Uranium Mining and Norm, a North American Perspective - 15545

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ABSTRACT

All soils and rocks contain naturally occurring radioactive materials (NORM). Many ores and raw materials contain relatively elevated levels of natural radionuclides and processing such materials can further increase the concentrations of natural radionuclides, sometimes referred to as technologically enhanced naturally occurring radioactive materials (TENORM). Examples of NORM minerals include uranium ores, monazite (a source of rare earth minerals), and phosphate rock used to produce phosphate fertilizer. Such activities have the potential to result in above background radiation exposure to workers and the public. Following a brief review of the sources and exposure from NORM in these varied industries, this paper will then present an overview of uranium mining and recovery in North America including discussion on the methods currently being used, production of NORM materials and wastes associated with these uranium recovery methods and the radiological composition of the NORM wastes produced by uranium recovery. The paper includes descriptions of the methods and associated NORM produced by both conventional (underground, open pit) and in situ recovery (ISR) mining methods and identifies the responsible government agencies in the US and Canada assigned the authority to regulate and control these NORM materials.

INTRODUCTION

People have always been exposed to natural background radiation and radioactivity from radionuclides found naturally in soils and rocks, from radioactivity in the food we eat, the water we drink, the air we breathe, and radiation from space. While the levels of radiation and radioactivity vary widely from place to place, by perhaps a factor of 10 or so, there is nowhere on earth that is not radioactive. According to the United Nations Scientific Committee on the Effects of Atomic Radiation [1], the nominal per caput annual dose from all natural sources of radiation is about 2.4 mSv with a range of (about) 1 mSv/y to 13 mSv/y.

WHAT IS NORM?

NORM is an acronym for naturally-occurring radioactive materials. Most NORM contains radionuclides from the so-called long-lived "primordial" decay chains, resulting from the decay of the U-238, U-235 and Th-232 and from other long-lived radionuclides, such as K-40. All soils and rocks are naturally radioactive and hence all ores are also radioactive. This is illustrated in Fig. 1 which shows the nominal range of radioactivity in soils and a variety of ores.

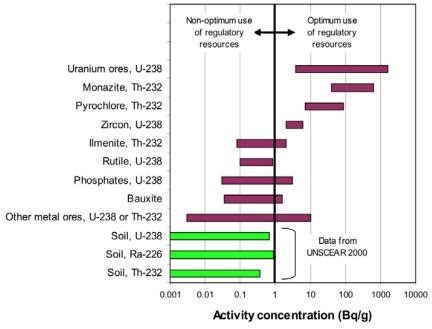


Fig. 1: Uranium and Thorium in Soils and Ores (after [2])

Elevated levels of natural background radiation are seen in many occupational settings, especially in the mining and processing of ores, the extraction of oil and gas, and the production of phosphate fertilizers. A few examples of radioactivity levels in a variety of NORM materials are summarized in Table 1, which illustrates the wide range of "typical" concentrations of uranium and thorium in NORM.

Source	Radionuclides (s) with highest activity concentration	<u>Typical activity concentration</u> (Bq/g)
Monazite sand	232Th series	40 - 600
Metal ores, e.g., Nb,Cu,Au	238U and 232Th series	Up to 10
Zircon sand	238U series	2 - 4
Phosphate rock	238U series	0.03 - 3
TiO2 feedstocks	232Th	0.001 - 2
Bauxite	232Th series	0.035 - 1.4
Red mud (alumina)	238U, 232Th	0.1 - 3
Phosphogypsum (H2SO4)	226Ra	0.015 - 3
Niobium extraction slag	232Th	20-120
Tin melting slag	232Th	0.07 - 15
Scale (oil and gas production)	226Ra	0.1 - 15,000
Residue (rare earth extraction)	228Ra	20 - 3,000
Scale (TiO2 pigment production	228Ra, 226Ra	<1-1,600
Scale (rare earth extraction)	226Ra, 228Th	1,000
Sludge (oil and gas production)	226Ra	0.05 - 800
Residue (niob. extraction)	228Ra	200 - 500
Coal	238U and 232Th series	0.01 - 0.025
Scale (coal mines with Ra rich inflow water)	226Ra, 228Ra	Up to 200

TABLE I. Examples of radioactivity levels in NORM (Source: Adapted from Table 1 of [3])

The radionuclides of interest for NORM are from the uranium and thorium natural decay series as depicted in Figure 2. As suggested by the foregoing, NORM is found in mining and mineral processing, as well as in many industrial waste streams among them, overburden, mine spoils and tailings from uranium mining, metal mining and processing wastes, rare earths mining and extraction, oil and gas production (including fracking), and the production of phosphate fertilizers. The remainder of this paper discusses NORM in association with uranium mining and milling only. The reader is referred to numerous other references that discuss occurrences of NORM in these other industries and associated health and safety considerations [4, 5, 6, 7, and 8].

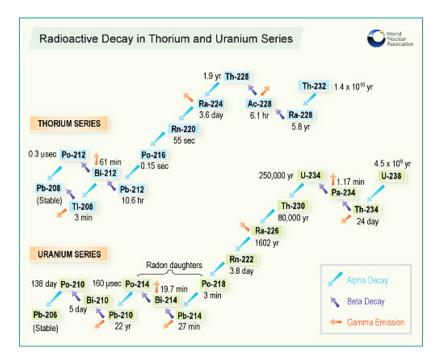


Fig. 2. Radionuclides of the Naturally Occurring Uranium and Thorium Series (from [9])

URANIUM MINING AND RECOVERY

According to the World Nuclear Association (10) uranium was produced in some 19 countries in 2013 through one or more of the three main methods of producing uranium: these methods are underground mining, open pit mining, and in-situ-leaching (ISL - sometimes referred to as insitu recovery or ISR). Conventional mines, either underground or open pit mines have a mill where the ore is crushed, grounded up and then leached¹ to dissolve the uranium and separate it from the host ore. At the mill of a conventional mine or the treatment plant of an ISL operation, the uranium which is now in solution is then separated by solvent extraction or ion exchange² before being precipitated, dried, and packed. This product, uranium oxide concentrate, is also referred to as yellowcake and mixed uranium oxides $- U_3O_8$. In addition, uranium can be

¹ Depending on the mineralogy of the ore, various processes including either sulphuric acid or alkaline (carbonate) leach are employed to liberate the uranium from the host ore. The method used to recover uranium has implications for protection of both workers and the environment.

² ISR mining uses ion exchange only.

recovered as a by-product from phosphate fertilizer production and from mining of other minerals including copper and gold where the ores contain economic quantities of uranium. In such situations, the treatment process to recover uranium may be more complex.

During uranium mining and processing, workers may be exposed externally to gamma rays emitted from the ores, process materials, products and tailings and internally through inhalation of aerosol particles (inhalable dust) containing long lived alpha activity and including radon progeny. Environmental exposures vary by region, type of mine and processing but include releases of dust and radon to the air, releases of uranium, radium and other radionuclides to the surface water and potential effects on groundwater. For modern mines and processing plants, most workers receive radiation doses well below the radiation protection guidance developed by the International Commission on Radiation Protection (ICRP). Annex B of UNSCEAR 2008 [11] provides considerable information on current levels of radiation seen by workers and the public arising from uranium recovery operations. Available information indicates that currently, average doses to workers involved with mining and processing uranium in the US, Canada and world-wide are less than 2-3 mSv/y [11, 12, 13, and 14].

History of Uranium Recovery in the United States

In the United States, the mining of ore that contains uranium goes back to the early part of the 20th century. At that time the interest was not in uranium per se, but in other minerals associated with it, namely vanadium and radium. Interest in uranium began in earnest in the years immediately following World War II with the passage by the U.S. Congress of the McMahon Act (more commonly known as the Atomic Energy Act [AEA], signed by President Truman in August 1946), which created the United States Atomic Energy Commission (AEC) and established the U.S. government as the only buyer of uranium (for the nuclear weapons program). The government's uranium ore procurement program sent thousands of prospectors crawling over the "Colorado Plateau" (the four corners area of Utah, New Mexico, Arizona, and Colorado). This ore was processed at a number of sites—collectively known as the "MED (Manhattan Engineering District) Sites"—and remediated decades later under the Formerly Utilized Sites Remedial Action programs still ongoing today. AEC incentives ceased in 1962, and mining and milling operations on a much larger scale than those early efforts were established by private companies.

As the commercial nuclear power industry developed in the late 1960s and early 1970s, the federal government was no longer the exclusive buyer of domestically produced uranium. U.S. production and uranium prices peaked in the early 1980s. Shortly thereafter, domestic demand for uranium ore declined as the commercial nuclear power industry fell far short of its expected growth and in response to, and low cost of, much higher-grade Canadian and Australian deposits that began to dominate world markets. Planning and construction of new U.S. commercial nuclear power plants came to a halt and the domestic price of uranium dropped dramatically, and the nation faced an oversupply of uranium despite the fact that demand remained about even through 2003. However, in the last 5-8 years the U.S. Nuclear Regulatory Commission (NRC) has issued a number of new licenses for ISR uranium recovery facilities and the State of Colorado issued in 2012 the first new license for a conventional uranium mill in the US in over 25 years. Additionally, several new nuclear reactors are under construction in the US. The uranium fuel cycle used in the

US is shown in Figure 3, which depicts the process from uranium mining and milling through several intermediate steps to ultimately produce the uranium fuel for nuclear reactors.

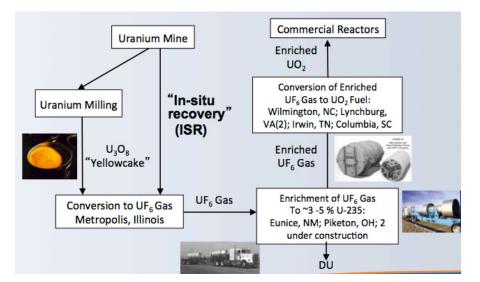


Fig. 3. The US Uranium Fuel Cycle

Mining Techniques

Conventional mining generally refers to open-pit and underground mining. Open-pit mining is employed for ore deposits that are located at or near the surface, while underground mining is used to extract ore, typically of higher grade (concentration of uranium in the ore expressed as weight percent or ppm), from deeper deposits. Conventional uranium mines are not regulated under the AEA since the raw ore is not considered "source material"1 under the Act and therefore is not a licensed material.³ The health and safety aspects of conventional uranium mines are regulated at the federal level by the Mine Safety and Health Administration of the U.S. Department of Labor and by respective state agencies with responsibility for health, safety, and environmental protection associated with mining.

Open-pit mining involves the surface removal of soil and rock overburden and extraction of ore. Open-pit mines are broad, open excavations that narrow toward the bottom and are generally used for shallow ore deposits. The maximum depth of open-pit mining in the United States is usually about 150 meters. Lower-grade ore can be recovered in open-pit mining, since costs are generally lower compared to underground mining. In open-pit mining, topsoil is removed and often stockpiled for later site reclamation (i.e., restoration). Overburden is removed using scrapers, mechanical shovels, trucks, and loaders. In some cases, the overburden may be ripped or blasted free for removal. Once the uranium ore-bearing horizon is reached, the ore is extracted. The extracted ore is stockpiled at the surface or trucked directly to a conventional uranium mill (see below) for processing into the U₃O₈ product (referred to as "yellowcake" due to its typical color).

³ However in Canada, the Canadian Nuclear Safety Commission (CNSC) regulates both uranium mining and milling

Deeper uranium ore deposits require underground mining in which declines or shafts are excavated and/or drilled from the surface to access the ore-bearing strata at depth. These deeper deposits may require one or more vertical concrete-lined shafts or declines large enough for motorized vehicles to reach the ore. Stopes (an underground excavation from which ore will be removed in a series of steps) reaching out from the main shaft provide access to the ore. Ore and waste rock generated during mining are usually removed through shafts via elevators or carried to the surface in trucks along declines. As with open pits, the extracted ore is stockpiled at the surface and subsequently transported directly to a conventional uranium mill. An overview of conventional uranium mining techniques is presented in Figure 4.

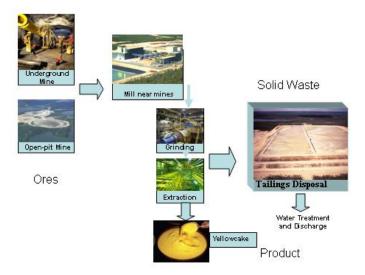
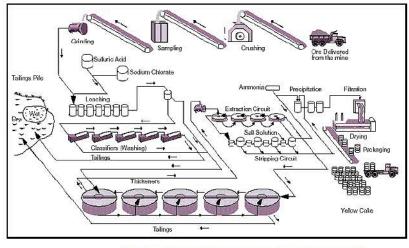


Fig. 4. Overview of Conventional Uranium Mining

Conventional Uranium Mills

Uranium mills (and in situ recovery facilities [ISRs]), are "licensed facilities" since they produce source material as defined under the US Atomic Energy Act (AEA). Accordingly, licensing requirements and management of uranium mills are defined in NRC's 10 CFR 40, *Domestic Licensing of Source Material*, and commensurate requirements of agreement state regulations. The generalized conventional uranium milling process is depicted in Figure 5.



Source: U.S. DOE/EIA, http://www.eia.doe.gov/cneaf/nuclear/page/uran_enrich_fuel/uraniummill.html

Fig. 5. Schematic if Conventional Uranium Milling Process

As shown in Figure 5, the initial step in conventional milling involves crushing and grinding of the raw ore to produce uniformly sized particles. Various mechanical mills grind the rock to further reduce the size of the ore. After the ore is ground and is put in the form of a slurry, it is then pumped to a series of tanks for leaching, either in an acid- or alkaline-based process. The uranium liquor is separated from residual solids and then dissolved into a solvent. These solids are the "uranium mill tailings" which must be managed in large surface impoundments as the major radioactive waste stream of a conventional mill. The uranium is then recovered (stripped) from the solvent-based liquor. The final steps consist of precipitation to produce yellowcake, followed by drying and packaging of the final U_3O_8 product.

A commercial-scale conventional mill processes on the order of 1,000 tons or more of ore per day and produces one to two million pounds per year of U_3O_8 . Over 95 percent of the ore mass constitutes radioactive wastes (tailings) and must be permanently impounded at or near the mill site in a highly engineered landfill ("tailings impoundment or pond"). This material, which is referred to as "11e.(2) byproduct material" as defined in the AEA, is regulated by the Canadian Nuclear Safety Commission (CNSC) and by the USNRC under the AEA. Radiologically, this material contains 99 percent of the uranium series radionuclides which occurred in secular equilibrium with the U-238 parent in the ore body, minus most of the uranium. For an ore grade of a few percent uranium, the tailings would contain an order of magnitude of a few 100s to a few 1000s Bq/g of each daughter in equilibrium.

In Situ Recovery Facilities

ISRs (also referred to as in situ leach or uranium solution mining) are rapidly becoming a preferred method around the world for uranium recovery (almost 50% of global production in 2013 was produced by ISR methods {10}). This is primarily because of lower capital costs, fewer manpower requirements for operations, smaller land-use footprints, and environmental advantages over conventional mines and mills. However, applicability of this technology is generally limited to very specific geological, hydrological, and geochemical conditions. Uranium

deposits typically amenable to in situ recoveries are usually associated with relatively shallow aquifers, about 30-150 meters subsurface, confined by nonporous shale or mudstone layers. The uranium was transported to these locations over geologic time as soluble anionic complexes by the natural movement of oxygenated groundwater. Deposition occurred in areas where the groundwater conditions changed from oxidizing to reducing, producing what is known as a "roll front deposit."

Accordingly, ISRs are typically used for recovery of uranium at ore grades below that associated with conventional mining (open pits or underground). Typical uranium ore grades associated with ISR roll-front deposits in the US are about 0.1 percent-0.2 percent (1,000-2,000 ppm uranium in the ore). ISRs, like conventional mills, are considered source material facilities under the AEA and therefore must be licensed and operated as such under NRC (e.g., 10 CFR 40) or commensurate agreement state regulations and requirements.

ISR processes in the United States⁴ typically involve the circulation of groundwater, fortified with oxidizing (typically gaseous oxygen) and complexing (e.g., carbon dioxide) agents into an ore body (referred to as "the lixiviant"), solubilizing the uranium in situ, and then pumping the solutions to the surface where they are fed to a processing plant (very similar to a conventional mill, without the need for ore crushing, grinding, and leaching). The uranium dissolved in solution returning from underground is first concentrated in an ion exchange circuit, stripped from the ion exchange resin via an elution process and then precipitated into yellowcake, dewatered, dried, and packaged as the final uranium oxide ($^{\circ}U_{3}O_{8"}$) product in an identical manner as in conventional mills. Figure 6 shows the basic approach to in situ uranium recovery.

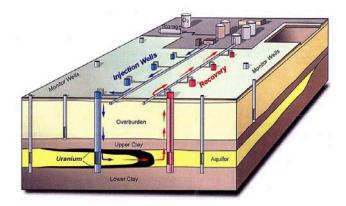


Fig. 6. Basic Concept of In Situ Recovery (ISR) Process

⁴ Acid leach processes are typically used outside the US, e.g., in Kazakhstan, which accounted for about 30% of global production in 2013, and in Australia

Since ISRs do not process large volumes of ore (rock), as do conventional mills, conventionaltype uranium mill tailings are not generated by these processes. However, ISRs do generate relatively small volumes of these NORM wastes related to the need to remove calcium compounds from the process to maintain formation and system permeability and remove impurities. Radium follows the calcium chemistry through the process. Measurements made in the 1970s and early 1980s suggested that 5-15 percent of equilibrium Ra-226 in the host formation ends up in this material [15, 16].

These NORM wastes, which are considered licensed 11.e(2) byproduct material by the USNRC (as is conventional uranium mill tailings) must be shipped off-site to a licensed uranium mill tailings impoundment or other licensed disposal facility authorized to accept it. Additionally, due to the need to extract several percent greater volume of solutions for hydrological control than is actually reinjected in the well fields ("over recovery"), large volumes of solutions must be impounded and managed at the surface. In modern designs, these fluids are disposed of via irrigation and/or injected in permitted deep-well disposal systems following treatment.

Currently Operating Uranium Recovery Facilities in North America

In the US, by the end of 2014, there was just one operating conventional uranium mill (Energy Fuels Resources. White Mesa Mill in Utah) and one licensed but not yet built conventional uranium mill (Energy Fuels Resources Pena Ridge project in Colorado). Additionally, there were seven operating in situ uranium recovery facilities in Wyoming and Texas and several other ISR projects that have been licensed by the USNRC and are in various stages of design and construction. Figure 7 presents the locations of the currently operating uranium recovery facilities in the US.



Fig. 7. US Uranium Recovery Production (2014)

In Canada, although potentially mineable deposits have been identified throughout Canada, the majority of recent and current uranium mining and milling activities are concentrated in northern Saskatchewan Province, in the region known as the Athabasca Basin. This area includes several large mine and mill complexes that produce about 16% of worldwide uranium supplies (10). The major Canadian uranium mines and mills are shown on Figure 8 and are operated by Areva, Denison Mines and/or Cameco.



Fig. 8. Major Uranium Mining and Milling Sites in Canada

NORM MATERIALS AND WASTES ASSOCIATED WITH URANIUM RECOVERY

The vast majority of NORM containing residual materials produced by the uranium recovery process is associated with the wastes ("tailings") that result following extraction of the uranium from the ore (conventional milling) and the much smaller volume of wastes produced primarily from filtration of solutions in ISR processes. Although the vast majority of the uranium has been removed by the extraction process, in conventional mill tailings essentially all of the uranium decay ("daughter") products remain in the waste. With ISR processes in which the ore itself remains underground in situ, only relatively small amounts of uranium decay products are mobilized and brought to the surface requiring management, primarily radium 226 [15,16]. In the US, since uranium recovery facilities must be licensed by the USNRC (or USNRC Agreement States such as Texas and Colorado), these wastes are also licensed material ("11.e (2) byproduct material"). Contaminated process equipment and other potentially contaminated consumable materials (PPE, cleaning wastes, etc.) may also need to be managed as 11.e (2) byproduct material.

Accordingly, in Canada and the US, the management and disposition of these wastes is highly regulated by the Canadian Nuclear Safety Commission (CNSC) and the USNRC respectively (e.g., USNRC in 10CFR40, Appendix A, *Criteria Relating to the Operation of Uranium Mills and the Disposition of Tailings or Wastes.* Figure 9 depicts aerial views of a conventional mill tailings impoundment (Cluff Lake, Saskatchewan) during mill operations (on left) with water cover vs. following decommissioning (on right) covered by clean soil on which native vegetation will be planted.



Fig. 9 Conventional Mill Tailings Impoundment During Mill Operations (Left) and Following Decommissioning (Right)

CONCLUSIONS

Uranium and thorium are ubiquitous in soil, rocks and ores. Thus, mining, milling and many industrial processes will have NORM as an integral part of their activities. The NORM industries that were the subject of this paper were the mining and milling of uranium ore although other metal mining and processing industries, rare earth elements mining and processing, phosphate fertilizer, oil and gas production, and fracking also involve NORM materials and can produce NORM containing wastes. Exposures and the specific radionuclides will vary between industries, but the most prominent NORM radionuclides arise from the uranium decay series and thorium decay series. The levels of uranium and thorium series radionuclides associated with NORM industries can vary quite widely. Even for industries where the initial concentrations of radionuclides at various locations in process streams or in wastes. Thus, the presence of NORM and consideration of the potential exposure pathways and levels of radiation exposure should be an integral part of health and safety planning.

Although this paper primarily addressed the circumstances of regulated "licensed" uranium recovery facilities, the national nuclear regulators in Canada and the United States do not regulate activities with uranium and thorium below 0.05% by weight (not licensed). However, in both Canada and the United States, guidance for radiation protection from NORM is available and, broadly speaking, is followed by the provinces and States. Both countries follow the guidance for transport of radioactive materials developed by the International Atomic Energy Agency (IAEA).

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