The New Generation of Uranium In Situ Recovery Facilities: Design Improvements Should Reduce Radiological Impacts Relative to First Generation Uranium Solution Mining Plants

S. H. BROWN, CHP, SHB INC., 7505 S. Xanthia Place, Centennial, Colorado USA 303 941 1506; shb12@msn.com

ABSTRACT

In the last few years, there has been a significant increase in the demand for Uranium as historical inventories have been consumed and new reactor orders are being placed. Numerous mineralized properties around the world are being evaluated for Uranium recovery and new mining/milling projects are being evaluated and developed. Ore bodies which are considered uneconomical to mine by conventional methods such as tunneling or open pits, can be candidates for non-conventional recovery techniques, involving considerably less capital expenditure. Technologies such as Uranium In Situ Leaching / In Situ Recovery (ISL / ISR - also referred to as “solution mining”), have enabled commercial scale mining and milling of relatively small ore pockets of lower grade, and are expected to make a significant contribution to overall world wide uranium supplies over the next ten years. Commercial size solution mining production facilities have operated in the US since the mid 1970’s.

However, current designs are expected to result in less radiological wastes and emissions relative to these “first” generation plants (which were designed, constructed and operated through the 1980s). These early designs typically used alkaline leach chemistries in situ including use of ammonium carbonate which resulted in groundwater restoration challenges, open to air recovery vessels and high temperature calcining systems for final product drying vs the “zero emissions” vacuum dryers as typically used today. Improved containment, automation and instrumentation control and use of vacuum dryers in the design of current generation plants are expected to reduce production of secondary waste byproduct material, reduce Radon emissions and reduce potential for employee exposure to uranium concentrate aerosols at the back end of the milling process.

In Situ Recovery in the U.S. typically involves the circulation of groundwater, fortified with oxidizing (gaseous oxygen e.g) and complexing agents (carbon dioxide, e.g) into an ore body, solubilizing the uranium in situ, and then pumping the solutions to the surface where they are fed to a processing plant (mill). Processing involves ion exchange and may also include precipitation, drying or calcining and packaging operations depending on facility specifics. This paper presents an overview of the ISR process and the health physics monitoring programs developed at a number of commercial scale ISL / ISR Uranium recovery and production facilities as a result of the radiological character of these processes. Although many radiological aspects of the process are similar to that of conventional mills, conventional-type tailings as such are not generated. However, liquid and solid byproduct materials may be generated and impounded. The quantity and radiological character of these by products are related to facility specifics. Some special monitoring considerations are presented which are required due to the manner in which radon gas is evolved in the process and the unique aspects of controlling solution flow patterns underground.

The radiological character of these processes are described using empirical data collected from many operating facilities. Additionally, the major aspects of the health physics and radiation protection programs that were developed at these first generation facilities are discussed and contrasted to circumstances of the current generation and state of the art of uranium ISR technologies and facilities.
INTRODUCTION

Much interest has again developed in alternative, i.e. non-conventional methods of Uranium extraction in recent years. Ore grades considerably below the economic demands of conventional techniques, i.e. underground and open pit mining, have become attractive. This is due primarily to the lower capital expenditure requirements, reduced manpower intensiveness and less environmental impact of these non-conventional methods. Uranium solution mining, or in-situ recovery, has received considerable attention and financial commitment from major international mining companies as well as the large number of “Juniors”, i.e., companies established in recent years to pursue Uranium development projects. Production facilities generating poundage from alternative Uranium recovery technologies have operated in the U.S. since the late 1960s, using ore-grade feed to milling processes as low as 6—8 ppm Uranium (Wyoming Mineral Corporation, 1977). The majority of historical solution mining interest in the U.S. has been associated with Uranium roll fronts in South Texas, Wyoming, Eastern Colorado and associated with historical conventional Uranium mining areas in New Mexico and Wyoming. The common denominator of many of these geologic settings is that the ore has been deposited by contact with reducing geochemical environments in shallow fluvial sandstone formations, confined by non-porous shale or mudstone layers above and below the uranium-bearing units.

STUDY POPULATION

The solution mining facilities from which operating data is presented in this paper used alkaline leach chemistries (ammonium carbonate and/or bicarbonate as the complexing agents) during the period of study (1975 – 1982). Development and progression of these facilities proceeded in a step-by-step fashion as a general function of wellfield size and corresponding processing plant equipment capacities and flow rates. Initial circulation of lixiviant for assessing general geochemical characteristics of the ore body usually began with 5—10 wells and a processing flow rate of approximately 100 liters / min (“test” plant). In some cases, an intermediate stage of development was used, i.e., “research and development” plant. Typical process flows were approximately 400 l/min. with several dozen to approximately 100 wells. The final stage of development was the commercial scale production facility with process flows in the range of 5000 - 12,000 l/min.and associated wellfields of several dozen acres (“production” plant). Table 1 presents the number and associated operational years of each developmental stage from which radiological monitoring data was accrued and incorporated for this paper.

<table>
<thead>
<tr>
<th>Plant development Stage</th>
<th>Number of Facilities</th>
<th>Total Study Years*</th>
</tr>
</thead>
<tbody>
<tr>
<td>Test</td>
<td>4</td>
<td>4</td>
</tr>
<tr>
<td>R&amp;D</td>
<td>2</td>
<td>3.5</td>
</tr>
<tr>
<td>Production</td>
<td>3</td>
<td>8</td>
</tr>
</tbody>
</table>

* sum of operational years under study for each facility in that development stage
**PROCESS COMPONENTS**

The solution-mining process consists of five major operational components: (1) ore body; (2) wellfield; (3) lixiviant; (4) Uranium recovery process; and (5) waste impoundment facilities and associated waste management strategies.

**Ore body**

Uranium deposits typically amenable to solution mining are usually associated with relatively shallow aquifers, about 30—150 meters subsurface, confined by non porous shale or mudstone layers. Uranium was transported to the 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 (Figures 1a and 1b). A vertical cross section of a typical uranium roll-front deposit showing the basic solution mining approach to uranium recovery is depicted in Figure 2.

![Conceptual Model of Uranium Roll Front Deposit](image)

**Figure 1a Redox Interface Showing Roll Front Deposit**
Figure 1b – Roll Front Geology

Figure 2 Roll Front Cross Section Showing Basic Mining Approach
**Wellfield**

The wellfield provides the means by which leach solution, known as the lixiviant, is circulated through the ore body to recover Uranium. Therefore, the wellfield design is crucial in maximizing the effectiveness of lixiviant confinement and utilization. The principal considerations are well spacing, injection / production well orientations and well completion methods. Figure 3 presents well completion considerations. Figure 4 illustrates a typical plant and wellfield layout and Figure 5 shows a typical “5 spot” pattern of four injection wells to one recovery (production) well.

Well spacing and orientation is influenced by the hydrologic and geochemical characteristics of the formation which limit the rate and efficiency of lixiviant circulation. Well-completion techniques contribute to vertical confinement and vertical sweep efficiency of the lixiviant through the mineralized zone. The ultimate number of injection and recovery wells comprising a wellfield is established by the desired rate of mining and the geohydrologic characteristics of the aquifer, in conjunction with the well patterns.

---

**Figure 3 Well Construction and Completion**
Figure 4 Plant and Wellfield Layout

Figure 5 Typical Five Spot Well Pattern
Lixiviant

The lixiviant reverses the chemical conditions which led to uranium deposition and thus solubilizes uranium as it is circulated through the mineralized formation. It consists of groundwater fortified with an oxidant and an anionic complexing agent. The oxidant converts Uranium from the +4 (reduced) to the +6 (oxidized) valence state, making it amenable to complexation and solubilization. In these alkaline leach solution mining projects, the lixiviant composition was 0.25 - 1.0 g/l H₂O₂ and 1.0—5.0 g/l HCO₃ at a slightly alkaline pH. Some early plants were operated at pH has high as 9. Gaseous oxygen was also used extensively in lieu of H₂O₂ as the oxidizing agent. An example of the basic mobilization chemistry in situ associated with these facilities is presented below. Other lixiviant chemistries were also investigated at that time involving, e.g. Sodium / Magnesium Carbonates and acid leach methods. In the current generation of ISR facilities, the preferred complexing agents are gaseous Oxygen with Carbon Dioxide or Sodium Carbonate (see below)

Alkaline Leach Based Facilities:
Oxidation: U⁺⁴O₂ + H₂O₂ > U⁺⁶O₃ + H₂O or U⁺⁴O₂ +O₂ > 2U⁺⁶O₃
Leaching: UO₃ + (NH₄)₂CO₃ + H₂O > (NH₄)₂UO₂(CO₃)₂ + H₂O₂

Current Generation of ISL/ISR:
Oxidation: 2U⁺⁴O₂ + O₂ (gaseous) > 2U⁺⁶O₃
Leaching: UO₃ + X(HCO₃)₂ > XUO₂(CO₃)₂ + H₂O
(where X is any monovalent or divalent cation, typically Na₂CO₃ or NaHCO₃)

These uranium-recovery processes consisted of four basic process circuits: (1) lixiviant/sorption circuit; (2) resin transfer circuit; (3) elution/precipitation circuit; and (4) product drying and packaging. ( see figures 6 thru 8)

Figure 6 Process Flow Diagram
Figure 7 Process Schematic – First Generation Alkaline Leach Plants

Figure 8 Process Schematic – Modern Designs Including Satellite Plant Concept
In the lixiviant/sorption circuit, Uranium is extracted from the recovered lixiviant by adsorption onto anionic resin. The lixiviant is then refortified and reinjected into the mineralized formation. In older plants operated at higher pH, some provision for calcium control was incorporated to reduce Calcium Carbonate precipitation in the fortified lixiviant which, if not removed, could plug up the wells and reduce formation permeability. In modern plants operated at about pH 7, calcium carbonate precipitation appears to be less problematic. The degree of Calcite precipitation is site-specific and is related to the geological formation and lixiviant chemistry chosen. More will be said about this Calcite byproduct later.

The lixiviant/sorption circuit and the elution/precipitation circuits of the first generation facilities were interconnected by means of a resin transfer system only. In the elution/precipitation circuit Uranium is chemically stripped from the resin and precipitated from solution (typical concentrations in the pregnant eluant are 8 – 20 grams / liter Uranium, and have been seen as high s 40 grams Uranium / liter – see U.S. Nuclear Regulatory Commission, 2001). In current designs, the resin may be eluted directly in the ion exchange vessel, or as was the case with these facilities, transferred to a separate elution column or tank. The Uranium precipitate, Ammonium Diuranate or Uranyl Peroxide, depending on the precipitation chemistry, may then be conveyed to a product drying/packaging facility where it is converted to the final UO$_3$ / U$_3$O$_8$ product. The barren supernate is refortified and recycled through the elution/precipitation circuit.

Currently, some process strategies involve a “final product” of loaded resin or an intermediate precipitate only (“satellite plant”), and then shipping this product to another facility for further processing. The final product may therefore be loaded resin, an intermediate product or slurry, rather than a UO$_3$ / U$_3$O$_8$ / UO$_2$ powder (Brown, S. and Blauer, M., 1980).

Waste Impoundment and Management

Various amounts of liquid and/or solid wastes may be generated by these processes. Potentially large volumes of liquid waste may need to be impounded resultant from over recovery in the wellfield and for process chemistry control. Over-recovery (bleed stream), i.e. recovery of several percent greater volume than is reinjected into the formation, is typically necessary to maintain a net inward movement of groundwater into the mineralized zone for solution control in situ. This results in continuous liquid bleeds from process which were impounded at the surface in large holding ponds. Surface impoundments were typically equipped with chlorinated polyethylene liners, depth gauges and underground leak-detection systems. The radiological character of the impounded liquid wastes is site specific but typically includes small amounts of residual Uranium and Radium 226 concentrations on the order of 20—110 Bq/l. Volumes were controlled via enhanced evaporation techniques, e.g., brine concentrators. Ultimate disposition of these fluids involved chemical treatment, e.g., reverse osmosis, to reduce parameters to near baseline values and re-injection during site restoration and decommissioning activities. Currently, an alternative method for disposition of these fluids is the use of deep well disposal into an aquifer that has been previously determined by regulators as unsuitable for drinking water use. Additionally, some facilities manage restoration fluids by disposal via irrigation as
shown in Figure 8 below.

Figure 8: Irrigation as Waste Management

Some facilities also generated a solid waste which was removed from the process and similarly impounded at the surface. Depending on site-specific formation characteristics and lixiviant chemistries, variable amounts of a Calcite (CaCO₃) precipitate was formed in the process. The degree of precipitation was site specific and related to the local importance of Calcium chemistry and choices of pH control if any. Modern in-situ operations operate at around pH seven (7) preventing calcite precipitation problems except at points in the process experiencing sharp pressure drops. Older high pH Ammonium Carbonate operations experienced calcite precipitation since they operated at a pH higher than that of the formation water. Use of high pH ammonium carbonate lixiviants caused initial precipitation of calcium carbonate unless the formation was pre-treated with water low in total dissolved solids (TDS). Modern in-situ uranium recovery operations operate at or near the formation water pH with less calcium carbonate precipitation.

As mobilized Radium will follow the Calcium chemistry in the process, this by-product, if important at a particular site, will invariably contain the majority of mobilized Radium 226 byproduct material as Radium Carbonates and Sulfates, coprecipitated with the Calcium Carbonates. Depending on site and process specifics, these precipitates may need to be removed from the process to prevent well plugging and reduction in formation permeability. They were typically impounded at the surface in ponds similar to the liquid waste evaporation ponds, in tanks, or drummed as produced. Although the total specific activity (Bq/g) of this material is typically less than conventional mill tailings, since it is almost exclusively Uranium and Radium 226, nonetheless the material must be considered as 11(e).2 byproduct material and requires disposition as such. In the facilities studied, the radiological character of this material typically involved several hundred ppm Uranium and 10-110 Bq/g Radium 226.
RADIONUCLIDE MOBILIZATION

A relatively small portion of the Uranium daughter products in the ore body are actually mobilized by the lixiviant. The vast majority of secular equilibrium radionuclides remain in the host formation. Table 2 presents typical concentration ranges for the facilities studied in the processing plant feed stream as well as the refortified tails being returned to the ore body. However, it should be noted that such values are probably process specific and may also be facility age dependent. It appears that the Thorium 230 will equilibrate and very little is actually removed by the process. The majority of the mobilized Radium 226 (80—90 percent) which is 5~15 percent of the calculated equilibrium Radium in the host formation, follows the Calcium chemistry in the process and resulted in Radium Carbonates / Sulfates in the Calcite slurry bleed stream.

| TABLE 2: Radionuclide Concentrations in Process Streams (Bq/l) |
|------------------|------------------|------------------|
| Th 230 | Ra 226 | Pb 210 |
| Pregnant Lixiviant (returning from underground) | 56-93 | 10 - 150 | <1 |
| Barren Lixiviant (being reinjected) | 48 -81 | 1.9 - 4.4 | <1 |

It appears that little, if any Lead 210 is mobilized as the Lead Carbonate complexes formed in situ are virtually insoluble in the alkaline lixiviant processes studied. In addition to the species just discussed, variable amounts of Radon 222 gas are brought up from underground dissolved in the lixiviant. This subject will be treated in some detail in a later section of this paper.

RADIOLOGICAL CONSIDERATIONS

The radiological circumstances, controls and monitoring requirements of Uranium solution mining processes are associated with four broad categories:

1. Hydrologic/groundwater controls and monitoring of solution flow patterns underground.
2. General environmental impact assessment and monitoring.
3. Special considerations for Radon evolution mechanisms.
4. Operational health physics and radiation protection programs

HYDROLOGIC/GROUNDWATER CONTROLS AND MONITORING

The geologic siting of the ore body, i.e. relatively permeable sandstone confined by shale or mudstone and the subsequent control of groundwater movement are basic to the mining operations and to environmental impact management and control. Injection and recovery well
flows must be balanced so that chemical solutions do not migrate out of the mining zone resulting in excursion conditions. As has been previously discussed, the radiological character of the solution returning from underground is composed of low-level concentrations of radioactive species. The average Uranium concentration is typically less than 0.1 weight percent (1000 ppm). Environmentally speaking, migration control is primarily for chemical control, rather than for radiological species, since parameters of concern include Selenium, Arsenic and Molybdenum.

To detect solutions that have potentially migrated out of the mining zone, monitor wells must be placed in the geologic unit being mined and in other geologic units that must be protected. These wells, encircling the wellfield within 100 – 200 meters of the mining unit, are used to detect potential migrations from the wellfield so that expedient detection and corrective actions can be taken to draw back the lixiviant into the mining unit. Additionally, trend wells are often drilled into the mining zone between the monitor wells and the mining unit. Trend wells are used to monitor solution and water table levels and detect potential migrations earlier. Should upper and/or lower aquifers exist that need to be protected, monitor wells are drilled into the first aquifer above and/or below the mining unit.

In general, migrations from the mining zone would be the result of well-field (hydrologic) imbalance, the presence of previously undetected faults or fractures resulting in undesirable communications between aquifers, or previously unidentified well-completion problems (cracks in casing; incomplete cementing; old and poorly abandoned bore holes, etc.). Once a possible migration is indicated, mining unit and trend monitors would be used to determine the extent of the migration, if any, by observing water level and chemistry changes. Under routine operating conditions, a bleed stream, which is often taken at the processing plant, is conveyed to a water reservoir or to tankage. This provides for a net inflow of groundwater into the wellfield aiding in hydrologic control. However, in the event of migration, several corrective action procedures are available to contain and correct migration of leachate. These methods may be applied to a few wells within a single production cell, or to the entire wellfield as the situation dictates. Examples are as follows:

**Overpumping:** This method involves adjusting pumping so that the rate of flow into the injection wells is exceeded by the flow from the production (extraction) wells. The net result is a general inward movement of native water.

**Reordering:** This is a variation of overpumping in that different ratios are applied to different areas in the wellfield. Hence, the inward movement of native water may be emphasized at one point or another. Reordering may further include direct pumping from one part of the field to another.

**Reducing injection:** This is the second way to adjust the ratio of recovery flow to injection flow. At the same time, it reduces the amount of leach solutions introduced into the production zone in the vicinity of the wells concerned.

**Ceasing pumping:** This method stops both the injection and recovery flows. Exclusive of the effects of natural forces (e.g. natural migration of groundwater, which are orders of magnitude smaller) this should arrest the further migration beyond the established boundaries.

**Beginning restoration:** Restoration essentially means returning natural groundwater to conditions which are acceptable for the long term to the appropriate regulatory agencies
and typically must be restored to conditions consistent with “original designated use” of the groundwater. It is currently performed in stages as well field sections become “mined out”. Additionally, it is of course a major component of final site restoration and decommissioning prior to license termination although it is an alternative available as a migration corrective action procedure.

Restoration techniques involve pumping residual fluids from the wellfield and applying conventional water-treatment processes. The fluids are chemically treated on the surface to concentrate contaminants into a small volume brine. The treated water may be returned to the aquifer or discharged. The radiological constituents of the brine, primarily Radium 226, can be removed via standard treatment methods (e.g. barium-sulfate precipitation, reverse osmosis, etc.) and/or disposed of as a small volume byproduct material (low level waste).

GENERAL ENVIRONMENTAL IMPACT ASSESSMENT AND MONITORING

The monitoring programs required to quantify effluents and environmental impact at these solution mining facilities were typical of the environmental requirements for conventional mills and were consistent with the requirements of U.S. NRC Regulatory Guide 4.14 (U.S. Nuclear Regulatory Commission, 1980b) and/or applicable state regulations and the conditions of the facility’s radioactive material license. Typical considerations in this regard included:

1. Radon releases from evaporation and surge ponds / tanks, top of ion exchange columns, in plant tank local exhaust system release points, etc.
2. Radionuclide particulates in air at site-boundary locations, specifically for Uranium (natural), Thorium 230, Radium 226 and Lead 210 (particularly if drying/calcining steps are involved), and passive monitoring for Radon 222.
3. Isokinetic type sampling of dryer / calciner scrubber exhaust and other particulate point source release locations.
4. Soil, vegetation, food crops in site environs for same species as for air particulates.
5. Radon sampling at site boundary locations.
6. Offsite sampling of surface and groundwater potentially impacted by site activities.
7. Direct radiation measurements at site boundary locations.

Note that several technical advances in recent designs are expected to reduce both the Radon and radionuclide particulate emissions relative to the observations and measurements made at these first generation ISL / ISR facilities. Specifically, current designs tend toward (1) use of “enclosed systems” in the recovery and ion exchange process steps thereby reducing opportunities for points of Radon release and (2) use of vacuum dryers (rather than higher temperature calciners) with little, if any expected particulate emissions.

RADON EVOLUTION MECHANISMS

A great deal of information has been published over the years on Radon gas and its daughter products associated with occupational exposure in underground mines and potential environmental impacts from surface tailings impoundments. In solution mining processes,
although the characters are the same, the relative roles they play can be different. It appears that the majority of Radon which is released at the surface is not, as at a conventional mill, a result of on-surface decay of Radium over time. The Radon is brought to the surface dynamically, dissolved in the lixiviant returning from underground. Just as dynamically, that portion of the total dissolved Radon which is above the solution's saturation value, is released when encountering atmospheric pressures and temperatures. The fact that the vast majority of Radon released results from dynamic plant operation rather than from surface impoundments was quantified and verified in Brown and Smith, 1982.

Although Radon is considered minimally soluble in water at standard conditions, the physical and geochemical environments in situ apparently enhance the amount of gas the fluids can carry by several orders of magnitude. An empirical model was developed to measure the Radon release rate and was applied to a commercial in situ leach facility (Brown and Smith, prev. cit.). Data for this facility indicated a source term of $10^{12} - 10^{13}$ Bq/yr at an average recovery flow rate of 3000 l/min. This parameter may be site-specific and is probably related to ore grade, formation characteristics and other factors. On a Bq released per Kg $U_3O_8$ recovered basis, this is approximately 50 percent of the model mill case described in the U.S. NRC Generic Environmental Impact Statement (GEIS) for Uranium Milling (U.S. Nuclear Regulatory Commission, 1980a). Table 3 presents Radon and Radium values as measured in various process components. The gas will evolve from solution in the early stages of the process as it becomes exposed to the surface environment. Depending on the limitations at a site, points of evolution could involve evaporation and/or surge ponds, in-plant surge tanks and tops of absorption columns, or combinations thereof. Monitoring is therefore necessary for environmental impact assessment, to define appropriate engineering controls (local exhaust systems e.g.), to assess occupational exposure conditions and to verify that exposures are maintained as low as is reasonably achievable (U.S. Nuclear Regulatory Commission, 2002)

<table>
<thead>
<tr>
<th>Process Stage/ Location</th>
<th>Ra 226 *</th>
<th>Rn 222*</th>
</tr>
</thead>
<tbody>
<tr>
<td>Circulating Lixiviant</td>
<td>3 - 20</td>
<td>300 - 7000</td>
</tr>
<tr>
<td>Calcite In Clarifiers</td>
<td>30 - 100**</td>
<td>N/A</td>
</tr>
<tr>
<td>Evaporation Ponds In Solution</td>
<td>20 - 30</td>
<td>Equilibrium Assumed</td>
</tr>
<tr>
<td>Evaporation Ponds, Sludge</td>
<td>30 - 45</td>
<td>Equilibrium Assumed</td>
</tr>
</tbody>
</table>

*Bq/l except ** which is in Bq/g CaCO$_3$*
OPERATIONAL HEALTH PHYSICS AND RADIATION PROTECTION PROGRAMS

Numerous historical assessments appear in the literature which describe the health physics monitoring and radiation protection programs associated with conventional Uranium mining and milling (International Atomic Energy Agency, 1976; Federal Radiation Council, 1967; Rock et al., 1971; American National Standards Institute, 1973; Schiager, Borak and Johnson, 1981). The radiation protection program requirements at a ISR facility are very similar and, in many cases, identical to those of a conventional mill (see Brown, 1982):

1. Airborne monitoring for long lived alpha emitters (U, Th) in appropriate process areas, primarily drying / calcining and packaging areas including combinations of grab sampling, breathing zone sampling and continuous monitoring techniques;

2. Surface area contamination surveillance and control throughout plant areas

3. Respiratory protection programs if necessary

4. Bio-assay (urinalysis) programs appropriate for the Uranium products to which employees are potentially exposed (product specific solubility characteristics can have metabolic implications for bioassay – see e.g., Cook and Holt, 1974; Eidson and Mewhinney, 1980; Brown and Blauer, 1980; U.S. Nuclear Regulatory Commission, 1988)

5. Work control and training via formalized procedures

6. Internal audit and quality control programs to ensure execution of safe work practices and regulatory compliance

7. Radon/daughter monitoring, particularly at front end of process where Radon is most likely to evolve from solutions returning from underground

8. External exposure monitoring primarily in areas in which large quantities of Uranium concentrates (ADU, Uranyl Peroxide, U₃O₈) are processed, packaged and/or stored. Additionally however, at the facilities comprising the study population of this paper, Radium 226 build-up occurred in resin columns, sand filters and clarifiers, etc., resulting in requirements for control and monitoring of external beta / gamma exposure during the maintenance of these systems.

Frequencies and details of program implementation are, of course, a function of applicable regulatory requirements, process specifics and necessary good health-physics practices (e.g., see Rich et al., 1988; U.S. Nuclear Regulatory Commission, 1983). For example, at some of the ISL facilities being developed today, the process will truncate at resin loading or at an intermediate precipitate with further processing to take place at another facility elsewhere. Accordingly, the radiation protection program should be commensurate with the degree of radiological hazard specific for the facility design and operational strategies.

As described previously, solubilized Radon will be brought up dynamically from underground with the Uranium bearing solutions. A portion of the dissolved gas may be released within the first few process areas, primarily surge ponds and tanks and/or at the tops of the absorption columns themselves. In the case of facilities in the warm southern U.S. (Texas, e.g.) out-of-doors, large surface area surge ponds were used and therefore very little gas remained in solution by the time the pregnant lixiviant reached in plant areas. On the other hand, severe Wyoming winters in the northern U.S. prohibited exposure of the solutions to the atmosphere.
out-of-doors and were piped directly from enclosed wellfield valve stations and surge tanks to in-plant recovery tanks. Local exhaust systems were required to remove the gas from in-plant vessels before it became an occupational exposure concern. Monitoring may therefore be required to evaluate engineering and health-physics needs within general plant areas, and particularly prior to entrance into enclosed areas and tanks which can be subject to significant radon daughter build-up. It should also be noted however, and as previously discussed, current designs tend toward use of enclosed systems for lixiviant recovery and ion exchange. Accordingly, less opportunity should be available for points of Radon release, either into the environment or within plant work areas.

In any individual facility the level of effort required to assess and control Radon and daughter exposure conditions, both in terms of monitoring frequencies and ventilation needs, must be evaluated on a site-specific basis and can change within a facility as design or operational conditions change. The mechanics of Radon solubilization in situ may involve a complex combination of factors including the geochemical characteristics of the ore body, lithographic texture of the sandstone, depth of, temperature and pressure in the ore zone, as well as the details of the lixiviant chemistry itself.

To summarize the Radon/daughter occupational exposure circumstances at these facilities, the following observations were made. Under differing circumstances and conditions, both Radon gas as well as its short lived daughter products (Po$^{218}$, Pb$^{214}$, Bi$^{214}$, Po$^{214}$) can be important concerns in occupational health physics programs. A great deal of information exists in the literature confirming that the daughters of Radon are considerably more important than the Radon parent in contributing to respiratory track dose (Coleman et al., 1956; Holaday et al., 1957; Jacobi, 1964; Altschuler et al., 1964; Schiager and Dahl, 1968). Extensive Radon and Radon daughter monitoring at these ISL facilities indicated that severe disequilibrium can be encountered between Radon and its daughters at both ends of the continuum. Situations were observed in which significant concentrations of Radon gas as high as $10^3 - 10^4$ Bq/liter in air) in the absence of significant levels of Radon daughters were measured in general plant areas. Conversely, situations were identified (e.g. within enclosed, poorly ventilated areas, tanks, etc) in which several tens of working-level concentrations of Radon daughters were measured in the absence of comensurate levels of Radon gas. Ventilation conditions obviously play a large role in determining this relationship. Monitoring considerations, particularly during startup phases and when operating conditions change, should include both the measurement of Radon daughters (for accurate assessment of potential dose commitments) as well as for Radon gas to adequately identify potential sources so that necessary engineering controls can be implemented.
IMPROVEMENTS IN MODERN DESIGNS

Modern ISR designs are expected to reduce radiological emissions and potential for employee exposure relative to first generation ISR facilities due to several important advancements:

1. Tendency toward enclosed systems at front end (lixiviant circulation, resin loading and elution) providing less opportunity for Radon gas release
2. Use of vacuum dryers rather than calciners should reduce effluent releases of yellowcake product to insignificant levels
3. The lower operating temperature of vacuum dryers relative to calciners is expected to produce a more soluble final Uranium product (more UO₃, less U₂O₈ in mixture) with less potential for longer term pulmonary retention and resultant dose. However, exposure limits based on chemotoxicity, rather than radiotoxicity, must be considered for the more soluble uranium products
4. Less concern or need for pH control of lixiviant relative to first generation alkaline leach processes since modern plants typically operate at pH of approximately seven rather than higher pH lixiviant chemistries of older plants. This should reduce the importance of Calcium and Radium chemistry and therefore less Radium should be mobilized from the host formation producing less volume with lower Radium concentrations of 11(e).2 byproduct material.

CONCLUSIONS

1. Special considerations are dictated by the in situ recovery technology related to hydrologic control of solutions underground and the management of large volumes of liquid at the surface.

2. Some unique radiological aspects of ISRs result from the characteristics of Radon solubility in – situ and resultant gas evolution mechanisms at the surface.

3. Conventional mill tailings are not generated by the ISR technology

4. However, solid LLW, “11(e). 2 byproduct materials”, can result, primarily from process specific aspects of Radium chemistry and mobilization

5. Health Physics and radiation protection programs required for ISRs are similar to conventional mills as well as any Uranium facility processing and manufacturing industrial U compounds with natural U enrichment. However, the degree of robustness and specifics should be dictated by individual facility designs.
SUMMARY

This paper has presented an overview of in situ Uranium recovery processes and associated major radiological aspects and monitoring considerations. Admittedly, the purpose was to present an overview of those special health physics considerations dictated by the in situ Uranium recovery technology, to point out similarities and differences to conventional mill programs and to contrast these alkaline leach facilities to modern day ISR designs. As evidenced by the large number of ISR projects currently under development in the U.S. and worldwide, non conventional Uranium recovery techniques, such as ISL / ISR (solution mining), can play a significant role in complimenting Uranium supplies during the next decade.

REFERENCES


Eidson, F and Mewhinney, J., 1980. In Vitro Solubility of Yellow Cake Samples from Four Uranium Mills and Implications for Bioassay Interpretation, Health Physics 39, 893-902


