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Radiological Aspects of Alkaline Leach Uranium In Situ Recovery (ISR) Facilities in the United States

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Summary

In Situ Recovery or In Situ Leach (ISR/ISL) uranium facilities, also referred to in the past as "uranium solution mining"have operated since the late 1960s in the US and in recent years have accounted for over 70 % of US production and internationally almost half of worldwide uranium supplies.

This extended abstract presents a summary of the radiological characteristics of typical ISR processes being employed in the United States today that have traditionally used alkaline based lixiviants. The paper describes the health physics and radiological monitoring programs necessary to adequately monitor and control radiological doses to workers based on the radiological character of these processes. Although many radiological characteristics are similar to that of conventional mills, conventional-type tailings as such are not generated. However, liquid and solid by-product materials may be generated and impounded which can result in sources of occupational exposure. Some special monitoring considerations are required due to the manner in which radon 222 gas is evolved in the process. The major aspects of the health physics and radiation protection programs that have been developed at these facilities over many years are discussed and include:

- Airborne monitoring for long lived radioactive dusts
- External exposure monitoring primarily in areas in which large quantities of uranium concentrates are processed and where radium precipitates may accumulate
- Surface area and personnel contamination surveillance
- Bio-assay (urinalysis) programs commensurate with the metabolic characteristics of the uranium species produced
- Radon/progeny monitoring, particularly at front end of process where radon is most likely to evolve from solutions returning from underground

Background

Uranium deposits in the U.S. typically amenable to ISR methods are usually associated with relatively shallow aquifers, about 30—150 meters subsurface, confined by non-porous shale or mudstone layers. Uranium was transported to present locations over geologic time as soluble anionic complexes by the natural movement of oxygenated groundwater. Uranium deposition occurred in areas where the groundwater conditions changed from oxidizing to reducing. This produced a roll front deposit with uranium concentrated at the interface between the oxidized and reduced sandstones. This interface is commonly known as the Redox Interface.

In the U.S., commercial scale recovery of uranium in ISR facilities is achieved through the use of alkaline solutions, known as the lixiviant, to mobilize the uranium in situ for recovery from wells (some historical R&D efforts have used acid based lixiviants and this is being further evaluated by one US operator today). Lixiviant solutions are stripped of uranium at the surface and the barren lixiviant is refortified and recycled through the process back into the well fields.

In the alkaline leach ISR process used in the US, groundwater is fortified with an oxidant (gaseous oxygen or hydrogen peroxide) and oftentimes an anionic complexing agent to solubilize the uranium within the ore body in situ. The oxidant converts uranium from the +4 (reduced) to the +6 (oxidized) valence state, making it amenable to complexation and solubilization. The lixiviant composition is usually maintained at a slightly alkaline pH, although early plants were operated at pH has high as 8 −9. The local geochemistry and the relative importance of calcium species establish these specifics. The uranium is extracted from the lixiviant by adsorption onto anionic resin.

The uranium is then chemically stripped from the resin and precipitated from the solution. In recent designs, the resin may be eluted directly in the ion exchange vessel or transferred to a separate elution column or tank. The uranium precipitate, formerly ammonium diuranate (e.g., using sodium or ammonium hydroxide) or more recently uranyl peroxide (using hydrogen peroxide) is conveyed to a product drying/packaging area where it is converted to the final uranium oxide product. At facilities using high temperature calciners (800 - 1000o C+), final products are typically U3O8 and/or UO2. In designs using lower temperature vacuum drying (e.g., 300 - 400 oC), the final products are typically uranyl peroxide (UO4) uranyl trioxide (UO3), their hydrates and/or combinations thereof (1,2). Some process strategies involve a final product of loaded resin or an intermediate precipitate only (satellite plant), and then ship this product to another uranium recovery facility for further processing. The final product may therefore be loaded resin, an inter¬mediate product or slurry or relatively dry oxide powder.

Radionuclide Mobilization and Associated Process Radiological Characteristics

Based on some early studies performed in the US at alkaline leach uranium solution mining plants (e.g., see 3) a relatively small percentage of the uranium progeny in the ore body is mobilized by the lixiviant and the majority of equilibrium radionuclides remain in the host formation (4). Note that such values may be process specific (e.g., alkaline vs. acid leach, pH, etc.) and may also be facility age dependent. In the US alkaline leach processes, it appears that the thorium 230 equilibrates and very little is removed by the process. The majority of the mobilized radium 226, 80-90%, estimated at 5-15% of the equilibrium radium calculated in the host formation, follows the calcium chemistry in these processes and results in radium carbonates / sulfates in calcite slurry bleed streams and associated wastes.

Additionally, the ion exchange (IX) resin used in US ISR facilities is specific for removal of uranium. Appreciable amounts of thorium and other progeny are not expected in the process downstream of the IX columns (e.g., elution, precipitation, and drying circuits). The radionuclide mixture that can potentially become airborne and result in personnel exposure and area or equipment contamination in the precipitation, drying and packaging areas would be expected to be primarily a natural uranium isotopic mixture with a relatively small progeny component. Although in growth of the first few short-lived progeny (e.g., thorium 234, protactinium 234) is occurring, the in process residence time is small relative to radionuclide half-lives and therefore time required for appreciable ingrowth. Accordingly, little contribution from these primarily beta emitters is experienced in the radiological aspects of in process materials.

In areas where solid wastes are processed, stored or during maintenance (resin tanks and columns, fabric and sand filters, clarifiers, etc.), mobilized radium 226 associated with calcium and carbonate chemistries may be an important external exposure and/or contamination source.

Additionally, during some maintenance activities when systems need to be opened and/or penetrated, aged process material may be encountered containing scale and/or precipitates in pipes, tanks, pumps, etc., which can exhibit elevated beta activity due to ingrowth of short lived thorium 234 and protactinium 234.

Additionally, large quantities of radon 222 gas can be dissolved in the lixiviant returning from under¬ground and is brought to the surface. That portion of the total dissolved radon which is above the solution's saturation value is released when encountering atmospheric pressures and temperatures and can also be released during the decay of radium contained in waste products (e.g., CaCo3 / gypsum) being processed and stored at the surface (4, 5, 6). However, despite potentially large quantities of the gas being evolved, it is "fresh radon"and the progeny equilibrium factors are typically quite low.

Principal Exposure Pathways and Associated Monitoring Requirements

The primary exposure pathways associated with ISRs were identified in the summary section and are discussed below.

Airborne monitoring for long lived radioactive dusts (LLRD) - Since ISRs are essentially an aqueous process until drying and packaging, control and containment of spills in process areas via design consideration is essential to reduce the risk of resuspension of LLRD (essentially yellowcake dusts) in these areas. Additionally, during operations, it is important to affect expedient wash down and clean up of spills to minimize dried material becoming an inhalation hazard via resuspended dusts.

Airborne monitoring for LLRD is necessary in back end process areas, e.g., beginning where the precipitate slurry is produced. Accordingly, LLRD exposure potential is primarily associated with the "yellowcake areas" of the process that include precipitation, drying and packaging. Applicable monitoring techniques include combinations of grab sampling, breathing zone sampling and continuous monitoring based on job functions and related radiological and work conditions.

External exposure monitoring - External exposure monitoring (via survey and personnel dosimetry) is required primarily in areas in which large quantities of uranium concentrates are processed, packaged and/or stored. Additionally, depending on importance of calcium chemistry in situ and therefore radium mobilization, radium build-up can occur in resin tanks and columns, filter membranes from reverse osmosis water treatment units, fabric and sand filters, clarifiers, etc., where large quantities of radium bearing calcite wastes are precipitated, processed and stored. This can result in requirements for control and monitoring of external exposure during work near these processes, during filter changes and / or maintenance of these systems.

External exposure (particularly extremity exposure) from short lived beta emitting uranium progeny (Th 234, Pa 234 e.g.) can occur during maintenance activities when systems are penetrated and / or opened. Accordingly, care should be taken and beta / gamma and/or beta exposure rate monitoring may need to be conducted on a case-by-case basis to assess degree of this potential hazard. Potential for exposure of hands and forearms during these activities must be considered, although these types of exposure events would be expected to be occasional and of relatively short duration (minutes or a few hours at a time).

Surface area contamination surveillance and control –ISRs are primarily aqueous processes until product drying and packaging. Accordingly, these back end areas are typically the most important sources of potential surface contamination and resuspension. Standard contamination controls (containment, ventilation, radiological survey [areas and personnel]) and expedient response to process upsets involving spills or other loss of containment events minimizes the potential for this pathway. Contamination surveillance and control is necessary throughout plant and ancillary areas including of personnel and for the release of equipment and materials for unrestricted use into the public domain.

Bio-assay (urinalysis) programs - As is the case with all uranium processing facilities, bioassay programs need to be designed commensurate with the metabolic characteristics of the uranium species produced. Modern ISRs in the US are producing peroxide-precipitated products dried by low temperature vacuum dryers. These products appear to be quite soluble and meet the ICRP 71 criteria for the Type F (fast) absorption category. For these products, chemical toxicity drives worker risk from intake - not radiation dose. (1,7,8,9). Accordingly, bioassay (urinalysis) programs at US ISRs involve frequent urinalysis sampling (can be weekly) and analysis for, in addition to uranium, the biomarkers associated with potential renal injury, e.g., glucose, lactate dehydrogenase (LDH) and protein albumen.

Radon and Radon Progeny - Exposure to radon gas evolving in front end process areas from uranium bearing lixivants returning from underground are typically controlled since (1) these areas are of low occupancy and typically well away from other work areas and (2) it is relatively "fresh radon"and therefore the progeny equilibrium factors are typically quite low with the potential for worker exposure also low since the vast majority of dose results from the short lived progeny and not the radon gas itself. Depending on design specifics, local exhaust systems on front end tanks and vessels are sometimes necessary to collect and remove the fresh radon gas before significant progeny ingrowth can occur in work areas. Most of the gas is released within the first few process areas, wherever first exposed to atmospheric pressure. Depending on design specifics, this can be at surge ponds and tanks, at the tops of the ion exchange columns and/or at the interface between resin loading and elution processes. Process tankage and piping may need to be enclosed and maintained under negative ventilation where practical. In warm climates such as ISR facilities in South Texas, out of doors surge ponds and/or open top ion exchange columns are often used and therefore most of the gas is released out of doors. In colder climates (Wyoming, Nebraska), the solutions are piped under pressure directly from enclosed well field valve stations and surge tanks to in plant recovery vessels including the IX tanks themselves. Some of the first generation ISR plants (1970s) used in plant IX surge tanks and up flow, open top IX columns requiring use of local exhaust systems to remove the gas from the vicinity of in-plant vessels before progeny ingrowth became an occupational exposure concern. Recent designs tend towards use of enclosed, pressurized systems for lixiviant recovery and ion exchange using local exhaust on the vessels themselves to remove radon prior to significant progeny in growth. This greatly reduces the potential for radon / progeny exposure in plant areas.

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