RADIOLOGICAL ASPECTS OF ALKALINE LEACH uranium In Situ Recovery (ISR) FACILITIES in the United States*

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* Health Physics- Operational Radiation Safety. July 2019. V117(1); 106-113.

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Outline

- Overview
- Process Summary
- Radionuclide mobilization
- Operational health physics and radiation protection programs
- Conclusions

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Overview

- In Situ Recovery or In Situ Leach (ISR/ISL) uranium facilities have operated in the US since the late 1960s
- In recent years have accounted for over 70 % of US production and internationally almost half of worldwide uranium supplies
- Paper presents summary of radiological characteristics of typical ISR processes in US - traditionally using alkaline based lixiviants
- Describes HP and radiological monitoring programs necessary to adequately monitor and control radiological doses to workers based on the radiological character of these processes
- The operational specifics of these facilities are contrasted to similarities and differences in radiological characteristics with conventional mills
- Terms In Situ Recovery (ISR), In Situ Lech (ISL) and Uranium Solution Mining can be used interchangeably

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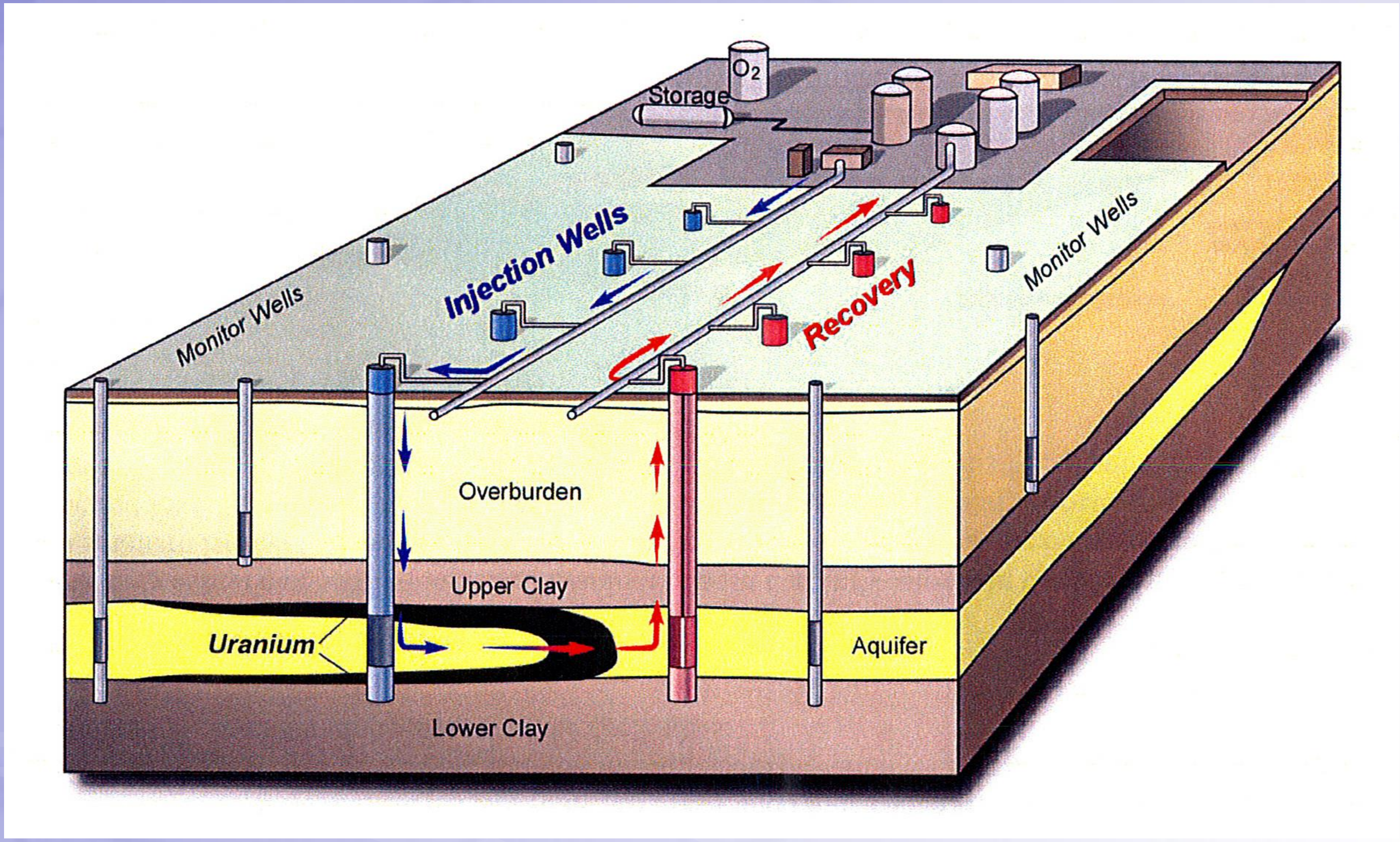
In Situ Uranium Recovery



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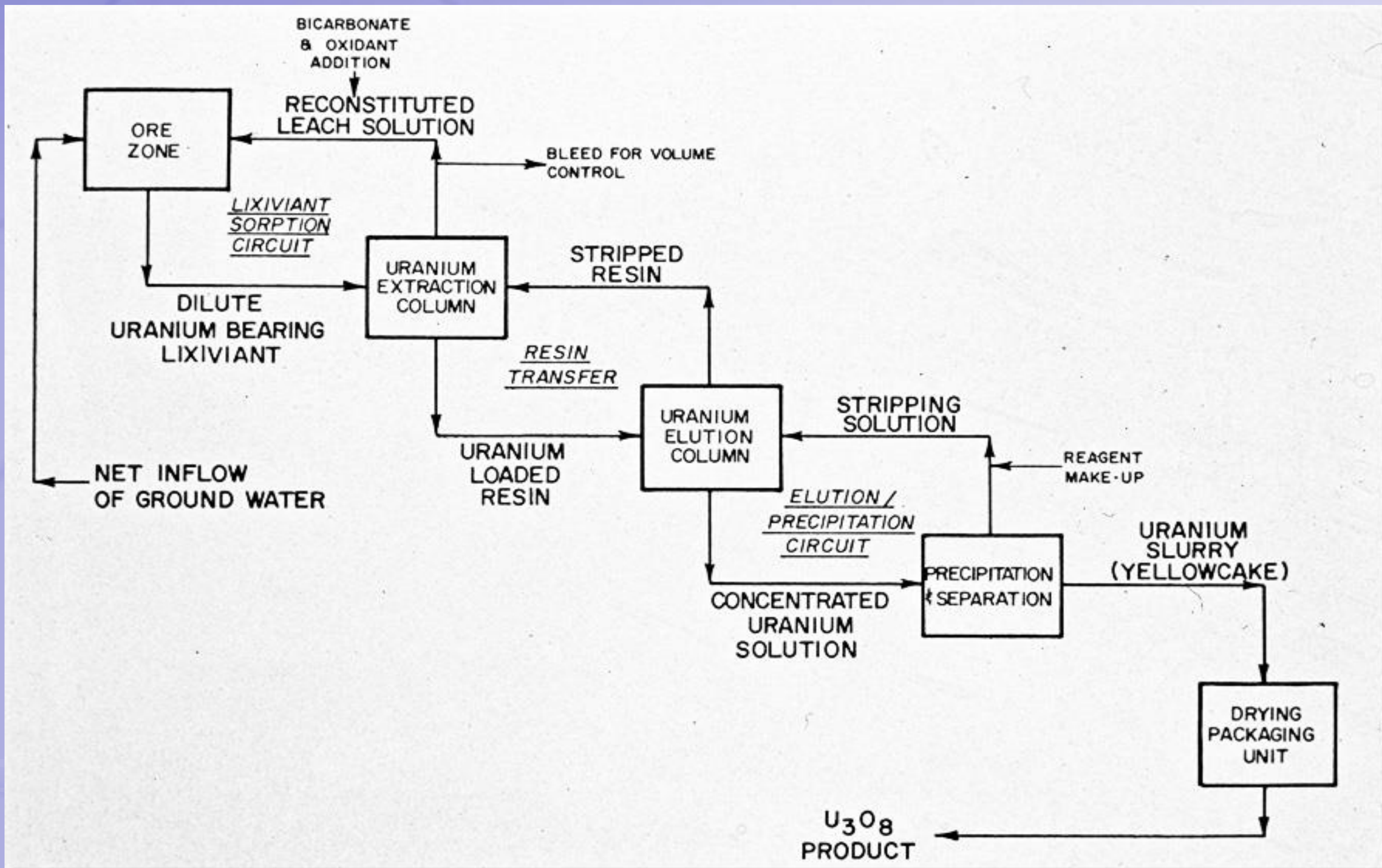
Process Flow Diagram



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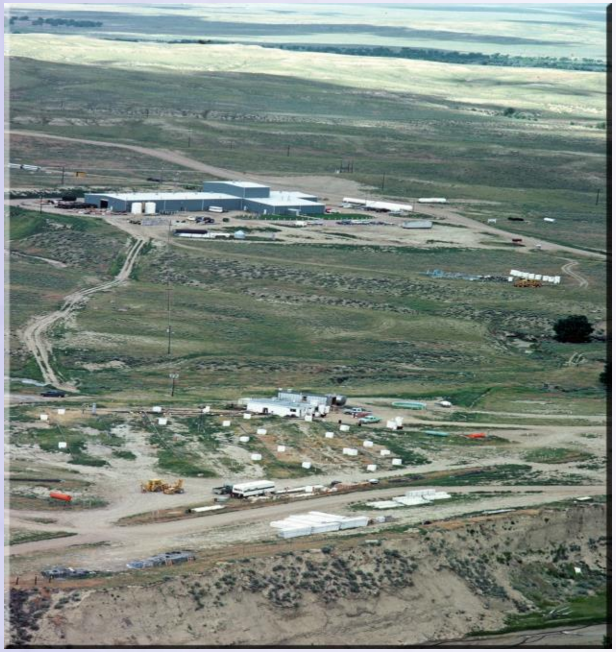
ISR Plants, Well Field and “Satellites”



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Radionuclide Mobilization (Alkaline Leach)*

- Relatively small % of U progeny in ore body observed to be mobilized by lixiviant
- But specifics may be process and facility age dependent
- Thorium appears to equilibrate in circulating lixiviant and little removed
- Lead carbonate complexes relatively insoluble, little mobilized
- Estimated that 5 -15% of calculated radium in host formation mobilized
- Radon gas evolution can be considerable and is discussed later

* Expect differences with low ph (acid) leach facilities, e.g., Kazakhstan, Australia

Radionuclide Mobilization - continued

- Ion exchange (IX) resin used in US ISR facilities is specific for removal of uranium
- Thorium and other progeny are not expected downstream of the IX columns (e.g., elution, precipitation, and drying circuits).
- LLRD in air typically only natural uranium isotopic mixture with a relatively small progeny component
- Where solid wastes are produced (resin tanks and columns, fabric and sand filters, clarifiers, etc.), mobilized radium 226 (calcium and carbonate chemistries) may be an important external exposure and/or contamination source.
- Large quantities of radon 222 gas is dissolved in lixiviant returning from underground and is brought to the surface.
- However this is “fresh radon“ “ – typically low equillibrium factor – low progeny WL.

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Radionuclide Mobilization and Typical Concentrations in Process

All values in Bq/l except * = Bq/g

(1 Bq = 27 pCi)

Process Location	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

Process Location	Ra 226	Rn 222
Circulating Lixiviant	3 - 20	300 – 7000+
Calcite In Clarifiers	30 – 100*	N/A
Evaporation Ponds In Solution	20 - 50	Equilibrium Assumed
Evaporation Ponds, Sludge	30 – 70*	Equilibrium Assumed

Reproduced from Brown 2009

More on Radon and ISRs

- Radon brought to surface dynamically dissolved in lixiviant returning from underground
- Physical and geochemical environments in situ apparently enhance solubility in lixiviant
- Significant quantities of gas can be released when solutions first exposed to atmospheric pressure - out of doors (surge ponds / tanks) or in plant areas (ion exchange and elution systems where open to atmosphere)
- Monitoring required to identify release points
- In plant air must be assessed for exposure potential from both radon gas as well as ventilation necessary to suppress in growth of radon progeny
- Empirical model developed by author based on measurements at commercial ISR = 10^{12} -10^{13} Bq/yr. Rn @ average recovery flow of 3000 l / min

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Operational Health Physics and Radiation Protection Programs

Similar to conventional mills and include:

- Airborne monitoring for long lived radioactive dusts
- External exposure monitoring in areas in which large quantities of uranium concentrates are processed and stored and where radium precipitates may accumulate
- Surface area and personnel contamination surveys
- 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

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External Exposure Monitoring- Area Surveys and Personnel Dosimetry

- Primarily in areas in which large quantities of uranium concentrates are processed and stored
- Radium precipitates can accumulate in resin tanks and columns, filter membranes from reverse osmosis water treatment units, fabric and sand filters, clarifiers, etc.
- Control and monitoring of external exposure during work near these processes, during filter changes and / or maintenance of these systems.
- Use of personal dosimeters (TLD/OSL) for all full time employees in restricted areas recommended
- Potential for extremity and eye exposure from short lived beta emitting uranium progeny (Th 234, Pa 234 e.g.- "aged yellowcake") during maintenance activities when systems are penetrated (occasional, of relatively short duration, use of PPE, annual dose limits 150 mSv eye, 500 mSv S and E)

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Airborne Monitoring for Long Lived Radioactive Dusts (LLRD)

- Essentially an aqueous process until drying and packaging
- Containment of spills in process areas via design is essential to reduce the risk of resuspension of LLRD .
- LLRD exposure potential is primarily associated with the back end "yellowcake areas" of the process that include precipitation, drying and packaging.
- Natural U mixture – gross alpha counting adequate following Rn progeny decay
- Applicable monitoring techniques include combinations of grab sampling, breathing zone sampling and continuous monitoring based on radiological and work conditions*.
- Results compared to DACs, ALIs and associated TEDE calculated*

* See Brown and Chambers. Importance of Uranium Recovery Facility Product Characteristics for Dose Assessment and Assignment. Health Physics. April 2018

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Area and Personnel Contamination Surveys

- Yellowcake areas typically the most important sources of potential surface contamination
- Standard contamination controls - containment, ventilation, radiological survey and expedient response to spills or other loss of containment minimizes potential for resuspension (inhalation) and ingestion
- Contamination surveys necessary throughout plant and ancillary areas including of personnel and for release of equipment and materials for unrestricted use into the public domain.

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Bio-assay (urinalysis) programs

- Designed commensurate with metabolic characteristics of uranium species produced
- Typically for yellowcake workers only
- US ISRs today producing peroxide-precipitated products dried by low temperature vacuum dryers
- Products appear to be quite soluble and meet ICRP 71 criteria for the Type F (fast) absorption category (similar to solubility "Class D" – ICRP 30 and 10 CFR 20) *.
- For these products, chemical toxicity also drives worker risk from intake - not just radiation dose
- Urinalysis programs involve frequent sampling (can be weekly) and analysis for U and if positive, biomarkers associated with potential renal injury, e.g., glucose, lactate dehydrogenase (LDH) and protein albumen.

* See Brown and Chambers, Worker Protection Implications of the Solubility and Human Metabolism of Modern Uranium Mill Products In the U.S. Health Physics November 2014.

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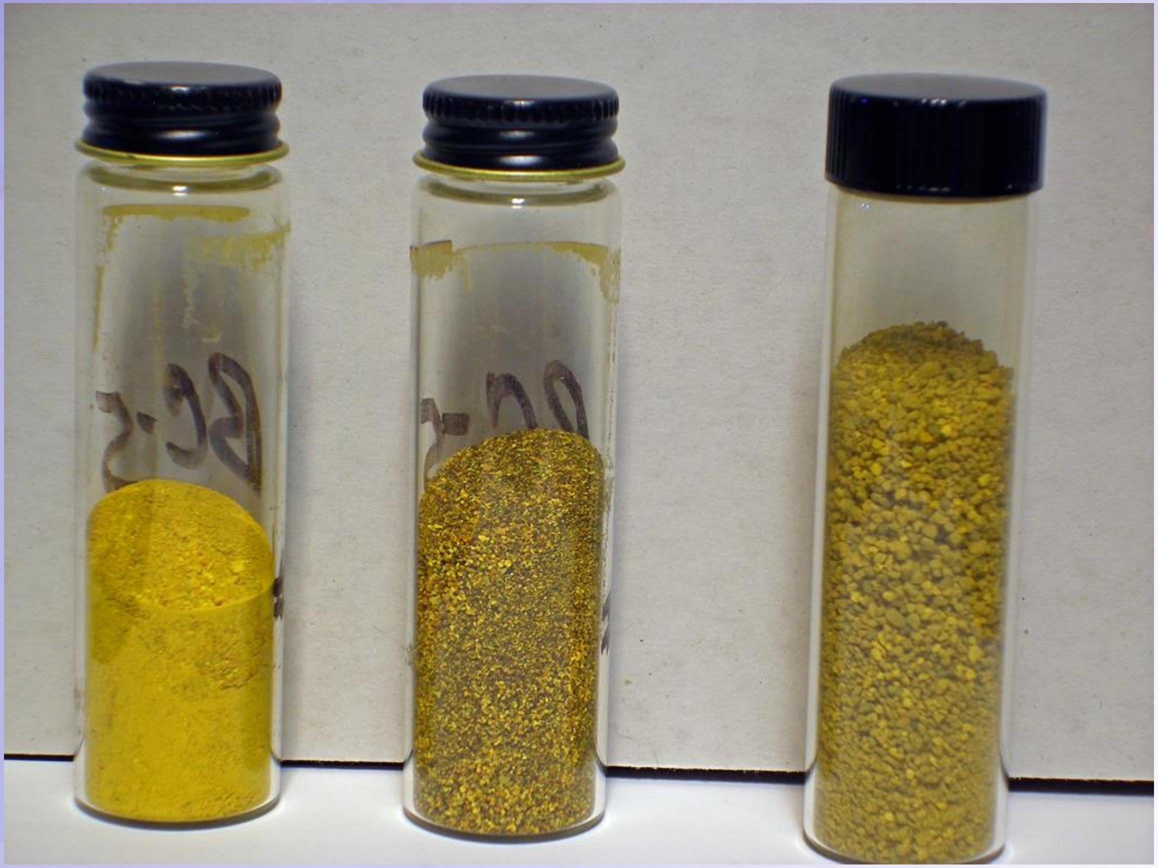
“Yellowcake” is Not All the Same



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Modern Yellowcake Products

- Uranium recovery facilities in the past typically used ammonia precipitation (ADU) and high temperature calciners producing relatively insoluble UO_2/U_3O_8
- Today's ISR facilities in the US use hydrogen peroxide precipitation and low temperature vacuum dryers (< 400°F*)
- XRF Studies conducted by two ISR licensees indicate products are combination of UO_4, UO_3 and their hydrates (e.g., UO_4 X H_20 where X= 1,2,3 ..)**
- Uranium content varied from 76 – 79%

* Willow Creek still older calciner ?

**Cameco Corporation, Solubility of Radionuclides in Simulated Lung Fluid. G. Tairova, M.Boucher ,K. Toews, et al. Proceedings of the 3rd International Conference on Uranium. Saskatoon 2010

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The Chemistry: % Uranium Content Based on Molecular Weight of Uranium Compounds:

Most Insoluble
Radiotoxicity Dominates
Brown / Black

Most Soluble
Chemotoxicity Dominates
(Modern US ISRs – Yellow / Green)

FORM	U wt %
UO_2	88.1
U_3O_8	84.8
UO_3	83.0
UO_4	78.8
UO_4*H_2O	74.0
UO_4*2H_2O	70.0

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Radon/Progeny Monitoring

- Large quantities of radon can be brought to surface dissolved in lixiviant
- However this is “fresh radon“ ; progeny equilibrium factors are typically quite low
- Potential for worker exposure low since vast majority of dose results from the short lived progeny and not the gas itself
- Local exhaust (closed) systems on front end tanks and vessels usually necessary to remove fresh radon before significant progeny ingrowth into work areas
- However, progeny buildup can occur in poorly ventilated areas

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Radon/Progeny Monitoring - continued

- Some of the first generation ISR plants ("U solution mining" - 1970s) used in plant IX surge tanks and up flow, open top IX columns requiring installation of local exhaust systems to remove the gas before progeny ingrowth created occupational exposure concerns.
- Radon levels of 10^3-10^4 Bq/l measured in process front end as well as radon progeny > 10 WL in poorly ventilated areas prior to installation of local exhaust systems
- Recent designs tend towards use of enclosed, pressurized systems for lixiviant recovery and ion exchange to remove radon prior to significant progeny in growth.

In Plant Surge Tanks and "Up Flow" Ion Exchange and Elution Columns – 1970s

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Modern Pressurized and Enclosed IX / Elution Systems

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Conclusions

- Radiological considerations dictated by the in situ recovery technology - hydrologic control and mobilization of radionuclides underground and management of fluids at the surface
- Some unique radiological aspects of ISRs result from mechanisms of radon evolution
- Conventional mill tailings are not generated
- However, solid LLW (11.e(2) byproduct material) can result primarily from process specific aspects of radium chemistry and mobilization – potential for external exposure
- Health physics programs similar to conventional mills or any uranium facility processing and manufacturing dispersible industrial U compounds at natural enrichment
- As for any uranium fuel cycle facility using Unat isotopic mixture, importance of radiotoxcity vs chemotoxicty must be considered based on metabolic implications of product characteristics

QUESTIONS AND DISCUSSION

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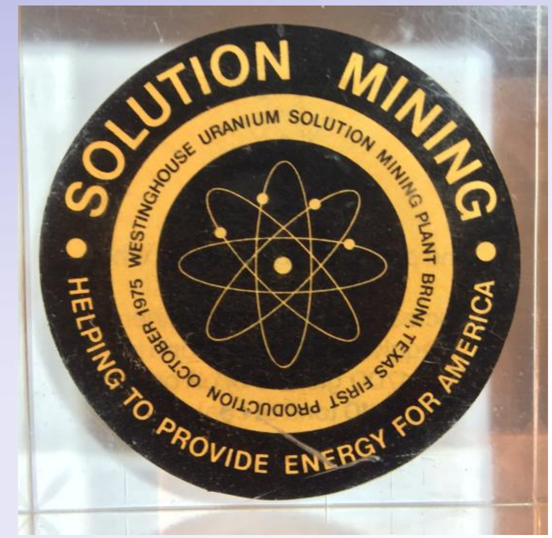

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