The Uranium Recovery Industry and the Current Nuclear Renaissance – A Health Physicists Perspective - 9040

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ABSTRACT

Concurrent with the recognition that nuclear generated electricity must play an increasing role in worldwide energy supply and in consideration of the new nuclear power plants ordered or planned, the demand for uranium needed to fuel these reactors has already outpaced supplies. Accordingly, the price of uranium (typically expressed as \$ per pound U3O8 equivalent) has increased significantly over the last two years. As a result, numerous new and reconstituted uranium recovery projects are being developed in the United States and in other countries that possess considerable uranium ore reserves (e.g., Canada, Australia, Kazakhstan, Mongolia, Namibia, and others).

It should be noted that in the United States, the current reactor fleet of 104 operating units, which generate 20 percent of the US's base-load electricity, requires approximately 55 million pounds of U3O8 per year, but only about 4-5 million pounds per year is produced domestically. That is, over 90 percent of current demand, ignoring anticipated increase in requirements in the near future as new plants come online must come from foreign sources. Domestic uranium production over the last 10 years reached a low of about two million pounds in 2003 and has been increasing steadily since then.

Uranium recovery as defined in this paper encompasses conventional uranium mining and milling as well as in situ recovery techniques and the recovery of uranium as a byproduct from other processes, such as phosphoric acid production. Following a brief history of uranium recovery in the US, the paper describes the basic methods and technologies associated with conventional uranium mining, conventional uranium milling and In Situ Recovery (ISR). The "health physicists perspective" is introduced into these discussions by providing summaries of the various radiological environmental monitoring and operational health physics programs that are required for these recovery techniques based on specific design and operational aspects of each. Applicable regulatory guidance and associated "best health physics practices" developed at these facilities are described.

INTRODUCTION

Uranium recovery encompasses conventional uranium mining and milling as well as in situ recovery techniques (Figure 1) and the recovery of uranium as a byproduct from other processes, such as phosphoric acid production.

Concurrent with the recognition that nuclear-generated electricity must play an increasing role in worldwide energy supply and in consideration of the new nuclear power plants ordered or planned, the demand for uranium needed to fuel these reactors has already outpaced supplies. Accordingly, the price of uranium (typically expressed as \$ per pound U3O8 equivalent) has increased significantly over the last two years. As a result, numerous new and reconstituted uranium recovery projects are being developed in the United States and in other countries that possess considerable uranium ore reserves (e.g., Canada, Australia, Kazakhstan, Mongolia, Namibia, and others).

This imbalance between supply and demand is depicted in Figure 2. Historical uranium prices are shown in Figure 3, including the recent significant price increase as a direct result of the supply/demand imbalance. It should be noted that in the United States, our current reactor fleet of 104 operating units, which generate 20 percent of our base-load electricity, requires approximately 55 million pounds of

U3O8 per year, but only about 4-5 million pounds per year is produced domestically. That is, over 90 percent of our current demand, ignoring anticipated increase in requirements in the near future as new plants come online must come from foreign sources. Domestic uranium production over the last 10 years reached a low of about two million pounds in 2003 and has been increasing steadily since then.

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). The AEC developed publications to assist prospectors in this regard (Figure 4). 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 Program (FUSRAP) still ongoing today. AEC incentives ceased in 1962, and private companies established mining and milling operations on a much larger scale than those early efforts.

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 demand remaining about constant through 2003.

As a result of the market conditions described above, the uranium recovery industry will benefit directly from the "nuclear renaissance" of today and into the near future. The U.S. Nuclear Regulatory Commission (NRC) Uranium Recovery Branch estimates that over the next few years, it expects to receive over 30 source material license applications for new and/or upgraded uranium recovery facilities (Camper 2008—see Table 1). Similar new project development is also taking place in the historical uranium recovery districts in NRC Agreement States (e.g., Texas and Colorado).

OVERVIEW OF CONVENTIONAL URANIUM 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." 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).

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.

CONVENTIONAL URANIUM MILLS

Uranium mills (and in situ recovery facilities [ISRs], see page 12) are "licensed facilities" since they produce source material as defined under the 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, and an aerial view showing the "footprint" of a conventional mill and associated tailings (radioactive waste) impoundment is shown in Figure 6.

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 U₃O₈ 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 pile or pond"). This material is referred to as "11e. (2) byproduct material" after the AEA paragraph which legally defines it. Radiologically, this material contains 99 percent of the uranium series radionuclides which occurred in secular equilibrium with the ²³⁸U parent in the ore body, minus most of the uranium. For an ore grade of a few tenths of percentage uranium, the tailings would contain an order of magnitude of a few 10s to a few 100s 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. 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 recovery 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 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 States 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 U3O8 product in an identical manner as in conventional mills. Figure 7 shows the basic approach to in situ uranium recovery. Figure 8 shows the footprint in an aerial view of a modern ISR.

Since ISRs do not process large volumes of ore (rock), as do conventional mills, conventional-type uranium mill tailings are not generated by these processes. However, ISRs do generate relatively small volumes of 11e. (2) byproduct material 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 indicated that 5-15 percent of equilibrium 226Ra in the host formation ends up in this material (NMA 2007, Brown 1982). This material 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.

Another special consideration associated with ISRs that has health physics implications is the manner in which 222 Rn gas is evolved by the process. At conventional mills, the "radon source term" is almost exclusively the result of the natural decay of 226 Ra in ore bins and the tailings impoundment. At ISRs, it was observed that more than 90 percent of the radon source term results from the dynamic release of radon dissolved in the lixiviant solution as it returns from the underground environment (Brown and Smith 1981). It appears that the temperatures and pressures in situ enhance solubility of radon, and much of the dissolved gas is released when the solutions are first exposed to atmospheric conditions. If this is inside the plant, local exhaust systems deployed at point(s) of release are often required to remove the radon from the work environment, thereby minimizing opportunity for progeny in growth. When inhaled, it is the particulate progeny, not the radon gas itself, that produce the majority of the pulmonary dose.

ADDITIONAL URANIUM RECOVERY TECHNOLOGIES THAT MAY BE REVISITED

In the 1970s and into the 1980s, uranium was also recovered as a byproduct of copper and phosphate production. I was the radiation safety officer for a plant that was colocated at the world's largest open-pit copper mine, near Salt Lake City, Utah. Our uranium plant received a portion of the copper recovery circuit liquor and, through ion exchange and subsequent traditional uranium milling processes as described above, produced 150,000-200,000 pounds per year of yellowcake. Similarly, I had corporate radiation protection oversight responsibility for one of the several uranium recovery facilities in the

phosphate lands of west central Florida. This facility received a portion of the phosphoric acid production plant stream and, through traditional uranium milling processes, also produced a similar rate of yellowcake. Regarding uranium's well-known occurrence in phosphate rocks, it seems reasonable to assume that uranium companies are again or shortly will be re-evaluating the potential uranium reserves inherent in this material and the associated economic viability of recovery.

ENVIRONMENTAL MONITORING AT URANIUM MILLS AND ISRS

Comprehensive environmental monitoring programs must be conducted at uranium recovery facilities to (1) establish the preoperational radiological baseline against which potential future impacts can be assessed and (2) demonstrate compliance during operations to public exposure standards (e.g., 1mSv/y per 10 CFR 20.1301) and to ensure effluent releases are maintained ALARA. These programs are typically performed in accordance with NRC Regulatory Guide 4.14 (NRC 1980).

Uranium and, therefore, its progeny are naturally occurring, and levels in environmental media can vary considerably from place to place depending on local geology, hydrology, and geochemistry. Accordingly, measurements are made of direct radiation (cosmic plus terrestrial) and of uranium-series radionuclides in air (long-lived alpha-emitting particulates and radon gas), in surface and groundwater, and in soil, vegetation, and meat, milk, and fish as may be applicable at a given locale. Key elements of the preoperational baseline program are continued during plant operations and also typically include effluent monitoring (radionuclide particulates and radon releases from ventilation systems and yellowcake dryer stacks).

OPERATIONAL HEALTH PHYSICS PROGRAMS AT URANIUM RECOVERY FACILITIES

Uranium Mines: The environment underground potentially exposes workers to two primary sources: (1) internal exposure from inhalation of ²²²Rn and its short-lived progeny in breathing air (the "radon daughters," ²¹⁸Po, ²¹⁴Bi, ²¹⁴Pb, and ²¹⁴Po) and (2) external exposure from close proximity to higher-grade uranium ore. Needless to say, ventilation and diligent air sampling programs are critical in maintaining internal exposure ALARA and, in higher-grade mines; occupancy times in some areas underground often must be managed and controlled. As indicated previously, the Mine Safety and Health Administration (MSHA) regulates worker health and safety in mines in the United States. MSHA

regulations currently require documentation of internal exposure (typically in working level² months [WLM] of radon daughter exposure relative to a standard of 4 WLM/y) and external exposure relative to a 5 rem/y standard. However, at the present time, MSHA does not require conversion of WLM of exposure to a committed effective dose equivalent (CEDE) nor the addition of internal and external exposure into an expression of the total effective dose equivalent (TEDE). At open-pit mines, internal exposure is usually minimized since excavation is in the open air and dust suppression technology is applied typical of large civil engineering construction projects.

Uranium Mills and ISRs: Operational health physics programs in conventional mills and ISRs are very similar and are generally consistent with any nuclear material facility that produces standard industrial uranium compounds of natural enrichment³ and include:

• Airborne monitoring for long-lived alpha emitters (uranium, thorium), primarily in ore crushing, drying, and packaging areas including combinations of grab sampling and breathing zone sampling.

• Radioactive material area ingress/egress control programs and surface-area contamination surveillance and control throughout plant areas.

• Respiratory protection programs if necessary, typically only necessary in ore crushing, product drying, and packaging areas.

• Bioassay programs appropriate for the uranium products to which employees are potentially exposed. It must be noted that product-specific solubility characteristics can have metabolic implications for bioassay

(NRC 1986; NRC 1988; Eidson and Mewhinney 1980). Higher solubility results in faster pulmonary clearance and, therefore, less pulmonary dose and vice versa. Typically, only urinalysis is performed with

in vivo lung counting in response to confirmed intakes above specified action levels.⁴

• Work control and training via formalized procedures.

- Internal audit and quality-control programs to ensure execution of safe work practices, regulatory compliance, and ALARA.
- Airborne monitoring for radon and progeny as dictated by specifics of facility design.

• External exposure monitoring, primarily in areas in which large quantities of uranium concentrates and/or byproduct material are processed, packaged, and/or stored.

Internal exposure is documented by recording the derived air-concentration hours (DAC-hrs) of exposure to long-lived alpha emitters (uranium, thorium, radium), exposure to radon progeny in working level months, and bioassay results. External exposure is documented from TLD results. CEDE resultant from internal exposures and the TEDE as the sum of internal and external exposure are typically calculated using methods described in, e.g., NRC Regulatory Guide 8.30, *Health Physics Surveys in Uranium Recovery Facilities*, 2002.

Over the years, the NRC has issued a number of helpful regulatory guides specific to uranium recovery facilities, providing a solid basis and foundation for "good health physics practice." Typically, the agreement states accept these as appropriate to demonstrate compliance to their own regulations commensurate with, e.g., NRC's 10 CFR 20 and 10 CFR 40. Examples include:

- 8.30 Health Physics Surveys in Uranium Recovery Facilities
- 8.31 ALARA Programs at Uranium Recovery Facilities
- 8.22 Bioassay at Uranium Mills
- 3.56 Emission Control Devices at Uranium Mills
- 3.59 Estimating Airborne Source Terms for Uranium Mills

CONCLUSIONS—OPPORTUNITIES FOR HEALTH PHYSICISTS IN THE EXPANDING URANIUM RECOVERY INDUSTRY

Hopefully, this broad overview above suggests that numerous opportunities for health physicists and radiological scientists are emerging as a result of the rapid ongoing expansion of the uranium recovery industry. Not only are there opportunities to support the health physics and related environmental-assessment and monitoring programs of operating plants, but the preoperational licensing process is arduous and can take several years. During this preoperational period, baseline radiological monitoring programs must be designed and implemented and source material license applications and numerous other permits must be prepared. These regulatory submittals must describe, in some aspects in considerable detail, the intended operational health physics and training programs and provide results of fate and transport modeling efforts to estimate off-site public exposure during operations, radiological design aspects to ensure incorporation of ALARA principles into the facility design and layout and for effluent control, and descriptions of the planned operational environmental monitoring program. After the doldrums of the last 20-

plus years, it is again an exciting time at the front end of the uranium fuel cycle.

FOOTNOTES:

¹ In general terms, "source material" means either the element thorium or the element uranium, provided that the uranium has not been enriched in the isotope 235 U. Source material also includes any combination of thorium and uranium, in any physical or chemical form, that contains by weight one-

twentieth of one percent (0.05 percent) or more of uranium, thorium, or any combination thereof that is processed for its uranium and/or thorium content.

² A working level (WL) is the total potential alpha energy dissipated in one liter of air from the decay of the short-lived daughters in equilibrium with 100 pCi/L of radon, equivalent to 1.3 x 10^5 MeV/liter of air; a working level month (WLM) is exposure to a concentration in air of one WL for a working month of 170 hours. It is generally assumed that 1 WLM = 12.5 mSv (1.25 rem) so that 4 WLM/y = 50 mSv (5 rem)/y. Note however, that ICRP 65 (ICRP 1994) equates 1 WLM to 5 mSv (500 mrem), which may be conservative.

³ Natural enrichment means the mixture of the three naturally occurring isotopes of uranium as it occurs in nature, which is, on a mass basis, 99.3 percent ²³⁸U, 0.72 percent ²³⁵U, and 0.005 percent ²³⁴U. Due to differing half-lives, and therefore different specific activities, on an activity basis these ratios are 48.9 percent 238U, 2.2 percent 235U, and 48.9 percent 234U. By "definition," the specific activity of natural uranium is 0.67 μ Ci/g (10 CFR 20, Appendix B, Table 1, footnote 3).

4 Over my career I have had the opportunity to have been the radiation safety officer at six different uranium recovery facilities that produced products of varying solubility depending on specifics of process chemistry and the drying temperatures used (e.g., Task Group on Lung Dynamics class D/W as well as Y—ICRP 1972). However, modern mill designs dry the final uranium product at much lower temperatures than in the past, producing more soluble products

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FIGURES AND TABLES



Figure 1. Uranium fuel cycle



Figure 2. U3O8 production versus demand www.uraniumproducersamerica.com/supply.html



Figure 3: Historical Uranium Prices

Facility	Quantity
New ISR* Facility	14
New Conventional Mill	7
Combined ISR-Conventional	1
ISR Expansion	7
ISR Restart	1
Conventional Restart	1
TOTAL	31

*In Situ Recovery Table 1. New source material licensing actions anticipated by NRC in next few years



Figure 4. AEC uranium prospecting booklets



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

Figure 5. Generalized conventional uranium milling process U.S. DOE Energy Information Administration



Figure 6. Aerial view—conventional uranium mill complex Photo courtesy Cotter Corporation



Figure 7 Basic approach to in situ uranium recover



Figure 8. Aerial view—in situ uranium facility Photo courtesy Wyoming Mining Association