MANAGEMENT OF WASTE FROM THE MINING AND MILLING OF URANIUM AND THORIUM BEARING ORES

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ABSTRACT

Mining and processing of uranium and thorium bearing ores for the extraction of uranium and other minerals gives rise to the production of various radioactively contaminated wastes and other re-cyclable materials. The wastes are characterised generally by their bulk nature , their low specific activity and the long radioactive half-lives of several of the radionuclides involved. The contaminated re-cyclable materials are mostly steels, timber and plastic and the contaminants can be of higher specific activity due to various chemical and physical concentration processes. The mining and minerals processing residues , essentially waste rock and tailings, present particular waste management problems. Their bulk nature precludes underground disposal and the long radioactive half-lives present problems of assuring their long term containment. In addition, the residues contain radium at elevated levels, corresponding generally to that in the original mined ore, and presenting a source of radon progeny. The nature of mining waste is such that they are often an inexpensive source of raw materials for use in building materials. Their diversion to such use must be considered in respect of both external and radon progeny exposure.

The paper describes the nature and characteristics of the wastes and re-cyclable materials arising from mining and minerals processing activities. It discusses the various management options adopted for these wastes in various countries and addresses the process of hazard assessment for these wastes and the associated safety criteria. The paper also discusses the consideration of these wastes in terms of the RADWASS programme and the principles of radioactive waste management embodied in the safety fundamentals which form the basis for the RADWASS programme.

1. The Nature and Characteristics of Radioactive Wastes From the Mining Industry

Uranium and thorium are present in the earth's crust at average mass concentrations of around ten parts per million. They can, however, be concentrated in mineral deposits and concentrations can range up to tens of percent. Such concentrations are exceptional, but extensive deposits are found with concentrations hundreds of times the average concentration. Both uranium and thorium are naturally occurring radioactive materials and in nature are comprised of several isotopes. The parent radionuclides, U-238, U-235 and Th-232 are primordial radionuclides with half-lives of 4 500 million, 700 million and 14 000 million years. They decay by alpha particle emission and give rise to subsequent radioactive decay series comprised of various radioactive progeny as illustrated in Table 1. The members of the decay series posses significantly differing physical and radiological characteristics which greatly influence the potential radiological hazards associated with wastes arising from mining and minerals processing activities.

Table 1. Natural Decay Series

URANIUM - 238 DECAY SERIES			THORIUM - 232 DECAY SERIES			URANIUM - 235 DECAY SERIES		
NUCLIDE	DECAY	HALF LIFE	NUCLIDE	DECAY	HALF LIFE	NUCLIDE	DECAY	HALF LIFE
U - 238	ALPHA	4,5 E9 y	Th - 232	ALPHA	1,4 E10 y	U - 235	ALPHA*	7,1 E8 y
Th - 234	BETA*	24 d	Ra - 228	BETA	6,7 y	Th - 231	BETA*	24,6 h
Pa - 234	BETA*	1,17 m	Ac - 228	BETA*	6,13 h	Pa - 231	ALPHA*	3,43 E4 y
U - 234	ALPHA*	2,4 E5 y	Th - 228	ALPHA*	1,9 y	Ac - 227	ALPHA*	22 y
Th - 230	ALPHA*	8 E4 y	Ra - 224	ALPHA*	3,64 d	Fr - 223	BETA*	21 m
Ra - 226	ALPHA*	1,6 E3 y	Rn - 220	ALPHA*	55 s	Ra - 223	ALPHA*	11,2 d
Rn - 222	ALPHA*	3,8 d	Po - 216	ALPHA	0,15 s	Rn - 219	ALPHA*	3,92 s
Po - 218	ALPHA	3 m	Pb - 212	BETA*	10,64 h	Po - 215	ALPHA	1,8 E-3 s
Pb - 214	BETA*	26,8 m	Bi - 212	BETA*	60,6 m	Pb - 211	BETA*	36 m
Bi - 214	BETA*	19,7 m	Po - 212	ALPHA	304 E-9 s	Bi - 211	ALPHA*	2,16 m
Po - 214	ALPHA*	164 E-6 s	Pb - 208	STABLE		Tl - 207	BETA*	4,79 m
Pb - 210	BETA*	21 y				Pb - 207	STABLE	
Bi - 210	BETA	5 d						
Po - 210	ALPHA*	138 d						
Pb - 206	STABLE							
* DENOTES ACCOMPANYING GAMMA EMISSION								

Since the advent of nuclear weapons development and the evolution of nuclear power generation, uranium as a mineral has been widely extracted. Thorium has been exploited to a much lesser degree, however, numerous other minerals are extracted from ore bodies containing elevated levels of uranium and thorium and giving rise to radioactive wastes. The latter include, inter alia, monazite, ilmanite, rutile, zircon baddelyite, phosphates, copper, lead, zinc, silver, flourospar, gold and coal. Radium rich mineral deposits have been exploited since the first quarter of the century and uranium mining activities commenced in the 1940's and accelerated considerably in the early 1950's. To date over thirty countries have produced in excess of 30 tonnes with six countries producing greater than 100 000 tonnes in the period up to the early 1990's as illustrated in Table 2. Greater than 1 800 000 tonnes of uranium have been extracted world-wide and projections are that by the turn of the century this will be in excess of 2 000 000 tonnes.

The mining and minerals extraction process entails mining of the ore body, either underground or by way of an open pit, crushing and milling the ore, possibly concentration of the minerals by various physical processes and extraction of the mineral being exploited by chemical processes. In addition, a limited number of in-situ leaching activities have been undertaken. More often than not these steps are followed by various purification processes. The outcome of the overall process is the mineral product together with various waste residues which will be combinations of:

- waste rock

  rock removed from the mine in winning the ore but which contains mineral concentrations too low to economically extract.

- tailings

  the crushed and milled rock from which the mineral has been extracted together with any chemicals and fluids remaining after the extraction process.

- chemical residues

  spent ion exchange resins, solvent extraction sludges, active charcoal etc.

- other waste

  In addition, various mine and plant consumables such as filters, catalytic saddles, timber etc. and scrap arisings such as metal and plastic become contaminated giving rise to radioactive wastes.

Table 2. World Production of Uranium up to 1992

COUNTRY	PRODUCTION 000 tonnes	COUNTRY	PRODUCTION 000 tonnes	COUNTRY	PRODUCTION 000 tonnes
USA	339	Namibia	53,1	Poland	1,00
Canada	258	Ukraine	45,1	Brazil	0,94
Germany	218	Zaire	26,6	Madagascar	0,77
South Africa	143	Bulgaria	21,9	Pakistan	0,66
Czech Rep.	102	Gabon	21,4	Belgium	0,51
Russia	101	Tadjekhistan	20	Slovenia	0,38
Kasakhstan	81,7	Hungary	19,9	Sweden	0,20
China	79,6	Romania	18,4	Japan	0,09
France	70	India	5,78	Mexico	0,04
Uzbeckhistan	61	Spain	3,78	Finland	0,03
Niger	56,8	Portugal	3,56		
Australia	54,2	Argentina	2,15	TOTAL	1 810

As a primary product, uranium bearing ores are generally exploitable at concentrations in excess of several hundred parts per million. As a by-product, however, uranium can be economically extracted at concentrations of around one to two hundred parts per million as in South African gold mines. At the extreme, uranium is extracted from copper and phosphate ore deposits at concentrations of around fifty parts per million. A representative average grade in the region of 0,1% or one thousand parts per million indicates that associated with the world wide production to the end of the current century will result somewhere in the region of 2 000 million tonnes of mill tailings from primary uranium producers.

The total arising of other wastes are less well documented, however, the estimated waste arisings for South African gold/uranium mining operations together with the characteristics of such wastes are indicated in Tables 3, 4 and 5.

Table 3. Illustrative Concentration Levels in a Selection of Waste Streams From South African Mining Operations

WASTE TYPE	TOTAL ACTIVITY $Bq\ g^{-1}$
Gold tailings	2 - 200 +
Calcine	30 - 2 000
Calcine Scale	200 - 300 000
Pyrites	< 1 - 400
Pyrite scales	1 - 25 000
ADU Section Scales	up to 30 000
Rubber lined components e.g. pipes	up to 200
Contaminated Soils	< 0,2 - 50 000
PMC scales	up to 350 000
Gypsum	up to 10
Waste rock	< 0,1 - 200

Table 4. Annual Waste Production Estimates From South African Mining Operations

MINES	TAILINGS M tonnes	WASTE ROCK M tonnes	SCRAP METAL SCO-I[2] tonnes	SCRAP METAL > SCO-I[3] tonnes
Gold (29)	121	37	15 000	2 000
Tailings re-cycling[3]	48	0,1	1 000	350
Other	60	Unknown	6 000	400

1. Reprocesses already existing tailings dams it does not add to the growing inventory of tailings.
2. Estimates of SCO-1 contaminated scrap are based on the indications that about 10% of all ferrous scrap is contaminated. More accurate statistics are not available at present. Use is made of the Surface Contaminated Object (SCO) classification criteria embodied in the IAEA Regulations for the Safe Transport of Radioactive Material.
3. Estimates of scrap contaminated above SCO-1 levels are based on actual data statistics in waste reports.

Table 5. Demolition wastes

> **Decommissioning and Demolition**
>
> There are a number of contaminated plants on mines awaiting demolition and site rehabilitation which will give rise to significant amounts of waste arisings in the future. The majority of these plants are associated with the gold mines and include acid, pyrite, calcine, uranium and gold plants. About 40 contaminated acid, pyrite, calcine, and uranium plants and their sites require to be demolished and rehabilitated. In many cases the plants have been completely or partially stripped but the sites have not been decontaminated and rehabilitated.
>
> Figures from recent demolition activities indicate the following approximate quantities per site:
>
> - metal SCO-1: up to 10 000 tonnes
> - metal > SCO-1: 100 - 200 tonnes
> - Rubber, resins, plastics : a few hundred tonnes
> - Contaminated soil and rubble (< 1 000 Bq g^{-1} LLA): up to 100 000 tonnes
> - Contaminated soil, rubble and scales (> 1 000 Bq g^{-1} LLA): a few hundred tonnes

In the undisturbed ore body, the uranium and thorium isotopes will exist in their natural ratios and the radioactive progeny will be present within the ore body in secular equilibrium with the parent radionuclides. If some separation has taken place for instance by the intrusion of water into the ore body, this state of equilibrium may have been disturbed. The mining and minerals processing activities significantly alter this state of equilibrium, primarily separating the uranium or thorium from their decay series progeny. Depending upon the extraction efficiency, the bulk waste arisings, i.e. the tailings, will contain only a few percent of the original uranium or thorium concentration. It will, however, still contain most of the radioactive progeny. In the case of uranium mining, this means that the radioactive content of the tailings will be dominated by Th - 230 with a radioactive half life of 80 000 years. Should the ore being processed contain elevated levels of uranium, which are, however, not economically viable for extraction at that time, clearly the majority of this uranium would be present in the tailings and the longer half life of U - 238 would in such cases predominate.

The various other wastes arise due to contamination of materials during the mining and extraction processes. This may be contamination by ore or can arise due to the selective concentration of particular species from the decay series by chemical and physical processes. In particular, radium is found to be concentrated and taken up in several waste arisings.

In terms of overall waste management, due consideration must also be given to the mine following closure. Both underground and surface mining operations result in the original ore body being in an altered state which may have given rise to exposure pathways that did not exist prior to mining activities. Such pathways require consideration in closing-out of the facility.

2. Radiation Hazards Associated With Mining Waste and Management Options

Possible Hazards

In view of the bulk nature of mining and processing wastes and the limited considerations that were originally given to their long term management, disposal options are generally limited to some form of in-situ stabilisation. Whilst there are some significant differences between uranium mining methods and extraction technologies and the resulting wastes, the radiation hazards associated with their management are generally dominated by Th - 230, Ra - 226, Rn - 222 and its short lived progeny and the longer lived radon progeny. In addition, the wastes associated with mining and processing of thorium bearing ores and other radioactive ores all have their own particular characteristics. However, consideration of uranium mining wastes covers all the potential issues associated with the latter.

There are numerous mechanisms by which radionuclides associated with mining and minerals processing waste can enter or give rise to exposure pathways to humans. These may arise during both the operational and post close-out phases and are elaborated below:

- Airborne dispersion from tailings piles and waste rock dumps of particulate material or radon emanating from the piles. This may arise during the operational phase and following close-out if cover material is eroded due to precipitation, wind or frost damage.

- Seepage from tailings and waste rock piles can transport dissolved radionuclides into ground water. The dissolution of nuclides is often enhanced by the presence of pyrite causing the wastes to be acid generating. Subsequent migration can cause the nuclides to enter exposure pathways involving the use of the groundwater or discharge of the groundwater to surface.

- Seepage at ground level may give rise to contamination of surface waters and is also a potential source for airborne dispersion.

- Intrusion on sites were mining and minerals processing residues are present by way of construction, agricultural and recreational activities. The associated exposure pathways include inhalation - both radon progeny and long lived alpha emitting species, direct exposure and ingestion due to uptake or contamination of feed stuffs.

- Diversion of mining residues from operating or closed-out sites. Tailings material often provides an inexpensive source of land fill or construction sand and waste rock an equally inexpensive source for the manufacture of aggregate for various construction purposes. Scrap materials including steel, timber and plastic are often desirable sources of raw material for re-cycling operations and give rise to a variety of exposure pathways depending on the re-cycling operation and the subsequent use of the re-cycled material.

- Closed-out mines may become flooded following operations and give rise to sources of enhance nuclide migration into surface and ground waters. They may also be a source of radon emanation into the atmosphere.

- The mechanisms of migration and transportation included above can also be affected by the impacts of natural phenomena such as earthquakes, floods etc. which may cause damage to engineered and natural containment barriers.

Waste Management Options

Waste management associated with operating mining and minerals processing facilities generally entails the siting, design and operation of tailings dams, siting and placing of waste rock piles, control of surface drainage, control and discharge of mine water, suppression of airborne dispersion from the site and management of consumable waste arisings such as chemical processing residues, scrap steel and timber.

At the time of mine closure, rehabilitation activities generally involve the dismantling of plant, demolition of buildings, stabilisation of tailings and waste rock piles, sealing of the mine and general site rehabilitation. Practice in tailings management has involved relocation, in-situ stabilisation and cover. The latter is aimed at preventing radon emanation, moisture infiltration, gamma radiation, oxidation, intrusion and erosion. Various methods of tailings cover have been used and depend on the local geological, climatological, and geographic characteristics. Often clay, sand and various grades of rock are used, vegetation is also appropriate in some locations and water cover has been used. Mines have been backfilled with mining residues and open pits have been backfilled, re-contoured or flooded.

In view of the limited attention paid to waste management on many sites during the early years of uranium mining, some countries have experienced the contamination of other properties in the vicinity of mining and minerals processing sites. This has been caused by the diversion of materials or from the uncontrolled dispersion of material from the sites. It has resulted in extensive rehabilitation efforts having to have been expended in the cleaning of these properties.

Where mining and minerals processing activities have given rise to the contamination of groundwater various engineered approaches have been adopted. These include isolation of the contaminated groundwater compartments and treatment of the contaminated water and increased natural flushing. In both instances, some institutional control on the site and its environs is necessary, such measures may be required over relatively long time scales - up to hundreds of years.

3. Hazard Assessment and Safety Criteria

Operating Phase

During the operating phase, consideration must be given to; which mining activities will be subject to regulatory control and which can be exempted, the control of effluent discharges from regulated facilities, the management of waste arisings stored or intended for disposal on the site and clearance of materials from the site. All these aspects can reasonably be addressed by deterministic hazard assessments and compared against laid down dose limits and subjected to optimisation assessments. The exception in this regard may be contamination of groundwater, where, due to the uncertainties in groundwater movement and transport of contaminants into and by the groundwater, elements of the assessment may have to be subject to probabilistic considerations.

Exemption gives rise to particular problems due to the existence of natural background levels of uranium and thorium, together with their progeny, and the variable nature of these background levels. This is compounded by the presence of radium, which, at relatively modestly elevated levels above average levels can, in building related scenarios, give rise to dose levels well in excess of recommended exemption levels of dose and in fact in excess of public dose limits. Exemption levels quoted in the Basic Safety Standards Ref [1] for natural uranium, natural thorium and radium-226 respectively are; 1 $Bq.g^{-1}$, 1 $Bq.g^{-1}$ and 10 $Bq.g^{-1}$. It is explicitly stated, however, that these values are not applicable to bulk materials. Control over building on land with radium levels greater than around 0,1 Bq $.g^{-1}$ of radium-226 are recommended in some countries Ref [2] although modelling of the resulting exposures is dependent on numerous factors including soil porosity, building construction method and building ventilation rates. Many countries adopt values in the range of 0,2 $Bq.g^{-1}$ as exemption levels. These correspond to the values laid down in the United States for clean-up of uranium mill tailings and contaminated vicinity properties associated with uranium mining and milling operations Ref [3, 4].

The concept of effluent discharge control in respect of mining and minerals processing facilities is complicated by the range of sources, the extended nature of some sources, such as tailings piles, the various transport pathways and mechanisms and the existence of discrete controlled discharges together with those less amenable to control such as surface run-off and wind-borne dispersion. Nevertheless, assessments can be carried out for comparison with laid down criteria and release control mechanisms are possible. Dose limits vary between different countries spanning the range from a fraction of 1mSv for discrete facilities to 5mSv Ref [5].

The management of waste on mining sites is generally dictated by the tailings and waste rock management programmes. The associated hazard assessment is more related to the eventual close-out of the mine and will be addressed later. One factor giving rise to problems in respect of on-site waste management is the handling of other wastes exhibiting concentrated levels of contamination. Mine tailings contain radium-226 concentrations generally up to around 10 $Bq.g^{-1}$ and in exceptional cases higher. Waste with concentrations up to tens of thousands of $Bq.g^{-1}$ can arise and pose particular waste management problems. Such material would generally not be acceptable for disposal in near surface repositories Ref [6], rather deep geological disposal being indicated. Practice in some mining operations has been to dispose of such materials within tailings dams or waste rock piles and, at this stage, there is no consistent management strategy for such materials. The classification of these types of waste also gives rise to problems, particularly when the waste is in the form of non-active, components contaminated with relatively thick contaminated surface deposits such as scales.

Clearance of materials from mining and minerals processing operations gives rise to many of the issues raised in terms of exemption. Materials which could be considered as candidates for clearance from mining operations could include tailings for land fill and construction purposes, waste rock for building aggregate and scrap steel, timber and plastic. On the assumption that the former materials if released will in fact be used for construction purposes, the selection of an appropriate dose criteria poses problems. Selecting a value at a fraction of the recommended public limit would result in a very restrictive derived levels for radium-226 in the material and basically preclude the clearance of any material with levels marginally above "average" levels of around 0,03 - 0,05 $Bq.g^{-1}$. The current practice being adopted in South Africa is to employ a process of conditional exemption whereby the end use of cleared material must be specified and must not be use in building construction. This process has many associated procedural and legal complications. Similar problems arise also with the "clearance" of former mining land.

The clearance of scrap materials is also complicated by the fact that a large fraction of contaminated materials arise from the deposition of scales on material and the concept of surface contamination as a criteria for clearance is questionable. Also, as a criteria for release, the concept of bulk contamination, taking into consideration the mass of the non-active object raises questions as invariably subsequent handling and processing of the material results in the scales being removed and giving rise to loose radioactive material of much higher unit density. These problems are presently addressed in South Africa by assuming that all surface deposited material is "loose" and by applying surface contamination levels for clearance at the lower end of the spectrum of values used in various countries Ref [7].

End of Mine Life

For many operations which commenced in the 1940's and 1950's, very little consideration was given to closure and to the potential radiological hazards that may be associated with the mining and processing residues. The potential hazards have become apparent within the past twenty years and hence the associated efforts to rehabilitate mines and sites and to implement engineered waste management solutions. The approach generally adopted is to demolish the buildings and to clear the site of contamination with a view to allowing unrestricted future use of the site. The tailings and waste rock piles are then effectively the resulting radioactive waste requiring disposal. The solutions adopted to date have included in-situ stabilisation, removal to engineered on surface containment cells, backfilling into mines and pits and disposal in water bodies. These solutions have necessarily involved the conduct of hazard assessments to demonstrate their adequacy. Such assessment has involved both deterministic and probabilistic modelling techniques both separately and complimentary. The most difficult challenges facing acceptance of the assessments is selection of the time periods required for demonstration of adequacy combined with predicting the performance of the engineered covers, migration barriers and erosion control features over long periods. The impact assessment of the closed mine poses similar problems.

Design criteria for performance demonstration have generally been limited to between two hundred and a thousand years and radiological criteria have often been set in terms of radon emanation rate from the surface of covers, gamma dose rates and ground water contamination levels. Typical levels being in the range of 700 $mBq.m^2$ s, 0,2 $\mu Sv.\text{-}h^{-1}$ and 0,2 $Bq.l^{-1}$ Ra-226 Ref [7].

Because of the need to prevent intrusion into the stabilised tailings , the need to prevent removal of the material for construction purposes and because of the extensive nature of the wastes, and the vulnerability of the covers and barriers, the need for ongoing institution control over these facilities appears to be inevitable.

4. Mining Wastes in the Context of the RADWASS Fundamentals

In view of the relatively low specific activity of mining wastes and the very large bulk volumes, mining wastes have often been viewed in a different light to radioactive waste arising from other parts of the nuclear fuel cycle. The approaches currently being adopted to their management, however, go a long way to respecting the fundamental principles of radioactive waste management adopted by the Member States of the International Atomic Energy Agency.

The principle of protecting human health is respected by adoption of safety standards in line with those recommended by ICRP Ref [8] and endorsed in the Basic Safety Standards Ref [1]. The second principle of protecting the environment is particularly pertinent when considering uranium mill tailings which are often acid generating and contain other non-radioactive potential pollutants such as arsenic, barium, cadmium, chromium, lead and several others. Again the rehabilitation and containment processes often take these other pollutants into account and provide for their isolation from the biosphere. Protection beyond national borders will generally be achieved by the engineered solutions being implemented, although, if mining facilities are close to national borders, the need for demonstration remains. The fourth and fifth principles dealing with protection of future generations and the burden on future generations do raise some questions with mining waste because of the need for perpetual institutional control and the inevitability of the eventual failure of containment structures over the time scale associated with the radioactive half-lives of the radionuclides involved. Nevertheless, the concerns are ameliorated to an extent by the relatively passive nature of the required institutional controls and the relative ease to repair any degradation of the covers.

Compliance with the sixth principle of managing the wastes within an appropriate national legal framework varies considerably between countries. Countries within which a nuclear industry operates often control mining wastes in terms of their nuclear regulatory programme. Such control, however, is often limited to uranium mining wastes and does not cover mining of radioactive ores from which uranium is not extracted. Principles seven and eight requiring radioactive waste generation to be minimised and interdependencies to be considered are features of more recent mining operations and those subject to good regulatory control regimes. The ninth and final principle addressing safety of waste management practise is less of an issue with mining operations due to the low specific activity and the generally chronic nature of the associated hazards.

In conclusion, mining wastes, whilst being characterised by relatively low specific activity and giving rise to chronic modes of exposure, can cause exposures well in excess of public dose limits and can give rise to substantial collective doses. Because of the attractive nature of mining residues for construction purposes and the utility of many of the waste arisings for re-cycling, and the disposal of such wastes often on the surface, particular attention needs to be given to control over their diversion. In addition, due to the relatively significant radiation doses that can arise from radon progeny clearance levels for bulk material must be carefully selected and administered. Because of the long radioactive half lives associated with mining waste, the concept of waste management options involving eventual removal of restrictions on the disposal sites cannot be realised. The need for consideration of all mining wastes involving elevated levels of natural radioactivity to be managed effectively and not only those associated with uranium mining require due consideration.

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