

**Radiological Aspects of U.S. Uranium In Situ Recovery Facilities (An Update)
and Some Current Health Physics Issues**

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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. In the US currently, there are five producing uranium milling facilities, of which four are In Situ Recovery (ISR). Additionally, the USNRC is currently or soon will be reviewing as many as 10 new license applications for uranium ISRs and an application for a new ISR is expected to be submitted in early 2010 to the Agreement State of Colorado. Commercial size ISRs (formerly referred to as in situ leach or uranium solution mining facilities) have operated in the US since the early 1970's.

Current designs are expected to result in less radiological wastes, less worker dose and smaller radiological emissions relative to "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, which used open to air recovery vessels and high temperature calcining systems for final product drying. Improved containment, automation and instrumentation control and use of lower temperature vacuum dryers in the design of current generation plants are expected to reduce production of secondary waste byproduct material, reduce radon and radionuclide particulate emissions and reduce potential for employee exposure to uranium concentrate aerosols at the back end of the milling process.

However, since there has been a very low level of activity in the US uranium recovery industry for several decades until recently, standards of health physics practice at uranium recovery facilities including ISRs have remained essentially unchanged since the early 1980s. Additionally, some of the more important USNRC regulatory guides that address "acceptable" operational health physics methods for these facilities have not been revised in many years.

This paper summarizes the operational aspects of modern ISRs, describes the radiological character of these processes and the major aspects of the health physics and radiation protection programs appropriate for these facilities. As license applications for new facilities are being prepared by applicants and reviewed by regulatory staff, several health physics aspects specific to the ISR technology are currently being revisited. These include considerations of the chemical and physical characteristics of ISR uranium products that define radiological properties including:

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- The expected radionuclide mixture mobilized and brought to the surface from the in situ environment and ultimately passed forward to uranium recovery and processing systems (i.e., degree of “radionuclide equilibrium” associated with the recovered uranium and its progeny).
- Chemical form of the uranium compounds produced (UO_4 , UO_3 , U_3O_8 , etc) and impact on establishing bioassay programs that adequately protect workers.
- Solubility of these products as related to human metabolism, dose delivery and potential chemotoxicity of the more soluble compounds.
- Appropriate airborne exposure concentration limits based on solubility characteristics, e.g., establishing appropriate Derived Air Concentration limits (DAC) and Annual Limits of Intake (ALI).
- Appropriate contamination limits for release of equipment and materials to unrestricted areas

The paper presents the limited data from operating plants that have been published in the literature going back over 30 years and related perspectives that result from “process knowledge” to support current (and longstanding) assumptions inherent in the above health physics issues. Suggested resolutions and their associated technical basis are provided for consideration by applicants, licensees and regulatory agency staffs.*

* The opinions and recommendations presented herein are exclusively those of the author and do not necessarily reflect the official position of the USNRC Uranium Recovery Branch nor the views of current source material licensees or applicants

INTRODUCTION TO THE URANIUM ISR MINING AND MILLING PROCESS

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 again 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 (ISR), has received considerable attention and financial commitment from major international mining companies as well as a large number of “Juniors”, i.e., companies established in recent years to pursue mineral development projects. Production facilities generating poundage from alternative uranium recovery technologies have operated in the U.S. since the early 1970s, 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, and 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.

Uranium deposits typically amenable to ISR methods are usually associated with relatively shallow aquifers, about 30—150 meters sub 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 (Figure 1). A schematic of a typical uranium roll-front deposit showing the basic solution mining approach to uranium recovery is depicted in Figure 2.

FIGURE 1: Redox Interface Showing Roll Front Deposit

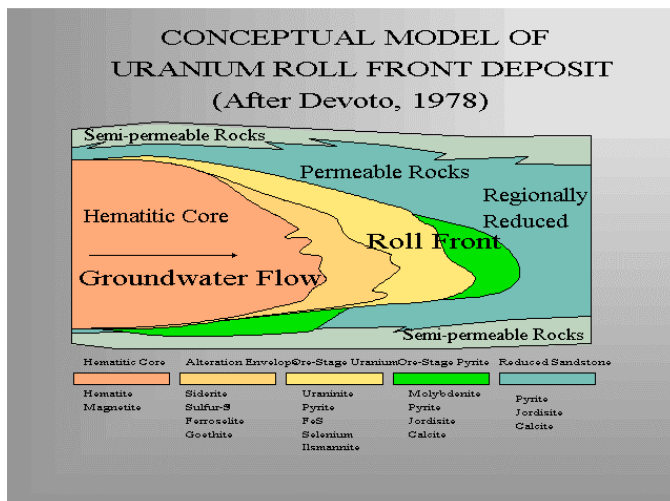


FIGURE 2: Basic Approach to Uranium In Situ Mining Method

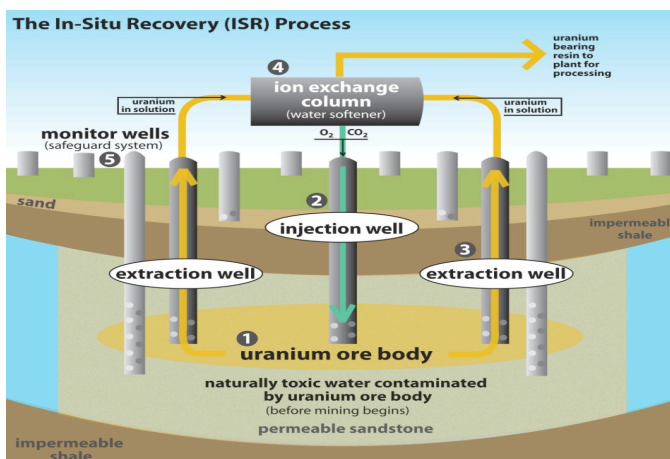


FIGURE 4: Uranium ISR Arial View



Radiological Waste Streams

Various amounts of liquid and/or solid wastes may be generated by these processes. Potentially large volumes of liquid waste may need to be dispositioned resultant from over recovery in the well field and for process chemistry control. Over-recovery (bleed stream), i.e. recovery of several percent greater volume than is re-injected 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 for which the preferred management method is deep well disposal (permitted by USEPA) into an aquifer that has been previously determined by regulators as unsuitable for drinking water use.

ISR facilities also typically generate a solid waste which must be removed from the process. Depending on site-specific formation characteristics and lixiviant chemistries, variable amounts of a calcite (CaCO_3) precipitate can be formed in the process. The degree of precipitation is site specific and related to the local importance of calcium chemistry and choices of pH control if any. Modern in-situ uranium recovery operations operate at or near the formation water pH with less expected calcium carbonate precipitation than was experienced by older first generation alkaline leach plants operating at higher pH and producing more calcite (Brown 2008, 2009)

As mobilized radium will follow the calcium chemistry in the process, this by-product will invariably contain the majority of mobilized radium 226 as radium carbonates and sulfates, co precipitated with the calcium compounds. The total specific activity (Bq/g) of this material is typically less than conventional mill tailings and is almost exclusively uranium (in natural isotopic abundances) and radium 226, Accordingly, the material must be considered as 11(e).2 byproduct material and requires

disposition as such under the Atomic Energy Act. In the first generation alkaline leach plants, the radiological character of this material typically involved several hundred ppm uranium and 10-110 Bq/g Radium 226 (Brown 1982)

RADIONUCLIDE MOBILIZATION

A relatively small percent of the uranium daughter products in the ore body is actually mobilized by the lixiviant. The vast majority of secular equilibrium radionuclides remain in the host formation. Results of measurements made of radionuclides of interest in various process streams are presented in Tables 1 and 2 (from Brown 1982). 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 was 5~15 percent of the equilibrium radium calculated to be in the host formation, followed the calcium chemistry in the process and resulted in radium carbonates / sulfates in the calcite slurry bleed stream and other associated 11(e).2 wastes.

TABLE 1: 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 re-injected)	48 -81	1.9 - 4.4	<1

TABLE 2: Typical Radium and Radon Concentrations in Process

Process Stage/ Location	Ra 226 *	Rn 222*
Circulating Lixiviant	3 - 20	300 - 7000
Calcite In Clarifiers	30 - 100**	N/A
Evaporation Ponds In Solution	20 - 30	Equilibrium Assumed
Evaporation Ponds, Sludge	30 - 45	Equilibrium Assumed

*Bq/l except ** which is in Bq/g CaCO₃

It appears that little if any lead 210 is mobilized as the lead carbonate complexes formed in situ were 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.

Radon Evolution Mechanisms

A great deal of information has been published over the years on the circumstances of radon gas and its progeny in conventional uranium recovery facilities associated with occupational exposure in underground mines (Coleman et al., 1956; Holaday et al., 1957; Jacobi, 1964; Altshuler et al., 1964; Schiager and Dahl, 1968) and potential environmental impacts from airborne effluents from mill tailings surface impoundments. (USNRC 1980). 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 (Brown and Smith 1982, Texas Department of Health 1982; NMA 2008, USNRC 2008). The occupational exposure implications of this are discussed below.

OPERATIONAL HEALTH PHYSICS PROGRAMS – HISTORICAL STANDARDS OF PRACTICE AND ASSUMPTIONS WHICH MAY REQUIRE REASSESSMENT, NEW DATA AND/OR UPDATES

Numerous examples appear in the literature which describe the health physics monitoring and dose assessment methods appropriate and necessary for 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, USNRC 2002a, 2002b). The radiation protection program requirements at an ISR facility are very similar and, in many cases, identical to those of a conventional mill (comparison in Brown, 2007) and are summarized below.

- Airborne monitoring for long lived alpha emitters in appropriate process areas, primarily drying and packaging areas including combinations of grab sampling, breathing zone sampling and continuous monitoring techniques
- Surface area contamination surveillance and control throughout plant and ancillary areas
- Bio-assay programs appropriate for the uranium products to which employees are potentially exposed since product specific solubility characteristics can have metabolic implications for bioassay
- Radon / progeny monitoring, particularly at front end of process where radon is most likely to evolve from solutions returning from underground
- External exposure monitoring primarily in areas in which large quantities of uranium concentrates are processed, packaged and/or stored. Additionally radium build-up can occur in resin tanks and columns, fabric and sand filters, clarifiers, etc., resulting in requirements for control and monitoring of external exposure during the maintenance of these systems.
- Work control through formal training, use of operating and radiation protection procedures including radiation work permit processes
- Internal audit and quality control programs to ensure execution of safe work practices and regulatory compliance

Table 3 identifies operational health physics program elements and issues for which historical measurement and calculational methods and associated technical basis are being revisited and/or re validated since the first generation of ISRs began operating in the 1970s and 80s. These “health physics issues” have been recently identified and are being discussed in the context of the source material license review process between USNRC staff and their license applicants. (Go to USNRC ADAMS data base and web site - search under specific source material license applicant or ISR project name). In the discussions which follow, each issue is discussed in the context of current

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practices and their associated technical basis. These form the precedent for what has been historically assumed necessary and sufficient to ensure compliance to the applicable sections of NRC regulations, provide an adequate “standard of care” and meet ALARA objectives. Conclusions, including suggestions for resolution of each issue are also presented.

TABLE 3: Current ISR Health Physics Issues

ISSUE	USNRC COMPLIANCE REFERENCE	CONCERN
1. Air sampling in process areas for long lived alpha emitters	10 CFR 20.1204(f)	Is analysis for just natural U in air adequate or do we need to consider “mixture” of Unat + Th 230 + Ra 226 + Pb 210?
2. Direct Radon 222 gas monitoring, in addition to progeny monitoring, in up front process areas (potential for severe disequilibrium with progeny)	10 CFR 20.1204(f)	Is this necessary in addition to traditional monitoring of particulate progeny via historical methods (e.g., Kusnetz 1956, Thomas 1972)?
3. Release / contamination limits for equipment and materials for “unrestricted” (public) areas	NRC Regulatory Guide 8.30, <i>Health Physics Surveys In Uranium Recovery Facilities</i>	Are higher limits for “uranium and associated decay products” adequately protective or are lower limits necessary for the radium 226 in 11(e). 2 byproduct wastes?
4. Solubility (metabolic) of uranium ISR products as related to appropriate worker airborne exposure limits and bioassay programs	10 CFR 20, App B 10 CFR 20.1204 10 CFR 20.1201(e)	What is proper solubility classification for ISR products? Are UO ₃ / UO ₄ products ICRP class F (D) or M (W) vs. S (Y) as were the “high fired” U ₃ O ₈ calcined products of first generation ISRs
5. Bioassay Programs for ISR products	10 CFR 20.1204	Related to issue # 4, does it need to be empirically demonstrated that ISR U products are “low fired” as defined in NUREG 0874 (i.e., “soluble” in lung - ICRP Class D/W) and therefore appropriate to use a monthly urinalysis frequency and associated action levels from Tables 1 of Regulatory Guide 8.22 w/o regular in – vivo lung measurements?
6. External exposure Monitoring	10 CFR 20.1201 Regulatory Guide 8.30, <i>Health Physics Surveys In Uranium Recovery Facilities</i>	Is there a “beta component” that also needs to be measured to assess shallow dose equivalent to workers in addition to routine monitoring of the deep dose equivalent from gamma exposure?

1. Air sampling in process areas for long lived alpha emitters

It is important to recognize the radiological environment of a modern ISR as related to the potential radionuclides of concern that could become airborne. Studies performed in the late 1970s and early 1980s of radionuclide mobilization from several ISRs and subsequent measurements at operating ISRs indicate a relatively small portion of the uranium daughter products in the ore body are actually mobilized by the lixiviant. (See previous discussion on radionuclide mobilization, Tables 1 and 2)

In addition to the fact that very little of these uranium daughter products are mobilized in situ, the ion exchange (IX) resin used in ISR facilities is specific for removal of uranium. Thorium compounds are not removed by the IX resin and are therefore not expected in the process downstream of the IX columns (e.g., elution, precipitation, and drying circuits). Accordingly, the “nuclide mix” that can potentially become airborne in the precipitation, drying and packaging areas of a modern ISR is expected to be almost exclusively U nat. In growth of the first few short lived daughter products (Thorium 234, Protactinium 234) takes approximately 4 months to reach equilibrium and therefore is not expected to be associated with relatively fresh product as would be the case in an operating ISR plant.

Additionally, it should be noted that in accordance with 10 CFR 20.1204(g), nuclides can be ignored in a mixture in air if the total activity in the mixture is used to determine compliance with 20.1201 and 20.1502(b) and any nuclides ignored are < 10% of the mixture and the sum of all nuclides ignored are < 30% of the mixture. For modern ISRs, these conditions are expected to be met.

Conclusion: In order to establish that natural uranium isotopes are the exclusive alpha emitting radionuclides of concern in airborne particulate samples at ISRs, it should be relatively easy and straightforward to collect composite samples (long sampling times to maximize collected material) from several representative air particulate monitoring locations. These sample locations should adequately characterize various points in the process (e.g., lixiviant, precipitation, and drying/packaging areas). Samples should be analyzed for U nat (total uranium) Th-230 and Ra-226. Then compare the results with the mixture exclusion conditions defined in 10 CFR 20.1204(g) to ensure that the appropriate Derived Air Concentration (DAC) limits from 10 CFR 20 Appendix B Table 1 are used. If a mixture is present greater than the 10 CFR 20.1204(g) exclusion, a “sum of fractions rule” can be applied to establish the appropriate DAC.

2. Direct Radon 222 gas monitoring, in addition to progeny monitoring, in up front process areas (potential for severe disequilibrium with progeny)

At ISRs, radon is 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, at the tops of the absorption columns if these components are used in a particular design and/or at the interface between resin loading and elution processes (resin shaker screens, e.g.). In the case of facilities in the warm southern U.S. (Texas, e.g.) out-of-

doors open top IX columns are still used and therefore most of the gas is released out of doors. On the other hand, severe Wyoming winters in the northern U.S. prohibit exposure of the solutions to the atmosphere out-of-doors and are typically piped under pressure directly from enclosed well field valve stations and surge tanks to in plant recovery vessels including the IX tanks themselves. In the first generation ISR plants that used in plant IX surge tanks and up flow, open top IX columns, local exhaust systems were required to remove the gas from in-plant vessels before it became an occupational exposure concern. In these early designs, general area monitoring for radon 222 gas, in addition to particulate progeny, was necessary to evaluate engineering and health-physics needs within general plant areas. Current designs tend toward use of enclosed, pressurized systems for lixiviant recovery and ion exchange and may also employ local exhaust on the vessels themselves to remove radon prior to in growth into its progeny and potentially become an occupational exposure concern.

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; Altshuler et al., 1964; Schiager and Dahl, 1968). Extensive radon and radon daughter monitoring at first generation ISR facilities indicated that severe disequilibrium can be encountered between radon and progeny (Brown 1982). 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 prior to implementation of local tank/vessel ventilation systems. Conversely, situations were identified (e.g. within enclosed and/or poorly ventilated areas) in which concentrations > 1 working level of radon daughters were measured in the absence of commensurate levels of radon gas. Ventilation conditions obviously play a large role in determining this relationship.

Conclusion: Monitoring considerations, particularly during startup phases and when operating conditions change, should include both the measurement of radon daughters (for accurate assessment of airborne concentrations and potential inhalation dose from progeny) as well as for radon gas to adequately identify potential sources so that necessary engineering controls can be implemented if necessary.

3. Release / contamination limits for equipment and materials for “unrestricted” (public) areas

Again it is important to recognize the radiological environment of a modern ISR as related to potential radionuclides of concern for which contamination surveys must be performed and unrestricted release limits established. This was discussed above as related to health physics issue # 1, air sampling in process areas for long lived alpha emitters.

Recommended surface contamination limits are defined in USNRC Regulatory Guide 8.30 in its Table 2 (USNRC 2002a) entitled “Surface Contamination Levels for Uranium and Daughters on Equipment to be Released for Unrestricted Use, on Clothing and on Non Operating Areas of UR Facilities”. A footnote to this table indicates the stated contamination levels are taken from Regulatory Guide 1.86 (USNRC 1974),

Termination of Operating Licenses for Nuclear Reactors and from an August 1987 USNRC document entitled *Guidelines for Decontamination of Facilities and Equipment Prior to Release for Unrestricted Use or Termination of Licenses for Byproduct Source or Special Nuclear Material*. It is also of interest to note that NRC Policy and Guidance Directive FC 83-23, *Termination of Byproduct Source and Special Nuclear Material* (1983) uses the 1982 version of *Guidelines for Decontamination of Facilities and Equipment*) as its ENCLOSURE 2 with the identical radionuclide categories and contamination limits as the 1987 version referenced in RG 8.30 as well as with RG 1.86.

Accordingly, FC - 83-23 including its Enclosure 2 (both the 1982 and 1987 versions of *Guidelines for Decontamination of Facilities and Equipment...*) use identical radionuclide categories and quantitative limits. Therefore the radionuclide categories, limits and intended application of FC 83-23, of *Guidelines for Decontamination of Facilities and Equipment....*(1982 and 1987), Regulatory Guide 8.30 and Regulatory Guide 1.86 are all consistent

Since the title of RG 8.30 Table 2 indicates applicability of the table's values to uranium *AND daughters* (emphasis added), it is reasonable to assume that it was clearly intended to be applied to uranium recovery facilities with expected varying degrees of equilibrium and ratios of natural uranium series radionuclides. Nothing in the historical documents referenced above provide any contradiction to or clarification of this interpretation. The use of the phrase “ and associated decay products” in e.g., FC 83-23 ENCLOSURE 2 (and subsequent 1987 revision) is not defined nor clarified in any historical documents nor is there any indication of distinctions made relative to the phrase “ and its daughters” as used in RG 8.30.

Another relevant and potentially useful approach to establish release criteria is dosimetric considerations. NRC presented results of a dosimetric assessment in *NMSS Handbook for Decommissioning Fuel Cycle and Material Licenses* (which unfortunately is no longer available). However, the results are summarized in Abelquist 2001. In that document, NRC presented the following results for each of the traditional nuclide groupings at the average surface activity guidelines of RG 1.86:

U-nat, U-235, and U-238 and daughters	13 mrem/yr
Ra-226, Ra-228, Transuranics	0.2 mrem/yr
Th-nat, Th-232, Sr-90	28 mrem/yr
Beta-gamma emitters	20 mrem /yr

It is interesting to note that three of the groupings are generally consistent with NRC's 25 mrem /yr criteria in 10 CFR 20 Subpart E and NUREG 1757 (NRC 2006) as used for the risk/dose based approach under the License Termination Rule. The estimated dose of 13 mrem / yr for “U-nat, U-235, and U-238 and daughters” is associated with the RG 1.86 and RG 8.30 contamination limits of 1000 dpm / 100 cm² removable alpha, 5000 dpm / 100 cm² total (fixed plus removable) alpha. The estimated dose of 0.2 mrem /yr for the radium / transuranic grouping suggests that the RG 1.86 guideline of 100 dpm / 100 cm² for those radionuclides should be increased by approximately a factor of 100 to yield the same dose! Admittedly, the exposure scenarios and modeling assumptions NRC used are unknown.

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Additionally, for ISR license applicants, NUREG 1569 (USNRC 2003), *Standard Review Plan for In Situ Leach Uranium Extraction License Applications* provides the current review guidance to NRC staff. It states that the applicant must ensure that “appropriate criteria are established to relinquish possession or control of equipment or scrap having surfaces contaminated with material in excess of the limits specified in Table 5.7.6.3” which is taken from Table 1 of Regulatory Guide 1.86). Furthermore, NUREG 1569 states (page 5-30) “The contamination control program is acceptable if it meets the following criteria:

- Radiation surveys of workers will be conducted to prevent contaminated employees from entering clean areas or from leaving the site in conformance with guidance in Regulatory Guide 8.30...
- The proposed contamination control program is consistent with the guidance on conducting surveys for contamination of skin and personal clothing provided in Regulatory Guide 8.30....
- Action levels for surface contamination are set in accordance with Regulatory Guide 8.30, Section 4.”

Conclusion: NRC has indicated that Regulatory Guide 8.30, *Health Physics Surveys at Uranium Recovery Facilities (prev. cit.)* is being revised and may address this issue. Until such time as this revision is officially promulgated the personnel contamination control guidance and surface contamination criteria for release of equipment and material to unrestricted areas as defined in RG 8.30 (and as referenced therein, the 1987 version of FC 83-23 ENCLOSURE 2) appears to represent the current, approved NRC staff position. NUREG 1569 similarly represents the currently approved guidance to NRC staff against which an ISR applicant’s source material license submittal is to be reviewed. Accordingly, until such time as NRC issues a revision to RG 8.30, the release limits defined in Table 1 of RG 8.30 and historical interpretation of the phrase “and associated decay products” as synonymous with “and its daughters” should be used since they have been previously determined by NRC to be adequately protective (1000 dpm / 100cm² removable alpha, 5000 dpm / 100cm² total alpha).

4. Solubility (metabolic) of uranium ISR products as related to appropriate worker airborne exposure limits and bioassay programs

Process knowledge suggests that uranium is present at modern ISRs exclusively in relatively soluble forms i.e., uranyl carbonates, (various forms) uranyl trioxide (UO₃), uranyl peroxide (UO₄) and their hydrates. The lixiviant uses oxygen and carbonate to dissolve and mobilize the uranium minerals in situ. Accordingly, the uranium goes into solution as a carbonate. If the uranyl carbonates formed were not very soluble, the in situ mining process could not work.

However, when acid is added to the precipitation cell the carbonate complexes are destroyed and

disassociate to form uranyl ions. When hydrogen peroxide is added to the precipitation vessel, the uranium is oxidized further to form uranyl peroxide ($\text{UO}_4 \cdot n\text{H}_2\text{O}$). When dried by the vacuum drier at relatively low temperature (250 – 300 degrees F), a combination of UO_4 , UO_3 and their hydrates are expected to result.

Although specific studies and references on solubility (e.g., in vitro solubility studies in simulated lung fluids, historical animal studies, etc.) for UO_4 are sparse (a few specific references are provided below), numerous studies appear in the literature regarding general solubility characteristics of industrial uranium compounds and are referenced herein. The UO_4 and UO_3 products are expected to be Task Group on Lung Dynamics (TGLD) Class D or W (most or moderately soluble – see ICRP 1972), which is equivalent to ICRP 66 class F or M (fast or medium dissolution – see ICRP 1994). It is also of note that ICRP 54, *Individual Monitoring for Intakes of Radionuclides by Workers* (ICRP 1988), which assigns Class W to UO_3 indicates “...there is evidence from animal studies that industrial uranium trioxide may

behave more like a class D material”. The issue of assumed solubility class is critical in establishing the appropriate DAC for air-monitoring parameters, for worker airborne exposure control and dose assessment.

The following provides support for a Class D or W designation for UO_3 and UO_4 :

- Regulatory Guide 8.30, *Health Physics Surveys in Uranium Recovery Facilities* (prev. cit.), Section 2.2: “Yellowcake dried at low temperature, which is predominantly composed of ammonium di-uranate, or in the new processes uranyl peroxide, both are more soluble in body fluids than yellowcake dried at higher temperature”
- Kathren R.L and Burklin R.K 2008: “ ...the more soluble compounds of uranium such as... and UO_4 are more quickly absorbed into the blood”
- Metzger R, Wichers D. et al 1997. In a solubility study performed for uranium products at a specific ISR, the authors concluded: “airborne uranium in wet process area = 97% with dissolution $T_{1/2} = 0.3$ days; airborne U in drum load out area = 97% with dissolution $T_{1/2} = 0.25$ days. X ray diffraction analysis of the final dried product indicated 79% $\text{UO}_4 \cdot 2\text{H}_2\text{O}$, 15 % UO_3 and 3 % CaCO_3 .

The results of this study indicate airborne U in both the wet process and drum load out areas of 97 % dissolution with half times <0.5 day and therefore clearly are solubility Class D (F).

- Moody JC, Birchall A et al 1997. In a solubility study performed with recycled UO_3 in rat lungs, the authors concluded “Using standardized criteria described in ICRP Publication 71 (ICRP, 1995) together with the data obtained in the study...for radiological protection, this UO_3 can be described as consistent with an absorption Type F compound, which is in line with studies on exposed workers.

Examples of additional references that specifically address solubility and solubility class of uranium mill and related uranium fuel cycle compounds are provided in the reference section. See in particular LANL 1985, Cook and Holt 1974, Eidson and Mewhinney 1980, Alexander 1984, Blauer, Kent and Dennis 1982, Brown and Blauer 1980, U.S. Uranium Registry 1984.

Conclusion: Although evidence suggests that both the wet process UO_4 and dried UO_3 products of modern ISRs will be ICRP 19 Class D or ICRP 66 Class F compounds, it may be prudent to assume them to be Class W / Class M for purposes of establishing the initial DAC upon plant startup. Studies on modern ISR products involving dissolution in simulated lung fluids and molecular analysis (e.g., X-ray diffraction) should be performed in accordance with the established protocols (well documented in the literature – examples provided) to validate these important assumptions and establish if Class D / Class F may be more accurate. This is appropriate to define not only the relevant Derived Air Concentration (DAC – see discussion above under health physics issue # 1), but also to verify assumptions of appropriate sampling frequencies and action levels for the facility uranium bioassay program as discussed in the next section.

5. Bioassay Programs for ISR products

Bioassay programs at uranium ISRs should be conducted in accordance with US NRC Regulatory Guide 8.22, *Bioassay at Uranium Mills* (USNRC 1988) and NUREG 0874, *Internal Dosimetry Model for Application to Bioassay at Uranium Mills* (USNRC 1986). This NUREG provides the technical basis for Regulatory Guide 8.22. In fact, frequencies of sampling based on solubility characteristics, associated action levels and recommended actions specified in, e.g. Tables 1 and 2 of RG 8.22 are from NUREG 0874 (Section 6 of NUREG 0874 compares action levels and bioassay frequencies recommended in the two documents).

Although there is some uncertainty at present regarding the applicability of TGLD solubility class D vs. Class W for modern ISR yellowcake products (see health physics issue # 4 above), the solubility characteristics of the less soluble class W can be well within the range of dissolution half times defined by NUREG 0874 for “low temperature drying” (see NUREG 0874 Table 1-3). Additionally, data from the technical literature indicates that the UO_3 , UO_4 and associated hydrates produced in modern ISRs are “low fired” and therefore relatively soluble as was previously presented.

Accordingly, urinalysis as has been used for > 30 years in uranium mills and ISRs is an appropriate bioassay method for detecting exposures to low fired, relatively soluble uranium products and a monthly sampling frequency for workers potentially exposed to product concentrates as recommended in RG 8.22 and NUREG 0874 is appropriate. However, special, ad – hoc samples, in addition to routine monthly samples, may need to be collected in response to potentially elevated airborne concentrations, as may be required by radiation work permits, whenever respiratory protection devices are found to be internally contaminated following use, in response to positive nasal and/or mouth swabs, etc. In such cases, it should be assumed that the exposure / intake occurred at a specific time related to the activities causing the potential intake and Figure 2 of RG 8.22 should be used to establish action levels.

Regarding the applicability of in vivo lung counting, it is also of note that footnote b of Table 1

of RG 8.22 defers to NUREG 0874 Section 6 for considerations of in vivo lung counting to detect intakes of more insoluble, high fired materials. The NUREG recommends that in vivo capabilities should be available “to guard against the unlikely, but possible, contingency that large intakes of Class W or Y transportability might go undetected”. In vivo capabilities as follow-up to confirmed urinalysis results in excess of action levels as specified in RG 8.22 Table 1 should be accessed as necessary. However, as discussed above, modern ISR yellowcake products appear to exhibit transportability characteristics typical of soluble, low-fired yellowcake (UO_3/UO_4 Class D/W) and the contingency alluded to in NUREG 0874 is not expected to be applicable to modern ISR yellowcake products. Given that there are only a few in vivo lung counting facilities in the United States with the appropriate equipment, software and experience to measure pulmonary deposition of natural uranium at the required detection limits (e.g. 9 nCi total pulmonary – see RG 8.22), urinalysis is the technically appropriate as well as the “practical” method of routine bioassay at uranium ISRs. Additionally, it is noted that this limited number of uranium in vivo lung counting facilities have been used over the years to assess deposition and dose by the uranium recovery industry in response to suspected “significant” intakes as based on confirmed urinalysis results.

6. External Exposure Monitoring

Historically, uranium ISRs have monitored all “radiation workers” for external exposure via personally assigned TLD ‘whole body’ dosimeters and/or area monitoring and results have typically been very low (unusual for any worker to receive annual deep dose equivalent > 2 mSv, i.e. 200 mrem). However, questions have been raised recently regarding the potential for extremity and/or skin dose from a possible beta component that may be associated with the ISR uranium products. As discussed several times elsewhere in this paper, the ISR process is “radiologically selective” and other than uranium itself, minor amounts (primarily radium 226) of uranium series radionuclides are mobilized in situ, loaded on IX resin or carried forward into the process. The “nuclide mix” that is associated with ISR products is expected to be almost exclusively U nat. In-growth of the first few short lived daughter products (e.g., the beta emitters Thorium 234 and Protactinium 234) takes approximately 4 months to reach equilibrium and therefore is not expected to be an important component of relatively fresh product in an operating recovery plant. Additionally, there is no routine need for workers at ISRs to be in close proximity to or “handle” ISR products for any extended time. Accordingly, “process knowledge” and anecdotal experience suggests there should be little concern with skin and/or extremity shallow dose equivalent at ISR facilities nor any special needs for extremity monitoring (e.g., use of finger or wrist TLDs). This can be easily and directly confirmed with beta exposure rate measurements within a few centimeters of product associated with precipitation, dewatering (filter press) and drying/packaging areas (“open vs closed shield” on standard GM probe) combined with studies of exposure times. Regulatory Guide 8.30 also provides guidance on estimating beta exposure rates as a function of product age and distance from it.

CONCLUSIONS

This paper has presented a summary of the radiological characteristics of in situ uranium recovery processes. The purpose was to present an overview of those special health physics considerations dictated by the in situ uranium recovery technology and to identify some recent health physics related issues that have been raised in licensing proceedings for new source material license applicants and that also potentially affect existing uranium ISR licensees. Given the many years since the first generation of ISRs began operation in the US and the fact that some of the standards of health physics practice and associated regulatory requirements and guidance had not been revised over much of that time, these current circumstances are not unexpected. This is appropriate and necessary as we move forward with numerous new ISR projects in the US. As health physicists, we must execute radiological protection programs for workers that ensure compliance to fundamental dose limits, provide a sufficient standard of care and maintain exposures ALARA. Additional characterization data on modern ISR uranium products should be relatively straightforward to obtain and will greatly assist in resolving these health physics issues and answer related questions. This data should address the relative radionuclide composition (“mix” of Unat, thorium 230 and radium 226 e.g.), molecular composition (UO₃ vs UO₄ vs. U₃O₈ etc.) as well as associated metabolic solubility characteristics. As evidenced by the large number of ISR projects currently under development in the US (and world wide), non conventional uranium recovery techniques, such as ISR, can play a significant role in complimenting uranium supplies during the next several decades contributing to America’s national security and energy independence.

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