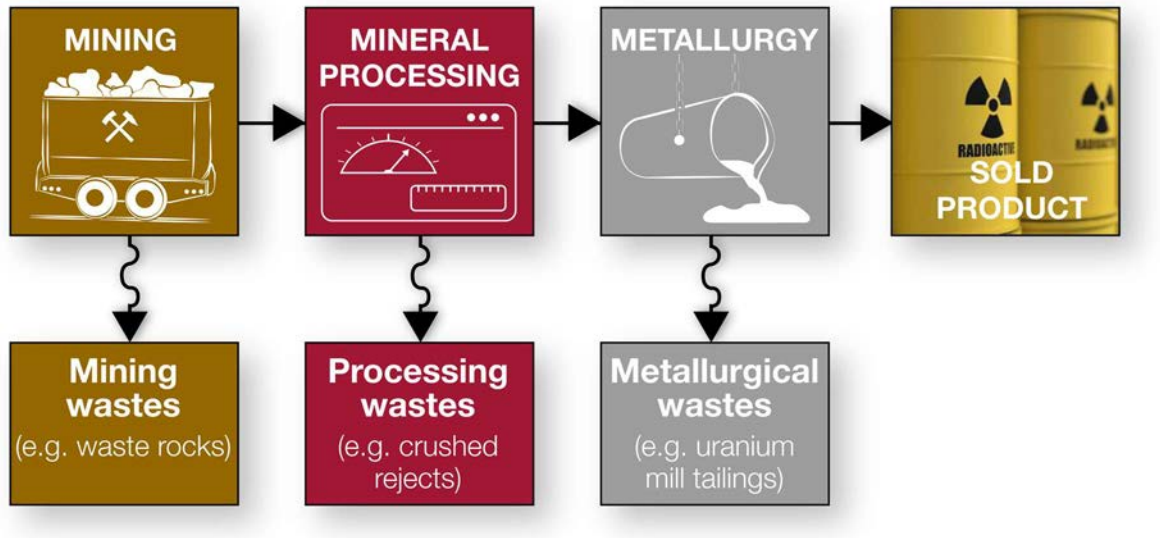

FIG. 3. Simplified flow chart of the mining, mineral processing and hydrometallurgical stages at a conventional uranium mine and mill that obtains ore from open pit or underground operations. The main waste products are illustrated for each stage.

3.2. EXTRACTIVE WASTE IN A REGULATORY CONTEXT

Notwithstanding the technical definitions of extractive wastes, the term ‘waste’ does have certain meanings and implications in different (national) regulatory systems. ‘Waste’ as a concept was introduced into (mainly) the environmental legislation around the world from 1970s onwards in order ensure the ‘orderly’ management and disposal of materials deemed as not useful to society at the time and to prevent its ‘wild’ deposition into the environment with ensuing contamination and public exposure issues.

The increasing life-cycle notion of materials now provides regulatory challenges in societies that increasingly consider reuse and recycle options with a view to keep i.e. geological materials, after extraction, as long as possible in the anthroposphere with the idea of the material having further beneficial uses. The disposal of 'waste' in most regulatory regimes requires a permit that also specifies the mode of disposal and the permissible emissions and releases (if any) from such sites. Depending on the regulatory regime, once a material has been declared 'waste' it may be very difficult, as practical experience has shown in some countries, to revoke this status. This puts up certain regulatory barriers to re-use and recycling. Similarly, in some regulatory systems the 'residues' of certain industrial processes automatically become 'waste' with a view to prevent a re-use that could be environmentally detrimental or be deposited at sites that do not provide adequate protection against adverse environmental effects e.g. emissions. Such regulatory provisions may constitute obstacles to reworking, re-use and recycling.

The European Union's Extractive Waste Directive (EWD) for instance

> "*covers the management of waste resulting from the prospecting, extraction, treatment and storage of mineral resources and the working of quarries*". According to the Directive "'*treatment' means the mechanical, physical, biological, thermal or chemical process or combination of processes carried out on mineral resources, including from the working of quarries, with a view to extracting the mineral, including size change, classification, separation and leaching, and the re-processing of previously discarded waste, but excluding smelting, thermal manufacturing processes (other than the burning of limestone) and metallurgical processes*" [35].

Thus, the residues (fly-ash, bottom ash) from burning hard coal or lignite in a power station are not 'extractive' waste. Likewise, slags from roasting ore, even when this happens at the site of the mine, are not 'extractive' waste. However, the residues from low-grade ore that had been subject to heap-leaching are considered 'extractive' waste.

Many countries aim to classify the different types of wastes, including those from the extractive industries, by attributing code to them codes. The European Commission established a set of six-digit waste codes on which also the reporting to Eurostat is undertaken [36]. A recent project undertaken for the European Commission's Directorate General on the Environment (DG ENV) on the status of implementation of the EWD (2006) and obstacles to implement the circular economy paradigm showed that the Eurostat statistics are neither detailed nor unambiguous enough for practical purposes [32]. It was found that different EU member states report different types of extractive waste under different waste codes and hence, the aggregation undertaken by Eurostat is confusing and not very useful for assessing recyclability. For the sake of maintaining continuity and comparability Eurostat is currently not considering any changes to their categorization, which unfortunately means that the current data is not that useful for practical purposes in enhancing circular economy in the EU mining industry.

3.3. MINING WASTES

Mining wastes are defined as assorted geological materials that comprise no valuable minerals or subeconomic contents of valuable mineral resources [37]. Mining wastes include overburden and particularly waste rocks extracted from surface and underground operations. In the case or uranium mining, such materials may, however, possess economic or sub-economic concentrations of other elements that are not extracted (e.g. Ni, Cu, Co, REE). At open pit

uranium mines, waste rocks are commonly placed into out-of-pit waste rock repositories, or waste-rocks are hauled into adjacent mined out pits. In underground uranium mines, there is minimal waste rock removal in comparison to open pit mining.

3.4. PROCESSING WASTES

Processing wastes are those portions of the crushed, milled, ground, washed or treated mineral resource with insufficient or no valuable mineral raw materials which could be economically processed. There may be concentrations of particular elements still present in these process wastes (e.g. leached Ni, Cu, Co, REE), but operators do not pursue their recovery. At uranium mining sites, processing wastes are typically coarse rejects as well as crushed and untreated wastes from the physical processing of uranium ores. In other non-uranium operations, rejects from coal washery plants and discarded materials from treated metal ores can also be considered processing wastes.

3.5. METALLURGICAL WASTES

In general, metallurgical wastes are defined as the discarded residues of a leached or smelted mineral resource, whereby metallurgical methods have been used to extract valuable resources (e.g. pyrometallurgy, hydrometallurgy, electrometallurgy, biohydrometallurgy). Such metallurgical wastes have no or insufficient valuable mineral raw materials or occur in mineral forms that are relatively refractory to leaching technology so that no further metallurgical extraction is economically justified. Materials may contain elevated concentrations of other elements that are not extracted (e.g. Ni, Cu, Co, REE in the raffinate stream). At uranium processing sites, hydrometallurgical wastes are typically uranium mill tailings and heap leach residues (Fig. 4).

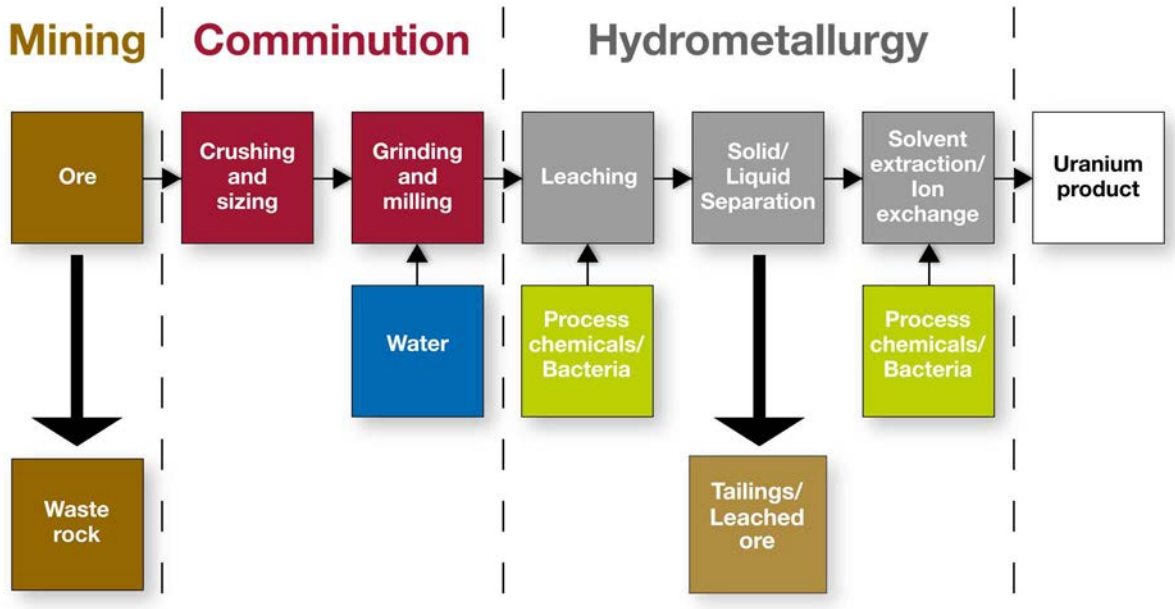

FIG. 4. Simplified flow chart at a conventional uranium mine and mill. Main throughputs and waste products are illustrated.

3.6. LIQUID WASTE STREAMS AND MINE WATERS

There are usually several liquid waste streams associated with uranium mining (i.e. mine water) and mineral processing (i.e. liquids or liquors). Mine waters are any waters that occur at a mine site including surface and groundwater. Mine waters include water from the dewatering of underground and open pit operations, which accumulate in the mine and must necessarily be brought to the surface. Moreover, seepage waters originating from tailings and waste rock dumps, abandoned heap leach piles and ore stockpiles as well as mine waters draining adits are waters that freely discharge, or are subject to treatment and subsequent controlled discharge to the environment. In addition, liquids or liquors rejected during milling and processing need to be collected, commonly into ponds or tailings storage facilities. In general, solutions containing spent solvents, leaching reagents, electrolytes and used oils from uranium mills require treatment or disposal. Considering that many of the mine waters and liquids are unwanted by-products of mining, mineral processing and metallurgical extraction, waters generated during uranium mining and solutions discharged from uranium mills are defined as wastes. At closed uranium mines, water generally continues to be an actively generated waste stream, well after active mining operations have ceased (e.g. seepage waters from waste repositories, decanting waters from flooded mines).

3.7. URANIUM LOSSES TO MINE WASTES

At uranium mines, losses of uranium occur during mining because of: (i) geological properties of the ore and waste; (ii) limitations or inefficiencies of the applied mining methods; and (iii) inherent incomplete recovery due to sub-optimal sorting/grading methods. For example, ore may be wrongly categorized as waste and directed to the waste rock dump. Alternatively, there may be ore dilution because waste is wrongly categorized as ore and sent to the mineral processing plant. Moreover, the mining method, such as room-and-pillar mining for example, does not allow complete extraction of the orebody. Such mining losses are generally lower for surface mines than for underground mines (Table 1).

In addition, at uranium mills there are metallurgical/milling losses (inefficiency in the recovery process) due to the leaching characteristics of the ore and the applied extraction procedure. Generic average recovery factors for various mining and processing methods from the OECD-NEA/IAEA publication ‘Uranium resources, production and demand’ [30] are provided in Table 1. Such mining and milling losses imply that a proportion of any identified uranium resource will remain in the ground or be lost to mine waste repositories, particularly tailings storage facilities. Here, the recovery rates of uranium in leach tanks and the associated losses of uranium to tailings storage facilities are influenced by a range of factors including: uranium mineralogy; residence time; particle size; leach temperature; acid concentration and pH; oxidation-reduction potential; presence of oxidizing agents; and pulp density.

TABLE 1. GENERALISED RECOVERY FACTORS FOR MINING AND MILLING [30]

Mining and milling method	Overall recovery factor (%)
Open-pit mining with conventional milling	80
Underground mining with conventional milling	75
In situ leaching (acid)	85
In situ leaching (alkaline)	70
Heap leaching	70
Block and stope leaching	75
Co-product or by-product	65
Unspecified method	65

4. WASTE TYPES OF CONVENTIONAL AND UNCONVENTIONAL URANIUM RESOURCES

Uranium resources can be broadly classified as either conventional resources or unconventional resources [30, 38]. These two resource types differ in their intrinsic properties and hence, uranium mine waste types are best presented using the IAEA classification system for uranium resources [38]. In the following, the different waste types of conventional and unconventional uranium resources are documented.

4.1. MINES WASTES OF CONVENTIONAL URANIUM RESOURCES

Conventional uranium resources have been defined as those resources that have an established history of production and generate uranium as a primary product, co-product or an important by-product [30, 38]. Conventional uranium resources are commonly exploited using established open pit or underground mining, mineral processing and hydrometallurgical extraction. Alternatively, in situ leaching (ISL) is applied to subsurface sandstone-type uranium ores.

Open pit mining methods of conventional uranium resources create the greatest diversity and quantity of mine wastes including waste rocks, heap leach residues, uranium mill tailings, mine waters and water treatment sludges. By comparison, underground mining methods may generate significant amounts of waste rock during mine development and then less significant waste rock during ore production. Tailings production of underground mining operations is still significant. Finally, active uranium mines using ISL techniques do not produce any waste rock or tailings and generate the smallest volume of mine waters and water treatment sludges. Yet, post-closure generation of contaminated water from ISL mines is often significant and can last for decades or even centuries. Therefore, considering the methods of uranium extraction, mine wastes of conventional uranium resources can be assigned to the following categories:

— Waste rock;
— Uranium mill tailings;
— Spent heap leach ores;
— Mine waters;
— Drainage treatment sludges.

4.1.1. Waste rock

Uranium ores are commonly enclosed by un-mineralized or poorly mineralized rock, which in open pit operations needs to be extracted to gain access to the ore (Fig. 5). Thus, waste rock can be defined as rock that has been excavated from open pits and is then transported out of the surface workings, because such rock does not have uranium concentrations of economic interest. Therefore at uranium mines, any mined waste rocks generally have very low or background uranium concentrations [37].

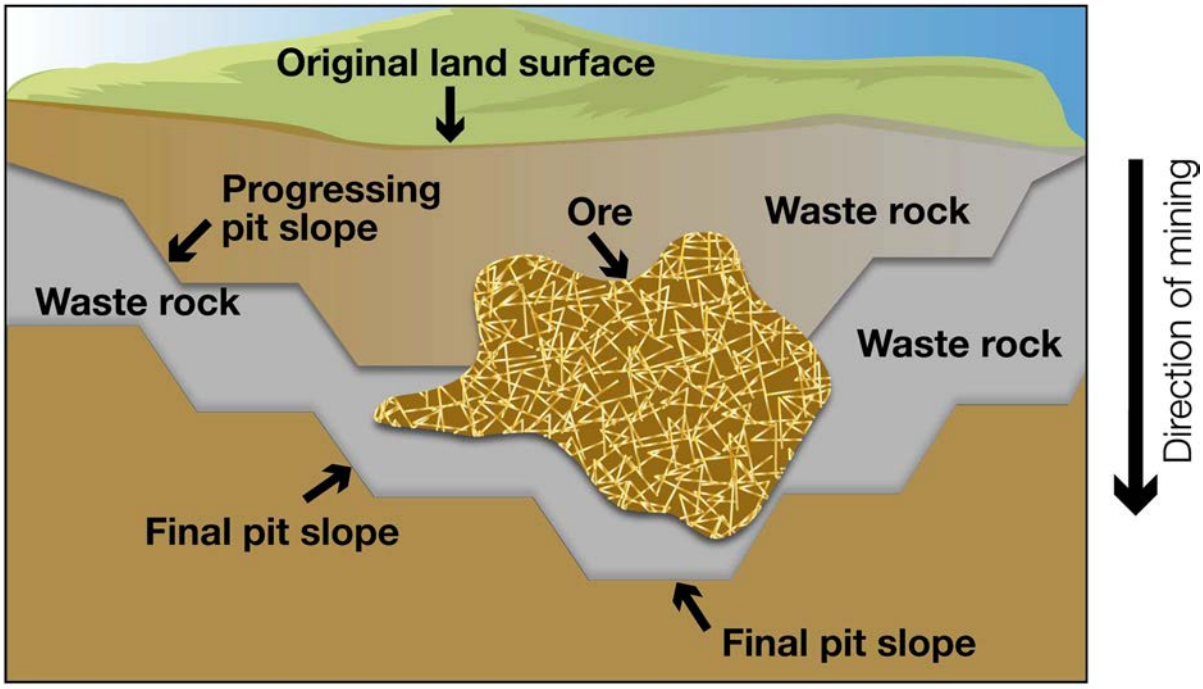

FIG. 5. Schematic cross-section of an open pit uranium mine. Extraction of uranium ore leads to the production of waste rocks that are discarded in out of pit waste repositories or transported into mined out pits.

4.1.2. Uranium mill tailings

Uranium tailings are hydrometallurgical wastes from a so-called uranium mill. A uranium mill (also referred to as metallurgical plant) is a chemical plant that extracts uranium from conventional uranium resources. A typical uranium processing flowsheet consists of mining followed by comminution, leaching, solid-liquid separation, solvent extraction and precipitation of uranium (Fig. 6). Uranium is extracted from its ores using sulfuric acid or alkaline solutions as leaching agent; impurities are taken out using solvent extraction or ion exchange; and uranium is precipitated with magnesium hydroxide, ammonium hydroxide or hydrogen peroxide to yield yellow cake.

After leaching of finely ground uranium ore in the mill, the solids are removed from the processing circuit, treated and pumped with excess process waters to the tailings' storage facility (Fig. 6). In order to alleviate issues with the stability of tailings storage facilities containing liquid tailings, accelerated dewatering of tailings by filtering or cycloning is sometimes practiced, resulting in less voluminous 'paste' tailings being deposited or used underground as ground support [39]. Such wastes are referred to as 'uranium tailings' or 'uranium mill tailings', which consist of solids and liquids. The solids are best further classified according to their particle size as sands (relatively coarse material) and slimes (fine grained), with each component having distinct properties depending on ore characteristics and applied mineral processing and hydrometallurgical extraction techniques (Table 2).

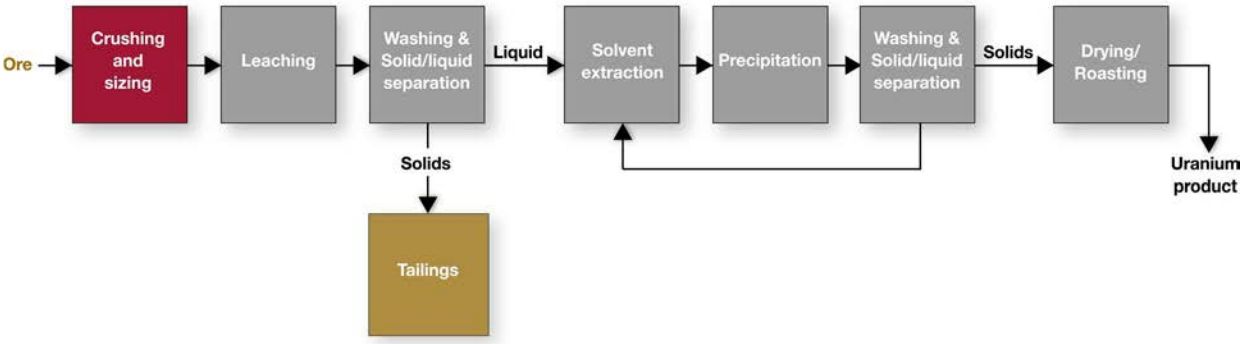

FIG. 6. Simplified flow diagram for the mineral processing and hydrometallurgical extraction of uranium.

TABLE 2. GENERAL PROPERTIES OF URANIUM MILL TAILINGS [40]

Component		Particle size	Chemistry	Mineralogy
Solids	Sands	>0.075 mm	Ore element signature	Mostly gangue minerals
	Slimes	<0.075 mm	Ore element signature	Mostly gangue minerals plus fine-grained clay minerals, oxides, fluorides, sulfates, amorphous phases
Liquids		—	*Acid leaching*: pH ~2, high SO_4^{2-}	—
			Alkaline leaching: pH ~10, high CO_3^{2-} & HCO_3^-	—

To date, there has been considerable attention towards the physical, geotechnical, chemical and radiochemical risks and resultant environmental impacts associated with uranium mill tailings [39]. Such focus is warranted because, for example, leaching of the powdered ore may also mobilise several other elements into the process waters (e.g. As, Fe, Mo, Ni, Pb, Se, V). If not properly treated, these environmental elements of concern may migrate, causing environmental damage or even toxicity to organisms.

Within uranium mills, the recovery of uranium from ores may vary significantly, yet many modern processing plants achieve on average 95% or better, depending on uranium ore mineralogy. Such extraction efficiencies imply that 5% of the original uranium ends up in the tailings. In fact, uranium concentrations in tailings typically range from several tens to hundreds of ppm uranium, caused by variable leaching efficiencies in the mill (Table 3). Historically, processes may have been less efficient. For example, at the Ranger uranium mine (Australia) a total of 19.78 Mt of uranium ore (laterite, weathered and fresh ore) were extracted from an open pit, containing an average 3 420 ppm U_3O_8 (equivalent to 2 744 ppm U) [41]. Between 1982 and 2000, the mine produced a total of 19.45 Mt of tailings at an average leaching efficiency of

89.2%. The mean uranium concentration in tailings from that period was 357 ppm uranium and thus higher than in many South African gold ores, from which uranium was co-produced [41].

TABLE 3. URANIUM CONTENT OF URANIUM MILL TAILINGS FROM DIFFERENT MILL SITES [2, 39, 42–48]

Country, location	Uranium concentration (μg/g)
Argentina, Sierra Pintada	100
Argentina, Los Gigantes	84
Australia, Radium Hill	149 to 1 600
Australia, Mary Kathleen	7 to 61
Canada, Rabbit Lake	152
Canada, Elliot Lake	25 to 65
Canada, Cluff Lake	6 700
France, Escarpiere	126
Poland, Kowary Podgórze	30 (up to 240)
Rössing, Namibia	64
USA, Slick Rock (acid-leached)	531
USA, Slick Rock (carbonate-leached)	350
Slovenia, Zirovski	~81

TABLE 4. CHARACTERISTICS OF URANIUM HEAP LEACH SYSTEMS [49]

Parameter (unit)	Value
Ore grade (U_3O_8 %)	<0.1
Tonnage (Mt pa)	1.4–36
Heap height (m)	6
Leach agent	H_2SO_4, Na_2CO_3
Leach agent consumption (kg/t)	15–40
Leach time (days)	40–100
Uranium extraction (%)	60–80
Irrigation rate (L/h m^2)	5–15
Issues	Fine crushing, oxidizing reagent possibly required
Capital costs (US$/t of ore to heap)	26–75

4.1.3. Spent heap leach ores

Heap leaching is a hydrometallurgical method, whereby crushed ores (typically low-grade <0.1 % U_3O_8) are piled over an engineered impermeable pad and leached with sulfuric acid or sodium bicarbonate as lixiviant under atmospheric conditions [50–52]. Uranium oxide minerals are thereby converted into water-soluble sulfates, and any leachate is gathered for uranium recovery. Many factors control heap leach efficiencies and therefore, the uranium concentrations of spent heap leach piles may vary. In particular, particle size and size

distribution, fluid flow through the media as well as mineralogical characteristics are important parameters determining leachability [49, 50]. Once the heaped material ceases to produce noteworthy uranium, the heap leach facility is rinsed and drained. The facility is then closed in situ, however, the leached ore may also be removed and placed in a separate lined repository. Regardless, the recovery of uranium in heap leach piles is variable, ranging from 60 to 80% (Table 4). Such extraction efficiencies imply that 20 to 40% of the original uranium remain in the spent heap leached ores.

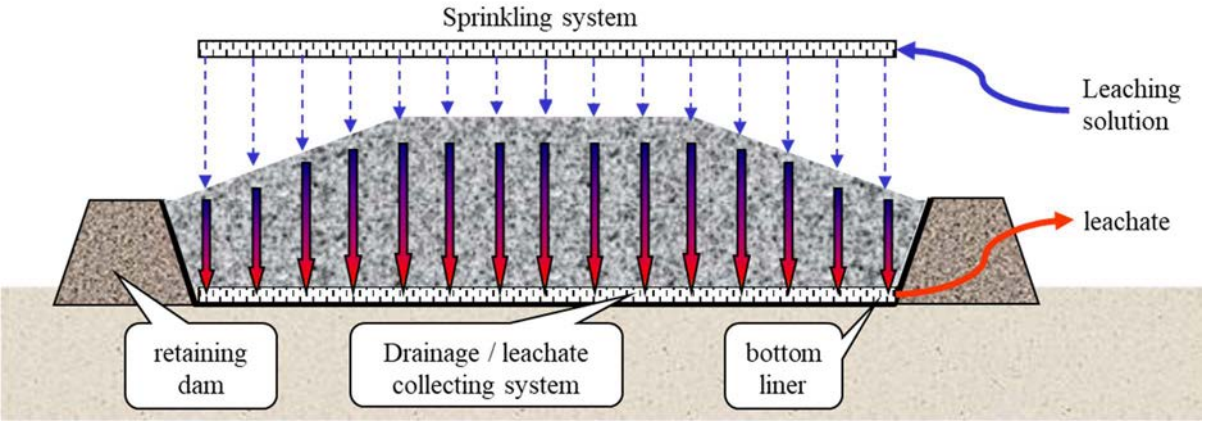

FIG. 7. Schematic cross-section of heap leach system [39].

4.1.4. Mine waters

Sulfide minerals like pyrite, marcasite or pyrrhotite are mineral forms in which the metal ion occurs in its reduced state. Accordingly, uranium ores that may contain these minerals also contain the reduced uranium specie. The presence of relatively large amounts of pyrite, marcasite or pyrrhotite in pit faces, underground workings, tailings, ore and waste rock piles may lead to acid mine drainage (AMD). AMD is a process, whereby iron-sulfide minerals are exposed and react with atmospheric oxygen, bacteria and water to form sulfuric acid which can reduce the pH conditions. The low pH conditions especially favour the dissolution of many metals and metalloids including uranium.

Uranium ore minerals can be categorized by the occurrence of uranium in the reduced (U^{4+}) and oxidized (U^{6+}) states. For example, uraninite contains uranium in its reduced (U^{4+}) state, whereas secondary uranium minerals formed during weathering and in surface environments are oxidized species (U^{6+}). Consequently, reduced uranium ores may contain abundant iron-sulfide minerals and, the formation of AMD waters poses severe environmental risks due to their high acidity as well as toxic metal and sulfate concentrations [4].

Possible resources that can be recovered from AMD waters include iron oxides and oxy-hydroxides, elemental sulfur, sulfuric acid and dissolved metals, uranium included [53]. In particular, there has been increasing interest in recovering metals from mine waters, and in addition to the obvious metals that may be recovered, the resulting ‘clean’ water itself can be considered as a potential product [54].

At some uranium mines, mine waters may show elevated uranium concentrations. Liquid effluent from uranium and non-uranium mines and mills during and after operation have long been known to contain mg/L concentrations of uranium [20, 22]. In conventional uranium

mining, this water is commonly recycled (particularly to the grinding/leaching circuit), where the uranium can be recovered. For any process water that is not recycled back to the grinding/leaching circuit, uranium may be recovered prior to wastewater treatment. AMD waters associated with uranium mines can contain mg/L concentrations of uranium, e.g. 15 mg/L U, Poços de Caldas, Brazil [55–57]. Uranium recovery from mine waters has been pursued since the 1970s, using sorption and ion exchange technologies [22, 23].

4.1.5. Drainage treatment sludges

At some uranium mines, metalliferous mine waters draining from tailings repositories, waste rock dumps or underground mine workings may require active or passive water treatment (e.g. uranium mine sites in Brazil, Canada, Germany and the United States of America [58, 59]). In particular, the oxidation of large amounts of pyrite, marcasite or pyrrhotite may lead to significant volumes of AMD waters that in turn require treatment. During acid water treatment, the sulfuric acid is consumed using a neutralizing agent (e.g. lime), and the dissolved sulfate and metals including uranium are removed from the mine water and precipitated as solids to form voluminous sludge. Such sludges typically comprise solid phases precipitated from the treated mine waters and includes hydroxides (e.g. iron phases), sulfates (e.g. gypsum, basanite, ettringite), carbonates (e.g. calcite) as well as amorphous and poorly crystalline material.

Any sludge generated over time ought to be taken from the mine water treatment system and placed into an appropriate waste impoundment. However, there are also various recovery, recycling and reuse options of sludge originating from mine water treatment. Possible valuable raw materials include iron oxy-hydroxides, metals, elemental sulfur, and calcium carbonate [60]. Sludges may also be reused as soil conditioner or fertilizer, substitutes for construction purposes, as cover material for tailings storage facilities to prevent sulfide oxidation, as innovative material to sequester carbon dioxide, and as raw material in the cement and pigment industries [60].

Unfortunately, there is very limited information available on the properties of sludge produced at individual uranium mine sites and the global quantities of sludge, because the properties of sludges, produced in these mine water treatment plants, are generally not systematically monitored and publicly documented [58]. However, some deductions can be made. Water treatment plants at uranium mine sites are designed to lower dissolved uranium concentrations and therefore uranium becomes concentrated in any sludge generated. Hence, at the Poços de Caldas uranium mine site for example, sludges may contain uranium concentrations high enough (2 420 ppm U) that uranium recovery becomes economically attractive [61]. In fact, uranium extraction from the Poços de Caldas sludge, which is continuously generated during AMD treatment, is possible via leaching with carbonate-based reagents [61].

4.2. MINE WASTES OF UNCONVENTIONAL URANIUM RESOURCES

Unconventional uranium resources are defined as: (i) those that are very low-grade resources; or (ii) those from which uranium is only recoverable as a minor by-product [30, 38]. A review of these sources and their potential utility can be seen in [62]. Mining, mineral processing and metallurgical extraction of unconventional uranium resources (e.g. phosphate rock, bauxite) yield in some cases the very same waste types as conventional uranium resources (i.e. waste rocks, tailings). However, mining and utilization of unconventional uranium resources generate additional wastes. These further waste types and classifications are described in the remainder of this chapter.

4.2.1. Bauxite residue

Bauxite is commonly refined by using concentrated sodium hydroxide (NaOH) to dissolve the aluminium ore minerals (i.e. the Bayer process). Any solids that do not dissolve in sodium hydroxide are extracted from the solution and discarded from the hydrometallurgical processing circuit. The generated waste product is referred to as 'bauxite residue', with the finer residue fraction called 'red mud' and the coarser fraction 'red sand'. Approximately 150 Mt of bauxite residue is produced worldwide every year [63].

Several alternatives to the storage and disposal of bauxite residues into waste impoundments have been proposed. Such reuse options include: cement production; raw material for making glass, iron and steel; manufacture of building materials such as ceramics or bricks; landfill capping; road construction; and soil amelioration [63]. In addition, red mud contains abundant valuable components, such as iron, aluminium, titanium, rare earth elements, thorium (20–30 ppm), gallium (60–80 ppm), yttrium (60–150 ppm), scandium (60–120 ppm), and uranium (5–-60 ppm) [64, 65]. Red mud is perceived as a valuable secondary resource of these trace elements and, sulfuric acid leaching and subsequent impurity removal by selective precipitation have been explored to extract these constituents [66]. Reprocessing bauxite residues may also have the added value of being able to redispose it in more stable forms after extraction of the metal value [67].

4.2.2. Tin slags

At tin smelters, smelting of cassiterite concentrates yields tin ingots as well as slags. Slags are known to be mineralogically and chemically diverse pyrometallurgical waste materials and comprise variable quantities of glass and crystallized phases as well as relict ore, gangue and flux minerals. The chemical composition of slags is largely a function of the ore and flux composition as well as the applied metallurgical processes.

Tin smelter slag originates from the smelting of cassiterite ores, which may possess significant amounts of trace elements including uranium (i.e. as cation substitutions within cassiterite) [68, 69]. Upon smelting, the uranium is rejected into the slag and therefore, tin slag may be considered as a possible source of uranium [70]. Recovery of uranium from tin slags is achievable using strong acid digests.

4.2.3. Coal bottom and fly ash

Substantial concentrations of uranium have been documented for lignite deposits of the United States of America and China and even more so in the fly ash produced from the use of lignite in power plants [71, 72]. Investigations of uraniferous coal from the Sweetwater County (USA) indicate that uranium is largely hosted by organic coal components [73]. Similarly, high uranium concentrations in the Rongyang coal (China) are associated with organic components as well as fine-grained inorganic minerals (e.g. clay minerals, pyrite) [74]. Here, uranium can partially be removed through gravity separation but not completely, due to its association with the organic components and fine-grained minerals [74].

Coal bottom and fly ash are coal combustion products that consist of particles of the burned fossil fuel. Depending on the composition of the coal, the ash varies considerably in its composition. The uranium in fly ash is primarily concentrated in the Al-Si surface of porous grains, which form part of the <0.05 mm sized fly ash [75]. Consequently, raw lignite and uranium-rich fly ash can be leached for uranium recovery [71, 76]. Added value and therefore

commercial viability may be derived from the extraction of not only uranium, but other metals of interest, e.g. germanium [77].

4.2.4. Phosphoric acid waste stream

At phosphate fertilizer plants, phosphoric acid production involves the reaction of mined and processed phosphate rock with sulfuric acid. The controlled reaction yields phosphoric acid (containing phosphate as well as trace elements including uranium) and phosphogypsum. Phosphogypsum is typically a non-uranium bearing hydrometallurgical waste product, which comprises solid gypsum particles, pore fluids as well as reaction products and residual phosphate ore minerals. Global production of phosphogypsum has been estimated as high as 1 239 Mt per year [78]. Recovery of uranium from phosphoric acid during phosphate fertilizer manufacturing has been pursued since the 1950s [19], and since then extensive research has been carried out not only on the recovery of uranium but also of other elements from phosphoric acid [70]. For example, the search for 'critical' raw materials in the EU has moved domestic phosphogypsum piles into the focus as potential sources of gypsum as well as critical elements such as REE. Any uranium, together with other radionuclides, are considered a nuisance and cost factor (for their disposal) [79]. Depending on the nuclear policies of the respective countries, such uranium could be fed into the fuel-cycle as by-product.

4.2.5. Process liquids of copper ores

Heap leaching of copper ore and waste with dilute sulfuric acid may generate not only copper-rich solutions, but also liquids with mg/L concentrations of uranium. Recovery of uranium from the leach solutions of copper ores has been pursued in the United States of America since the 1960s [20, 21]. Especially during the 1970s and 1980s, copper leach solutions were used as sources of uranium and two by-product uranium extraction installations were operative commercially in the United States of America (Kennecott Bingham Canyon, Utah; Anamax Twin Buttes, Arizona). For the Kupferschiefer (copper shale) deposits of Poland, uranium can be leached from copper ores or tailings and it can be recovered subsequently using ion exchange sol-gel techniques, as long as mining and leaching operations continue [80]. It was estimated in IAEA-TECDOC-1849 [81] that 1 000 to 2 000 t/a of uranium could be recovered from heap leaching solutions from the copper industry in Chile.

5. MINE WASTES OF MAJOR ORE DEPOSIT TYPES

Geological mineral deposit classifications use fundamental ore-forming processes and host rocks to categorise uranium ore deposits, resulting in diverse ore deposit types (Table 5). Uranium ore deposits of the same type have formed from the same genetic processes considering host rock, structure, metasomatic alteration and surficial processes [38]. Hence, uranium ores of the same type have common geological settings as well as lithological, tectonic, mineralogical and geochemical properties.

These common characteristics of uranium deposits (e.g. the type of uranium ore mineral, amount and type of trace metals and metalloids enriched in the ore, type of country rocks associated with the ore) also determine the mineralogical and geochemical characteristics of mine wastes. Consequently, uranium ores of the same ore deposit type have similar geochemical compositions, similar ore and gangue minerals and consequently similar kinds of wastes. Mining of the same ore deposit type generally produces comparable waste types. Hence, uranium mine waste sites are best presented using the IAEA classification system for uranium ore deposits (Table 5). The remainder of this chapter comprises selected examples of uranium deposit types and their wastes.

TABLE 5. URANIUM DEPOSIT TYPES ACCORDING TO IAEA [38]

Type	Deposit type
1	Intrusive
2	Granite-related
3	Polymetallic iron oxide breccia complex
4	Volcanic-related
5	Metasomatite
6	Metamorphite
7	Proterozoic unconformity
8	Collapse breccia pipe
9	Sandstone
10	Palaeo quartz-pebble conglomerate
11	Surficial
12	Lignite
13	Carbonate
14	Phosphate
15	Black shale

5.1. POLYMETALLIC IRON OXIDE BRECCIA COMPLEX DEPOSITS

Iron oxide copper-gold deposits (IOCG) or polymetallic iron oxide breccia complex deposits are accumulations of valuable copper-gold ores hosted within iron oxide dominant gangue minerals (e.g. Olympic Dam, Prominent Hill, Australia). Economic concentrations of other trace elements may also be present such as uranium, bismuth or rare earth elements. Host rocks of IOCG deposits have variable mineralogical compositions and include aluminosilicate, silicate, sulfide, oxide, sulfate, carbonate, phosphate, fluoride and uranium minerals [82, 83]. Uranium minerals (uraninite, coffinite, brannerite) are typically fine-grained and largely

responsible for elevated uranium concentrations. Minor to trace amounts of uranium are accounted for by hematite, thorite-uranothorite, thorianite, thorite, crandallite, xenotime, zircon, REE-group minerals (i.e., florencite, bastnaesite, monazite, synchysite) as well as sulfides (i.e., pyrite, chalcopyrite, bornite, chalcocite) [83]. Uranium grades of IOCG deposits vary considerably, depending on the abundance of uranium-bearing minerals. The Olympic Dam deposit contains on average 230 ppm uranium [38].

Mining of IOCG ores is commonly pursued by recognizing, differentiating and extracting distinct ore and waste units, with each lithology having different uranium concentrations. The uranium concentrations may or may not be high enough to be of economic interest. Regardless, mined lithological units (i.e. ores, waste rocks) and the processed mineral raw materials (i.e. mineral concentrate, tailings) of IOCG deposits commonly have anomalous uranium concentrations that exceed the crustal abundance of uranium (3 ppm) [84].

While the mineralogical abundance of uranium minerals primarily controls the uranium concentration in IOCG mine wastes, there are other parameters that influence uranium contents of IOCG mine wastes. For example, variable uranium concentrations have been reported for the mineral concentrate of IOCG deposits depending on flotation parameters of polymineralic grains [85]. Here, the uraninite particles that reported to the final concentrate were either locked up with chalcopyrite or entrained as liberated particles of less than 5 μm in size.

Recovery of uranium from IOCG ores is also influenced by the uranium mineralogy. Brannerite is a refractory uranium mineral from which it is very challenging to extract uranium using conventional leaching techniques [86]. Consequently, in mineral processing operations, brannerite is frequently directed to the waste stream. Several large uranium ore deposits have major proportions of brannerite and hence, processing and extraction for uranium using conventional hydrometallurgical extraction methods lead to poor recoveries. For example, the ore of Olympic Dam contains on average 54% coffinite, 34% brannerite and 12% uraninite [87], leading to a historical recovery rate of only 67.1% [88]. Here, most of the uranium is dissolved from the coffinite and uraninite, while only a very small fraction of the uranium produced is generated from the leaching of brannerite. Consequently, uranium contents of ore and waste rock units, mineral concentrates as well as tailings of IOCG deposits are site-specific and influenced by the abundance, grain size and textural arrangement of uranium ore minerals in the primary ore and the behavior of different uranium ore minerals during mineral processing and hydrometallurgical extraction.

5.2. PHOSPHATE ROCK DEPOSITS

Phosphate rock is obtained from the mining of sedimentary phosphorite and igneous phosphate deposits. Phosphate ore deposits commonly have elevated concentrations of valuable elements other than phosphate. In particular, uranium contents of phosphate rock vary from mineral deposit to mineral deposit, with sedimentary phosphorites having higher uranium concentrations than igneous phosphate ores (Table 6). Organic matter is the likely mineralogical host in most phosphorites, but the uranium may also be hosted by an oxide or another stable mineral phase [89]. Uranium may also be incorporated in sedimentary phosphorite ores through ionic substitution into the carbonate–fluorapatite crystals.

TABLE 6. CONCENTRATIONS OF MAJOR RADIONUCLIDES FOR DIFFERENT PHOSPHATE ROCKS [90]

Country	Uranium		Thorium		^{226}Ra	^{228}Ra
	(Bq/kg)	(ppm)	(Bq/kg)	(ppm)	(Bq/kg)	(Bq/kg)
Former Soviet Union	44–90	—	78–92	—	30–70	—
United States of America	259–3 700	62–156	3.7–22.2	—	1 540	—
Florida	1 500–1 900	—	16–59	—	1 800	—
South and Central Florida	847–1 980	—	—	—	881–1 980	—
North Florida	241–981	—	—	—	229–884	—
Idaho	1 850	—	30	—	300	—
Wyoming	2 300	—	10	—	1 200	—
Brazil	114–880	27–71	204–753	55–185	330–700	350–1550
Chile	40	—	30	—	40	—
Algeria	1 295	—	56	—	1 150	—
Morocco	1 500–1 700	—	10–200	—	1 500–1 700	—
Senegal	1 332	—	67	—	1 370	—
South Africa	163–180	—	483–564	—	—	—
South Africa	100–200	—	—	—	—	300–500
United Republic of Tanzania	5 000	—	—	—	5 000	—
Togo	1 360	—	110	—	1 200	—
Tunisia	590	—	92	—	520	—
Egypt	1 520	—	26	—	1 370	—
Israel	1 500–1 700	—	—	—	—	—
Jordan	1 300–1 850	—	—	—	—	—
Australia	15–900	—	5–47	—	28–900	—

Mineral processing and metallurgical extraction of phosphate rock is used in the production of phosphoric acid, which is then converted to produce phosphate fertilizer. The processing of phosphate rock also causes the liberation of trace elements into the processing circuit and associated product and waste streams.

There are several processes to convert phosphate rock into phosphoric acid which is the intermediate step for fertilizer production. In each of these processes the uranium partitions differently into products and residues, compared with [90]. The processing is preceded by a beneficiation that consists of washing and screening the raw rock to remove accessory clay and

sand fractions, particularly from sedimentary phosphates. The beneficiation removes a large amount of the radium that may also be associated with the raw phosphate [91].

The two main processes for processing phosphate rock are: (i) acid leaching (using either strong H_2SO_4, HCl or HNO_3) to produce phosphoric acid (H_3PO_4); or (ii) the reducing thermal process resulting in elemental phosphorous. Leaching of e.g. apatite with acid sulfuric acid proceeds according to the following generic reaction:

$$3\ Ca_3(PO_4)_2 \cdot CaF_2 + 10\ H_2SO_4 <=> 10\ CaSO_4 + 6\ H_3PO_4 + 2HF$$

The partitioning of the radionuclides, including uranium, during the sulfuric acid leaching process is illustrated in Fig. 8. During the wet sulfuric acid process, much of the uranium present in the phosphate rock is dissolved into the phosphoric acid, whereas other elements like radium become concentrated in phosphogypsum [92] (Fig. 8). An extraction of uranium and other elements is possible from the phosphoric acid and phosphogypsum, using acid leaching or the application of organic solvents. Liberated elements may then be recovered using ion exchange technologies. Thus, mineral processing and hydrometallurgical extraction of phosphate rock produce phosphoric acid with variable uranium concentrations, largely dependent on the uranium content of the primary phosphate ore and fertilizer production process [90].

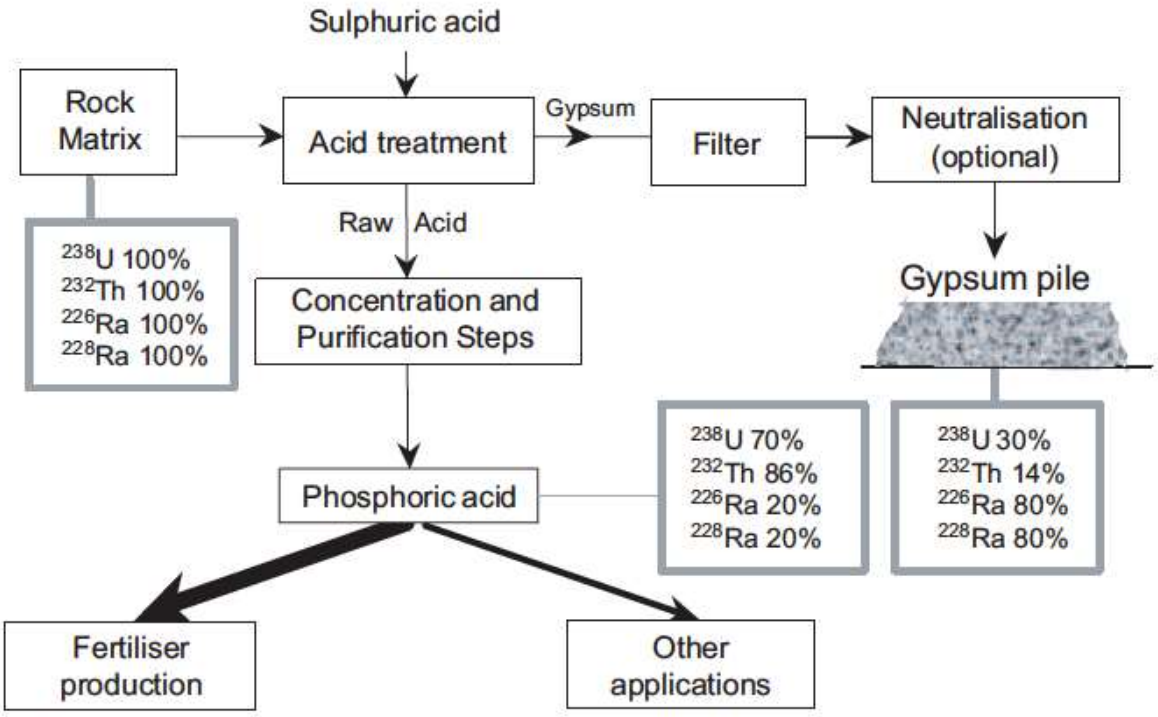

FIG. 8. The partitioning of uranium into products and residues during the sulfuric acid leaching process of phosphate rock [90].

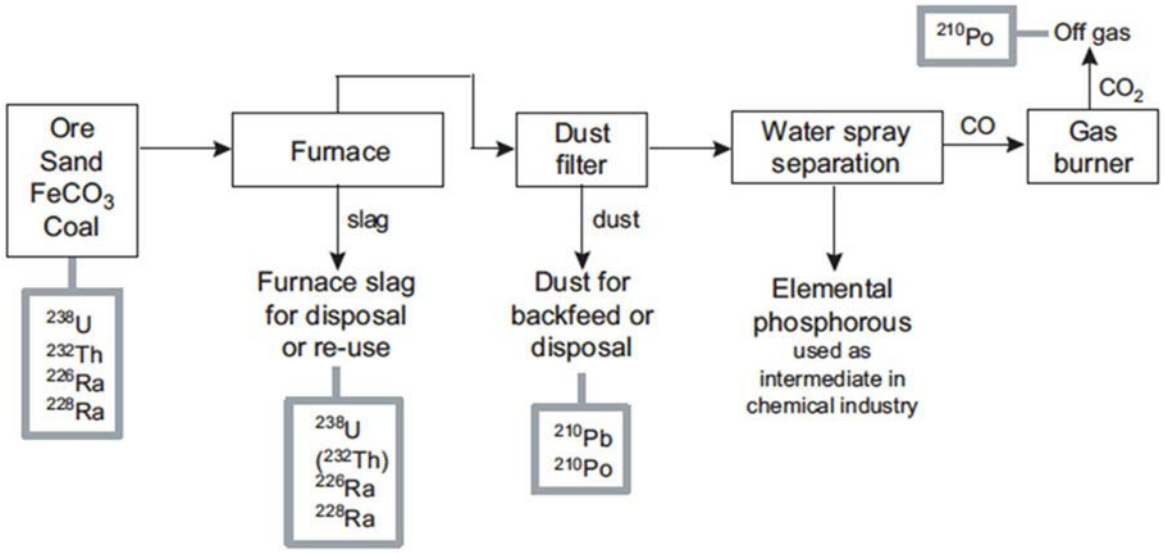

FIG. 9. Thermal production of elemental phosphorous and radionuclide partitioning [90].

Unlike in the acid leaching processes, the reduction of phosphate ore in a furnace at 1400°C with coal, silica sand and siderite ($FeCO_3$) as additives, results in virtually all of the uranium (together with most of the other radionuclides) ending up in the silicate slag and not in the phosphoric acid (Fig. 9). However, the sulfuric acid leaching process is the most commonly used around the world.

At some phosphate fertilizer plants, the uranium content is perceived to be high enough to be of economic interest. Therefore, uranium has been recovered during fertilizer manufacturing at several locations [93]. Also, several authors have proposed that a significant proportion of the uranium required for peaceful purposes worldwide could be obtained during phosphate fertilizer production [62, 92, 94]. In fact [94] propose that >15% of the globally required uranium could be derived from phosphate fertilizer manufacturing. Presently, the European Union imports large quantities of phosphate rock and phosphate fertilizer, and the uranium contained in these materials (334 t uranium) could have supplied 2.1% of the EU's uranium demand for nuclear power plants in 2017 [95].

For more details on the fate of uranium during phosphate fertilizer production see [90] and [96]. From these processing schemes, one can conclude that the most promising secondary sources would be the extraction of uranium during phosphoric acid production. In addition, the vast phosphogypsum piles, which have accumulated at various places around the world, offer reprocessing opportunities for other resource ingredients (e.g. gypsum, radium).

Phosphate rock is also a potential source for REE elements. REE have been identified by the EU as 'critical' elements [97], owing to the dominance of a few producer countries (mainly China). Extracting REE from phosphate processing residues within the EU is currently being considered seriously to augment supply security. While many consider uranium a 'nuisance', its utilization in this way could avoid some waste management issues.

Regardless of the significant research efforts since the 1950s [19] and the various commercial ventures to extract uranium from phosphate rock and during fertilizer manufacturing, many of the suggested reprocessing, reuse and recycling routes of phosphate mine wastes and the possible extraction of uranium during fertilizer manufacturing have not been taken up by

industry. Therefore, the uranium still migrates in the mining value chain from 'pit-to-product' and gets concentrated in the waste stream or the fertilizer product. Upon application of phosphate fertilizer to agricultural soils, topsoils and underlying aquifers may become contaminated by fertilizer-derived uranium and other elements [92, 98].

5.3. BLACK SHALE DEPOSITS

Black shale is a fine-grained, clastic sedimentary rock, which is comprised of clay minerals as well other rock-forming phases (quartz, carbonates, sulfates) plus in some shales there are appreciable amounts of organic matter and pyrite. At some locations, these shales are hosts to economic concentrations of base and precious metals. In addition, the shales may have elevated values of valuable trace elements including uranium (e.g. Kupferschiefer deposits of central Europe; Alum shales, Sweden; Okchon deposit, South Korea) [30, 38]. In these black shales, host minerals of uranium include uraninite, brannerite, thucholite, uranothorite, francevillite and torbernite [99, 100]. For example, the Polish Kupferschiefer copper deposits in the Lubin-Sieroszowice region contain approximately 60 ppm uranium [80]. Black shales are renowned for their uranium enrichment, and it is estimated that the black shales around the Baltic Sea collectively contain the largest known uranium resource in the European Union [101].

Considering that the uranium content of black shale deposits is highly variable, the unwanted mine wastes of black shale deposits have also variable, yet enriched uranium concentrations (Table 7). At Kvarntorp (Sweden), pyrolysis of the alum shale for oil production during WW2 to the 1960s led to the production of leached and burnt shale residues (in total 40 M m^3) that today still have significant temperatures and uranium contents [102, 103]. The inventory of valuable metals in the Kvarntorp waste pile has been estimated at 4 000 t uranium as well as 10 000 t vanadium and 4000 t molybdenum [103]. In future, waste pile cooling will lead to a greater release of uranium to drainage waters. Recovery of uranium from the leachate is achievable through ammonia addition [103].

Similarly, the recovery of uranium is possible from leach solutions at operating mines using solvent extraction technologies (e.g. Talivaara, Finland) [104]. Alternatively, black shale ores or their flotation tailings may be leached for uranium recovery [80].

TABLE 7. URANIUM CONCENTRATIONS OF MINE WASTES FROM BLACK SHALE DEPOSITS [80, 100, 102]

Location	Waste type	Mean uranium concentrations in mine waste or byproducts (ppm)
Talvivaara, Finland	Production waste	58
Kupferschiefer, Germany	Theisen sludge (i.e. flue sludge)	22
Kupferschiefer, Poland	Tailings	4.5
Ranstad, Sweden	Leached, burnt shales	64
Kavarntorp, Sweden	Leached, burnt shales	235

6. CASE STUDY: WITWATERSRAND TAILINGS, SOUTH AFRICA

The Witwatersrand deposits of South Africa are famous for their gold content and gold production. These ore deposits have been assigned using the IAEA classification scheme [38] as Precambrian quartz-pebble conglomerate deposits, which in turn can be subcategorized as: (i) uranium-dominant associated with gold and rare earth elements (e.g. Elliot Lake, Canada); and (ii) gold-dominant associated with uranium (e.g. Witwatersrand, South Africa). The deposits typically have an ore mineralogy dominated by native gold, uraninite, brannerite, Fe-Ti oxides, organic matter, pyrite and other sulfides within an aluminosilicate matrix.

Mining of quartz-pebble conglomerate ores have not only yielded significant gold production, but the extraction of uranium and reprocessing of tailings for the recovery of both, gold and uranium, have been pursued since the 1950s. In fact, mine wastes of quartz-pebble conglomerate deposits offer other commercial opportunities. For example, at the former mine Quirke Lake mine in Canada, mine waters from the quartz-pebble conglomerate deposit have elevated uranium concentrations, suitable for passive underground stope leaching [105]. Also at the Driefontein mine (South Africa), the recovery of uranium from mine water circuits is possible using ion exchange technologies [106]. Most of all, large quantities of uraniferous tailings remain in the Witwatersrand region.

6.1. THE WITWATERSRAND GOLD AND URANIUM DEPOSITS

The Witwatersrand basin is host to the world's largest gold deposit and has contributed about to 40% of the total historic gold production [107]. Since 1886, six major and several smaller gold fields were developed in the Witwatersrand region [108]. The most important gold fields are associated with the northern and the western margins of the basin, where mining of gold-bearing horizons occurs within 4 000 m from the surface [109].

The gold deposits occur in the form of an auriferous quartz-pebble conglomerate that was formed about 2.8 to 3 billion years ago. Most gold is concentrated in so-called reefs of 20–50 cm thickness in layered, siliciclastic sedimentary rock, termed the Witwatersrand Sequence, part of the up to 7.5 km thick Witwatersrand Supergroup. Two main theories explain the ore formation in the sedimentary strata. The first theory is based on sedimentary processes that include: (i) the erosion of granite-greenstone basement; (ii) sediment transport by large river systems; and (iii) final deposition with minor remobilization in a delta or an inland sea in the Witwatersrand basin [110, 111]. By contrast, the second theory suggests a hydrothermal origin of the ore with precipitation occurring as a result of hydrothermal fluids migrating into the basin and interacting with shale-derived hydrocarbons [109].

The conglomerate presents not only the most important horizon in terms of gold, but also in terms of uranium resources. The rock composition consists of a variety of quartz pebbles and vein quartz imbedded in a matrix of sand grain-sized quartz and sulfides, in particular pyrite (Fig. 10). Other sulfide minerals are present in minor amounts. Further minor mineral phases include muscovite, pyrophyllite, chlorite and carbon granules. Heavy oxide minerals occur in the form of chromite, zircon and leucoxene and trace constituents comprise not only economic concentrations of native gold, but also notable amounts of uranium minerals [81].

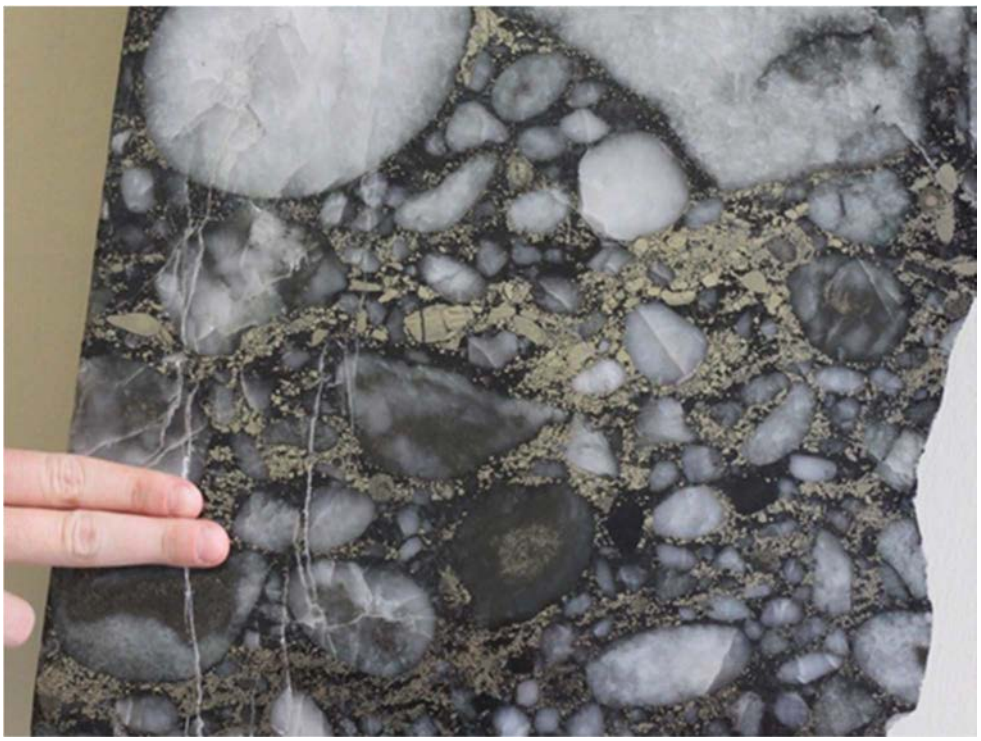

FIG. 10. Precambrian quartz pebble conglomerate from the Witwatersrand basin (displayed at Stellenbosch University, South Africa, 2015).

The close association of gold with uranium in the quartz-pebble conglomerate makes the rock type a significant uranium resource [107, 112]. Uranium occurs mainly in primary and secondary forms of uraninite. Primary uraninite crystals are typically rounded and occur in a range of sizes from 77–100 μm and the secondary uraninite minerals occur often as enclosures or replacements of the initial crystal [112, 113].

Besides uraninite, different proportions of other secondary uranium-bearing minerals are present in the Witwatersrand reefs [113]. These mineral phases include pitchblende, brannerite, coffinite, U-Ti phases (i.e. uraniferous leucoxene, euxenite), U-Th phases (uranothorite) and thucholite [114–116]. The chemical composition of these minerals and exemplary occurrences are listed in Table 8.

TABLE 8. URANIUM MINERALOGY OF THE WITWATERSRAND BASIN. DATA AND CHEMICAL INFORMATION BASED ON [113, 116, 117] IF NOT INDICATED OTHERWISE (NS = NOT SPECIFIED)

Mineral phase	Chemical formula or composition	Exemplary geographic occurrence
Brannerite	UTi_2O_6	e.g. Gauteng province: Far West Rand, West Wits Line; North West: Klerksdorp; Areas with relatively high metamorphic grades (West Wits)
Coffinite	$U(SiO_4)\ nH_2O$	e.g. Western areas and Randfontein
Euxenite [114]	$(Y,Ca,Ce,U,Th)(Nb,Ta,Ti)_2O_6$	e.g. Western deep levels
Pitchblende	Amorphous mixture of UO_2 and U_3O_6	NS
Thucholite	Uraninite-bearing kerogen	e.g. Free State (Carbon Leader Reef) [118]; Gauteng: Johannesburg District (Central Rand) [119]
Uraniferous leucoxene	Uranous titanates: UTi_zO_2	e.g. Free State [120]
Uraninite	UO_2	Most abundant
Uranothorite	$(Th,U)SiO_4$	NS

The relative abundance of the uranium-bearing minerals varies across the Witwatersrand strata. Consequently, also the uranium mineralogy of the material recovered at a specific mine site — and the wastes and residues produced — varies geographically across the Witwatersrand Basin. The highest uranium concentrations, for example, are associated with kerogen. For the Carbon Leader Reef, for example, maximum uranium concentrations of 58 000 ppm (5.8%) were reported [113].

Average uranium concentrations in the conglomerates are several magnitudes higher than those of gold. The relatively low gold-to-uranium ratios of 1:10 to 1:100 in the rock exemplify the ample amounts of uranium that gold mining transports to the surface in the Witwatersrand Basin [121]. When perceived as invaluable residue or waste, the uranium is subsequently directed to tailings storage facilities.

6.2. WITWATERSRAND GOLD TAILINGS

The term ‘tailings’ usually describes the fine-grained waste materials resulting from ore processing activities. While economic commodities (e.g. gold) are removed from milled ore through physical and/or chemical processes, the residual constituents are discharged as slurry to tailings dumps or dams, where waters evaporate. The resulting material consists of solid mineral particles and pore water [122, 123]. These residues can be furthermore categorized

according to their size fractions as follows: (1) slimes with particle sizes of 75% passing 74 µm; and (2) sands with particle sizes of 10–20% passing 74 µm [124].

The disposal of uranium in the form of tailings from gold mining and processing leads to significant uranium stocks in the Witwatersrand gold tailings. The tailings locations are indicated in Fig. 11. While reprocessing the tailings for both gold and uranium has become an established practice in the Witwatersrand Basin, tailings are still not an official source of uranium as a by-product, as defined by the IAEA [81]. A by-product is a commodity sourced from either 'unconventional deposits' or 'other metal production' including gold, copper or nickel. Although the gold-dominant Witwatersrand deposit is categorized in the UDEPO database as an 'unconventional uranium resource', the Witwatersrand tailings remain undefined. This missing recognition of the gold tailings as a source of uranium as a by-product highlights the necessity for detailed descriptions and evaluation of the tailings in the Witwatersrand area with regards to the occurrence of uranium in ores and wastes.

FIG. 11. Overview map showing the locations of the Witwatersrand gold tailings.

6.2.1. Tailings volumes and masses

In the Witwatersrand Basin, tailings cover an area of approximately 400 km^2 [125]. The total number of tailings sites in the Witwatersrand area is difficult to quantify due to ongoing mining and reclamation activities. However, there are likely several hundred sites, since the number of tailings heaps around Johannesburg alone amounted to 270 in 2015, according to media reports [126]. More reliable estimates are possible for tailings volumes and masses. Given ongoing

production of gold tailings over 20 years, data extrapolations based on the South African Chamber of Mines (1999), indicate roughly 7 000 Mt of tailings material in 2020. Figure 12 shows the distribution of tailings volumes across different parts of the Witwatersrand Basin, based on the same data. The dimensions of single tailings heaps can reach extraordinary dimensions. For example, the Driefontein mine no. 3 tailings dam has been described as being 2 km long, 500 wide and 73 m tall [127].

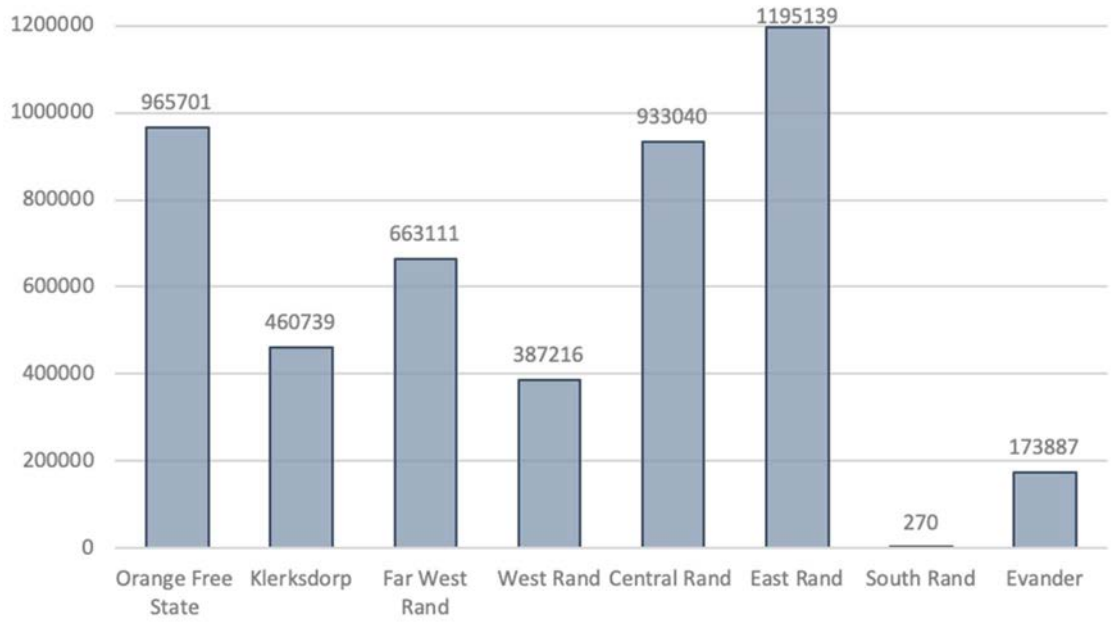

FIG. 12. Tailings masses (t) associated with the Witwatersrand gold fields [128].

The Witwatersrand gold tailings have an average uranium concentration of 100 ppm. Besides potential resource recovery, these concentrations are also significant in terms of environmental risks [113, 121, 129, 130].

6.2.2. Environmental impacts

The environmental impacts of the Witwatersrand tailings are intrinsically linked to their mineralogical and geochemical composition and may present long-term environmental and radiological hazards. The perceived risks associated with the tailings include potential processes such as: (1) Acid Mine Drainage (AMD) development; (2) erosion or failure of waste material; and (3) water and soil pollution due to heavy metal and radionuclide release [113, 121, 129, 130]. Contaminant release to the environment may occur either through mobilization processes acting on the tailings dumps themselves (e.g. wind and water erosion, leaching, slump failure), or through their geochemical footprint left behind after waste removal.

As modeled by Netshiongolwe [130], the following elements: arsenic, lead, copper and zinc (besides uranium) may be released from Witwatersrand tailings at elevated concentrations to ground and surface waters. The author [128] also emphasizes the AMD potential of tailings due to sulfide oxidation and the resulting acidic pH conditions that enhance metal and metalloid mobility. Topsoil samples from tailings footprints typically reflect these acidic conditions with pH values ranging from 3.5 to 3.9 and elevated arsenic, lead, copper and zinc levels. The pH values were determined on surface waters of tailings ponds and solid tailings sampled along depth profiles. The pH value of oxidized tailings near or at the surface (0–5 cm) is 3.5 and increases to 7.3 in reduced zones below surface (25–30 cm), reflecting sulfide oxidation at

surface and acid generation potential in the tailings below [130]. Thus, Witwatersrand tailings dumps and their legacy footprints may pose significant environmental risks.

6.2.3. Tailings mineralogy and geochemistry

The composition of the mining and mineral processing residues determines both environmental impact and potential resource recovery and generally reflects the composition of the mined rock material. Typical Witwatersrand tailings comprise quartz (70–80%), mica (10%), chlorite and chloritoid (9–18%) as well as sulfide minerals, in particular pyrite, and their oxidation product jarosite (0.5–2%) (Fig. 13). Furthermore, individual tailings dams may also contain pyrophyllite and K-feldspar in trace amounts [131–133]. Gold, uranium, zirconium and chromium are trace constituents which are generally reported at minor concentrations. Uranium levels average 100 ppm but can occasionally surpass 200 ppm; thereby notably exceeding average natural background levels of <2 to 4 ppm [121, 134]. Gold generally occurs well below 1 ppm [135].

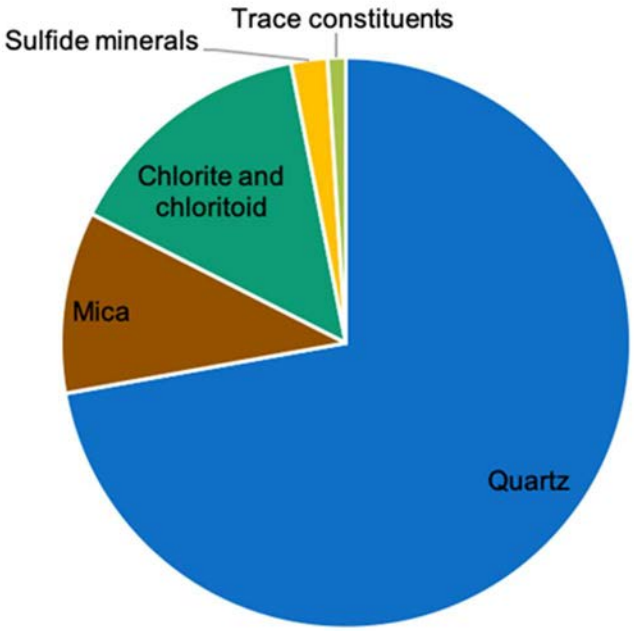

FIG. 13. Typical mineralogy of Witwatersrand tailings. Major components are quartz (70–80%), mica (10%), chlorite and chloritoid (9–18%), sulfide minerals (i.e. pyrite) (0.5–2%). Ore elements are present in traces and include uranium (occasionally exceeding 200 ppm) and gold (generally below 1 ppm) [133].

Subtle differences in the mineralogical and geochemical composition of tailings from the East Rand, West Rand, Central Gauteng and Free State regions are documented by the Institute of Waste Management of Southern Africa [135]. Results from X-ray diffraction based investigations (Table 9) show similar bulk mineralogical compositions for samples from all regions, with quartz ranging from 70.7 to 81%, pyrophyllite from 12.2 to 16.2%, serpentine from 0.6 to 2.2%, mica from 3.8 to 9.9%, gypsum from 0.9 to 2.6 % and aluminite from 4.1 to 5.3% [135].

TABLE 9. TYPICAL MINERALOGICAL COMPOSITION OF WITWATERSRAND TAILINGS [133] VALUES FOR JAROSITE AND CHLORITE ARE BASED ON [133] (NS = NOT SPECIFIED)

Mineral / Sulfide species	Chemical formula	East Rand	Central Gauteng	West Rand	Free State	Unspecified
		%	%	%	%	%
Quartz	SiO_2	81	76.4	71.8	70.7	—
Pyrophyllite	$Al_2Si_4O_{10}(OH)_2$	12.2	13.7	14.7	16.2	—
Mica	$KAl_2(Si_3Al)O_{10}(OH,F)_2$	4.6	3.8	5.9	9.9	—
Gypsum	$CaSO_4•2H_2O$	1.26	0.9	1.4	2.6	—
Aluminite	$Al_2(SO_4)(OH)_4•7(H_2O)$		5.3	4.1		—
Serpentine	$Fe_{2-3}Si_2O_5(OH)_4$	0.9	NS	2.2	0.6	—
Sulfide	NS	0.41	0.25	0.5	0.9	—
Sulfate	NS	0.5	0.01	0.3	0.4	—
Jarosite	$KFe^{3+}{}_3(SO_4)_2(OH)_6$	—	—	—	—	0.2–5.0
Chlorite	NS	—	—	—	—	3.0–10.0

Similar mineralogical compositions were reported for both samples taken from tailings piles and samples from the remaining base layer of relocated tailings dumps (i.e. footprints) in the Central Rand Basin. Quartz is the major mineral, while primary pyrite, chalcopyrite, pyrophyllite, chlorite and mica and secondary weathering related minerals such as goethite, melanterite and gypsum are minor phases [130].

Trace element composition is crucial for both potential resource recovery and contaminant release. Geographically discerned values are given for the East Rand, West Rand, Central Gauteng and Free State provinces in Table 10 [135]. Average gold contents range from 0.27 to 0.41 ppm, and uranium from 19 to 65 ppm. A regional distribution of uranium concentrations in tailings could be observed. Tailings from the West Rand and Free State regions possess the highest average uranium contents, whereas tailings from the East Rand and Central Gauteng provinces have the lowest average uranium values. Sulfide values range from 0.41 to 0.9% [135]. The levels of cobalt, nickel, zinc and chromium presented in Table 10 are comparably low compared to other studies, e.g. [133].

TABLE 10. TRACE ELEMENT COMPOSITION OF TYPICAL TAILINGS SAMPLES FROM SEVERAL PROVINCES OF THE WITWATERSRAND REGION [135] (ND = NOT DETERMINED)

Element	Unit	East Rand	Central Rand	West Rand	Free State
Ti	%	0.25	0.18	0.17	0.13
V	%	<0.05	<0.05	<0.05	<0.05
Cr	%	<0.05	<0.05	<0.05	<0.05
Mn	%	<0.05	<0.05	<0.05	<0.05
Fe	%	3	2.32	3.1	1.86
Co	%	<0.05	<0.05	<0.05	<0.05
Ni	%	<0.05	<0.05	<0.05	<0.05
Cu	%	<0.05	<0.05	<0.05	<0.05
Zn	%	<0.05	<0.05	<0.05	<0.05
Pb	%	<0.05	<0.05	<0.05	<0.05
As	%	<0.05	<0.05	<0.05	<0.05
Au	ppm	0.27	0.32	0.35	0.41
U	ppm	19	ND	59	65

6.2.3.1. Environmental implications.

The active oxidation zones that develop on the surface of individual tailings dams through the exposure of suflide minerals to atmospheric conditions represent the main sources of contaminant release. These zones can potentially reach several meters of depth from the surface and are characterized by the formation of acidic pore water from sulfide oxidiation. Sampling campaigns along depth profiles revealed trace metal concentrations along oxidized, transition and unoxidized tailings zones of individual tailings dams. Oxidized zones and transition zones were defined between 1.5 and 3.5 m and 4.5. to 9 m from the surface, respectively. Values for chromium, copper and zinc are shown in Table 11, while uranium levels were not published in the study [133].

TABLE 11. TRACE METAL CONCENTRATION RANGES ALONG DEPTH PROFILES FROM OXIDIZED ZONES NEAR THE TAILINGS SURFACE TO UNOXIDIZED ZONES AT GREATER DEPTHS (ND = NOT DETERMINED) [133]

Element	Unit	Oxidized zone	Transition zone	Unoxidized zone
Cr	ppm	202−579	201−452	185−456
Cu	ppm	24−68	22−65	25−58
Zn	ppm	70−127	55−123	89−217
U	ppm	ND	ND	ND

6.2.3.2. Resource implications

According to estimates, about 600 000 t of U_3O_8 have been excavated through gold mining activities and deposited in form of tailings in the Witwatersrand area [136]. Many slime dams of the area show average uranium concentrations of 100 ppm and higher, representing concentrations that are distinctly elevated compared to some conventional uranium mines [136]. In addition, current gold mining adds anually about 6 000 t of uranium to tailings facilities [121]. The significant uranium concentrations have enabled uranium recovery projects to operate during periodically high uranium world market prices.

The extraction of uranium during tailings reclamation projects may provide two main benefits. Firstly, it may eradicate long-term environmental impacts of the tailings and secondly, potential revenues from the production of a valuable commodity may be generated. The following section discusses the uranium occurrences in the tailings of the Witwatersrand region, future opportunities, and constraints faced in tailings reprocessing and recovery of uranium and other commodities.

6.3. COMMODITY EXTRACTION FROM TAILINGS

While mining non-renewable minerals and metals is not a sustainable practice in the strict understanding of the term "sustainability" (i.e. it takes options from future generations), it is recognized that mining can contribute to sustainability in the broad sense of the term, namely by creating long-term benefits. This may be achieved when an industrial activity is embedded in an environment of poverty alleviation, good governance, transparency and stakeholder engagement and also when the 'polluter-pays' principle applies and wealth created from the mining operations contributes to its remediation [137].

Mine wastes like the Witwatersrand tailings offer significant opportunities for remining and reprocessing and, these waste types can contain one single or several mineral commodities. While the reprocessing for a single commodity may be practiced due to targeted market realities, reprocessing tailings for more than one commodity entails several other opportunities: (i) cross-financing of remediation activities; (ii) solving long-term environmental problems; (iii) acting in the interest of the local community and stakeholders; and (iv) using resources necessary for remining, such as water and energy, only once and hence, more efficiently. Thus, the common paradigm in economic geology to 'disturb the ground only once' [81], can also be applied to tailings recovery projects. The following section will thus explore potential opportunities and constraints in the recovery of uranium from the Witwatersrand gold tailings.

6.3.1. Opportunities for uranium recovery from tailings

South Africa has a relatively long history in the reprocessing of Witwatersrand tailings for both gold and uranium. Full-scale operations of uranium production as a by-product started in the 1950s [81, 138]. However, due to a fall and the subsequent rise of the uranium price, reprocessing of tailings began to decline in the 1980s and; remining activities focused primarily on gold extraction [135, 139]. In the Far West Rand and the Witwatersrand regions, for example, uranium was not extracted, with the tailings estimated to contain 600 000 t of U_3O_8 [136]. Uranium recovery re-gained momentum in 2003 and continued to increase until 2007; when ten new tailings reclamation projects were in the planning phase [136]. In 2017, South Africa counted 29 ongoing uranium projects, predominantly in the Witwatersrand region [136, 138].

In South Africa, two types of commodity recoveries, as illustrated in Fig. 14, have been pursued. Some historic and present projects in the Witwatersrand aim at poly-commodity recovery, i.e. the simultaneous extraction of gold, uranium and/or pyrite utilising sulfuric acid production [124, 127, 140, 141]. Other projects focus on mono-commodity recovery, i.e. the sole recovery of gold, leaving uranium, other heavy metals and sulfides in the reworked tailings behind [141–143].

Operations pursuing mono-commodity or poly-commodity recoveries may also generate 'cleaned' tailings for alternative purposes. For example, clay and sand-rich tailings may be recycled for brick manufacturing [144]. In this case, however, it is critical to note that remnants from the uranium decay series can release radon gas, if not removed from the tailings material beforehand. The noble gas is a well-known pollutant in indoor settings with important implications for human health, i.e. lung cancer [145]. Other suggestions for alternative usage of the tailings material include as aggregate for road base, as cement additive or for backfilling mine shafts. Informal activities include agriculture or activities for recreational or educational purposes [146]. Finally, indirect usages of the tailings for green electricity production have also been proposed. This includes the erection of solar power plants on the tailings and the cultivation of crops for biofuel production [147].

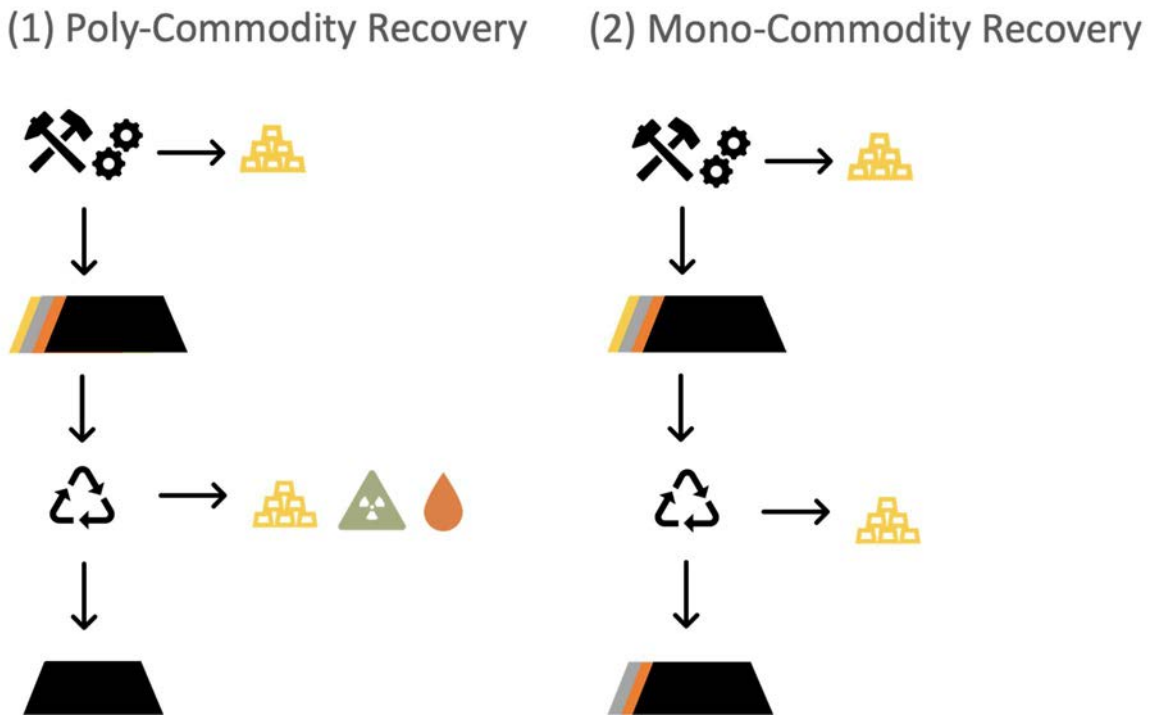

FIG. 14. Two types of commodity recovery. Poly-commodity recovery extracts more than one commodity during tailings reclamation projects. Mono-commodity recovery extracts one commodity leaving the others in the tailings. For this example, yellow is gold, green is uranium and orange is solvent or acid.

6.3.2. Regional uranium occurrences and recovery examples

Data records from the South African Chamber of Mines (1999) state were used to obtain estimates on the approximate uranium masses in the gold tailings of individual regions in the Witwatersrand basin [128]. A depiction of the uranium ranges possibly contained in the tailings

of selected regions (n = 82) is presented in Fig. 15. The illustration is based on the statistics shown in Table 12.

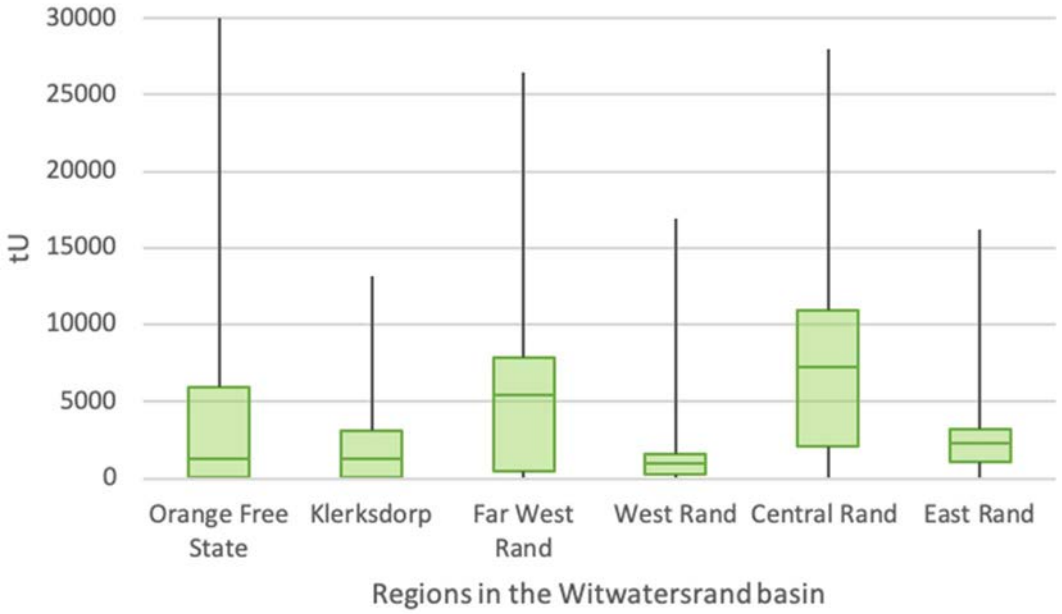

FIG. 15. Estimated uranium (y axis=tU) contained in gold tailings of the Witwatersrand Basin according to the South Africa Chamber of Mines state records from 1999. Note that the maximum value for the Orange Free State (tU = 73 914) lies outside the y-axis range. Data source: [128].

Total uranium masses contained in the tailings range from insignificant amounts to a maximum of approximately 74 000 t uranium (Orange Free State) (Fig. 15). On average, tailings dumps of the Witwatersrand Basin contain several thousand tonnes of uranium, as indicated by a median value of 1 675 t uranium for the entire data set (n = 91).

TABLE 12. STATISTICS (tU) ON POSSIBLE URANIUM IN TAILINGS OF INDIVIDUAL MINES IN SELECTED REGIONS OF THE WITWATERSRAND BASIN. DATA SOURCE: [128]

	Orange Free State	Klerks-dorp	Far West Rand	West Rand	Central Rand	East Rand
Number of mines	*12*	*8*	*12*	*9*	*12*	*29*
Minimum	0	78	377	111	183	60
Q1	66	145	836	358	2 270	1 117
Median	1 260	1 389	5 836	1 068	7 452	2 393
Q3	5 980	3 132	8 232	1 725	11 064	3 221
Maximum	73 914	13 279	26 813	17 066	28 224	16 216

Approximating uranium masses is also possible for individual mine sites. Estimates by the South African Chamber of Mines (1999) indicate approximately 5 000 t uranium in the tailings at Stilfontein in the Klerksdorp goldfied, for example [128]. At Stilfontein, a tailings dam with 24 million m^3 contained, on average, about 0.45 ppm gold. Upon closure, the tailings dam was purchased along with existing surface infrastructure. During reprocessing, primarily gold was extracted and an annual gold production of about 1 738 kg was achieved, leaving uranium in the reworked tailings.The remining project managed to guarantee small net returns for the shareholders despite short-term setbacks and also addressed environmental concerns of the local community. Finally, the abandoned mine waste material was moved into engineered waste repositories [143].

The Randfontein and Driefontein regions host some of the largest tailings remining activities in South Africa. Uranium masses of about 17 000 t were reported to be contained in the Randfontein tailings [128]. Remining activities are mostly performed by DRD Gold and Sibanye Stillwater Company. Tailings volumes are considerable. For example, the Driefontein no. 3 tailings dump occupies an area of 72.26 ha, holds a volume of 10 million m^3 and contains, on average, 0.8 ppm gold. The three commodities gold, uranium and sulfuric acid were extracted [127]. Similar to other projects [142], the recent emphasis remains on gold recovery, leaving currently uneconomic uranium within the tailings. Uranium contents of 6 000 to 12 000 t of uranium are possibly present in the tailings of the Driefontein mine sites. One important aspect with regards to this project is the West Rand Tailings Relocation Project (WRTRP) that aims to relocate reworked tailings from their initial location on dolomitic karst to a major new tailings storage facility [148]. The project's objective is to create benefits for all stakeholders, to explore potential uranium and sulfuric acid recovery, and to dispose process waste safely [149].

In the East Rand, the ERGO operations extracted gold, sulfuric acid and periodically uranium from tailings. Existing processing facilities with excess capacities allowed the retreatment of tailings at marginal costs [124]. According to the IAEA [81], cumulative uranium production amounted to 2 150 t U_3O_8 from 1978 to 1991. Benefits of tailings reprocessing were that tailings remining increased land availabilty for urban development and removed environmental legacies in the form of waste dumps [124]. Also, reprocessed tailings were secured in more modern waste storage facilities.

6.3.3. Constraints on uranium recovery

Various constraints may impede the recovery of uranium during the reprocessing of Witwatersrand gold tailings. However, the predominant factor that constraints the recovery of uranium as a by-product is economic feasibility. The uranium price fluctuates depending on resource demand and/or subsidies. As mentioned previously, South African uranium recovery from the tailings ceased, in particular during the 1980s, when uranium demand declined worldwide. Despite other potential benefits that may be associated with poly-commodity recovery (e.g. the mitigation of environmental impacts), incentives for uranium recovery are often limited due to the market reality. Only when the costs can be covered, may flowsheets for reclamation processes include processes for additional commodities (e.g. sulfide or uranium removal) [135]. Additional costs can also arise from infrastructure development (i.e. water and energy) [149] or from legal compliance with environmental law, monitoring obligations or financing rehabilitation [150, 151]. Although these additional costs are not covered in detail by this study, one can summarize that inadequate economic feasibility coupled with market realities have often limited the remining activities of tailings to only gold recovery, which leaves uranium and sulfide minerals with AMD potential in the tailings.

Economic uncertainties can be reduced when more precise estimations of uranium tonnages and distributions in the tailings exist. Because ore grades can vary throughout both the mined ore and the tailings dump, unbiased sampling and analysis are of prime importance for reliable resource estimation. A first impediment may be the so-called wall effect that arises from sulfide oxidation at the tailings surface. Samples from the oxidized zones at a tailings surface may not be representative of the materials at greater depths in the tailings. The wall effect is negligible, however, when sulfur contents are low [133]. Other factors relate to the initial construction of the dam, for example, the presence of reinforcement structures or foreign materials (e.g. larger rocks or municipal waste). The removal of these materials may entail higher costs in the reprocessing of tailings. Moreover, the physical and chemical state of uranium in the tailings is often uncertain. This complexity underlines the need for accurate and precise sampling and conservative extrapolations, if necessary, to achieve reliable resource estimation. Inaccuracies may arise furthermore from biased sampling grids and borehole increments, sample cross-contamination and insufficient analytical accuracy and precision [124]. The final reclamation method depends also on the particle size of the tailings material (sand, slime or mixture) and moreover, on the uranium mineralogy.

Restraints on uranium recovery can also be of a technical nature. The mineral form in which uranium occurs does not only determine environmental mobility, but also uranium extraction efficiency during leaching processes. While uraninite gives relatively high yields, other mineral phases such as brannerite, coffinite or euxenite (Table 8) are relatively refractory to leaching technologies. For example, leaching efficiency of uranium was reported to vary between 30 to 64% when relatively more refractory brannerite is present alongside uraninite [117]. Another mineral that is more refractory in leaching relative to uraninite is euxenite. Better leaching results were reported for coffinite, but recovery rates were still lower when compared to those of uraninite. Consequently, the potential presence of relatively refractory uranium-bearing minerals may require advanced processing techniques at some mine sites in the Witwatersrand basin. These techniques may include elevated temperature and pressure conditions in leach-based uranium recovery [114, 116].

Beyond uranium recovery, important other constraints occur with regards to the tailings' material and the legacy landforms. As previously mentioned, the tailings material can comprise radionuclides and elevated concentrations of trace metals that may adversely affect exposed biota. Consequently, alternative usages may ideally guarantee safe material properties (e.g. no radon emissions from bricks) within clear legal frameworks, in addition to markets for these products [146]. Moreover, tailings reprocessing, and relocations activities often leave so-called tailings 'footprints'. These residues can exhibit toxic levels of trace metals and radiological properties thereby impeding future land use (e.g. for urban development or agriculture) and require adequate management [139].

6.4. CONCLUSIONS

Mining for gold has led to the presence of numerous tailings dumps in large areas of the Witwatersrand Basin, with the waste residues still containing gold, uranium and sulfide minerals. Although many of these tailings dumps have been remined in the past, most extraction activities were limited to one commodity, in particular gold. Consequently, other mineral commodities still remain in the reworked tailings. In general, uranium has not been recovered due to poor economics relating to commodity price fluctuations or insufficient cost coverage. Moreover, many uncertainties prevail with regards to the physical and chemical occurence of uranium in the tailings. Mono-commodity recovery is thus often the case in the Witwatersrand

area, although poly-commodity recovery could entail important synergies for both efficient use of resources and environmental remediation. To date, such partial extraction of mineral commodities from Witwatersrand tailings provides opportunities for future uranium remining and reprocessing activities. This study highlights the importance to describe, quantify, evaluate and report on uranium occurrences in Witwatersrand tailings in detail. A better and more complete understading of the waste could provide many benefits to the minerals industry and society including: (i) the integration of tailings in uranium resource databases; (ii) informed management decisions on remining and reprocessing of Witwatersrand tailings for uranium; and (iii) enhanced environmental protection in the area.

7. CASE STUDY: URANIUM TAILINGS IN MAILUU SUU, KYRGYSTAN

The Central Asian states of Kazakhstan, Kyrgyzstan, Tajikistan and Uzbekistan hosted about 30% of the former Soviet Union's uranium production. Following their independence in 1991, awareness increased on the magnitude of uranium mining legacies from the Soviet era in these states. Numerous sites had been abandoned without sufficient environmental management which resulted in long-term environmental burdens in the form of mining and processing wastes containing significant amounts of uranium [152]. The total radioactive waste volume in the region associated with uranium mining is estimated to amount to more than 400 hundred million tonnes. Tailings from uranium ore processing are estimated to amount to approx. 370 million tonnes. The territory affected by both ore mining and processing comprises more than 60 km^2 in the region [153]. Fig. 16 presents a geographic overview of uranium-bearing tailings deposits in Central Asia.

FIG. 16. Uranium-bearing tailings deposits in Central Asia. The location of Mailuu Suu is indicated by red color.

Uranium ore was not only processed from local sources but also from international sources. Consequently, the volume of processing residues (i.e. tailings) is not comparable to the volume of waste rock excavated by local mining activities. The import of uranium ore occurred primarily from Eastern European countries. The largest extent (about 75%) can be attributed to Eastern Germany. Other countries of origin include Czech Republic, Bulgaria and Romania

[152]. Following independence in 1991, observations were made on the increasing deterioration of the mining and processing sites. This lead to concerns with regards to: (i) potential human exposure among the population living in the vicinity of the sites; and (ii) downstream transport of contaminants and broader dispersion into the environment. Another aspect is the proximity of the sites to neighbouring countries making the mining legacies a subject of international debate.

A comparison of the situation of Soviet era uranium mining legacies in eastern Europe with those in Central Asia reveals that the majority of legacies in Europe have been made safe and have been remediated to acceptable environmental standards. Unremediated legacies remain in Bulgaria and Romania as both countries became member states of the European Union (EU) in 2007 [154]. In Central Asia — despite sharing a similar mining history — the situation is quite different.

A number of international aid organizations started to address the issue in the 1990s by providing assistance to the Central Asian countries. In particular, international assessments (TACIS, INTAS, IAEA, NATO, ENVSEC, etc.) were conducted to identify the legacy sites that were least secure and posing the highest risks. Deficient waste containment structures, if even existent, and geotechnical risks (erosion, landslides, seismic activities) were generally identified as important factors during these assessments.

Enabled by its relative wealth, Kazakhstan launched a state-funded national programme for remediation of former uranium production in 2001. However, the program was discontinued in 2010 and missed longer-term outcomes. This might be explained by the fact that remediated sites or objects had not been placed under long-term institutional control [155]. In Kyrgyzstan, Tajikistan and Uzbekistan most legacies have remained unremediated since 1990. A few international initiatives have achieved limited progress in this regard. The most prominent example is a Disaster Hazard Mitigation Project funded by the World Bank that supported the remediation of high risk objects at Mailuu-Suu in the period 2004–2013 [156].

Attempts to recover uranium from reprocessing of uranium mill tailings have been made in Tajikistan and Kyrgyzstan. An example is an initiative in the north of Tajikistan, where in 2008 the state enterprise Vostokredmet began research on the possibility of uranium extraction from mill tailings [157]. In Tajikistan, attempts were also made to valorize another form of uranium waste, namely uranium concentrate produced during the treatment of mine waters, to international costumers. This option was restrained, however, by insufficiently defined regulations impeding transport and export licensing [158].

In 2008, the company Nimrodel Resources Ltd. announced that its Kyrgyz subsidiary had been issued a license to allow prospecting for the reprocessing and extraction of uranium and other metals from tailings deposits in Mailuu-Suu. In 2015, a discourse was held between the Chinese BSF Group Corporation and the Kyrgyz Minister of Emergency Situations on a cooperation in the processing of uranium tailings [159]. Until now, none of these initiatives had commercial success. This may also relate to the lack of a pronounced domestic mining industry in the countries, which distinguishes them very much from, for example, South Africa.

As the main outcome, the international assessments identified deterioration of the legacies left by uranium mining and processing activities in the area. Deterioration was critical in particular for those wastes and residues that were located in areas of seismic activity or in the direct vicinity of rivers — thereby posing environmental and radiation risks. As an international response, a first event on ‘Uranium Tailings in Central Asia: Local Problems, Regional

Consequences, Global Solution' took place in Geneva on 29 June 2009. Agreements on "additional financial and technical resources to manage and maintain the uranium tailings sites at a safe level" were achieved, in addition to the "continued support by the international community" [160]. The resolution by the UN General Assembly [160] bears furthermore witness to the relative importance that was given to the Central Asian legacy sites in the international community. A second forum with the title 'Uranium Tailings in Central Asia: Joint Efforts in Risk Reduction' was held, devoted to the 'dangers associated with former uranium mining activities in Central Asia'. Concurrently to these events, the IAEA developed a baseline document that — taking into the account the outcomes of previous international assessments — prioritized sites for environmental remediation by ranking their risk. In addition, the Coordination Group for Uranium Legacy Sites (CGULS) was formed as a platform to exchange information, provide technical assistance and coordinate actions for IAEA Member States in Central Asia.

In the period 2008–2012, a concept for a Commonwealth of Independent States (CIS) inter-state targeted programme on remediation of member state territories affected by uranium mining industries was developed for the territories of EurAsian Economic Community (EurAsEC) Member States. It was approved by a decision of the EurAsEC Interstate Council in 2012. Funding is being provided by those CIS Member States participating in the programme. The CIS Programme commenced in 2013 and is currently in the second phase of implementation (2017–2023). Remediation at Kadji-Say in Kyrgyzstan was completed in 2019. Future remediation works are foreseen for Min-Kush tailing ponds in Kyrgyzstan and the 'Yellow Hill' and tailings piles 1–4 at Istiklol in Tajikistan.

In 2010, with funding from its Instrument for Nuclear Safety Cooperation (INSC), the European Commission funded a programme for remediation preparation and planning for Kyrgyzstan, Tajikistan and Uzbekistan. Kazakhstan was not included, based on its financial situation. As a result of this programme, remediation projects at seven sites — Min-Kush (for objects other than tailings), Shekaftar, Charkesar, Yangiabad, Istiklol, Degmay and Mailuu-Suu — are now ready for implementation and are part of a Strategic Master Plan (SMP). This SMP was developed by the IAEA in cooperation with regional experts and partners from international organisations and presents the framework under which remediation is to be performed. The overall aim of the SMP is to ensure coordination, timeliness, cost-effectiveness and sustainability of the activities [155].

In 2015, the European Commission (EC) initiated the Environmental Remediation Account for Central Asia (ERA) to finance remediation. The launch of the account reflected the understanding that funds for the remediation can only be generated by a multilateral approach. The total cost of remediation is currently estimated at around 100 million € and the management of the ERA occurs under the European Bank for Reconstruction and Development (EBRD). It is noteworthy that this is the first time that such a multilateral Mining Remediation Fund has been established — it is internationally unique. Remediation activities are foreseen to start in Kyrgyzstan (Min-Kush, Shekaftar) in 2020 and a Project Management Unit (PMU) for the implementation of remediation was set up.

7.1. URANIUM PRODUCTION AT MAILUU SUU, KYRGYSTAN

Among the Central Asian states, Kyrgyzstan was one of the first to start uranium mining. One famous example is Mailuu Suu, a mining town with 25 000 inhabitants in the Fergana valley, part of the Mailuu Suu river valley. Located at an altitude between 900 and 1000 m a.s.l, the

area is prone to erosion and landslides and part of the Syr Darya that discharges into lake Aral. The distance to the Uzbek border is approx. 25 km.

Mining and milling of uranium ore commenced in Mailuu Suu in 1946 and ended in 1968. Two ore processing techniques were applied to extract uranium from the from mined or shipped material: (i) ion exchange techniques; and (ii) alkaline leach. Material was also delivered to Leninabad in Tajikistan where further processing took place. It is estimated that in Mailuu Suu, more than 10 000 tonnes of uranium were produced from around 9.1 million tonnes of ore [161]. However, the activities were not restrained to local uranium occurrences. Imports and processing by local enterprises is recorded for ores from from Eastern Germany (Erzgebirge), Chekhoslovakia (Yakhimov), Bulgaria (Bukhovo) and Tadjikistan (Taboshar, Adrasman) until the mid 1950s. The former German Democratic Republic contributed the majority of the foreign ore with about 75% [162]. These imported ores are noteworthy due to their differing properties when compared to the Kyrgyz ore. The imported ores showed higher contents of lead and arsenic and also exhibited higher levels of radiation than the local counterpart — characteristics that may also be reflected in the processing residues and tailings [163].

Economic ore accumulations (Carnotite- and Tyuyamunite) were first discovered in Mailuu Suu around 1935 in the surface outcrops of Paleogene lime stones. Increasing exploration efforts for uranium started in 1945 cumulating in important uranium ore discoveries in 1946, 1948 and 1949. The local deposits can be described as the type “bituminous limestone”. Uranium mineralization in the hydrocarbon-bearing carbonates is furthermore structure-related. The ores are usually bound to faults and fractures in specific horizons of the limestone’s Syn- and Anticlines. The grades of uranium in the ore varied between 0.03% and more than 0.5%. Local uranium mineralogy can be distinguished in oxidized zone minerals (Carnotite and Tyuyamunite) and non-oxidized zone (Pitchblende, nivenite, and sooty pitchblende) [164]. The local uranium deposits are primarily Pitchblende and the oxidation products Carnotite ($K_2(UO_2)_2V_2O_8 \cdot 3H_2O$) and Tyuyamunnite ($Ca(UO_2)_2V_2O_8 \cdot 5\text{-}8H_2O$). Orthobrannerite ($U^{4+}U^{6+}Ti_4O_{12}(OH)_2$) and Uranotallite $Ca_2(UO_2)(CO_3)_3 \cdot 11H_2O$ are of minor significance in the area [165].

7.2. MINING LEGACIES IN MAILUU SUU, KYRGISTAN

Seven priority sites for remediation were identified for Central Asia, while Mailuu Suu became the most prominent one having international attention since the early 1990s. Until today, the mining town is often used as an reference site for environmental impacts of Soviet uranium production and its wastes, being representative for most of the environmental problems of similar sites in Central Asia. Attention by international media, the public and political stakeholders helped to raise international funds for the environmental legacies at Mailuu Suu and at other sites.

The town of Mailuu Suu is located in proximity to the tailing impoundments and mine waste rock depositories. With respect to uranium tailings, it is the largest site, occupied by 23 tailings and 13 mine waste dumps. Overall, these wastes and residues cover an area of about 44 ha. The total volume is estimated to account to approximately 3 million m^3 while the volume of tailings ponds constitutes about 2 million m^3 [161].

From 1966 to 1968, right after mining terminated, the 23 tailing impoundments were secured complying with the standards of the time while the 13 mine waste dumps remained unmanaged. Subsequent monitoring of the tailings and mine waste dumps was continued until 1992. After this time, reconstruction and repair work was conducted, yet with limited effects on the

uranium-containing mining and processing wastes, their potential deterioration and environmental impacts [166].

7.2.1. Geochemistry and recovery of tailings in Mailuu Suu

Nimrodel Resources Ltd undertook first pass core drilling on the 17 largest of the 23 tailings dams, with 94 holes drilled, taking 482 samples for analysis by ICP-MS at the Central Scientific research Laboratory at Kara Balta Uranium Processing Plant. Multiple significant intercepts exceeding 100 ppm were recorded in tailings dams 2, 3, 4, 5, 7, 13 and 14 (Fig. 17). Initial leach tests undertaken at Kara Balta indicated recoveries of up to 90% U, giving encouragement for further mineralogical and metallurgical testwork. A conceptual processing scheme of screening and repulping of material on site, addition of alkali carbonate leach agent to the slurry, and transport to a processing plant for ion exchange or solvemt extraction recovery of uranium were considered. It was also considered that discharge of the relatively neutralised tailings from the processing plant would occur in the adjacent re-engineeered Tailings Dam 15 on site. The project was never progressed further due to a change in company priorities during 2009. In contrast to the established practice of tailings reprocessing in the Witwatersrand Basin, the scoping study on possible reprocessing (see above) in Mailuu Suu did not proceed to the commercial reprocessing of tailings at that time [167]. Varying physical and chemical properties as well as material volumes and footprints were reported for different tailings storage facilities in the area.

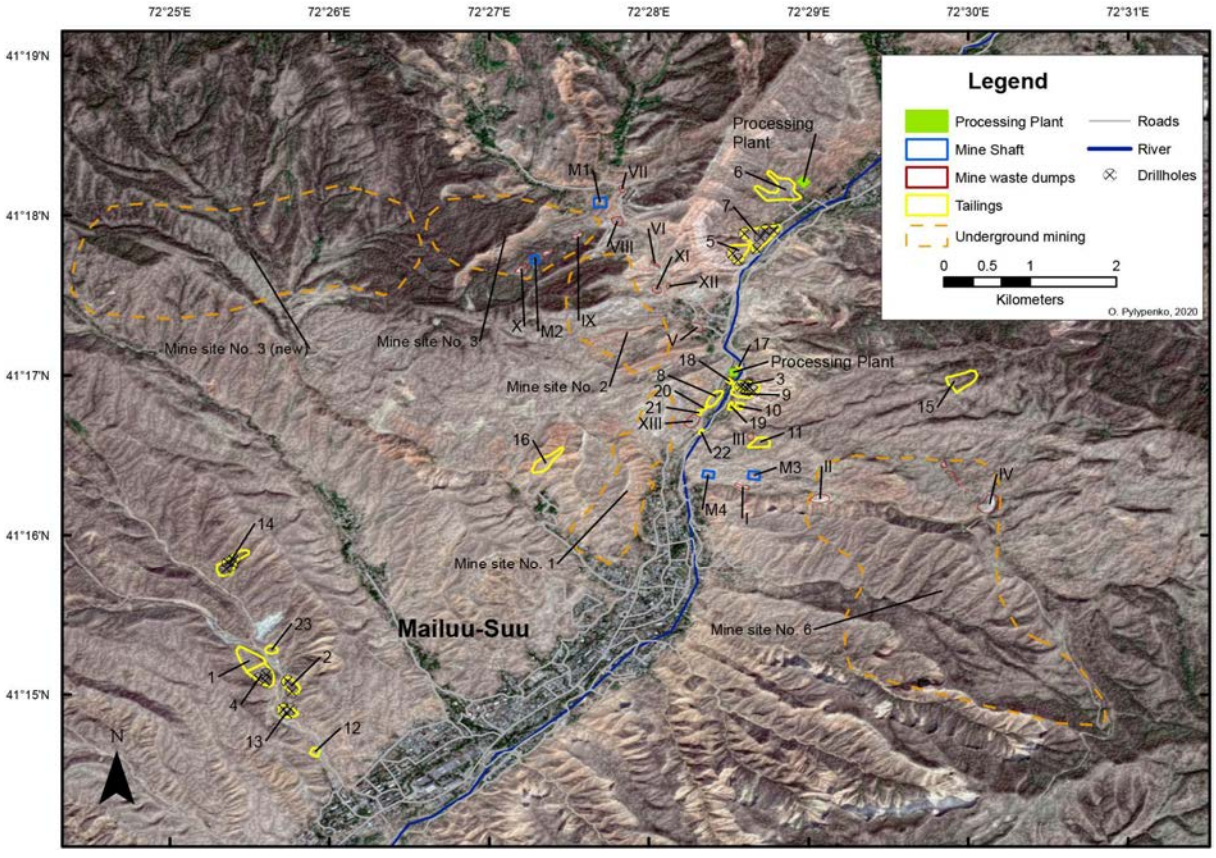

FIG. 17. Copernicus Sentinel hyperspectral image on digital terrain (2020, processed by ESA) of the Mailuu Suu area showing location of tailings dams, waste rock dumps, mine shafts, original processing facilities, surface trace of underground mine workings and Nimrodel Resources Limited drillholes.

The difference in the chemical characteristics of four tailings impoundments can be seen in Table 13. While the uranium concentration reaches relatively high values (600 ppm) in tailing pond (TP) 3 with an average uranium concentration of around 250 ppm [159], it is low in TP 7. TP 5 and TP 8 have intermediate concentration ranges. Other trace metals such as Ni, As or Pb exhibit a similar variability. This illustrates that the processing of imported ore was a main contributor to uranium production which lead to variable geochemical tailings characteristics. The significance of the ore imports is also exemplified by the fact that the total volume of tailings in Mailuu Suu is twice as high as total volume of waste dumps.

The variability of the uranium concentration is also an indication that the extraction efficiency of milling either varied and/or that in the early years of processing the extraction efficiency was lower than in later years. Processing of uranium ores of variable origin is challenging during milling and extraction efficiency is sometimes not optimal. The same would hold for the reprocessing of tailings derived from various ore sources. This may be an additional reason for non-profitability of tailing reprocessing in Mailluu Suu even though the uranium content is high compared to tailings from the Witwatersrand region. It is notable that especially TP 3 with an average uranium content of 250 ppm (0.025 %). It is notable that especially TP 3 with a volume of 110 500 m^3 and an average uranium content of 250 ppm (0.025 %) was comparable to the lower economic limit for uranium recovery from the Mailuu Suu deposit. This limit corresponds to an ore grade of 300 ppm (0.03 %). All other trace element concentrations presented in Table 13 are all below economic thresholds.

TABLE 13. COMPOSITIONAL RANGES OF TAILINGS ACCORDING TO THE TACIS REPORT [166] (NA = NOT AVAILABLE)

Elements	Tailing #3	Tailing # 5	Tailing # 7	Tailing # 8
Si (%)	0.02 – 0.1	0.16 – 0.17	0.04 – 0.08	0.05 – 0.08
U (ppm)	48 – 600	95 – 132	3 – 16	27 – 178
Ra (Bq/kg)	277 – 303 770	1 114 – 2 512	777 – 24 446	8 606 – 21 959
Th (Bq/kg)	393 – 13 831	NA	570 – 28 235	1 961 – 16 413
Ni (ppm)	17 – 22	29 – 138	23 – 50	10 – 40
V (ppm)	15 – 230	17 – 27	10 – 83	44 – 72
Mo (ppm)	1 – 2	1	1 – 3	2 – 10
Cd (ppm)	1 – 3	1 – 2	1	1
Pb (ppm)	4 – 136	5 – 10	7 – 268	5 – 6
Mn (ppm)	198 – 12 130	392 – 700	140 – 475	375 – 639
Co (ppm)	2 – 40	5 – 9	2 – 9	4 – 9
Ba (ppm)	22 – 275	137 – 167	21 – 261	33 – 88
Cr (ppm)	8 – 52	20 – 36	8 – 23	6 – 19
Hg (ppm)	< 5	< 5	< 5	< 5
Sb (ppm)	≤ 3	≤ 3	≤ 5	≤ 5
Se (ppm)	1 – 34	< 2	1 – 152	23 – 75
As (ppm)	1 – 53	2 – 5	2 – 61	2 – 3

7.2.2. Environmental situation and impact of the tailings

The Tailings facilities may cause environmental impacts when eroded by wind, water or landslides or when destabilised by seismic events in the geotechnically instable region. In these circumstances, environmental and health risks can arise.

The environmental hazard that appears most prominent is erosion of the tailings due to precipitation, wind, snow melt or other processes working on the tailings. Another factor relating to erosion is the fact that some tailings are deposited on the banks of the Mailuu Suu river and its tributaries from where material may get transported downstream by the river. Moreover, structural stability of tailings may be compromised by seismic loads, i.e. the build-up of eroded material from other sources on the tailings top. This might be the case when loamy alluvial material is eroded from hillslopes, eventually accumulated on the tailings surface. Another direct and indirect hazard are landslides. While landslides and mudflows may have a direct impact, also indirect impacts may occur from rising river water levels due to river blockages by landslides. In this latter scenario, inundations may affect the waste dumps and tailings situated upstream the blockage [156].

Thus, it is recognized that natural processes act on the Mailuu Suu uranium tailings. This presents an environmental risk since these processes have the potential to disperse uranium and other tailings constituents from the tailings stocks into the broader environment.

7.2.3. Tailings remediation measures

Mailuu Suu is also among the first sites where internationally funded remediation measures were carried out. With an investment of $ 12 million, the World Bank enabled the “Disaster Hazard Mitigation Project” (DHMP) that was conducted from 2004 to 2012. One of the project objectives was to reduce the exposure of flaura and fauna to radionuclides from abandoned uranium tailings. Based on existing environmental data and additional investigations, an iterative evaluation and decision-making procedure was carried out to categorize waste facilities in the area according to their risks. Throughout the process, the risks associated with erosion and geotechnical failure played important roles. Radiological risks were also adressed. Regarding remediation, 14 objects were identified with high priority. Another priorization was elaborated for remedial actions consistent with the budgetary and timely limitations of the DHMP.

First intervention methods comprised: (i) placing inert cover materials of a specific thickness on the tailings surface; and (ii) the restauration of drainage and water channels. Moreover, complementary measures took place to inform the public, such as awareness and information campaigns and the erection of warning signs. These intervention methods were perceived as most simple, rapid and cost-effective.

More complex interventions concerned the stabilisation of the tailings against erosion. One proposed solution was the relocation of the most eroded tailings ponds to safer disposal areas. This relocation was initially considered for a total of 7 tailings and waste rock dumps, however, only realized for the tailing pond with the highest risk (TP3) due to cost and time restraints. This was performed under the condition that any potential contamination of pristine land by the disposal of waste should ideally be avoided. In coordination with relevant stakeholders (regulators and public) and by ensuring long-term aims, such as stability, technical feasability and risks, minimum impact on local infrastructure, the tailings material of TP3 was eventually added to an exisiting tailings pond (TP6) [156].

As an important project oucome, the DHMP project identified the need to continue remedial activities for five other tailings ponds. The project funding had been insufficient to cover the associated costs for these sites at the time. The DHMP was moreover successful in increasing the visibility of Central Asia and its uranium mining legacies in the international community. This led to a call for enhanced international support in managing the instable, uranium-containing tailings ponds from mining and processing both local and imported ore. As a response, both, the EC-funded programme for remediation preparation and planning as well as the Environmental Remediation Account (ERA) to finance remediation in the region emerged (see above).

The ongoing INSC Project Mailuu Suu (KG 4.01/14) which is the last of the 4 major engineering projects funded by the INSC in the region will address the remaining needs identified in the DHMP. It will result in a remediation strategy for Mailuu Suu — agreed with all relevant stakeholders — including an engineering designs and an environmental impact assessment that underwent regulatory review and permitting, so that the remediation work can be tendered, contracted, and executed under the umbrella of ERA.

7.3. CONCLUSIONS

Mining for uranium has led to the presence of numerous tailings dumps containing significant amounts of uranium in large areas of Central Asia, in mostly low-income countries. The lack of a pronounced mining industry, the complex logistics of uranium production and economic uncertainty may present some of the reasons why attempts on reprocessing tailings have not been economically successful until now. However, the environmental problems associated with these tailings continued to prevail. Recognizing that the supranational economic ties of the Soviet era led to serious environmental consequences in the region points to a two-fold fact: First, these environmental problems are not only national. Second, dealing with them is consequently a question of international cooperation. In the early 2000's, it became increasingly clear that these mostly low-income countries are in the need of an international initiative to solve the problem of uranium in mining and processing wastes with associated environmental risks. This has led to the establishment of the first international and multilateral mining remediation fund that will finance the onset of remediation in the region in 2020.

8. ACCOUNTING OF URANIUM IN TAILINGS

Operational data of individual metal mines show that the mass of tailings generated largely reflects the total mass of ore processed [10]. Therefore, a mill extracting 1000 t of metal ore per day using tank leaching will also produce the near same tonnage of tailings per day. The dry mass of tailings generated is almost equal to the dry mass of raw material processed. Aside from the uncertainties related to input data (original resource estimates, production rates, mining dilution, etc.) it should be noted that lack of data is a significant limitation in these calculations. For example, chemical analysis for waste material may be so scarce as to be only broadly representative.

Similarly, uranium mills extract only the small valuable proportion of uranium from the ore during mineral processing and hydrometallurgical extraction and the remainder of the mass reports to the tailings facility. Conventional uranium mills receive an ore feed, produce a uranium product (uranium production) and discharge uranium-bearing tailings to waste impoundments. Thus, when uranium ore is fed into a uranium mill there are only two uranium-bearing materials generated, the uranium product (ammonium diuranate or uranium oxide) and the waste product (uranium mill tailings).

The performance of uranium mills can be evaluated based on the cost of operation, quantity of ore processed, purity of concentrate produced, and the recovery rate of the extracted uranium from the processed ore. Thus, metallurgical accounting is commonly used to understand the operation at every stage. Mathematical equations are thereby applied to identify and calculate material balances, similar to other published process calculations [34]. The procedure uses the inputs and outputs of the process plant. The inputs and outputs of a conventional uranium mill are simple (mass in = mass out plus in-circuit inventories). The input is the ore fed to the uranium mill, whereas the output amounts to the uranium concentrate and the uranium mill tailings. Hence, a simplified material balance equation for the overall mass flow process of a uranium mill can be expressed as follows:

$$M_F = M_T + M_P \quad \text{(eq. 1)}$$

where,
M_F = mass of ore fed to the mill (t)
M_T = mass of tailings (t)
M_P = mass of drummed uranium production (t)

A component balance for any element including uranium is therefore:

$$M_F \times C_F = (M_T \times C_T) + (M_P \times C_P) \quad \text{(eq. 2)}$$

where,
M_F = mass of ore fed to the mill (t)
C_F = concentration of uranium in ore fed to the mill (%) (head grade)
M_T = mass of tailings (t)
C_T = concentration of uranium in tailings (%)
M_P = mass of drummed uranium production (t)
C_P = concentration of uranium in drummed uranium production (%)

A balance for the mass of uranium moving through a uranium mill can be calculated using the following simplified material balance equation:

$$U_F = U_T + U_P + \text{in-circuit inventory or } U_T = U_F - U_P - \text{in-circuit inventory} \quad \text{(eq. 3)}$$

where,
U_T = mass of uranium in tailings (t U_3O_8)
U_F = mass of uranium fed to the mill (t U_3O_8)
U_P = mass of uranium in drummed uranium production (t U_3O_8)

Mass balancing and uranium accounting may be performed on a shift or day/week basis to act and react in the processing plant. However, cumulative long-term data (i.e. annual accounting) are more representative of mass balances and uranium mass flows than short-term results (i.e. daily accounting). Therefore, quantitative assessments of the uranium masses (t) present in tailings can be pursued following two different accounting methods:

(i) Direct accounting methods based on quantitative data obtained on the tailings (i.e. tailings mass, uranium concentration in the tailings mass), or;
(ii) Indirect accounting methods that use existing production data on the milled and treated ores (ore milled, head grade of milled ore, uranium production, recovery rate).

So, the difference between these two accounting methods (direct and indirect accounting) is that the direct assessment relies on measurements and analyses of tailings, whereas the latter is based on uranium production data. Both assessment methods use mathematical equations to quantify the uranium masses in tailings. Because these computations are entirely independent of the sampling, analyses, record keeping and publication of the externally sourced data; the calculated results are only as accurate as the sampling, field and laboratory analyses, record keeping and publication activities applied to the data by others. Moreover, this metallurgical accounting considers dry materials, disregards the addition of chemicals and assumes the presence of a closed system, ignoring any material losses, for example, due to spillages.

In this chapter, accounting of uranium losses to tailings have been pursued as follows:

(a) Direct accounting of uranium in tailings for selected mine sites using tailings data (Section 8.2);
(b) Indirect accounting of uranium in tailings for selected mine sites using yearly ore processing data (Section 8.2);
(c) Indirect accounting of uranium present in several tailings storage facilities using cumulative ore production data (Section 8.3);
(d) Indirect accounting of uranium in tailings repositories globally using cumulative tailings masses and average uranium contents (Section 8.4).

8.1. METALLURGICAL ACCOUNTING METHODS

8.1.1. Direct accounting

Analyses of uranium concentrations in tailings and data on the tailings mass allow a direct calculation of the uranium mass present in the tailings. A simplified equation for the calculation of uranium stocks in tailings (t U_3O_8) can be written as follows:

$$U_T = M_T \times C_T \quad \text{(eq. 4)}$$

where,
U_T = mass of uranium in tailings (t U_3O_8)
M_T = mass of tailings (t)
C_T = concentration of uranium in tailings (% U_3O_8)

The annual mass of uranium stock accumulating in tailings can be calculated as follows:

$$U_{T(a)} = M_{T(a)} \times C_{T(a)} \quad \text{(eq. 5)}$$

where,
$U_{T(a)}$ = mass of uranium in tailings (t U_3O_8) per annum
$M_{T(a)}$ = mass of tailings (t) per annum
$C_{T(a)}$ = concentration of uranium in tailings (% U_3O_8) per annum

However, few industry reports and publications document these key variables for individual uranium mills on a monthly/quarterly or annual basis, which in turn would allow direct accounting of uranium in tailings. Moreover, data on waste production are commonly not published by the mining industry. In this case, indirect accounting is required to estimate the quantity of uranium in tailings.

8.1.2. Indirect accounting

Indirect accounting relies on mass balance equations that use ore processing and production data. Hence, these balances represent indirect estimates and are therefore only proxies to actual uranium quantities in mill tailings. Equation (6) is a material balance equation for solids, which allows deductions on the amount of uranium transferred to tailings, provided that the amount of material fed to the mill and the uranium production data are available.

$$U_T = U_F - U_P \quad \text{(eq. 6)}$$

where,
U_T = mass of uranium in tailings (t U_3O_8)
U_F = mass of uranium fed to the mill (t U_3O_8)
U_P = mass of uranium in drummed uranium production (t U_3O_8)

If parameters are measured at a regular interval (e.g. yearly), then the annual amount of uranium present in tailings ($U_{T(a)}$) is the difference between the annual quantity of uranium fed to the mill ($U_{F(a)}$) and the production of drummed U_3O_8 ($U_{P(a)}$) per year:

$$U_{T(a)} = U_{F(a)} - U_{P(a)} \quad \text{(eq. 7)}$$

where,
$U_{T(a)}$ = mass of uranium in tailings (t U_3O_8) per annum
$U_{F(a)}$ = mass of uranium fed to the mill (t U_3O_8) per annum
$U_{P(a)}$ = mass of uranium in drummed uranium production (t U_3O_8) per annum

There are two possible approaches to solve equation (7) using either (a) data on the mass of ore fed to the mill and the mill head grade (i.e. the mill feed), or (b) data on the mill recovery rate.

Mill feed

In this case, the annual amount of uranium mass in the tailings ($U_{(T(a))}$) can be calculated from the amount of ore fed to the mill and the concentration of uranium in the ore fed to the mill (i.e. the reported mill head grade) and uranium production for that year, whereby:

$$U_{T(a)} = (M_{F(a)} \text{ x } C_{F(a)}) - U_{P(a)} \quad \text{(eq. 8)}$$

where,
$U_{T(a)}$ = mass of uranium in tailings (t U_3O_8) per annum
$M_{F(a)}$ = mass of ore fed to the mill (Mt) per annum
$C_{F(a)}$ = concentration of uranium in ore fed to the mill (% U_3O_8) per annum (head grade)
$U_{P(a)}$ = mass of uranium in drummed uranium production (t U_3O_8) per annum

Mill recovery rate

Industry may report the recovery rate for uranium mills; however, this parameter can be calculated via different approaches depending on the data available. Unfortunately, the approaches used to establish mill recovery rates are generally not given in published industry reports. In this study, all stated mill recovery rates for uranium were assumed to represent the ratio of the weight of uranium annually recovered in the concentrate to 100% of uranium in the annual feed to the uranium mill, expressed as a percentage whereby:

$$R = 100 \text{ x } (U_{P(a)} / U_{F(a)}) \text{ or } U_{F(a)} = 100 \text{ x } (U_{P(a)} / R) \quad \text{(eq. 9)}$$

where,
R = mill recovery rate (%)
$U_{P(a)}$ = mass of uranium in drummed uranium production (t U_3O_8) per annum
$U_{F(a)}$ = mass of uranium fed to the mill (t U_3O_8) per annum

Consequently, the total amount of uranium mass in tailings can be estimated using uranium production data and the mill recovery rate:

$$U_{T(a)} = U_{F(a)} - U_{P(a)} \quad \text{(eq. 10)}$$

and

$$U_{F(a)} = 100 \text{ x } (U_{P(a)} / R) \quad \text{(eq. 11)}$$

therefore,

$$U_{T(a)} = (100 \text{ x } (U_{P(a)} / R)) - U_{P(a)} \quad \text{(eq. 12)}$$

where,
$U_{T(a)}$ = mass of uranium in tailings (t U_3O_8) per annum
$U_{P(a)}$ = mass of uranium in drummed uranium production (t U_3O_8) per annum
R = average mill recovery rate (% U_3O_8)

Theoretically, calculations of these three different accounting techniques using the given mass balance equations (eq. 10, 11 and 12) should yield very similar uranium quantities in mill tailings (t U_3O_8) for the same year, providing the input data are correct and any assumptions are valid.

8.2. TAILINGS CASE STUDIES

8.2.1. McClean Lake, Canada

Production from the McClean Lake JEB mill (Canada) resumed in 2014 to process low-grade ore from the stockpile and high-grade ore from the Cigar Lake mine. Published annual production data for the McClean Lake uranium mill are limited and include only information on annual uranium production, annual milled ore tonnages, average mill feed grade and uranium recovery for the years 2014 to 2016 (Table 14).

An indirect assessment of the annually generated uranium mass in the McClean Lake tailings is possible using published production data as stated in [168]. Applying the stated mill recovery rate and ore production (eq. 8.9), the annual uranium losses to the tailings repository range from 6 to 61 t U_3O_8 in per year in the period between 2014 to 2016 (Table 15). Alternatively, an indirect assessment of the annual uranium mass in McClean Lake tailings is possible using uranium production figures, milled ore tonnages and mill head grades (eq. 8.7). Applying such data, the annual uranium losses to the tailings repository range from negligible amounts to 181 t U_3O_8 in the years 2014 to 2016 (Table 15). Thus, estimates of the two calculation methods reveal dissimilar annual uranium losses to the mill tailings, however, both methods indicate that uranium has ended up in the McClean tailings and amounts to several tens of tonnes of U_3O_8 per annum since 2015.

TABLE 14. PRODUCTION DATA FOR THE MCCLEAN LAKE MILL AS DOCUMENTED BY CNSC [168]

Year	Uranium production (t U_3O_8)	Mill ore feed (t)	Average mill feed grade (% U_3O_8)	Mill recovery rate (%)
2014	235	7 830	3.00	97.54
2015	5 071	25 520	17.56	98.99
2016	7 866	37 200	18.08	99.10

TABLE 15. CALCULATED ANNUAL URANIUM LOSSES TO TAILINGS AT THE MCCLEAN LAKE URANIUM MILL BASED ON DATA GIVEN BY [168]

Year	Annual loss of uranium to tailings (t U_3O_8) based on known uranium production and mill recovery rate (eq. 8.9)	Annual loss of uranium to tailings (t U_3O_8) based on known uranium production, milled ore mass and mill head grade (eq. 8.7)
2014	6	—
2015	44	181
2016	61	56

8.2.2. Rössing, Namibia

Rössing Uranium Limited (RUL) operates a large, low-grade open pit uranium mine and processing plant in the Erongo region of Namibia. Ore is mined, crushed and milled and uranium is then extracted and recovered from the ore via sulfuric acid leaching, ion exchange, solvent extraction, precipitation of yellow cake and finally calcination to form a final uranium oxide product. The mill has been producing since 1976. Uranium is hosted by various different primary minerals, including uraninite, uranophane, betafite and traces of coffinite, brannerite and carnotite as well as secondary uranium bearing minerals [169]. Thus, the Rössing orebody contains some refractory uranium minerals, i.e. betafite and brannerite. The presence of these refractory uranium minerals in 'run-of-mine' ore to the leach plant can influence the amount of uranium extraction, if the leach is run at ambient conditions [169].

Published annual production data for the Rössing uranium mill (Namibia) are limited and include only information on annual uranium production, annual milled ore tonnages and annual tailings tonnages for the years 2010 to 2018 (Table 16). Data were obtained from information published by the company [170, 171]. Uranium concentrations in Rössing tailings have been stated as 64 ppm uranium (= 75 ppm U_3O_8) [48], with different grain sizes of the residues having various uranium concentrations [169].

TABLE 16. PRODUCTION DATA OF THE RÖSSING URANIUM MILL AS DOCUMENTED BY [170, 171] (ND: NOT DOCUMENTED)

Year	Uranium production (t U_3O_8)	Processed ore (t)	Tailings disposed in tailings storage facility (t)	Waste rock placed into waste rock dumps (t)
2010	ND	ND	11 594 430	40 022 450
2011	2 149	ND	10 370 362	39 608 654
2012	2 700	ND	12 152 173	33 749 173
2013	2 395	ND	11 261 619	25 332 432
2014	1 543	7 040 000	7 040 277	15 954 100
2015	1 245	6 876 000	6 875 719	12 522 652
2016	1 850	9 194 000	9 194 439	16 467 097
2017	2 110	9 000 000	8 962 923	15 109 738
2018	2 479	8 851 000	8 851 288	11 459 319

A direct assessment of the annual uranium mass present in tailings is possible using analyses of uranium concentrations in tailings and data on the tailings mass produced (equation 5). Assuming an average uranium concentration of 75 ppm U_3O_8, then annual uranium losses to the tailings repository range from 519 to 917 t U_3O_8 in the years 2010 to 2018 (Table 17).

An indirect assessment of the uranium mass present in Rössing tailings is possible using published uranium production figures (Table 16) and a mill recovery rate of 85% as stated in [30] (eq. 8.9). Assuming such a constant recovery rate, then annual uranium losses to the tailings repository range from 220 to 476 t U_3O_8 in the years 2011 to 2018 (Table 17).

An alternative indirect assessment of the uranium mass present in Rössing tailings is possible using uranium production figures, milled ore tonnages and mill head grades (eq. 8.7). Mill head grades of the Rössing plant have been given as 322, 317, 244 and 218 ppm uranium, depending on ore type, and 300 ppm uranium has been assumed for leach optimization studies [169]. Thus, mill head grades are not available on an annual basis. For the indirect calculations of uranium in tailings, head grades were assumed to amount to 0.03% U_3O_8 for the years 2014 to 2018. Assuming this average head grade, annual uranium losses to the tailings repository range from 176 to 908 t U_3O_8 in the years 2014 to 2018 (Table 17).

Estimates of the three calculation methods reveal slightly dissimilar annual uranium losses to the mill tailings; however, overall the annual losses to the Rössing tailings storage facility amount to several hundred tonnes of U_3O_8 per year (Table 17). The obvious discrepancies in the determined annual uranium losses to tailings between the three calculation methods are likely due to flawed assumptions made in the calculations (eq. 8.5, 8.7 and 8.9), which assume a constant uranium concentration in tailings (75 ppm U_3O_8) as well as constant mill recovery rates (85%) and head grades (0.03% U_3O_8) over several years.

TABLE 17. CALCULATED ANNUAL URANIUM STOCK LOSSES TO TAILINGS AT THE RÖSSING URANIUM MILL BASED ON DATA GIVEN BY [28, 48, 170, 171] (ND: NO DATA)

Year	Annual loss of uranium to tailings (t U_3O_8) based on known tailings mass and assumed 75 ppm U_3O_8 in the tailings (eq. 8.5)	Annual loss of uranium to tailings (t U_3O_8) based on known uranium production and assumed mill recovery rate of 85% (eq. 8.9)	Annual loss of uranium to tailings (t U_3O_8) based on known uranium production, milled ore mass and assumed mill head grade of 0.03% U_3O_8 (eq. 8.7)
2010	875	ND	ND
2011	783	379	ND
2012	917	476	ND
2013	850	423	ND
2014	531	272	569
2015	519	220	818
2016	694	326	908
2017	676	372	590
2018	668	437	176

8.2.3. Ranger, Australia

Energy Resources of Australia Ltd (ERA) operates the Ranger uranium mine, which has been producing since 1981 and is located in the Ranger Project Area approximately 250 km east of Darwin, Australia. The Ranger operation is a conventional milling, leaching, and solvent extraction circuit that extracts ore from open pits and treats it on-site to produce uranium oxide. Uranium is hosted by uraninite with accessory coffinite and brannerite [38].

Published annual production data for the Ranger uranium mill (Australia) were used to calculate the annual mass of uranium lost to tailings. Data provided by industry for the Ranger mine are more detailed than those for the Rössing operation and include annual drummed uranium production, mill recovery rates, milled ore tonnages and mill head grades for the years 1982 to

2019 (Table 18). Data were obtained from annual company reports published by Energy Resources Australia [172]. The annual reports vary in their detail and do not always provide all variables necessary for the calculations (i.e. sometimes information on mill recovery rates, head grades, or the amount of ore milled is missing). Regardless, indirect assessments of the uranium mass present in Ranger tailings are possible using: (i) uranium production figures and mill recovery rates (eq. 8.9); and (ii) uranium production figures, milled ore tonnages and mill head grades (eq. 8.7). Depending on the calculation method, uranium losses range from 147 to 797 t U_3O_8 and 230 to 783 t U_3O_8 per year, respectively. Thus, the two calculation methods reveal slightly dissimilar annual uranium losses to mill tailings, however, both methods indicate that the overall annual losses to the Ranger tailings storage facility generally amount to several hundred tonnes of U_3O_8 per year (Table 19). Since the beginning of operation in 1981 and over the last 38 years, total losses to tailings total 16 911 and 16 766 t U_3O_8, respectively (Table 19).

TABLE 18. PRODUCTION DATA OF THE RANGER URANIUM MILL AS DOCUMENTED BY ANNUAL REPORTS FROM [172] (ND: NOT DOCUMENTED)

Year	Mill recovery rate (%)	Uranium production (t U_3O_8)	Milled ore (t)	Mill head grade (% U_3O_8)
1982	88.50	3 110	ND	ND
1983	90.35	3 000	ND	ND
1984	89.92	3 098	ND	ND
1985	92.45	3 037	ND	ND
1986	92.00	3 067	860 000	ND
1987	93.05	3 076	800 000	ND
1988	91.95	3 041	782 000	0.423
1989	91.06	3 595	975 000	0.408
1990	90.10	3 084	1 089 000	0.314
1991	90.78	2 908	1 090 000	0.295
1992	89.83	2 980	986 000	0.324
1993	ND	1 335	426 000	0.348
1994	ND	1 461	437 000	0.389
1985	ND	1 548	578 000	0.345
1996	ND	3 453	1 201 000	0.349
1997	ND	4 236	1 571 000	0.311
1998	ND	4 161	1 843 000	0.269
1999	ND	4 374	1 827 000	0.267
2000	ND	4 144	1 500 000	ND
2001	91.3	4 203	1 579 000	0.306
2002	89.74	4 470	1 784 000	0.281
2003	88.3	5 065	2 067 000	0.281
2004	88.8	5 137	2 086 000	0.278
2005	88.3	5 910	2 293 000	0.288
2006	87.5	4 748	2 072 000	0.261
2007	88.2	5 412	1 900 000	0.31
2008	88.2	5 339	2 000 000	0.30
2009	88.3	5 240	2 300 000	0.26
2010	87.2	3 793	2 400 000	0.19
2011	87.9	2 641	1 600 000	0.18
2012	86.2	3 710	2 600 000	0.17
2013	84.8	2 960	2 300 000	0.15
2014	81.5	1 165	1 300 000	0.11
2015	82.0	2 005	2 500 000	0.10
2016	84.9	2 351	2 700 000	0.10
2017	84.7	2 294	2 600 000	0.10
2018	86.6	1 999	2 500 000	0.09
2019	86.8	1 751	2 500 000	0.08

TABLE 19. CALCULATED ANNUAL URANIUM STOCK LOSSES TO TAILINGS AT THE RANGER URANIUM MILL BASED ON DATA GIVEN IN ANNUAL REPORTS OF [172] (ND: NO DATA)

Year	Annual loss of uranium to tailings (t U_3O_8) based on mill recovery rate and uranium production (eq. 8.9)	Annual loss of uranium to tailings (t U_3O_8) based on mill head grade, amount of milled ore and uranium production (eq. 8.7)
1982	404	ND
1983	320	ND
1984	347	ND
1985	248	ND
1986	267	ND
1987	230	ND
1988	266	267
1989	353	383
1990	339	335
1991	295	308
1992	337	215
1993	ND	147
1994	ND	239
1985	ND	446
1996	ND	738
1997	ND	650
1998	ND	797
1999	ND	504
2000	ND	ND
2001	401	629
2002	511	543
2003	671	743
2004	648	662
2005	783	694
2006	678	660
2007	724	478
2008	714	661
2009	694	740
2010	557	767
2011	364	239
2012	594	710
2013	531	490
2014	264	265
2015	440	495
2016	418	349
2017	414	306
2018	309	251
2019	266	249
Sum to date	16 911	16 776

8.3. TAILINGS STORAGE FACILITIES

Quantitative assessments of total uranium stocks present in entire tailings impoundments can be based on cumulative ore and uranium production data. In this study, the uranium stocks left

in tailings storage facilities of selected uranium mines were calculated using cumulative ore und uranium production data as given by [88] (Table 20). The total mass of uranium left in tailings storage facilities was established using two mass balance equations: (a) cumulative uranium production and average mill recovery rate (eq. 8.10), and (b) cumulative milled ore mass, average mill head grade and cumulative uranium production (eq. 8.11).

$$U_{T(c)} = (100 \times (U_{P(c)} / R) - U_{P(c)} \quad \text{(eq. 8.10)}$$

where,
$U_{T(c)}$ = cumulative mass of uranium in tailings (t U_3O_8)
$U_{P(c)}$ = cumulative mass of uranium in drummed uranium production (t U_3O_8)
R = average mill recovery rate (% U_3O_8)

$$U_{T(c)} = (M_{F(c)} \times C_{F(c)}) - U_{P(c)} \quad \text{(eq. 8.11)}$$

where,
$U_{T(c)}$ = cumulative mass of uranium in tailings (t U_3O_8)
$M_{F(c)}$ = cumulative milled ore mass (Mt)
$C_{F(c)}$ = average concentration of uranium in ore fed to the mill (% U_3O_8) (head grade)
$U_{P(c)}$ = cumulative mass of uranium in drummed uranium production (t U_3O_8)

Both calculation methods (eq. 8.10 and 8.11) reveal similar cumulative uranium stock losses to tailings repositories, with the exception of the McLean Lake deposit, Canada (Table 21). Also, the mass balance calculations demonstrate that the quantity of uranium stocks present in the various tailings repositories is site specific, with values ranging from a minimum of 4 t U_3O_8 (Rockhole, Australia) to a maximum of 26 670 t U_3O_8 (Olympic Dam, Australia) (Table 21). Overall, tailings of the database of 32 mines contain an additional ~10% uranium (70 497 t and 73 187 t U_3O_8, respectively, Table 18) compared to the cumulative uranium production of all examined uranium mills (719 818 t U_3O_8) (Table 21).

TABLE 20. CUMULATIVE PRODUCTION DATA OF SELECTED URANIUM MINES AND MILLS [88]. URANIUM MINES WERE ASSIGNED TO URANIUM DEPOSIT TYPES ACCORDING TO [38]

Country	Mine	Deposit type	Operating	Total amount of milled ore (Mt)	Mill head grade (% U_3O_8)	Cumulative U_3O_8 production (t)	Mill recovery rate (%)
Australia	Rum Jungle	Proterozoic unconformity	1954–71	0.682	0.33	1841	81.13
Australia	Radium Hill	Intrusive	1956–61	0.969	0.117	852.1	75.15
Australia	Rockhole	Proterozoic unconformity	1959–62	0.013	1.107	139.7	94.07
Australia	Upper South Alligator Valley Group	Proterozoic unconformity	1959–64	0.095	0.483	392.7	85.55
Australia	Mary Kathleen	Metasomatite	1958–63	2.668	0.172	4092	89.22
Australia	Mary Kathleen	Metasomatite	1976–82	6.3	0.1	4801	76.20
Australia	Nabarlek	Proterozoic unconformity	1980–88	0.669	1.65	10 875	98.31
Australia	Ranger	Proterozoic unconformity	1981–2012	45.672	0.288	116 005	88.86
Australia	Olympic Dam	IOCG	1988–2012	148.379	0.063	66 809	71.47
Canada	Bicroft–Macassa	Intrusive	1956–63	2.318	0.094	2025	93.12
Canada	Pronto	Palaeo quartz-pebble conglomerate	1956–58	1.327	0.121	1374	85.46
Canada	Faraday	Intrusive	1957–64	2.683	0.104	2629	94.36
Canada	Elliot Lake, Denison	Palaeo quartz-pebble conglomerate	1957–92	62.771	0.112	65 809	93.71
Canada	Elliot Lake, Quirke	Palaeo quartz-pebble conglomerate	1957–90	32.337	0.116	35 333	94.58
Canada	Elliot Lake, Nordic	Palaeo quartz-pebble conglomerate	1957–90	34.971	0.112	37 355	95.28
Canada	Elliot Lake, Lacnor	Palaeo quartz-pebble conglomerate	1958	2.239	0.096	1 977	91.87
Canada	Elliot Lake, Dyno	Palaeo quartz-pebble conglomerate	1958–60	0.807	0.062	479	95.63
Canada	Elliot Lake, Stanrock	Palaeo quartz-pebble conglomerate	1958–63	2.432	0.094	2 122	92.74
Canada	Elliot Lake, CanMet	Palaeo quartz-pebble conglomerate	1958	0.664	0.096	584	91.64

Country	Mine	Deposit type	Operating	Total amount of milled ore (Mt)	Mill head grade (% U_3O_8)	Cumulative U_3O_8 production (t)	Mill recovery rate (%)
Canada	Port Radium	Metamorphite	1958, 60	0.156	0.59	889	96.14
Canada	Gunnar	Metamorphite	1960–61	1.256	0.185	2 207	95.20
Canada	Elliot Lake, Stanleigh	Palaeo quartz-pebble conglomerate	1960, 1983–96	12.509	0.09	10 326	91.28
Canada	Beaverlodge	Metamorphite	1962–63, 1977–82	2.526	0.202	4743	93.13
Canada	Elliot Lake, Milliken	Palaeo quartz-pebble conglomerate	1964	0.481	0.112	531	94.50
Canada	Bancroft	Intrusive	1978–82	1.646	0.081	1 269	94.84
Canada	Rabbit Lake	Proterozoic unconformity	1979–2012	12.329	0.73	83 290	93.84
Canada	Cluff Lake	Proterozoic unconformity	1982–2002	3.228	0.85	26 849	97.36
Canada	Key Lake, McArthur River	Proterozoic unconformity	1983–2012	6.78	3.03	199 596	97.11
Canada	McLean Lake	Proterozoic unconformity	1999–2010	1.526	1.49	22 095	99.77
Malawi	Kayelekera	Surficial	2009–12	3.379	0.127	3 208	74.94
Mongolia	Dornod	Volcanic-related	1988–96	0.946	0.107	920	90.79
Namibia	Langer Heinrich	Surficial	2007–12	11.728	0.09	8 401	79.75
			TOTAL			719 818	

TABLE 21. CALCULATED CUMULATIVE URANIUM STOCKS LEFT IN TAILINGS IMPOUNDMENTS OF SELECTED URANIUM MINES AND MILLS USING CUMULATIVE PRODUCTION DATA OF [88]

Country	Mine	Amount of uranium in tailings repository (t U_3O_8) calculated based on mill recovery rate and uranium production (eq. 8.9)	Amount of uranium in tailings repository (t U_3O_8) calculated based on mill head grade, amount of milled ore and uranium production (eq. 8.7)
Australia	Rum Jungle	428	410
Australia	Radium Hill	282	282
Australia	Rockhole	9	4
Australia	Upper South Alligator Valley Group	66	66
Australia	Mary Kathleen	494	497
Australia	Mary Kathleen	1 500	1 499
Australia	Nabarlek	187	164
Australia	Ranger	14 543	15 530
Australia	Olympic Dam	26 669	26 670
Canada	Bicroft-Macassa	150	154
Canada	Pronto	234	232
Canada	Faraday	157	161
Canada	Elliot Lake, Denison	4 417	4 495
Canada	Elliot Lake, Quirke	2 025	2 178
Canada	Elliot Lake, Nordic	1 850	1 813
Canada	Elliot Lake, Lacnor	175	172
Canada	Elliot Lake, Dyno	22	21
Canada	Elliot Lake, Stanrock	166	164
Canada	Elliot Lake, CanMet	53	53
Canada	Port Radium	36	31
Canada	Gunnar	111	117
Canada	Elliot Lake, Stanleigh	986	932
Canada	Beaverlodge	350	360
Canada	Elliot Lake, Milliken	31	8
Canada	Bancroft	69	64
Canada	Rabbit Lake	5 467	6 712
Canada	Cluff Lake	728	589
Canada	Key Lake, McArthur River	5 940	5 838
Canada	McLean Lake	51	642
Malawi	Kayelekera	1 073	1 083
Mongolia	Dornod	93	92
Namibia	Langer Heinrich	2 133	2 154
	TOTAL	70 497	73 187

8.3.1. Mary Kathleen, Australia

The quantitative estimates rely on accurate production and processing data for individual mill sites. Yet, there are instances where accurate data are not available and production figures for the same uranium mill have been stated by different sources differently. For example, a number of authors provide cumulative production data for the Mary Kathleen uranium mill (Australia), including the amount of ore milled, the mill head grade, and the drummed uranium production (Table 22). The reports differ on the cumulative amount of ore milled, stating 7 Mt [173] as well as 9.2 Mt [174] and 9.168 Mt [88], the latter two amounts being essentially the same. However at the Mary Kathleen mine site, there is only one single tailings storage facility containing approximately 7 Mt of tailings [46, 173], which has been confirmed by recent investigations[175]. By contrast, the production data of the World Nuclear Association [174] and Mudd [88] exceed the existing tailings mass (7 Mt) by ~30%.

This highlights the fact that auditing is required to confirm the milling and production data for individual sites (e.g. the amount of milled ore, mill head grade, mill recovery rate, and tailings tonnages). Consequently, waste accounting and quantitative assessments of uranium stocks in tailings impoundments, using published production data, can only represent approximations of the real conditions that prevail at individual tailings storage facilities.

TABLE 22. CUMULATIVE PRODUCTION DATA FOR THE MARY KATHLEEN URANIUM MINE (AUSTRALIA) AS DOCUMENTED BY VARIOUS SOURCES WITH REFERENCE SOURCE INDICATED IN SQUARE BRACKETS (ND: NO DATA)

	Unit	[173]	[176]	[174]	[88]
Ore processed 1958–63	Mt	ND	2.947	2.9	2.668
Ore processed 1975–82	Mt	ND	ND	6.3	6.3
Total ore processed	Mt	7	ND	9.2	9.168
Cumulative tailings	Mt	7	ND		
Ore grade processed 1958–63	U_3O_8 %	ND	0.15	0.15	0.172
Ore grade processed 1975–82	U_3O_8 %	ND	ND	0.10	0.10
Cumulative production U_30_8 1958–63	t	ND	4 080	4 080	4 092
Cumulative production U_30_8 1975–82	t	ND	4 802	4 802	4 801

8.4. GLOBAL ASSESSMENT

As part of the current project, a data compilation of uranium in mine waste will be included in the IAEA Uranium Deposit Database (UDEPO). Over 900 records were compiled worldwide (Fig. 18) and a representative sample is shown in Table 23. The global stockpile of uranium mill tailings is significant and growing, with estimates on the total uranium mill tailings present ranging from 100–200 Mt in 1980 [177], 938 Mm^3 in 2004 (Table 24), to 8 718 Mt in 2019 (Table 25). The latter two and most recent global estimates are likely conservative and remain only approximations of the actual conditions today, because both global estimates are incomplete. For example: (i) the assessment of 938 Mm^3 does not include uranium-bearing

tailings in African countries (Table 24); and (ii) the estimated mass of 8 718 Mt does not include data on tailings from some Asian and African countries (Table 25).

A direct comparison of these two documented tailings databases (Tables 24, 25) cannot be accomplished because: (i) there is a 15-year gap between reporting dates; and (ii) the 2004 database reports tailings in cubic metres and the 2019 database reports tailings in tonnes. Although bulk densities of most mine waste rocks fall within the range of 1-5-2.5 t/mw (with tailings commonly being slightly higher), without knowing the accurate bulk density of tailings at each site, it remains impossible to convert the reported tailings volumes to tonnages and vice versa. Regardless, if one considers the uranium-bearing gold tailings in South Africa, then the great majority of uranium-bearing tailings worldwide is present in Africa (Fig. 18).

The total mass of uranium contained in tailings globally can be calculated using the documented global tailings mass (Table 25) and an assumed average uranium content for all tailings globally using the following equation:

$$U_{T(g)} = M_{T(g)} \times C_{T(g)} \qquad \text{(eq. 13)}$$

where,
$U_{T(g)}$ = cumulative mass of uranium in tailings (t U_3O_8) globally
$M_{T(g)}$ = cumulative mass of tailings (t) globally
$C_{T(g)}$ = average concentration of uranium in tailings (% U_3O_8) globally

Assuming that a global tailings mass of 8 718 Mt (compare with Table 24) contains on average 100 to 200 ppm U_3O_8 (compare with Table 3), then at least 0.87 to 1.74 Mt U_3O_8 are contained in mill tailings worldwide. Such calculations represent estimates and can only indicate the extraordinary scale of uranium stocks available in uranium mill tailings globally. In this case, the estimate is ca. 17 to 35 years of global uranium supply at recent rates of requirements (approximately 50 000tU per annum).

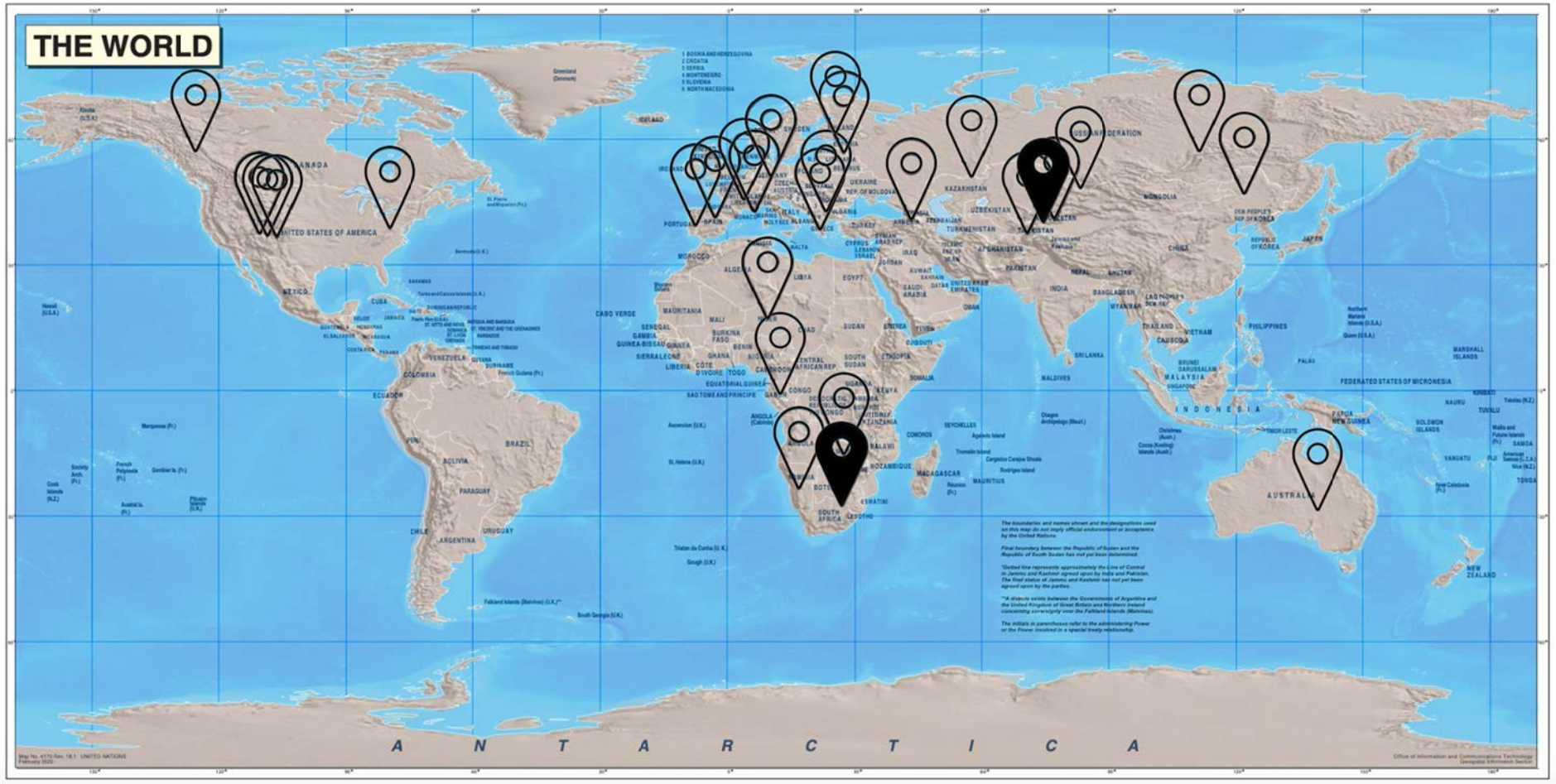

FIG. 18. A preliminary inventory of uranium-bearing mining and milling wastes. Data availability sufficient for detailed case studies (South Africa, Kyrgyzstan) indicated by black color.

TABLE 23. SELECTED ENTRIES OF THE PRELIMINARY DATA COMPILATION ON URANIUM IN MINE AND PROCESSING WASTE BY MINE SITE AND THEIR REPORTED URANIUM MASSES OR CONCENTRATIONS [28] (NA: NOT AVAILABLE)

Region	Country	Mine site	Waste type	Waste name	Waste mass (tonnes)	U in waste	Unit
Africa	South Africa	City Deep	Tailings	NA	67 463	161	µg/g
Africa	South Africa	CMR	Tailings	NA	68 582	284	µg/g
Africa	South Africa	East Champ d'or	Tailings	NA	6 560	201	µg/g
Africa	South Africa	East Driefontein	Tailings	NA	52 005	136	µg/g
Africa	South Africa	East Geduld	Tailings	NA	53 141	127	µg/g
Africa	South Africa	Elsburg	Tailings	NA	4 881	164	µg/g
Africa	South Africa	French Rand	Tailings	NA	1 116	146	µg/g
Africa	South Africa	Harmony	Tailings	NA	180 116	102	µg/g
Africa	South Africa	Leeudoorn	Tailings	NA	2 740	164	µg/g
Africa	South Africa	Loraine	Tailings	NA	47 235	110	µg/g
Africa	South Africa	Randfontein	Tailings	NA	106 469	198	µg/g
Africa	South Africa	St Helena	Tailings	NA	78 595	129	µg/g
Africa	South Africa	St Helena (Beisa)	Tailings	NA	1 236	391	µg/g
Africa	South Africa	Virginia	Tailings	NA	21 242	203	µg/g
Africa	South Africa	Vlakfontein	Tailings	NA	18 201	173	µg/g
Africa	South Africa	West Driefontein	Tailings	NA	89 624	155	µg/g
Africa	South Africa	Western Areas	Tailings	NA	92 233	443	µg/g
Africa	South Africa	Zandpan	Tailings	NA	4 640	136	µg/g
Asia	Japan	Ningyo-toge	Tailings	NS	54 000	54 000	t
Asia	Kyrgyzstan	Mailuu Suu	Tailings	Tailing 2	NA	121–247	µg/g
Asia	Kyrgyzstan	Mailuu Suu	Tailings	Tailing 3	NA	210–606	µg/g
Asia	Kyrgyzstan	Mailuu Suu	Tailings	Tailing 4	NA	139–191	µg/g
Asia	Kyrgyzstan	Mailuu Suu	Tailings	Tailing 5	NA	128–182	µg/g
Asia	Kyrgyzstan	Mailuu Suu	Tailings	Tailing 7	NA	114–275	µg/g
Asia	Kyrgyzstan	Mailuu Suu	Tailings	Tailing 8	NA	129 ± 58	µg/g

Region	Country	Mine site	Waste type	Waste name	Waste mass (tonnes)	U in waste	Unit
Asia	Kyrgyzstan	Mailuu Suu	Tailings	Tailing 13	NA	200–353	µg/g
Asia	Kyrgyzstan	Mailuu Suu	Tailings	Tailing 14	NA	101–123	µg/g
Asia	Kyrgyzstan	Mailuu Suu	Tailings	Tailing 15	NA	128 ± 79	µg/g
Europe	Germany	Helmsdorf tailing disposal	Tailings	NA	NA	80	t
Europe	Germany	Heubachtal	Mine products and waste (unprocessed)	NA	300 000	2.7	µg/g
Europe	Germany	Mansfeld smelter	Metallurgical residues, slags	NA	50 000 000	50	µg/g
Europe	Germany	Wittichen 1	Mine products and waste (unprocessed)	NA	25 000	229	µg/g
Europe	Germany	Wittichen 2	Mine products and waste (unprocessed)	NA	5 000	20	µg/g
Europe	Greece	Ayia Paraskevi	Slag	NA	2 475 000	4.6	µg/g
Europe	Greece	Fougara	Slag	NA	NA	3.9	µg/g
Europe	Greece	Kavodokanos	Slag	NA	NA	3.5	µg/g
Europe	Greece	Kiprianos	Slag	NA	NA	2.5	µg/g
Europe	Greece	Komobil	Slag	NA	NA	6.6	µg/g
Europe	Greece	St Phillip (Kirki)	Mine waste	NA	180 000	7.5	µg/g
Europe	Portugal	Baixa Mine	Tailing	NA	NA	318	µg/g
North America	United States	Former Feed Materials Production Centre, Fernald, Ohio	NS	Silo 1 and 2	NA	28	tU
North America	United States	Unspecified Mine in New Mexico	Waste pile		NA	189	µg/g
South America	Brazil	Poços de Caldas (Minas Gerais)	Sludge		NA	0.25	% U_3O_8

TABLE 24. URANIUM MILL TAILINGS PILES BY CONTINENT AND COUNTRY AND THEIR REPORTED VOLUMES [39] (ND: NOT DOCUMENTED)

Continent	Country	Tailings volume ($x10^6$ m^3)
Africa	Gabon	ND
	Namibia	ND
	Zaire	ND
	South Africa	ND
North America	Canada	~ 30
	United States of America	120
South America	Argentina	ND
	Brazil	2.17
Australia	Australia	48.6
Asia	China	ND
	India	ND
	Japan	0.03
	Kazakhstan	209
	Kyrgyzstan	ND
	Uzbekistan	30
Europe	Bulgaria	18.5
	Czech Republic	46.8
	Estonia	8
	Finland	0.04
	France	47.3
	Germany	161
	Hungary	20.4
	Poland	0.114
	Portugal	3.5
	Romania	4.5
	Russian Federation	54.1
	Slovenia	0.7
	Spain	2.4
	Sweden	1
	Ukraine	130
TOTAL		938

TABLE 25. URANIUM MILL TAILINGS PILES BY CONTINENT AND COUNTRY AND THEIR REPORTED MASSES [178] (ND: NOT DOCUMENTED). THE MILL TAILINGS TONNAGE FOR SOUTH AFRICA HAS BEEN TAKEN FROM [128] AND FOR GERMANY FROM [179]

Continent	Country	Tailings mass (Mt)
Africa	Botswana	ND
	Chad	ND
	Democratic Republic of the Congo	ND
	Egypt	ND
	Gabon	6.5
	Mali	ND
	Mauritania	ND
	Namibia	350
	Niger	17.2
	South Africa	7 000
	United Republic of Tanzania	ND
	Zambia	ND
North America	Canada	202.13
	United States of America	235
	Mexico	NA
South America	Argentina	0.7
	Brazil	2.45
Australia	Australia	79
Asia	China	ND
	India	8
	Israel	ND
	Japan	0.054
	Kazakhstan	165
	Kyrgyzstan	32.3
	North Korea	ND
	Pakistan	ND
	Uzbekistan	60
Europe	Bulgaria	16
	Czech Republic	89
	Estonia	4
	Finland	0.04
	France	29.318
	Germany	240
	Hungary	20.4
	Poland	0.25
	Portugal	4
	Romania	6.8
	Russian Federation	56.85
	Slovenia	0.7
	Spain	1.408
	Sweden	1.5
	Ukraine	89.5
TOTAL		8 718.10

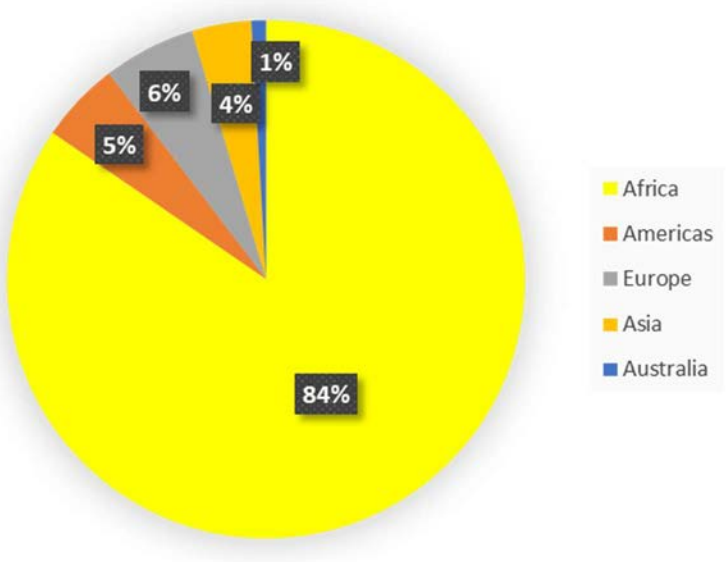

FIG. 18. Proportional distribution of total uranium mill tailings (8 718 Mt) generated up until 2019 across the various continents. Source data: [128, 178, 179]. The boundary between Europe and Asia is not clearly defined, Member States like the Russian Federation and Kazakhstan were only partially located in Europe in the past and hence were assigned to the Asian part only.

9. LIMITATIONS AND POSSIBLE SOLUTIONS OF URANIUM RECOVERY FROM WASTE

9.1. CHALLENGES TO THE ASSESSMENT OF (URANIUM) MINING RESIDUES

Geological mineral resources are commonly defined as natural concentrations or occurrences of materials that are, or may become, of potential economic interest due to their intrinsic properties. Mineral resources are further sub-classified, in order of increasing geological confidence, into inferred, indicated and measured categories [30]. By contrast, mineral reserves are valuable resources known to be economically and technically feasible to extract. Mineral reserves are also sub-classified, in order of increasing geological confidence as well as mining, metallurgical, environmental, legal, political, social and economic factors into probable and proven categories. Thus, sufficient information and level of confidence in geological knowledge as well as other considerations are required to make public statements about the potential extraction of uranium from mineral deposits. Furthermore, public reporting, i.e. reporting with respect to the needs of potential investors, of exploration results, mineral resources and ore reserves generally follows professional codes of practice (e.g. [180–183]). The viability of a potential project to rework mining residues not only depends on the scientific-technical aspects of the material as such, but also on the regulatory, legal, economic, and societal context and these aspects will be discussed in more detail in Section 9.3. The codes, such as JORC, consider this in specific sections, for example in the case of JORC in its extensive 'Table 1' [184]. For an operating mine these contextual aspects may be of less relevance, though feeding material back into the productive materials cycle may face obstacles in some jurisdictions once these materials have been declared as 'waste'.

Akin to mineral deposits, there needs be an increased knowledge and level of confidence in the properties and amounts of residues as well as technical, environmental, social and economic considerations to make 'public statements' about the likely potential of remining and reprocessing of the residues. Similar to the professional codes of practice for geological resources and reserves, uranium mining and milling wastes need to be further defined to indicate that they are potentially valuable, and for which reasonable prospects exist for eventual economic extraction once market conditions improve. Uranium waste may be characterised and classified as identified and undiscovered resources based on an increasing level of knowledge and confidence on geological knowledge (specifically in this case, physical, chemical and mineralogical properties) and quality and quantity of grade, tonnage and volume data. Such information on waste needs to be recognized and communicated using agreed assessment methods, classification frameworks and reporting standards, some of which already have provisions for this purpose (e.g. Clause 41 in JORC [180]).

In this section, available assessment methods, classification frameworks and reporting standards for uranium in wastes are compared with actual requirements for establishing reliable estimates of uranium inventories in mine waste. A particular focus is on uranium mill tailings. Such a gap analysis also provides insights into what activities could be undertaken to ensure uranium stocks in wastes are identified reliably and extracted in a responsible manner using state-of-the-art technologies.

9.2. URANIUM MILL TAILINGS REPOSITORIES

9.2.1. Assessment methods

Conventional and unconventional uranium resources are assessed and quantified using established geological, mineralogical, geochemical, metallurgical and technical tools. These tools rely on sampling and direct analysis or indirect measurement of ore properties to establish the concentration and distribution of recoverable uranium in mineral ores using three-dimensional modelling.

Similarly, quantitative and qualitative data are needed to establish a database and three-dimensional model of the uranium to be extracted from tailings. In particular, the mineralogy, geochemistry, volume and deposit geometry of the wastes have to be known, and continuities, boundaries and grade variability of different waste types within a tailings repository need to be established because they are crucial components of any project that aims to mine existing waste dumps [185]. Tailings in particular may display rapid changes in grain size or density due to sorting upon sedimentation or hydrocycloning. At or near the surface, sulfidic tailings may also be partially or completely oxidized due to oxygen ingress over time, resulting in a displacement of the uranium, particularly in older tailings. As a consequence, uranium mill tailings repositories commonly represent a heterogeneous sedimentary mass.

Prior to any remining, reprocessing and uranium extraction, a database on the chemical and physical properties of tailings and the spatial distribution of these properties needs to be established as per the reporting codes (refer to Section 9.3). This may also include information on their current status, land use, ownership and resulting accessibility and may distinguish tailings deposits largely left in their original state (including subsequent erosion), from those which have already been expensively remediated (e.g. through dewatering, reshaping and capping).

The assessment strategy would probably follow best the strategies that have been developed for the assessment of contaminated sites over the past few decades. These assessments are faced with a similar problem, namely the randomness (from a geological perspective) of the distribution of constituents in a volume of often unknown extent. These assessments, apart from establishing the overall properties of the contaminated volume, seek to identify, in particular, ‘hot spots’ of elevated contamination levels. In an analogous way, the assessment of mining residues seeks to establish areas of uranium concentration that are of interest for remining.

Since the late 1980s an iterative scheme for the assessment of contaminated sites became established as the most cost-effective approach, as it helps to focus project resources to the real problem (see also [186], p. 47). Broadly speaking this iterative approach begins with collating the available historical information, including anecdotical evidence from (former) employees, as a basis for planning non-invasive surveys using, for example, geophysical techniques. The analysis of historical aerial photographs (if available) and of operational records (if available) would be useful to identify the location, timing, and quantities of tailings accumulation or other materials.

9.2.1.1. Geophysics

Geophysical investigations can establish (or verify if detailed pre-emplacement surveys are available) the bottom topography of the tailings pond or residue heap. Geoelectrics, ground-penetrating radar (GPR) and/or hammer-seismic can also map out stratifications, if there is

sufficient contrast between the layers, e.g. [187]. They may help to identify, at least, whether other materials other than tailings have been deposited in the tailings pond. It is not uncommon to find drums, scrap etc. in the ponds, which can cause problems during drilling and last, but not least, the profile of the dam, as constructed, within a tailings pond can be delineated.

The geophysical investigations can be complemented with a traditional topographic survey or more advanced techniques, such as ground-based or air-borne LIDAR, to establish the topographical surface of the mining or milling residue deposit. These two data sets allow to construct a 3D-model of the volume of the deposit.

The geophysical survey will also help to identify the absence or presence of any bottom liners and fractures and faults in the underlying rocks, which may act as conduits for leaching solutions. The latter can be important for the decision-making on the most appropriate re-mining techniques. The absence of impermeable layers underneath the extractive wastes and the presence of faults will rule out, for instance, the application of heap-leaching techniques.

9.2.1.2. Sampling strategy

The results from these surveys help to establish a first sampling plan, ruling out areas that are not likely to be of interest. On tailings ponds and mining residue heaps etc., the sampling is likely to consist of drilling down and collecting samples at regular intervals from the cuttings or coring (preferred and if possible) to the bottom so that material for determining the uranium content and its matrix can be collected. A detailed sampling plan listing various requirements will be defined (e.g. number and spatial distribution of drillholes, sample locations, drilling/sampling technique, subsample types, sample sizes, sampling equipment, sample storage, analytical techniques, etc.). As noted earlier, the deposition of liquid tailings results in a grain-size fractionation away from the discharge point. For this reason, it may be important to understand with which grain-size fraction and which minerals the uranium is mainly associated with. The core/sample material collected can also be used to carry out leachability tests. The drill holes are arranged in an irregular triangular pattern, widely spaced at the beginning. Once the samples from the first campaign have been analysed, further campaigns can be undertaken using a closer spaced regular triangular pattern for areas that show higher concentrations in order to delineate better these areas. Cone-penetrometer tests or similar geotechnical investigations will help to determine the geotechnical properties of tailings and hence indicate the method of recovery or where stability problems might be expected.

9.2.1.3. Sampling

A wide variety of sampling and drilling techniques are available, each with different advantages and disadvantages and limitations. The sampling and drilling technique have to be compatible with the questions to be answered. Certain drilling techniques can be fast, do not introduce contaminants or lead to cross-contamination, but can lead to grain size fractionation and will disturb geochemical equilibria. Most drilling techniques that disaggregate the material at the drill-head (e.g. augering, reverse circulation (RC) drilling and its variants fall into this category). Stratigraphically and geochemically intact samples can only be obtained by one of the core-drilling techniques. Test trenching is an option in stable materials but can generate considerable amounts of ‘waste’ that need to be managed safely. It allows, however, to retrieve intact samples. The procedures for mixing and dividing samples with a view to reduce the amount of analyses to be undertaken, while maintaining representativeness has been intensively studied and some countries have issued guidelines for that purpose, e.g. [188].

Porewater sampling and the in-situ determination of easily changed parameters, such as pH and Eh, will help to understand the geochemical evolution of the tailings and whether residual uranium may have been solubilized and migrated through the tailings mass. A good picture of the hydrogeochemical environment, together with the assessment of the physical boundary conditions and the mineralogy, will allow a decision as to whether any in situ techniques, in particular bioleaching (see section 9.2.1.5. Assaying), might be an option for recovery.

9.2.1.4. Representativeness of samples

A difficult to resolve problem is the development of drilling/sampling strategies that are statistically representative. When exploring primary mineralizations the layout of a sampling or drilling grid is iteratively developed as the knowledge of the geology grows. Knowledge of geological features such as bedding, faulting and folding allows to make certain predictions as to the 3D structure of the mineral occurrence. Although certain stratifications due to the method of deposition will be present in tailings and other residues, their lateral extent may vary unpredictably due to changes of discharge points or later redistribution by earth moving machinery or mass slides that have occurred after deposition. One can look at the strategies used in contaminated site assessment, where contamination also may be distributed more or less randomly and for which guidelines have been developed e.g. in various European countries [188–192].

Related to this is the strategy for representative sampling. A number of European countries faced with the problem of remediating mining and industrial residues have developed guidelines for this, though in general the total volume to be sampled is typically much smaller and solid mining residues tend to more inhomogeneous than tailings, e.g. [188]. Minnitt et al. [191, 192] have tried to capture mathematically the 'fundamental sampling error' (such as the size of the sample), which is important among the ten factors that can contribute to the non-representativeness of samples as identified by Pitard [193]:

(1) In situ Nugget Effect;
(2) Fundamental sampling error;
(3) Grouping and segregation errors;
(4) Long-range heterogeneity (quality) fluctuation error (shifts and trends);
(5) Long-range periodic heterogeneity (quality) fluctuation error (cycles);
(6) Increment delimitation error;
(7) Incremental extraction error;
(8) Weighing error;
(9) Preparation error;
(10) Analytical error.

Abzalov and Newman [190] describe an example of developing such strategies and protocols to minimise these errors for a tailings pond. However, there are still no agreed upon assessment methods and procedures for many parameters, for example: (i) how to sample wet unconsolidated and dry consolidated tailings; (ii) how to sample different tailings types (e.g. thickened, paste and dry stacked tailings); (iii) what sampling techniques are appropriate; and (iv) how to consider tailings that have changed over time and have oxidized. The challenge remains to outline best practices and agreed assessment methods for uranium mill tailings deposits for remining and reprocessing.

Based on field investigations a 3D-model of the deposit and the distribution of uranium minerals therein can be developed [190, 194, 195]. If the model building is integrated into the iterative site investigation process, it can be progressively refined and also used to guide the refinement of the sampling grid in those areas of particular interest. However, there does not appear to be a universally accepted model for estimating the resources uncertainty, as may be required by the reporting codes (e.g. JORC, SAMREC, PERC). Relevant geostatistical methods include [194, 196]:

— Kriging variance;
— Estimates of the kriging efficiency;
— Estimation variance of the resource blocks using;
— Conditional simulation;
— Probability estimates;
— Extension variances.

9.2.1.5. Assaying

Common methods include, after appropriate sample preparation, to digest the sample in various stages in increasingly strong and oxidizing acids or dissolution after peroxide fusion. The staged dissolution gives an idea of how easy or difficult extraction may be, but as it is known a priori, that the uranium mill tailings will mainly contain those uranium minerals that were refractory in the previous milling process, the assay may need to be tailored to this aspect. So, in addition to determining the total uranium content, also some (bio)leaching tests may be required.

9.2.1.6. Environmental impacts

It is understood that these site investigations should ideally not unduly compromise closure and remediation measures undertaken for the mining and milling residues. Care has to be taken not to penetrate bottom liners (if in place) and any covers that have been drilled through need to be sealed again. These precautions are to be taken in case the repository is considered not economic to remine, and its long-term physical and chemical stability has to be ensured.

9.2.2. Recovery technologies

9.2.2.1. Geometallurgical assessment of tailings

The mineralogy of uranium deposits influences the possible leaching, extraction and recovery of uranium from its ores [105, 197]. Consequently, mineralogical and geometallurgical assessments are recommended, even at an early stage of mine development, to select, design and optimize a suitable extraction scheme. Given the fact that much of the uranium in tailings will be bound in mineral phases that were refractory in the previous processing steps, it may be advisable, as has been pointed out earlier, to include all previous and current geometallurgical assessments and chemical assays as part of the site assessment. This is mandated in any case by the reporting codes (JORC, SAMREC, PERC).

Geometallurgical investigations are studies that combine mineralogical and geostatistical information with mineral processing and extractive metallurgical data investigations to create a spatially predictive model for the feed to the mineral processing plants. Such activities are pursued in the metal mining industry to design, optimize and monitor mineral processing plants. Many scientific publications on advanced recovery techniques have not investigated in detail

the ore mineralogy or, this was not reported in the respective papers, so that their applicability to refractory uranium minerals is difficult to judge.

Similarly, geometallurgical information is needed for tailings repositories. Tailings repositories contain those uranium fractions of the milled and treated ore that could not be recovered using the applied milling and metallurgical techniques. The recovery of uranium from ores depends on many factors including the uranium mineralogy, with many modern processing plants achieving on average 95% or better for uranium ores containing uraninite, carnotite or coffinite. Such a high recovery rate is due to the fact that many uranium minerals, such as uraninite, carnotite and coffinite, are readily soluble in acidic or alkaline solutions (particularly in the presence of an oxidant such as pyrolusite, oxygen or hydrogen peroxide). By contrast, other uranium minerals, such betafite, brannerite and davidite, have a very low solubility in acidic or alkaline solutions, and brannerite-rich ores are seen as refractory ores (e.g. Elkon deposit, Russian Federation) [86, 198]. These uranium minerals are only soluble under very aggressive conditions (i.e. extremely strong acids, high temperatures and/or high pressures). Consequently, the leaching efficiency of uranium in processing plants is influenced by the uranium mineralogy and refractory uranium minerals (e.g. brannerite, betafite, uranophane) typically report to the uranium mill tailings. Also uranium mill tailings that derive from the processing of silica-rich ores, commonly contain colloidal silica, which can be an issue in hydrometallurgical circuits [199]. Thus, the mineralogical composition of uranium mill tailings can cause major impediments to the extraction and recovery of uranium using conventional acid or alkaline leaching technologies.

9.2.2.2. Novel extraction procedures

Extraction of uranium from tailings by sulfuric acid leaching with oxidants represents a possible recovery route [200]. However, some uranium ores contain multiple oxide type uranium minerals and are generally difficult to leach. Tailings generated from such refractory uranium ores will preferentially contain proportionately more refractory uranium ore minerals than the ore from which they are derived. Minerals such as brannerite and betafite are refractory and would require leach processes with high temperatures and high reagent concentrations for effective uranium leaching to take place [201, 202]. Though leaching under autoclave conditions is quite feasible [203], the energetic requirement would need to be critically assessed and put into the overall context of the energy and CO_2-footprint of nuclear energy systems. The same applies to proposed leaching processes using more aggressive fluids and changing the mineralogy of the ore by a preceding calcination step, e.g. [204] and references therein. The life-cycle energy requirement and CO_2-footprint as well as risks of using certain reactants needs to be taken into consideration. It should be noted also that most of the proposed processes reported have not actually been tested at an industrial scale. Therefore, innovative methods for the extraction and recovery of uranium from mill tailings are needed, particularly for refractory wastes [205].

In fact, novel extraction procedures have already been proposed for the recovery of uranium from low-grade uranium ores, tailings and mine waters. For example, resin-in-pulp (RIP) technologies could be used for the treatment of tailings [206, 207]. With the right resin, the RIP technology can also be used in alkaline systems, for which e.g. [208] received a Russian patent. Here, remined tailings would be leached to solubilize uranium, followed by the resin-in-pulp treatment, solvent extraction and uranium recovery [209]. The RIP or solvent extraction processes do not actually solubilize the uranium from the refractory ore, but prevent its re-reduction and reprecipitation or sorption, thus facilitating its separation from the suspension (pulp). These techniques can be combined, as long as resins or solvents are compatible, with a

variety of solubilisation processes. Also, carrying out the leaching in steps with different reactants and reaction times can reduce the 'poisoning' effect of certain minerals or elements in the gangue [210].

9.2.2.3. Bioleaching

Bioleaching procedures for the extraction of residual metals including uranium contained in sulfide-rich tailings wastes is another possibility [211, 212]. Heap/dump, in stope and in situ leaching have already been applied to uranium ores [213], and the biohydrometallurgical technology may also be adapted to uranium mill tailings. According to [214–216], most uranium minerals, with the exception of coffinite, respond well to bioleaching (Table 26).

TABLE 26: RESPONSE OF URANIUM MINERALS TO BIOLEACHING [212] AFTER [214–216]

Mineral	Formula	Relative response to bioleaching
Uraninite	UO_2	easy
Brannerite	$(U,Ca,Ce)(Ti,Fe)_2O_6$	easy
Davidite	$(Fe,Ce,U)2(Ti,Fe,V,Cr)_5O_2$	easy
Coffinite	$U[(SiO_4),(OH)_4]$	difficult
Autunite	$Ca[(UO_2)(PO_4)]_2 \cdot 11\ H_2O$	easy
Torbernite	$Cu(UO_2)_2(PO_4)_2 \cdot 12\ H_2O$	easy
Carnotite	$K_2[UO_2\|VO_4]_2 \cdot 3\ H_2O$	variable
Uranospilite	$[(UO2)_6(SO_4)O_2(OH)_6] \cdot 14\ H_2O$	easy

The feasibility of bioleaching depends on the gangue mineralogy as well as the geohydraulic conditions, if undertaken in situ. Tailings deposited in a pond may not be sufficiently permeable for this purpose. Acid consuming gangue minerals, such as carbonates, will impede the establishment of suitable conditions, while acid producing minerals, such as sulfides, will foster the establishment of suitable microbial communities. The presence of sulfides is a major factor in the establishment of bioleaching environments containing sulfur-oxidising bacteria (e.g. Thiobacillus spec.). However, other nutrients may need to be added [212]. In consequence, a wide variety of operational variables (e.g. solid/solution ratio, temperature, addition of sulfides or nutrients, etc. may need to be adjusted simultaneously, which can lead to a multitude of preliminary experiments and evaluation methods for these multi-dimensional approaches, e.g. [217].

It is, therefore, necessary to also investigate the presence of suitable microbial communities that otherwise may need to be introduced, e.g. [218]. Certain microbial communities thrive in anaerobic conditions, while others require aerobic conditions. The presence and role of fungi may also need to be investigated. Metalloid compounds in the mining residues, such as arsenates can also be toxic to many microbial species and thus impede the establishment of bioleaching systems. Table 27 gives an overview of the various species that can be used in bio-leaching, the leaching mechanism and the respective environmental requirements.

TABLE 27. MICROORGANISMS USEFUL FOR THE BIOLEACHING OF URANIUM ORES [212]

Organism	Biological role	Aeration	Characteristics	Temperature	pH
A. thiooxidans	S^0, thiosulfate and tetrathionate oxidation	aerobe	chemolithotroph	25°C – 30°C	0.5–3.0
A. ferrooxidans	Fe^{+2}, S^0, U^{+4}, thiosulfate, S^{-2} oxidation	aerobe	chemolithotroph	25°C – 35°C	1.5–2.5
Thiobacillus acidophilus	S^0, organic compounds oxidation	aerobe	facultative autotroph	25°C – 30°C	2.5–5.0
Leptospirillum ferrooxidans	Fe^{+2}, pyrite oxidation	aerobe	obligate chemolithotroph	20°C – 40°C	1.5–4.5
Sulfolobus thermosulfidooxidans	Fe^{+2}, S^0, S^{-2} oxidation	aerobe	facultative autotroph	20°C – 50°C	1.5–3.0
Sulfolobus acidocaldarius	Fe^{+2}, S^0 oxidation	aerobe	facultative autotroph	55°C – 85°C	2.0–5.0
Acidianus brierleyi	S^0 oxidation	aerobe	facultative chemolithotroph	45°C – 70°C	1.0–6.0
Desulfovibrio desulfurican, and a few other SRBs, Geobacter	Removal of dissolved U from effluents and reduction of U^{+6} to U^{+4}	anaerobe	heterotroph	25°C – 30°C	4–7
Pseudomonas spp.	Intracellular accumulation of U and other metals oxidation	aerobe	heterotroph	25°C – 30°C	6–8.5
Penicillium spp.	Accumulation of U, The, Ra in the cell wall, oxidation	aerobe	heterotroph	15°C – 30°C	5–7
Rhizopus spp.	Accumulation of U, The, Ra in the cell wall, oxidation	aerobe	heterotroph	24°C – 27°C	4–6

Depending on the species, most microbial species have a certain preferred temperature window, mostly above 20°C and below 40°C, in which they thrive. Some species are also thermophilic. This can make the operation of heap-leach systems intermittent in regions with temperate or even boreal climate. On the other hand, sulfide oxidation reactions within mine residues are exothermic, so that microbial communities to a certain degree can create a suitable environment for themselves, as long as heat dissipation due to infiltrating meteoric waters and gas migration remains sufficiently low.

The use of fungi and their metabolic products appear to be another interesting route [219] and references therein. However, few authors seem to have investigated in detail the mineralogy of the uranium ores used, or at least this was not reported in many papers reviewed for this study. This lack of fundamental information makes the drawing of general conclusions difficult, thus a case by case assessment will be needed.

Uranium in ore minerals is tetravalent (Table 26), but it needs to be oxidised in order to become soluble, which then implies that relevant microbial communities must be aerobic (see Table 27 for an overview of suitable species). Therefore, oxic conditions must be maintained throughout the system to ensure the elution of uranium. Bacterial oxidation of ferrous iron helps to maintain these oxic conditions. The routes of bacterial oxidation of uranium, pyrite and ferrous iron, however, are complex [220]. Mining residues that do not contain sulfide minerals may need to have these added to provide the necessary energy source.

While anaerobic environments in general favour the immobilization of uranium in its tetravalent state, anaerobic oxidation of U^{4+} to U^{6+} with nitrate as electron acceptor may be a route of interest in certain types of environments [221]. Nitrate would also act as a complexing agent to enhance the solubility of uranium.

Francis and Nancharaiah [222] reviewed various processes that can lead to the solubilization of uranium. Namely heterotrophic bacteria and fungi excrete metabolic and decomposition products that can act as complexing and chelating agents, thus increasing the solubility of uranium and lowering the pH in the pore solutions. However, after an initial interest in study of these mechanisms in the 1970s, it seems to have waned. Perhaps at the time conventional milling processes were considered sufficiently effective and cheap, while bioremediation by extraction was not yet on the agenda. Also, later bioremediation studies were more focused on immobilization, rather than solubilization.

Francis and Nancharaiah [222] also reviewed the use of citric acid to extract uranium. The uranyl ions form stable mono-, bi- and tridentate and binuclear complexes with citric acid. While citric acid and mono- and bidentate complexes are biodegradable, tridentate complexes are not, but decompose quickly in sunlight. These properties can be used to extract and separate the uranium and then recover it from the complexes. The advantage of citrate extraction over acid or alkaline extraction is that the system is buffered and does not result in disposal problems with strong acids or alkaline solutions. As citrates complexes solubilize a variety of other divalent metals, this can be of added economic interest in order to add value to the treatment of tailings beyond the recovery of uranium.

All these factors will determine the feasibility and kinetics of bioleaching systems and need to be investigated in the context of assessing the feasibility of uranium bioextraction from residues containing refractory uranium minerals.

The kinetics of the bioleaching process also depend on the location of the precipitation of reaction products such as ferric oxyhydroxides and jarosite that may cover the surface of uranium minerals or of the pyrite that is an important energy source for the active microbial communities. Heap-leaching pads may experience a decline in productivity through the formation of secondary minerals, which would be otherwise a desirable effect in a remediation context. Fixed-film or fluidized-bed reactors can be a solution under such circumstances but require a more substantial mill infrastructure and therefore capital expenditure (CAPEX).

The actual role microbial communities play in leaching processes in situ is still not very well understood [212], requiring further preliminary laboratory experiments and in situ tests to establish the treatability. It may be noted that the majority of uranium bioleaching publications of recent years seem to originate from China and the Middle East, which may reflect the level of interest in new nuclear energy systems and the accessibility to primary uranium ores.

In summary, one may note that the systems discussed seem to offer interesting perspectives for the recovery of uranium, even from minerals that are considered refractory in most inorganic processes but have not been tested yet in industrial-scale applications. For a given case, one may need to develop the right procedure with autochthonous or seed microbiology and the appropriate nutrients and growth conditions in situ or ex situ to determine optimum system conditions for each case.

9.2.2.4. Stope- and heap-leaching versus reactors

As noted above the operational performance of bioleaching systems depends on many different factors, notably the hydrodynamics of the system and the kinetics of the reactions. It is important that as much as possible of the ore surface is exposed to the percolating fluids and that these fluids can transport oxygen (in aerobic systems) and nutrients to the ore surfaces and carry away dissolved uranium. Secondary mineral precipitation on ore surfaces would also inhibit the system. This can mean that stope- and heap-leaching may experience a significant decline in performance over time, as pores clog up due to filtration effects and secondary mineral formation. Providing for complexing agents that suppress secondary mineral formation and keep uranium in solution would ameliorate this situation. In general, agitation of the tailings or mining residues would continuously expose fresh mineral surfaces and prevent clogging. It would also ensure an efficient mixing of solids and (fresh) solutions, as static system dead-ends and preferential flow paths may develop. Agitation also ensures that acidic and oxic conditions are maintained across the whole volume, to ensure that the uranium is maintained in the hexavalent state and to prevent reprecipitation in dead-ends etc.

Pulping the materials in stirred tanks or fluidized-bed reactors leads to faster and more complete uranium removal, albeit at higher CAPEX and operational expenditures compared to stope- or heap-leaching. Treating the tailings in a reactor would also allow to extract the solubilised uranium into a non-aqueous phase for separation. One may note, however, that the CO_2- and energy footprint of such industrial systems is likely to be higher than that of stope- or heap-leaching arrangements due to a more sophisticated infrastructure and the need to move the tailings masses into and out of the reactors. Where such operations would take place in (sub)tropical environments, some of the operational energy requirements could be met with decentralized photovoltaic or thermal solar systems (see e.g. [147]), rather than fossil fuels, probably resulting in lower CO_2-footprints. Minimizing the CO_2-footprint and life-cycle energy requirements of nuclear fuel production is a major discourse in comparing the environmental impacts from different energy conversion systems in the public debate [62].

9.2.2.5. Extraction of co- and by-products

Conventional and unconventional uranium resources may be enriched in other metal minerals [81]. Therefore, the extraction of co- and by-products other than uranium from mill tailings has been proposed for some time. For example, uranium mill tailings from the United States of America containing 0.33% V_2O_5 and 0.045% U_3O_8 were acid leached, with recoveries of 65% for vanadium and 78% for uranium [223]. Also, uranium mill tailings from Cluff Lake (Canada) have been treated with cyanide to recover gold [43]. Most recently, the extraction of critical metals (V, Bi, Sb, Co) as by-products has been suggested for the Eureka uranium deposit, Portugal [224].

In the context of ensuring supply security and striving for more independence from monopolistic suppliers, the EU is actively searching for domestic REE and other metal resources that are deemed critical [97]. Particularly, REE mineralisation may also contain

uranium and thorium. While these radioactive elements are considered undesirable 'waste' in various countries, their inclusion into the nuclear fuel cycle will provide added value to the respective extractive operations. A challenge is the decomposition of minerals that may be refractive in traditional milling processes and the subsequent separation of the dissolved metals. While it is possible to decompose virtually every mineral with aggressive procedures (e.g. pressurised digestion in hydrofluoric acid) at the laboratory scale, such procedures will be impractical or it may be difficult to obtain operating licenses due to the inherent risk of high-pressure, high-temperature plants containing aggressive fluids. Another factor to consider is the life-time CO_2-footprint per unit recovered metal that increases significantly with more aggressive procedures. Considering this background, novel extraction technologies (e.g. resin-in-pulp, biosorption, bioleaching) are needed, that will have to be tailored to the specific mineral assembly and the targeted metals. Considering also the strategic supply and sustainability aspects for certain countries or associations of countries (such as the EU), purely market economic consideration may be overridden in their interest. Thus, total extraction of the metal value may be of interest in a sustainability context, although no immediate market may exist for certain metals. While a hundred years ago perhaps only a handful of elements were used, we now make use of virtually the whole periodic table [17]. This is likely to lead to the increased availability of uranium as a by-product with little extra energy expenditure. A multi-disciplinary approach of chemistry, extractive metallurgy, engineering and biotechnology is required to realize this ambition.

9.2.2.6. Extraction from liquid effluents

Mines and mining residues often give rise to drainage waters, particularly when they have not been properly closed and remediated. Also, during the period of mine flooding, such drainage will arise. Typically, such drainage is collected and treated in order to bring contaminant levels to acceptable levels and other properties, such as pH, to levels fit for release into surface water courses. In many cases the drainage is acidic (acid rock or acid mine drainage, ARD/AMD) due to pyrite oxidation and upon neutralization most of the metals, including the uranium, precipitate or co-precipitate. Otherwise, the effluents can be subject to the same procedures of metal separation as used in the milling process, for instance ion-exchange, solvent extraction, and other methods. Currently, several tonnes of uranium are extracted in the context of ongoing uranium mine remediation projects in e.g. the Czech Republic, Hungary and France (see the OECD-NEA/IAEA 'Red Book' for data [30]).

However, in most cases the formation of ARD or AMD is undesirable and considered a transition phase to more stable closure conditions. Remediation aims to prevent the formation of ARD by reducing the vector, namely water percolation through mining residues e.g. by capping. Similarly, AMD formation will be reduced by limiting the access of oxygen to open mine workings.

There are examples, however, where ARD formation has been fostered in order to accelerate the removal of reactive pyrite and the metal value associated with it as part of a remediation programme [225]. This accelerated pyrite oxidation uses the same concept as the heap leaching discussed earlier.

9.3. CLASSIFICATION FRAMEWORKS AND REPORTING STANDARDS

9.3.1. Implications of established reporting standards

Existing classification frameworks (CRIRSCO, Committee for Mineral Reserves International Reporting Standards) and the regional standards, such as JORC [180], NI43-101 [181], SAMREC [182], or PERC [183], allow the classification of tailings as mineral resources and reserves and are perceived as applicable to mine waste repositories and their contained uranium waste stocks [226]. Geological mineral commodities are categorized into resources and reserves, using national and international classification frameworks and reporting standards. These classification schemes require a solid knowledge of geological continuities, ore boundaries, deposit geometry, and grade variability. Geological, geophysical and geochemical data thereby allow certain generalizations and predictions, extrapolation and interpolation between data points, and three-dimensional modelling of ore and rock occurrences. However, there is still a lack of agreed assessments methods for tailings as has been discussed previously in this document. In addition, a wide range of 'modifying factors' are taken into consideration that pertain to the actual exploitability of the mineralization, notwithstanding the economic value, such as the feasibility of extraction from the mined ore (ore mineralogy), or the regulatory and the socio-political context.

9.3.1.1. Applicability of reporting standards

Mining and milling residues as potential resources are specifically referred to in Clause 41 of JORC [180], "*The term 'Mineral Resource' covers mineralization, including dumps and tailings, which has been identified and estimated through exploration and sampling and within which Ore Reserves may be defined by the consideration and application of the Modifying Factors.*", and Section 9 and 16 of PERC [183], 'Reporting of Mineralized Fill, Pillars, Low Grade Mineralization, Stockpiles, Dumps and Tailings'.

9.3.1.2. Modifying factors

While the reworking of residues as part of an on-going mining and milling operation may not pose particular problems from a regulatory and social licensing perspective, the situation for historic residues is more akin to opening a new operation. The socio-political and societal context will be a decisive factor, as (environmental) regulators and local or regional stakeholders may not be in favour of such operations. At least in Europe, many such residues tend to be close to settlements or settlements have encroached upon such mining and milling residues. In some areas, there may also be cultural heritage issues to consider, as both residues and remaining mining infrastructure may have been declared as protected sites under national regulations or international conventions (e.g. the UNESCO World Heritage Centre — https://whc.unesco.org, which is generally averse to extractive activities).

9.3.1.3. Assessment methods

JORC [180] points out that "*where untested practices are applied in the determination of reasonable prospects, the use of the proposed practices for reporting of the Mineral Resource must be justified by the Competent Person in the Public Report*." This can pose certain challenges, as a combination of methods are required to assess residues that are less common in the assessment of primary mineralization. Scoping and feasibility studies using an iterative approach will be particularly important. Table 1 in [180] and comparable sections in the other resource code reporting schemes list a wide range of criteria that need to be fulfilled. However,

in the context of mining and milling residues, these need to be adapted to this special situation. Therefore, the competent person will need to explain in detail what methods had been used and why.

9.3.1.4. Competent person

The expert signing off the reporting for the mineralization, i.e. the 'competent person', plays a key role in the process and is subject to a range of requirements in order to qualify as 'competent'. While the assessment of tailings and other residues from still operating mines may not pose particular difficulties and can be undertaken, in particular with existing operational records, the situation may be different for historic residues. Here competences akin to those required for the assessment of contaminated sites may be required. To this end the 'competent person' may require the assistance of suitably qualified experts, who can demonstrate their competences through their accreditation with a suitable national body, e.g. in Germany the accreditation as 'Altlastensachverständiger' (i.e. expert on legacy sites). These experts would be trained in the development of site investigation and sampling programmes for industrial and other residues.

9.3.2. Uranium mill tailings

Geological, geophysical and geochemical data for primary mineralization allow certain generalizations and predictions, extrapolation and interpolation between data points, and three-dimensional modelling of ore and rock occurrences. As has been pointed out earlier, these geological models are not applicable to mining and milling residues. Here, information on waste homogeneity or heterogeneity, waste continuities, waste boundaries, dump geometry, and grade variability is needed to establish a database and three-dimensional model of the uranium to be extracted. Homogeneity and waste material continuities in tailings dumps cannot be assumed and, unlike geological lithologies, continuities are impossible to predict due to random waste disposal, grain-size separation processes during dumping, settling and slumping, and post-depositional oxidation processes within the repository. Therefore, tailings repositories may be much more heterogeneous in their physical and chemical properties than their corresponding geological ores.

Considering the challenges of creating a three-dimensional model of uranium in waste dumps, any reported potential economic quantity and quality of uranium in waste repositories may be perceived conceptual in nature. In some cases, there could be insufficient information to estimate a uranium resource, and it may be uncertain, if further exploration will result in a feasible estimation of a uranium resource. Hence in public statements, it is currently more appropriate when describing the activities related to remining and reprocessing projects of uranium mine wastes to refer to these as 'exploration targets', with ranges given for tonnages and grades.

Notwithstanding these difficulties, it may be possible to determine upper limits of total uranium content in the tailings based on knowledge of the ore grade, mill output and/or recovery rates. This could make it possible to classify many tailings ponds as probable reserves, if such data are available.

9.3.3. Liquid waste streams and mine waters

As noted before, liquid waste streams and mine waters of conventional and unconventional uranium ores may contain elevated uranium concentrations. Economic extractions of such dissolved uranium require an assessment of available resources and reserves. Australian JORC [180] and Canadian CIM [227] guidelines are available for resource and reserve estimation of brines, from which valuable elements in solution may be economically recovered. These procedures are primarily meant for the evaluation of lithium brines and they do not consider key parameters that control the mobility and concentration of uranium in mine waters (e.g. pH, Eh, presence of complexing agents). Therefore, the existing guidelines cannot be simply applied to uranium-bearing liquid waste streams and mine waters.

The main issue will be that the vector for acidification and dissolution, the infiltrating oxygenated meteoric water, is not normally controlled and is subject to climatic changes. The solubilization of uranium and other metals of interest depends on the flowrates, the water balance and the resulting hydrogeochemical conditions. Therefore, the 'production' rate will be difficult to predict. As already noted, in many instances the production of ARD and AMD is a transitory phenomenon that remediation actions aim to end. Thus, any remediation strategies will have to be considered as modifying factors.

Considering these caveats, the JORC [180] and CIM [227] guidelines could be applied to such situations, considering fluid properties (e.g. pH, Eh) and a comprehensive understanding of the geohydrological and geochemical regime as a basis for elaborating modifying factors (e.g. flow rates through mines and mine waste dumps) relevant for uranium-bearing effluent and water. Otherwise, an application of the brine guidelines in their present format could result in under- or overstating the potential of uranium-bearing liquid waste streams and waters.

10. SUSTAINABLE URANIUM MINING

Current mining operations manage mine waste based on linear economy thinking ('*take-make-dispose*') [228]. As a result, there are large tonnages of mine wastes disposed with various sources providing insights into the scale of waste production [229]. The current extractive mine waste problem could be an opportunity to recover raw materials and to help the mining industry to move towards the circular economy thinking ('*make-use-return*'). For the development of a circular economy model in mining, the extraction of mineral resources needs to be optimized and mine wastes could be used as raw material resource. Indeed, the European Extractive Waste Directive [35] requires operators to explore further uses of residues, before these can be declared waste and deposited.

While some authors state that the mining industry is apart and excluded from restorative circular loops (Fig. 2) [228], the efforts made by mining companies' contributions to a circular economy have been overlooked. For example, there have been approximately 75 major tailings remining projects that aim to extract gold, diamonds and copper [230]. These remining activities do not only provide mineral resources, but they also conserve finite mineral resources and reduce environmental impacts of waste repositories and mine sites. Therefore, in this section, the potential role of the uranium mining industry in a circular economy and the contribution of uranium recovery from mine wastes to the United Nations Sustainable Development Goals are explored.

10.1. THE ROLE OF THE URANIUM MINING INDUSTRY IN A CIRCULAR ECONOMY

For remining and reprocessing projects of uranium-bearing mine wastes to occur, an overall approach needs to be tailored to be appropriate for each uranium mine site. Such an activity would include performing resource assessments of existing uranium waste impoundments as well as changes to operating uranium mills with the aim of minimizing tailings production and optimizing uranium extraction. In particular, uranium mill tailings offer significant cost savings and resource opportunities. These wastes have already been crushed and ground, reducing the reprocessing costs of any further extraction activities.

Sensor-based sorting, upgrading existing comminution facilities, and improving mineral beneficiation plants may help to minimize tailings production. Also, if reprocessing of previously generated tailings is being pursued, then long-term bioleaching of spent heap leach ores or tailings impoundments may be an option (Fig. 8). In this case, the recovery of uranium from waste repositories can be pursued through the installation of impermeable liners at the base of the waste impoundments. Spent heap leach piles and uranium mill tailings impoundments could be considered low-grade and very low-grade uranium ore resources, respectively (Fig. 18). Both of these approaches, waste reduction and waste reprocessing, would help to conserve uranium ore reserves and produce a more sustainable uranium mining industry in a circular economy that generates economic profits, minimizes waste and has less impact on the environment.

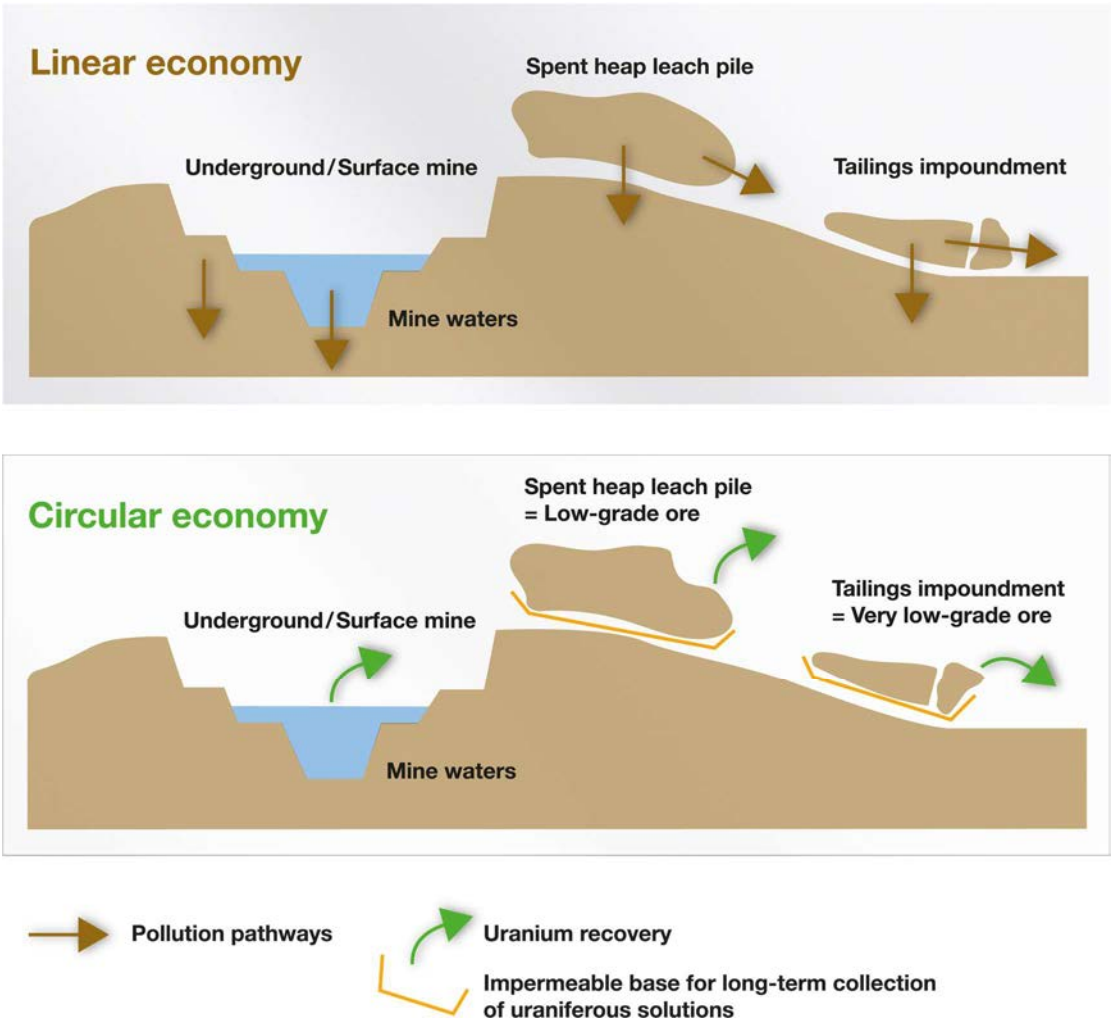

FIG. 18. Simplified cross-sections of an underground/surface uranium mine and its spent heap leach and tailings impoundment. In a linear economy, waste management potentially leads to the pollution of environmental media. By contrast, waste management in a circular economy aims to minimize wastes and to recover uranium from waste repositories through the installation of impermeable liners at the base of the waste impoundments, adapted from [231, 232].

10.2. CONTRIBUTION OF URANIUM RECOVERY FROM WASTES TO THE UNITED NATIONS SUSTAINABLE DEVELOPMENT GOALS

The Sustainable Development Goals (SDGs) and the 2030 Agenda for Sustainable Development represent the world's plan of action for social inclusion, environmental sustainability and economic development [233]. Within this framework there are opportunities for mining companies to contribute to the achievement of the Sustainable Development Goals. In particular, remining of uranium waste and reprocessing of mine wastes for uranium recovery are aligned with SDG12 (responsible consumption and production). Through the recovery of uranium from mine wastes, the needs of nuclear energy are supported and, reprocessed 'cleaned' wastes could be transformed into valuable residues for other purposes and industries (e.g. clay-rich tailings may become raw materials for the building industry).

11. CONCLUSIONS

11.1. KEY FINDINGS

A number of key findings were identified. These findings relate to the individual waste types and general recovery principles:

(1) Uranium has been recovered from phosphorite ores and from copper leach solutions since the 1950s and 1960s, respectively. Also, sorption and ion exchange technologies have been used to extract uranium from mine waters since the 1970s and, reprocessing of uranium mill tailings has been pursued since the 1980s. Thus, uranium mine wastes have been and still are a portion of the total world uranium supply, albeit at very low levels currently amounting to <50 t uranium/year [30];

(2) Uranium ores of the same ore deposit type have similar geochemical compositions, similar ore and gangue minerals and consequently similar kinds of wastes. Mining of the same ore deposit type produces comparable waste types. Hence, uranium mine waste sites are best classified using the IAEA classification system for uranium ore deposits;

(3) In the past and today, the largest masses of uranium-bearing mine waste has been and still is produced at those mines that extract conventional uranium resources and use traditional open pit mining and hydrometallurgical extraction methods. Uranium mill tailings thereby represent the largest waste fraction, compared to the mined uranium ores with relatively low uranium concentrations;

(4) Metallurgical accounting allows for an assessment of uranium quantities in mill tailings at a single uranium mill, as long as the necessary mass flow data are available. Such metallurgical accounting indicates that the global mass of uranium mill tailings contains at least between 0.87 to 1.74 Mt U_3O_8;

(5) Uranium is also present in significant concentrations in spent heap leach piles and drainage treatment sludges of conventionally mined uranium resources. In addition, a multitude of metallurgical residues from unconventional uranium resources are known to contain elevated uranium concentrations, including bauxite residue, metallurgical tin slags, lignite fly ash, phosphoric acid waste streams, and process liquids of copper ores;

(6) Mine waters of active and historical uranium mines may contain uranium concentrations that are of possible economic interest and could be extractable using established technologies;

(7) The potential of solid mine wastes, mine waters and liquors to yield uranium stocks is highly variable. This variability is due to the fact that each mine generates its own unique waste because: (a) each mine has different criteria for distinguishing between ore and waste; (b) there are mineralogical and geochemical differences in the mined ore and waste; and (c) there is a great diversity of applied mining, mineral processing and metallurgical methods and practices. Therefore, every waste type and each waste site will require their own waste characterization and appraisal for potential uranium recovery;

(8) Geometallurgical assessments of mine waste impoundments are crucial components of any project that aims to remine existing waste impoundments for uranium;

(9) Uranium ores and wastes rich in refractory uranium minerals require new processing and metallurgical extraction technologies. This particularly applies to ores and wastes rich in brannerite, betafite and uranophane;

(10) There is a strong need for innovative recovery technologies that are capable to solubilise refractory ores and keep the dissolved uranium in solution so that it can be separated out. At the same time less hazardous, less aggressive and less energy-intensive solution

could be given preference for economic and environmental protection reasons. More novel techniques include e.g. resin-in-pulp, biosorption, and/or bioleaching systems. A multi-disciplinary approach of chemistry, extractive metallurgy, engineering and biotechnology is required to realize this ambition;

(11) Given the typically low concentrations of elements in mining and milling residues, added value may be created by extracting all or most of the potentially valuable constituents together with the uranium; this applies in particular to those constituents that are deemed critical raw materials for the sustained and sustainable development of our socio-economic systems;

(12) If recovery of uranium from mine waste impoundments is planned, then the potentially valuable uranium resources need to be quantified using agreed upon and internationally accepted assessment methods, classification frameworks and reporting standards. The existing methods and tools were, however, originally designed for the definition of geological resources and reserves. Similar to the assessments of geological ores, an increased knowledge and confidence on mine waste properties is needed to make valid statements about the potential economic extraction of uranium from mine wastes. Considering the heterogeneity of solid mine wastes and the lack of agreed best practices for assessment methods, it appears far more appropriate that current remining and reprocessing projects of uranium mine wastes are referred to in public statements to 'exploration targets', with ranges given for tonnages and grades;

(13) Reprocessing of the Witwatersrand tailings (South Africa) for uranium is achieving significance because of the available mineral processing and hydrometallurgical treatment technologies, which in turn could lead to significant financial revenues and address environmental concerns;

(14) Recovery of uranium from mine wastes may ultimately initiate the valorization of uranium resources, support total resource use of uranium ores and help the transformation towards a circular economy in uranium mining.

11.2. RECOMMENDATIONS FOR FUTURE WORK

A number of recommendations can be made based on the key findings as well as limitations and constraints of this study. These recommendations may be pursued to support better waste management practices in industry and efficient recovery of uranium from mine wastes:

(1) At operating uranium mine and mill sites, mine wastes could be systematically monitored, measured, analysed and documented for their chemistry, mineralogy, volume and mass;

(2) National mineral waste databases could be extended to include wastes at uranium mines and mills;

(3) For closed uranium mine sites, auditing would facilitate the production data of former mills and the properties of mine wastes and their repositories;

(4) Assessment methods and best practices for the economic evaluation of uranium mill tailings repositories could be developed, considering in particular also the socio-political context as an important modifying factor;

(5) Guidelines for resource and reserve estimation of uranium liquors and mine waters could be established, considering geochemical conditions (e.g. pH, Eh) and modifying factors (e.g. flow rates through mines and waste impoundments);

(6) Future studies could be conducted on other non-uranium critical raw materials from U deposits (e.g. REE, base metals like nickel or vanadium, etc.).

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ABBREVIATIONS

AMD	acid mine drainage
ARD	acid rock drainage
CAPEX	capital expenditure
CRIRSCO	Committee for Mineral Reserves International Reporting Standards
EC	European Commission
ENVSEC	Environment and Security Initiative
ESA	European Space Agency
EU	European Union
Eurostat	European Statistical Office
g/t	grams per tonne
IAEA	International Atomic Energy Agency
INTAS	International Association for the Promotion of Cooperation with Scientists from the Independent States of the former Soviet Union
ha	hectare, 10,000 square metres
h	hour
JORC	Joint Ore Reserves Committee
LIDAR	light radar
M	mega, one million
m^3	cubic metre
micron	micrometre, 10^{-6} metre
NATO	North Atlantic Treaty Organization
NI43-101	(Canadian) National Instrument 43-101
OECD	Organisation for Economic Co-operation and Development
PERC	Pan European Reserves and Resources Reporting Committee
ppm	parts per million
RIP	resin-in-pulp
SAMREC	South African Mineral Reporting Codes
t	tonne, 10^3 kg
TACIS	Technical Assistance to the Commonwealth of Independent States and Georgia
%	percentage

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