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Inherently Safe In Situ Uranium Recovery

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Abstract

Expansion of uranium mining in the United States is a concern to some environmental groups and sovereign Native American Nations. An approach which may alleviate some problems is to develop inherently safe *in situ* uranium recovery (“ISR”) technologies. Current ISR technology relies on chemical extraction of trace levels of uranium from aquifers that, once mined, can still contain dissolved uranium and other trace metals that are a health concern. Existing ISR operations are few in number; however, high uranium prices are driving the industry to consider expanding operations nation-wide. Environmental concerns and enforcement of the new 30 ppb uranium drinking water standard may make opening new mining operations more difficult and costly. Here we propose a technological fix: the development of inherently safe in situ recovery (ISISR) methods. The four central features of an ISISR approach are: 1. New “green” leachants that break down predictably in the subsurface, leaving uranium, and associated trace metals, in an immobile form; 2. Post-leachant uranium/metals-immobilizing washes that provide a backup decontamination process; 3. An optimized well-field design that increases uranium recovery efficiency and minimizes excursions of contaminated water; and 4. A combined hydrologic/geochemical protocol for designing low-cost post-extraction long-term monitoring. ISISR would bring larger amounts of uranium to the surface, leave fewer toxic metals in the aquifer, and cost less to monitor safely – thus providing a “win-win-win” solution to all stakeholders.

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I. Introduction:

In the last few years a sharp increase in the price of uranium has sparked a renewed interest in uranium mining around the world, including the western United States. However, most of the (non-stockpile) uranium consumed in American reactors presently comes from foreign sources, which in turn reflects the fact that the highest grade, most inexpensively produced uranium deposits lie outside the United States. In a free market economy one must anticipate that imports from these sources will remain important. At the same time it would also be smart planning to build a “buffer” mechanism into the emerging nuclear energy strategy which minimizes the impacts of foreign uranium suppliers on our domestic economy.

Fortunately, the United States has significant indigenous uranium deposits. These might developed to provide a steady feed of domestic uranium or held in readiness as a stop-gap against adverse policies initiated by foreign uranium suppliers. However, earlier “uranium booms” have left a legacy of cultural and environmental concerns with some stakeholders who would be most directly impacted by a resurgence of uranium mining in (or near) their communities. In situ uranium recovery (“ISR”) is a technology allowing for uranium extraction while avoiding many of the environmental impacts of previous mining activities. In principal, it is as simple as circulating a uranium-leaching fluid underground into the ore deposit down one well, and then bringing it back to the surface through a second well (or array of wells). Only a small chemical treatment plant is located at the site to recover the uranium, and the only waste that cannot be removed when the site closes is the residual fluid left underground in the aquifer.

In the desert southwest, groundwater resources are a matter of ever increasing importance to stakeholders, communities and regulators. Consequently, being able to demonstrate that an ISR-impacted aquifer could be remediated reliably should be a major selling point in being allowed to set up an ISR activity in a community. Additionally, any technologies which might improve the efficiency or ease of the extraction process would be viewed favorably by the mining industry. By virtue of Sandia’s unique interdisciplinary nature we are in a position to make major contributions to improving both the public acceptance and efficiency of ISR activities. The following short report summarizes several approaches which might be followed to achieve these ends.

II. Origin and Characteristics of “Sandstone” uranium deposits:

Park and MacDiarmid (1975) broadly divide uranium into two general categories: those of hydrothermal origin and those hosted in sedimentary rocks (principally sandstones) showing no evidence of having originated at elevated temperatures. Hydrothermal uranium deposits are similar to the more traditionally recognized “vein” deposits of copper, silver, lead, gold, etc., in which the metals were deposited from hot, often saline fluids, heated by the emplacement of igneous (typically granitic) rocks. Such deposits have a restricted occurrence in the United States, will not play an immediate role in the Nation’s nuclear future, and are not the subject of this white paper.

Sandstone-hosted deposits (variously referred to as “roll-front” and “channel deposits”) are widely distributed in Mesozoic and Tertiary rocks across the western US. In the past these supplied significant amounts of uranium for both civilian and military

applications. They formed where groundwaters moved along ancient buried stream channels (or other high porosity sedimentary structures) and encountered localized concentrations of organic matter, hydrogen sulfide or pyrite, which created reducing conditions and precipitated minerals (Fig. 1) such as pitchblende (AKA uraninite, $\sim\text{UO}_2 - \text{UO}_3$) and coffinite ($\text{U}(\text{SiO}_4)_{1-x}(\text{OH})_{4x}$). The uranium deposits are clearly much younger than the rock units which host them and the fluids responsible for their origin may, in fact, have originated in rather distant deep hydrothermal systems which cooled and mixed with normal groundwaters prior to forming the ore body. Or, they may have originated when normal groundwaters acquired trace amounts of uranium from the vast amounts of weathering volcanic ash they encountered while infiltrating into the proximate sedimentary units – or both.

Precambrian “unconformity” deposits were probably formed by processes similar to those that formed the younger sandstone-hosted deposits. That is, they are typically found where U-enriched groundwaters apparently percolated along discontinuities between ancient Precambrian crystalline basement complexes (Archean and early Proterozoic age) and thick, overlying accumulations of younger (mid-Proterozoic) sandstones (all of which have suffered later metamorphism, obscuring the exact details of how the ore bodies formed). These deposits are confined to the Precambrian shields (notably in Canada) and, unlike the sandstone deposits of the United States with ore grades of a fraction of a percent uranium; these deposits may have ore grades in the tens of percent range (Macfarlane and Miller, 2007).

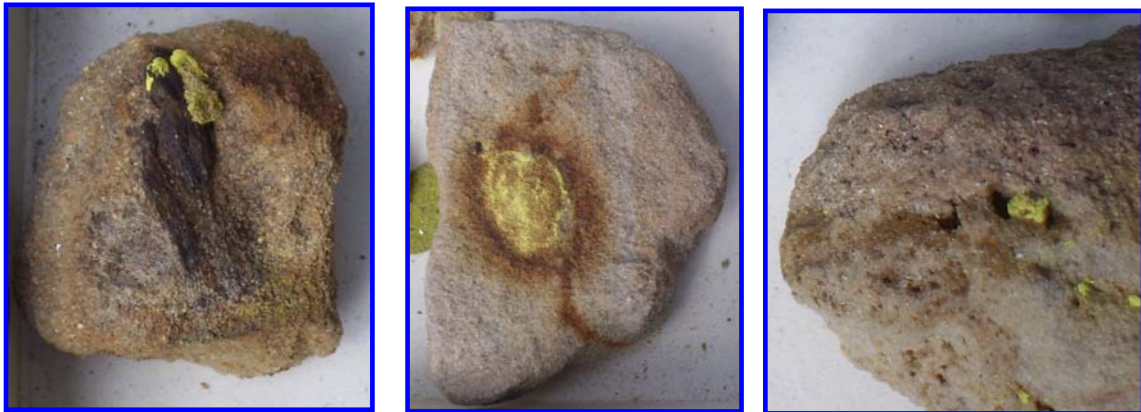


Fig. 1: Uranium mineralization from a sandstone-type deposit, Carlisle Mine, Wyoming. Left photograph illustrates uranium associated with organic matter (lignite), while the middle and right photographs emphasize co-occurrence with iron pyrite. Samples were taken from a mine dump where exposure to the air had oxidized the original black uranium (IV) minerals to form the bright yellow uranium (VI) phases, and converted the pyrite to “rust”.

III. In Situ Recovery:

In situ recovery (“ISR”) is an attractive alternative because it avoids the expense and environmental implications of traditional mining practices (underground or open pit workings). The first step is to introduce a chemical agent (“lixiviant”) into a mineralized aquifer to dissolve the uranium minerals. Uranium-laden formation water is then brought to the surface and uranium is recovered at a chemical processing plant (Table 1).

Table 1: U-Rich Lixiviant Water Chemistry (Air-CO₂-NaHCO₃ type leach)

| Constituent | Typical Concentration Range |
|------------------------------|------------------------------|
| Calcium | 100 - 350 - mg/l |
| Magnesium | 10 - 50 - mg/l |
| Sodium | 500 - 1600 - mg/l |
| Potassium | 25 - 250 - mg/l |
| Bicarbonate | 0 - 500 - mg/l |
| Sulfate | 100 - 1,200 - mg/l |
| Chloride | 250 - 1,800 - mg/l |
| Silica (SiO ₂ aq) | 25 - 50 - mg/l |
| Total Dissolved Solids | 1,500 - 5,500 - mg/l |
| Uranium | 50 - 250 - mg/l |
| 226 radium | 500 - pCi/l |
| Conductivity | 2,500 - 7,500 - μ S/cm l |
| pH | 7-9 |

From: Pelizza, 2008.

Since the process involves neither actually digging a shaft or running a conventional ore milling operation there are never large piles of waste left exposed on the surface at a site. Instead, the principal environmental impact of this activity resides underground in the formation from which the uranium was leached. A secondary concern is the disposal, or treatment, of excess process waters once the uranium has been stripped. Ultimately this problem can be addressed by re-injecting them underground or by land surface application once the water has been purified by reverse osmosis (RO) or chemical treatment. However, before final disposal becomes an issue this water will probably be re-circulated through the formation numerous times.

During the early post-war years, traditional uranium ore dressing processes graduated from an alkali-carbonate-ore roasting technology to a more efficient sulfuric acid leach treatment (ERDA 76-43, 1976). The earliest ISR processes in the US were undertaken at the time when sulfuric acid was in use for traditional ore dressing, so it was logical that it was one of the first reagents tried for ISR. The extremely low pH produced by the acid was often all that was needed to dissolve the uranium. Occasionally, however, oxidants (hydrogen peroxide, sodium chlorate, and occasionally nitrate or nitric acid) were also added to facilitate the process. Alternatively, merely circulating air-saturated solutions containing dissolved ferric iron can remove economic levels of uranium. The sulfuric acid process can generally remove between 70% and 90% of the uranium (Taylor et al., 2004) but leaves aquifers difficult to remediate. Acid leaching was (and is) employed to

a much greater extent in the former Soviet Block countries and Australia, than in North America. Because of its environmental consequences, it seems unlikely that this approach will be employed again in the US.

Modern ISR technologies in the United States depend on coupling oxidation of the uranium ore (initially a mix of the U(IV) hydroxide and silicate minerals) with complexing of the resulting hexavalent uranyl ion (UO_2^{++}) with carbonate ion to form soluble $\text{UO}_2(\text{CO}_3)_3^{-4}$ in solution. Typical extraction efficiencies are in the 60-70% range (Taylor et al., 2004). The simplest chemical combination presently being employed to affect this process is a mix of carbon dioxide and air (analogous to what caused the oxidation in Fig. 1). Yields increase when hydrogen peroxide or oxygen is substituted for air, and a carbonate or bicarbonate salt is substituted for carbon dioxide. A limitation on this latter switch is that the uranyl ion forms sparingly soluble salts with most alkali and alkaline earth elements. Consequently, for example, if the formation waters already contain significant dissolved sodium, one may not actually get enhanced leaching by using sodium carbonate since solid sodium uraninite may precipitate. In some instances, ammonium carbonate is used (rather than sodium carbonate) to avoid this problem. But, the ammonium left in the groundwater then presents additional remediation problems. (Appendix 1, from Taylor et al., 2004, compares the relative benefits and drawbacks of acid and alkaline ISR processes).

IV. Potential Pollution Sources:

The neutral to mildly basic pH established underground by modern ISR reagents is far less likely to produce the whole scale heavy metal mobilization caused by older acid-wash technologies. However, elements such as selenium, arsenic, vanadium and molybdenum, which form oxy-anions (Garrels and Christ, 1965) and usually are found with the uranium in sandstone-type deposits, are still mobilized by acid-free ISR technologies (Schoeppner, 2008; Erskint and Ardito, 2008). Also, with drinking water standards moving to 30 ppb U, the residual dissolved uranium itself can be problematic. The uranium-series decay daughter products (notably radium and radon) are another concern since they are hazardous at concentrations which are considerably below levels where mineral precipitation will limit their mobility. A final concern is that ISR activities may increase the salinity of potential drinking water sources, which is unlikely to be viewed as a positive development by regulatory agencies charged with protecting potential drinking water sources. This is of particular concern on the Colorado Plateau and in Wyoming where, respectively, the Morrison and Wind River Formations are both important host formations for ore deposits – and also some of the few important regional sources of low salinity water (Hovorka et al, 2000; and Zelt et al., 1999 respectively). An additional concern are organic pollutants which may originate as a consequence of the leach fluids interacting with the nominally insoluble organic constituents indigenous to the ore, or because organic chemicals are added during uranium recovery operations, that are then transported underground when fluids are re-circulated to recover additional uranium. However, to date we have not found published accounts stating that such constituents constituted a significant pollution concern.

V. Chemistry-Based Improvements to the ISR Process for Uranium Recovery:

Because Sandia played such a large role in the WIPP and Yucca Mountain nuclear waste repository programs our laboratory has unique capabilities for predicting *and potentially manipulating*, the mobility of uranium (as well as other radionuclides) in the subsurface. Approaches can generally be divided into improving chemical processes or better application of hydrologic principals, though occasionally cross over issues are encountered. Consider, for example, the detrimental impact that carbonaceous constituents in the ore may have on ISR technologies. Unlike the ores of Texas and Wyoming (where most of the organic content exists as fragments of lignin-like material, Fig. 1) the porosity in some of New Mexico's highest grade ores may be plugged by a degraded, insoluble, kerogen like material called "humate" (Holen and Hatchell, 1986). In a first order sense the problem is mechanical; the leach fluids simply can't get to most of the uranium. The key to developing such deposits economically may lie in: (1) better hydrologic models that optimize access to what uranium is still available, and, or (2) use of novel surfactants that allows the leach solution to access the coffinite/pitchblende which is sequestered beneath the organic coatings.

In the realm of strictly chemical approaches we considered two objectives; improving the efficiency of uranium removal and providing technologies that would make it easier to remediate an aquifer once the ore body was exhausted. Below we outline five potential topics for study derived from Sandia's past experience in modeling radionuclide migration which address both objectives.

1. *Better complexing agents*; the uranyl carbonate complex is indeed strong and it may be difficult to beat from an economic standpoint. However, other stronger complexes (such as citrate or EDTA) may be applicable in settings where current technologies leave too much of the uranium behind. Developing a complexing agent that is both specific to uranium and capable of complexing uranium in its tetravalent form would also be a significant advance. Isosaccarinic and gluconic acids (or similar compounds) may be capable of this and should be considered (Warwick, 2004). With such a development, extracting the uranium would no longer depend on imposing an oxidizing condition on the formation. It follows, then, that many potential pollutants (As, Se, V, Mo etc.) would not form oxyanions and hence remain fixed in indigenous minerals rather than be mobilized into the groundwater.
2. *Readily degraded complexing agents*; once it has been introduced into the subsurface in an ISR operation, bicarbonate typically remains present for a very long period of time. This may result in the long term elevation of dissolved uranium concentrations. A complexing agent which would degrade over time (by bacterial agents or as a consequence of injecting a second reagent) might lower long-term residual dissolved uranium concentrations to levels acceptable to regulators. (Any of the common organic complexing agents, citrate etc., when exposed to excess oxidizing agents - or possibly a mixture of additives designed to stimulate microbial activity - could fulfill this role).
3. *Oxy-anion pollutants (As, Se, V, Mo, U)*; the ease of site remediation would also be improved by developing a technology that would reduce (e.g., lower the oxidation state) of pollutants dissolved as oxyanions back to their initial (insoluble) condition at

a predictable rate. This technology might also focus on providing a substrate conducive to accelerating the precipitation reactions. The various sulfide, polysulfide or thiosulfide ions are obvious candidates for reducing agents since the formations initially contained abundant sulfide minerals (Garrels and Christ, 1965). Consequently, such a technology would not introduce a new constituent into the formations that would further complicate the job of evaluating the overall environmental impact of the remediation process. Alternatively, introducing Fe^{++} or Mn^{++} could also reestablish reducing conditions, but would lack the sulfide component needed to re-precipitate some indigenous formation minerals. As an adjunct to adding iron or manganese, or as a stand-alone activity, one might also develop technologies to stimulate sulfate-reducing bacteria so that the indigenous sulfate in the groundwater could provide the sulfide needed to precipitate insoluble phases from pollutants such as As, Se, V and Mo.

4. Alternatively, it might be possible to identify chemicals that would not interfere with the removal of uranium but would *suppress the liberation of the other pollutants*. Aside from suppressing the oxidization of the indigenous pollutant-containing minerals (see #5, below) and having the principal complexant decay with time (see #2 above), we might target the specific chemistries of various pollutants. In conjunction with various additives, it is possible to form a host of sparingly soluble phases: calcium molybdate, calcium copper arsenate, apatite group minerals $[\text{Ca,Pb,Ba})_5(\text{VO}_4, \text{AsO}_4, \text{SeO}_4)_3(\text{Cl, OH, F})]$, as well as enhance the sorptive properties of many mineral surfaces.
5. Similarly, it might be possible to identify additives that would *suppress oxidation of some constituents* in the formation (notably pyrite and other sulfide minerals), while still allowing uranium-containing minerals to be oxidized. Phosphate, for example, is attractive in this regard because of its documented ability to suppress pyrite oxidation (Nyavor and Egiebor, 1995). Secondly, the first association constants (log units) of common inorganic ligands with the uranyl ion increase in the order: fluoride - 5.16, sulfate - 6.58, carbonate - 9.91, and phosphate - 13.2 (Guillaumont, 2003) so phosphate additions might also facilitate leaching of uranium. One concern is that a uranyl phosphate phase might also precipitate, but some preliminary geochemical modeling indicates that this problem is avoidable. Further modeling is needed to fully evaluate the possibilities presented by varying the pH and Eh of the system, in addition to manipulating phosphate concentration. A variation on this theme would make use of the various organophosphates developed by the detergent industry. These might also passivate sulfide mineral surfaces (with respect to oxidation) while having a negligible impact on uranyl ion solubility.

VI. Hydrogeologic Improvements for ISR Technologies:

A typical ISR well field consists of a regular network of production and injection wells, with each injection well surrounded by four to six recovery wells (5-spot or 7-spot patterns) 15 to 50 m away. The goal of such a design is for all the lixiviant injected in the injection wells to migrate toward the nearest production well(s), dissolving uranium and sweeping the entire ore body as it moves. Because of geologic heterogeneity, however, the ideal flow pattern rarely develops. Preferential flowpaths are typically present,

resulting in uranium being left behind in lower permeability sections of the aquifer and undesirable “excursions” of lixiviant occurring beyond the well field. Tracking and remediation of excursions represents a major expense when ISR operations are terminated. Optimization of the well-field design could lead to increased efficiency of uranium removal as well as decreased excursions requiring remediation.

In addition to the production well field, ISR operations also require a perimeter of monitoring wells within the ore horizon but beyond the production wells, as well as monitoring wells in aquifers above and below the ore horizon. The purpose of these wells is to detect excursions and other unanticipated hydraulic effects of ISR operations. Optimization of the monitoring well network can lead to enhanced confidence that excursions will be detected while reducing the cost of the network itself.

Well-field optimization has two primary components that coincide with specific Sandia areas of expertise: hydrogeologic characterization of the ore body and surrounding rock, and modeling to define the optimum design. Sandia possesses unique hydraulic and tracer testing capabilities, built around two Mobile Integrated Aquifer Testing and Analysis (MIATA) trailers and specialized well-test and tracer-test analysis codes (nSIGHTS and STAMMT-R). The MIATA trailers contain all the mechanical and electronic equipment needed for Sandia to perform any type of pumping or tracer test, with full pressure (head) and flow-rate data-acquisition capabilities.

Well testing is performed to define the three-dimensional heterogeneous distribution of permeability controlling lixiviant movement in the subsurface. To obtain the needed information, a well test must be properly designed with observation wells completed in appropriate horizons at appropriate distances, and a test duration adequate for interpretable responses to be produced. The test design must be based on an understanding of the geologic setting to ensure that heterogeneity is properly characterized. The Sandia code nSIGHTS is uniquely suited to analyze well tests in heterogeneous media because it is not based on analytical solutions that require unrealistically ideal conditions and does not require an initial assumption of radial flow to a well. It also defines the uncertainty associated with the parameter estimates it provides.

Tracer testing is performed to define and characterize the specific pathways by which water moves through a formation. It can identify the areas that are not well connected to a pumping well, which are areas where uranium might be left behind or where the residence time of injected lixiviant might be especially long. This information can be used to design a production well field that maximizes uranium extraction. The Sandia tracer-test-analysis code STAMMT-R provides estimates of the aquifer properties controlling transport of dissolved species.

With the information provided by hydraulic and tracer tests, recently developed methods for identifying and weighting alternative geological conceptual models in inverse groundwater flow model calibration can be applied to the specific problems of simulating lixiviant flow for ISR of uranium, minimizing the likelihood of excursions, and detecting any excursions that might occur. These probabilistic methods can be adapted to the specific fluvial stratigraphic systems that host uranium ore bodies that are commonly amenable to ISR to optimize uranium production and quantify investment and environmental risks by mining companies and regulatory bodies. ISR of uranium in the U.S. is licensed by the NRC, and these probabilistic methods would be consistent with the NRC’s desire to see that uncertainty in risk is quantitatively evaluated by license

applicants. With these methods, Sandia can design a production pattern for an individual ISR site that will optimize recovery efficiency and minimize environmental impacts, as well as an optimized monitoring network to detect excursions.

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Appendix 1 (from Taylor et al., 2004)

Table 1 Relative Features of Acid and Alkaline I situ Leaching

Acid leaching

Acid leaching achieves a high uranium extraction, typically 70-90%.

Acid leaching yields faster dissolution of uranium, requiring 40 to 70 pore volumes.

Increased concentration of dissolved solids (TDS) in recycled leach solutions (10-25 g/L).

High acid consumption for carbonate-bearing ores.

Mandatory use of corrosion resistant equipment and pipelines.

Addition of oxidant not always required because of presence of iron in recycled solutions.

Possibility of recovering by-products.

Additional processing on surface may be required to produce contaminant "free" product.

Risk of deterioration of permeability due to chemical and gaseous plugging.

Restoration to baseline levels requires an extended treatment period. Such restoration has only been demonstrated at one pilot site¹.

Seepage beyond bore field is unlikely due to formation of chemical precipitates that reduce porosity, and given natural attenuation due to reaction of contaminants with adjacent barren rock and unaffected groundwater.

Alkaline Leaching

Extraction from alkaline leaching is low(er), typically 60-70%.

Slower kinetics of uranium dissolution. Alkaline leaching requires typically more pore volumes than acid leaching.

Insignificant increase in groundwater TDS

Potential to treat ores containing high levels of carbonate.

Common material and equipment can be used.

Addition of oxidant always required.

Leaching chemistry is very selective for uranium.

Product solution from ion exchange should produce product of required quality.

Formation of carbonate or sulfate precipitates also a concern that can lead to plugging of formation.

Restoration of water to pre-mining baseline water quality has been demonstrated for some sites.

Potential for residual solutions to spread beyond the contours of areas being treated.

Note, for many acid ISL sites, restoration to pre-mining water quality has not been a requirement, because of the poor quality of the pre-mining groundwater.

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