University of Cincinnati

Date: 7/2/2013

I, Grant Spitler, hereby submit this original work as part of the requirements for the degree of Master of Science in Mechanical Engineering.

It is entitled:

In vitro dissolution of uranium contaminated soil in simulated lung fluid containing a pulmonary surfactant

Student's name: Grant Spitler

This work and its defense approved by:

Committee chair: Henry Spitz, Ph.D.

Committee member: Sam Glover, Ph.D.

Committee member: Thomas Richard Huston, Ph.D.

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In vitro dissolution of uranium contaminated soil in
simulated lung fluid containing a pulmonary surfactant

A thesis submitted to the
Graduate School
of the University of Cincinnati
in partial fulfillment of the
requirements for the degree of

Master of Science

in the School of Dynamic Systems
of the College of Engineering and Applied Science
by

Grant M. Spitler

B.S. Applied Physics, Xavier University
May 2010

Committee Chair: Henry Spitz, Ph.D.

Abstract

Uranium contamination in the dirt floor of an abandoned metal rolling mill was collected to determine whether it presents a significant exposure risk to workers when the facility is decontaminated. The contamination was present in the soil for over 50 years and subjected to rain, snow, ice and animal infestation while the building was abandoned. Multiple 100 mg samples of this dirt were exposed for increasing periods of time in simulated lung fluid to determine the dissolution rate. A pulmonary surfactant was added to half of the samples to determine whether the surfactant would affect the dissolution rate of uranium. Results indicate approximately 50% of the uranium dissolves quickly, within the first day, while no more than 15% of the uranium was found to dissolve during the intermediate time period, leaving 35-40% of the uranium undissolved, or constrained to a slow dissolution rate. The rapid dissolution of a significant fraction of the uranium contamination is atypical of most forms of uranium in soil and should be considered for safety controls for workers who may be exposed to resuspended uranium contamination during remediation. The addition of the pulmonary surfactant made no impact to the dissolution rate of uranium in the dirt.

Acknowledgements

First of all, I would like to thank DOE and DHS for their funding through the Nuclear Forensics research grant at the University of Cincinnati. I would also like to thank my academic advisor and committee chair, Dr. Henry Spitz, for the inspiration to pursue this research, as well as incredible guidance in planning all aspects of this work. There are also many people who helped me develop my laboratory abilities for the duration of this project. Lastly, I cannot go without mentioning my gratitude towards my wife, Elisabeth, who supported me during this endeavor.

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1.0 Introduction

The goal of this research was to determine the dissolution rate of uranium in simulated lung fluid and whether the presence of pulmonary surfactant affects the rate of uranium dissolution. Samples of dirt contaminated with uranium were collected from the dirt floor of an abandoned metal rolling mill that rolled approximately 30 million pounds of natural uranium metal ingots between 1948 and 1957 (Figure 1). A description of this legacy site was reported by Glassford et al. (2013). The solubility of uranium is an important factor in predicting the fate of the contamination in the environment and potential risk to humans. A series of experiments were performed using a standard formula for simulated lung fluid (SLF) modified by the addition of a lung surfactant to determine whether the dissolution rate would be affected by the presence of the surfactant. The surfactant was added to investigate whether inhaled uranium particles would interact with lysosomes found in pulmonary alveolar cells. The research study was conducted as a part of the nuclear forensics academic and research program at the University of Cincinnati, College of Engineering and Applied Science.

1.1 Background

Throughout most of the second half of the twentieth century, the U.S. Department of Energy (DOE) established multiple sites around the United States to develop nuclear materials for defense (Elless 1997). Activities at these sites ranged through multiple steps to fabricate nuclear weapons, starting with uranium ore through manufacturing nuclear fuel and operating nuclear reactors to produce materials for nuclear weapons components. Many of the legacy sites have been retired, including those used for uranium metal processing and fabrication. During the very early period of the Manhattan Project, regulations to control emissions of radioactive materials,

including uranium, were less rigorous than those of today. The United States Environmental Protection Agency (USEPA) has cited more than 1,500 hazardous waste sites within the United States on the National Priorities List (NPL), also known as Superfund waste sites, some of which were part of the nuclear weapons complex (EPA 2011).

While a few legacy facilities have been decontaminated and plans have been developed for others, residual uranium metal contamination remaining at a few facilities requires evaluation of risk to humans and the environment should this material be released or resuspended during site remediation. An important route of exposure is inhalation of uranium particles that would deposit in the respiratory tract and become distributed in other organs and tissue depending upon the solubility of the particles. This legacy contamination is also an excellent surrogate material for nuclear forensics analyses training since the processes that generated the contamination are well known.

The literature describes results of many studies involving the solubility of uranium in simulated lung fluid and environmental media to elucidate risk to human health (Elless et al. 1997; LaMont et al. 2001; Dennis et al. 1982; Heffernan et al. 2001). This research has focused on a unique form of uranium metal contamination that has been exposed for approximately 50 years to the effects of weathering that are believed to affect the rate at which inhaled particles would dissolve in the respiratory tract should the material be resuspended (Elless et al. 1997).

1.2 Lung Solubility and Alveolar Surfactants

Many studies have been performed to determine uranium dissolution in the respiratory tract (Heffernan et al. 2001; Mercer 1967; Cooke and Holt 1974; Chazel et al. 1998; Eidson and Mewhinney 1980; Eidson and Griffith 1984; Eidson 1994). Likewise, many clinical studies

have been performed regarding the significance of pulmonary surfactants for treatment of respiratory disease, especially their effect on trace metals (Wright 2003; Whitsett et al. 2010; Devendra and Spragg 2002). One hypothesis adopted in this research was to determine whether a pulmonary surfactant would affect the dissolution of uranium particles in the respiratory tract because evidence has been documented of lysosomes in certain surfactants concentrating and interacting with heavy, toxic metals (Berry et al. 1988).

1.2.1 Lung Dissolution Experiments

The focus of reports on the dissolution of uranium in the respiratory tract described in the literature include studies of (1) different uranium compounds (Mercer 1967; Cooke and Holt 1974; Chazel et al. 1998), (2) *in vitro* dissolution of uranium produced at uranium mills (Eidson and Mewhinney 1980; Eidson and Griffith 1984), and review articles of *in vivo* and *in vitro* dissolution rates of uranium compounds present at industrial sites (Eidson 1994). Metzger et al. (1997) studied the solubility of airborne uranium compounds found in a uranium processing plant. Heffernan et al. (2001) measured airborne uranium collected on filters at the former Fernald plant. There are no studies on uranium solubility that have explored the role of pulmonary surfactants found in the deep lung. Consequently, for this research, the conventional recipe for simulated lung fluid was modified to incorporate pulmonary surfactant that is commonly used to treat pneumothorax and acute atelectasis, especially in neonates (Lee 2012; Wedding and Gylys 2005).

1.2.2 Composition and Mechanisms of Pulmonary Surfactant

The alveolar region of the lung is where the majority of oxygen and carbon dioxide gas exchange between the lung and blood stream occurs (Wright 2003). The thin epithelium coats the surface of the alveolar region of the lung is lined by type I and type II alveolar epithelial cells that are in contact with respiratory gases. These cells provide a region for air-liquid interactions (Whitsett et al. 2010). Type I epithelial cells comprise the wall of the alveolus, while Type II epithelial cells are scattered within the alveolar walls and are found at intersecting points of multiple alveoli where they serve as a reserve for producing and repairing damaged surfactant (Caceci 2012).

Surfactants are surface active agents that, in the general sense, are molecular agents that are usually amphiphilic organic compounds. Consumer products containing surfactants include soaps and detergents that are used to clean hydrophobic substances in a manner similar to a pulmonary surfactant. One end of the surfactant molecule exhibits a hydrophilic nature and adheres to areas where gases and liquids meet. The other end of the surfactant molecule is hydrophobic, usually consisting of hydrocarbons, and interacts with other hydrophobic substances, such as bacteria or oil-like material, in air (Furse 2012).

Pulmonary surfactant is an essential coating on the walls of the alveoli, enabling expansion and contraction by reducing surface tension. Without surfactant present at the air-capillary barrier, the collapsing forces of the lung would render the alveoli non-functional (Whitsett et al. 2010). Pulmonary surfactant is a lipid rich material that maintains lung volume while reducing surface tension of the aqueous film surrounding the cells and lessens the intermolecular attraction of water molecules (a hydrophobic barrier). In addition, various proteins are found in surfactants

that also provide defensive measures against foreign bodies and pulmonary inflammation. Some of these proteins interact with macrophages to carry out these duties through phagocytosis (Wright 2003). Macrophages are free cells within alveolar lumen, which serve as phagocytic sterilizers in the deepest part of the lung. When the lung is diseased, the number of macrophages increases due to the mobilization of monocytes.

The respiratory tract is comprised of an elaborately branched, tubular structure, which culminates into roughly 300 million alveolar sacs. Type II epithelial cells are a critical provider for surfactant fabrication and provide repairing capabilities following injury. Type II epithelial cells also generate type I epithelial cells, which reside over the majority of the gas-exchange region of the alveoli (Whitsett et al. 2010). Pulmonary surfactants have three general components; phospholipids (80%), proteins (10%), and neutral lipids (10%). Phospholipids include phosphatidylcholine (PC) and phosphatidylglycerol, with contributions from phosphatidylinositol and sphingomyelin (Whitsett et al. 2010; Devendra and Spragg 2002). The surfactant proteins (SP) are comprised of four components, SP-A, SP-B, SP-C, and SP-D, each having a specific task (Wright 2003). SP-A and SP-D provide defense properties, while SP-B and SP-C are incredibly hydrophobic, interacting with the phospholipids in surface tension stabilization (Devendra and Spragg 2002). The host defense proteins, SP-A and SP-D, are known to guard against infection and inflammation by facilitating phagocytosis through macrophages and monocytes (Wright 2003). SP-A has a mechanism to bind to lipopolysaccharides and numerous microbial pathogens that augments alveolar clearance (Whitsett et al. 2010). SP-D plays a role in the structure formation of surfactant and regulating alveolar surfactant pools and reuptake (Ikegami et al. 2005). The roles of SP-A and SP-D, along

with some of the physical properties of surfactant in general, may have a role in affecting the dissolution rate of particles deposited in the respiratory tract.

1.2.3 SURVANTA®

SURVANTA® (beractant) (Figure 2), the pulmonary surfactant that was added to the conventional recipe for simulated lung fluid, is commonly used to treat Infant Respiratory Distress Syndrome (IRDS) and Acute Respiratory Distress Syndrome (ARDS) (Blanco and Perez-Gil 2007). It is a natural bovine lung extract that contains phospholipids, neutral lipids, and surfactant-associated proteins found in pulmonary surfactant. Although SURVANTA® does not contain SP-A, one of the key proteins associated with alveolar defense, it does contain SP-B and SP-C (SURVANTA® 1999). Since this surfactant is manufactured to prevent and control RDS, the presence of surface tension active protein rather than alveolar defense proteins is understandable. However, the hydrophobic proteins may not fully mix with the high water volume of the SLF.

1.2.4 Toxic Materials in the Lung

Berry et al. (1988) has studied whether toxic mineral elements, such as chromium, cerium, uranium, or other readily and slightly soluble materials become concentrated in specific areas of the body, especially the pulmonary region of the respiratory tract following inhalation. It was reported that toxic heavy metals are not transported from deposition sites in the alveolar region of the lung into the blood stream, but accumulate within the lysosomes found in pulmonary alveolar cells (Berry et al. 1988).

There are several mechanisms that describe interactions of toxic metals, such as uranium, in the alveolar region of the respiratory tract. A mechanism of interest involves clearance of material isolated by lysosomes from the alveoli. Pulmonary lysosomes, such as type I pneumocytes, macrophages, and lysosomal hydrolases, will concentrate toxic substances. The manner in which they treat a foreign substance plays a pivotal role in the fate of the toxic material within an organism (Berry et al. 1988). One or more of these mechanisms may affect dissolution of the uranium metal contamination in samples of dirt collected at the rolling mill, should it be inhaled after resuspension.

Toxicity of a material is significantly dependent on its solubility. Insoluble materials reaching the pulmonary alveoli may also be engulfed by macrophages. Alternatively, the material may cross the alveolar-capillary barrier and be transported to other areas of the body which have an affinity for heavier materials (e.g. bone) (Berry et al. 1988).

A second group of very small, clay-like inhaled insoluble particles can reach the alveoli and concentrate in the macrophages. The difference occurs when lysosomal hydrolases slowly dissolve the minerals that may be released and transported through alveolar clearance. Some toxic elements bind to cerebral tissues, causing incurable cerebral lesions (Roberts 1987).

Inhaled water-soluble toxic particles, such as iodine, diffuse rapidly across the alveolar-capillary barrier and into target organs. Alternatively, alveolar clearance of such particles would involve being captured by lysosomes of alveolar macrophages where the dissolved particles concentrate. If the toxic substances concentrate in the macrophages, they cannot spread to target organs, but instead are slowly eliminated through alveolar clearance (Berry et al. 1988).

The complex, diverse physical arrangement of the respiratory tract makes it challenging to describe the transport and dissolution of inhaled particles. If SP-A and SP-D treat inhaled particles of uranium metal as foreign bodies in the alveolus, then dissolution of uranium should be affected by these protein interactions. It becomes a question whether the surfactant traps and isolates toxic metals, like uranium, or expels them through an alveolar clearance mechanism. It is also necessary to consider how well a surfactant will interact with components of the conventional SLF.

1.3 Scope and Aims of Experiment

This study was designed to investigate the dissolution rate of uranium metal contamination in simulated lung fluid made using a conventional recipe that has been augmented by the presence of a pulmonary surfactant. The uranium metal contamination was generated between 1948 and 1957 by rolling hot forged uranium metal billets into rods at a rolling mill that was first opened in 1911. The mill was closed and essentially abandoned after 1958 and has not been decontaminated. The dirt floor of the mill is contaminated with particles of uranium and other metals and is subjected to direct environmental effects from contact with water from rain, snow, ice, and animal infestation since the roof and windows are in disrepair.

This study investigated the dissolution rate of uranium metal particles present in dirt by sequentially exposing approximately 100 mg of dirt to simulated lung fluid for increasing periods of time from 1 hour up to 30 days. The recipe for SLF was modified by the addition of a pulmonary surfactant. The measured rate of dissolution was evaluated using categories adopted by the International Commission on Radiation Protection (ICRP60 1991). These categories are (F-fast), (M-moderate), and (S-slow), and are closely analogous to former categories: (D-days),

(W-weeks), and (Y-years), established by ICRP30 (1982). ICRP Publication 66 provides parameter values for the F, M, and S absorption/dissolution types and discusses the fate of uranium upon entering the blood stream and removal half-times from the body as a whole (ICRP66 1994).

The F category represents materials that readily absorb into blood (within a few days), corresponding to the D inhalation class. This accounts for a significant amount of material being absorbed from the extrathoracic and bronchial regions of the lung. A fraction of these materials are trapped in mucus and swallowed, while the remaining fraction is absorbed through the epithelium. For F class material, the removal half time, $t_{1/2}$, is about 10 minutes (ICRP66 1994).

The M category characterizes materials with an intermediate absorption rate, corresponding to the W inhalation class. In this category, 10% of the inhaled fraction is considered to dissolve quickly (ICRP66 1994).

Category S denotes comparatively insoluble materials and corresponds to the Y inhalation class. This represents the particle transport rate of the slow clearing alveolar interstitial (AI) compartment. Even for this category, there is a very small contributing factor of rapid uptake after inhalation, often considered a negligible amount (ICRP66 1994).

Determining whether pulmonary surfactant in SLF affects the dissolution rate of uranium metal contamination is the second focus of this research. Any differences will be determined by measuring the amount of uranium dissolved between SLF with and without surfactant, for each dissolution fraction.

2.0 Materials and Methods

Four samples of approximately 100 mg of uranium-contaminated dirt collected from the dirt floor of the abandoned uranium rolling mill were prepared and used in this dissolution study. Initial uranium measurement of each 100 mg sample of uranium-contaminated dirt was performed using gamma spectrometry. A high purity germanium detector (Canberra model GC10021) and computer based gamma spectrometry and analysis software (Genie 2000) were used to measure each sample arranged on the face of the detector. The standard recipe for simulated lung fluid is shown on Table 1 (Moss 1979). The pulmonary surfactant, SURVANTA®, was added to half of the samples at the beginning of the experiment to determine whether the surfactant affects the rate of dissolution of uranium. Table 2 lists the detailed characteristics and aliquot sizes for each of the 8 individual samples that were processed in this research project. The uranium content in each sample was determined by gamma spectrometry prior to dissolution testing since the uranium contamination is not homogeneously distributed in the bulk material (Glassford et al. 2013).

2.1 Simulated Lung Fluid Fabrication

Table 1 lists the components of the SLF used in the research at recommended and measured concentrations used (Moss 1979). All components of the SLF, as well as the containers must be sterilized to minimize mold and bacterial growth. The SLF was sterilized by filtration using a 0.22 µm syringe filter. The surfactant was originally sterilized by the manufacturer, but not filtered prior to use because it was too viscous. All materials and equipment that came into contact with the SLF and surfactant was sterilized using an autoclave to minimize bacterial contamination (Figure 3).

2.2 Dissolution Procedure

Approximately 100 mg of uranium-contaminated dirt and 100 mL of simulated lung fluid (as the solvent with and without surfactant) were added to a 250 mL polyethylene bottle. This size bottle and volume of solvent were selected to insure good surface to volume contact between the sample and solvent. SLF containing surfactant was a mixture of 98% conventional SLF with 2.0% SURVANTA® surfactant. The pH of the solvent was controlled between 7.2-7.4 with the addition of CO₂ as needed. A pH meter was used to monitor the solution during the dissolution period. All sample bottles were immersed in a shaking water bath maintained at 37 °C with the rotational set to 125 rpm (Figure 4).

After each dissolution period, the sample and solvent were separated using a centrifuge at 2,500 rpm for 30 minutes (Figure 5) (Briant and James 1990). Immediately after centrifuging, a pipet was used to extract 90% of the supernatant on top of the sample. The supernatant was acidified with a concentrated HNO₃ at 10% of the total solution to retain the dissolved uranium in solution (Figure 6). Fresh SLF was added to each of the decanted samples to begin the next period of dissolution. No additional surfactant was added since contact with the uranium was expected to be complete within the first hour (Figure 7).

The dissolution period adopted for each of the fractions was 1 hour, 4 hours, 8 hours, 24 hours, 7 days, and 30 days. These periods were selected because they are consistent with observing the type F and M soluble fractions adopted by Briant and James (1990). Any remaining undissolved uranium after 30 days was considered as type S. All undissolved uranium remaining after 30 days was totally dissolved for analysis using a rigorous potassium fluoride and pyrosulfate fusion procedure (Sill 1987).

3.0 Analysis

The precise content of uranium in each of the dissolution fractions was determined by using anion exchange chromatography, electrodepositing the uranium onto a stainless steel planchet, and measuring uranium on the planchet using alpha spectrometry. The selection of aliquot size was determined by measuring the gross alpha content of a 1.0 mL aliquot of each fraction using liquid scintillation counting (LSC). Excessive uranium mass in a sample will degrade the resolution of the alpha spectrometer. Thus, the size of the aliquot selected is based upon a volume of the dissolution fraction that contains less than 50 disintegrations per minute (dpm) of uranium alpha activity measured by LSC. The rate of uranium dissolution was determined by the percentage of total uranium that dissolved in increasing intervals of time. The quantity of total uranium is determined by the sum of uranium measured in each fraction plus any residual uranium that did not dissolve after 30 days. The undissolved uranium was brought into solution using a modified potassium fluoride and pyrosulfate fusion (Sill 1987; UCRADLAB-100 2011). It was unnecessary to fuse the sample blanks, as there was no sample to fuse.

3.1 Liquid Scintillation Counting for Aliquot Determination

Prior to using the LSC, 1.0 mL of sample aliquot was added to 20.0 mL of LSC cocktail solution (Perkin Elmer®, Ultimate Gold™ LLT, High Flash-point LSC Cocktail) in a glass vial. A natural certified uranium solution is used as a reference for a gross alpha calibration standard for comparison with each dissolution sample aliquot. The LSC gross alpha protocol detects alpha particles in a range between 4-5 MeV, corresponding with the uranium decay chain (Figure 8).

3.2 Potassium Fluoride (KF) and Sodium Bisulfate (NaHSO₄) Fusion

Dissolution refers to dissolving a substance in a desired liquid at low temperatures, sometimes with a chemical reaction. However, sometimes more vigorous methods involving molten flux to force materials to decompose are required to assure that complete dissolution will occur, especially for soils containing high silica content (Bock pp. 64 1979). Decomposition of undissolved solids in a melt is typically conducted in a small crucible, highly resistive to the flux being used. Platinum is the most popular crucible since it has a high melting point of 1772⁰C and remains unaffected by any commonly used acids, except for concentrated phosphoric acid at high temperatures and other combinations of undiluted acids (Bock pp. 9 1979). The EPA MARLAP training guide offers suggestions on the type of crucible, the fusion flux, as well as the necessary temperature to perform a fusion (MARLAP 2004). Sill (1987) has also described a method involving multiple steps of adding melt material and performing multiple fusions in one procedure should undissolved material remain after a traditional fusion. Boron and lithium fusions can also be used, although sodium mixtures will typically react faster than lithium and lithium borate fusions which require higher melting temperatures than a sodium or potassium fusion (Bock pp. 63 1979; MARLAP 2004).

Silicates are almost always present in soil. Therefore, it is necessary to remove silica from samples because they will degrade alpha spectra (Sill 1987). A potassium fluoride decomposition method was found to be suitable for dissolving the remaining sample in the post 30 day fractions. Residual aluminum and iron oxides, which were present in the dirt from the rolling mill, were treated by using a pyrosulfate fusion. Fortunately, the uranium in the dirt collected was highly soluble once disassociated from other soil contents, especially in a nitric acid solution.

Approximately 150 mL of 8M HNO_3 was added to the undissolved fraction as a preservative and to insure that the uranium remains in solution. The undissolved fraction was vacuum filtered using a Whatman #42 filter to remove any residual particles. The filtered fraction, along with two distilled water rinses was transferred to a 500 mL polyethylene bottle. The filter and its contents were then ashed slowly in a platinum dish until the entire contents exhibited a dark, soot-like appearance. The filtered sample was subjected to 0.5 mL of concentrated nitric acid with low heat to aid in ashing and to insure the sample was retained within the platinum dish. Approximately 3.0 g of anhydrous potassium fluoride was added to the sample in a platinum dish and heated slowly over a Meker burner until a clear fusion melt was achieved. The sample in the dish was then cooled to room temperature and 4.0 mL of concentrated sulfuric acid was added to the platinum dish and heated gently, until excessive fumes of sulfuric acid subsided and a thick melt formed. An additional 0.5 g of anhydrous potassium fluoride was sprinkled over the melt and quickly swirled to volatilize any remaining silica. Four drops of concentrated sulfuric acid was added with heat to volatilize hydrogen fluoride, silicon tetrafluoride and water. The melt was allowed to cool and solidify into a brown-olive colored cake, with some darker, undissolved pieces still remaining.

The remaining soil was dissolved by adding 2.0 g of sodium bisulfate to the cooled cake and heating until a clear pyrosulfate fusion was observed. The fusion was then swirled around the sides of the dish as it cools to produce a uniformly thin solid layer which was easily removed from the walls by flexing the dish (Figure 9). The cake was removed from the dish by addition of very dilute HCl (35:1) and rinsed into a beaker. The beaker was heated over a burner to dissolve any undissolved cake, leaving a bright green solution that was filtered by vacuum and

added to the pre-fusion portion. Any remaining solid material was considered as a metal or alloy other than uranium and discarded.

3.3 Tracer Standard for Sample Recovery

The uranium concentration in each of the dissolution fractions is determined by alpha spectrometry. A known quantity of a ^{232}U isotopic tracer, not exceeding 10 dpm in activity, is added to each fraction as a means to determine the chemical yield. Any losses of sample through the procedure can be corrected by determining the recovery of tracer.

3.4 Anion Exchange Chromatography

Before the uranium concentration of each sample can be determined, it must be chemically purified via anion exchange chromatography. The procedure used in this research was adapted from Lamont et al. (2001). Ten anion exchange columns are processed at one time that include eight dissolution fractions, a tracer blank, and a natural uranium standard solution as a quality control (Figure 10). The uranium separation process was performed seven times; six iterations included the dissolution fractions and sample blanks, while a seventh included the fused fractions with no sample blanks (Table 2). Any detected uranium in a blank sample above the minimum detectable activity (MDA) of approximately 0.030 dpm for each alpha detector was considered contamination. Each sample aliquot was traced with the ^{232}U , dried on a hot plate, and dissolved with 5.0 mL concentrated HNO_3 and repeated once. After drying, the samples were converted to chloride form by adding 5.0 mL of concentrated HCl and dried two additional times to insure that all the uranium was in the same oxidation state.

Each aliquot was further dissolved in 25.0 mL of 9M HCl prior to loading onto a column containing 10.0 mL of BioRad AG 1x4, 100-200 mesh (Cl^- form) resin. The resin in the column was rinsed with 5 column volumes of 1M HCl. Each column was conditioned for sample loading with 5 column volumes of 9M HCl. After loading the samples, the sample beakers were rinsed with 5 column volumes of 9M HCl (split into two separate 2.5 column volume rinses) and used to elute thorium from the columns. Thereafter, the columns were rinsed with 3 column volumes of 8M HNO_3 to elute the iron. Uranium was eluted using 7 additional column volumes of 8M HNO_3 .

3.5 Electrodeposition Procedure

Each sample was electrodeposited onto a 5/16 inch stainless steel planchet. Following UC RADLAB Protocol 510 (2011), 2.0 mL of 0.36M NaHSO_4 was added to the extracted uranium solutions, and dried. Subsequently, the samples underwent dry-downs with 5 drops of concentrated H_2SO_4 in 5.0 mL concentrated HNO_3 . Samples were dried down once more with 5.0 mL of concentrated HNO_3 and several drops of 30% H_2O_2 , and wet-ashed twice using 5.0 mL concentrated HNO_3 . The dried samples were dissolved in 5.0 mL of 0.75M H_2SO_4 and 4 drops of thymol blue indicator. The samples were transferred into the electrodeposition cell with two additional 2.5 mL rinses of 0.75M H_2SO_4 for each sample beaker (Figure 11). The solution in the cell was adjusted to a pH between 2.0-2.3 using concentrated NH_4OH . A platinum wire anode was inserted into the cell and electrodeposition was conducted for one hour at a constant current of 0.75 A, and a starting voltage set between 10-11 V. After one hour, the samples were quenched for one minute with 10% NH_4OH in order to keep the acidic solution from eroding

the deposited sample. The cell was disassembled and the electrodeposited planchet was rinsed with DI water and dried under a heat lamp.

3.6 Alpha Analysis

Alpha spectrometry measurements were performed within three days after separation to avoid interference from the ingrowth of thorium (especially from the ^{232}U tracer) and other progeny due to the following decay schemes:

Alpha spectrometry measurements were performed with each planchet was measured for 1000 minutes and analyzed using Alpha Analyst software (CANBERRA). The planchet was placed on shelf #2 at a distance of 5.0 mm (Figure 12). A thin collodion film was placed over each planchet to minimize recoil nuclei from contaminating the face of the detector (Inn et al. 2008; Sill and Olson 1970).

4.0 Data Analysis

The data selected for analysis was based on the isotopes of natural uranium, ^{238}U , ^{234}U , and ^{235}U , and the ^{232}U tracer exhibiting distinct alpha energy peaks at 4.197 MeV, 4.776 MeV, 4.398 MeV, and 5.320 MeV, respectively (Figure 13). The spectra contain all three natural uranium

isotopes plus the ^{232}U tracer representing four distinct, well-resolved alpha energy peaks. The relative quantities of ^{238}U , ^{234}U , and ^{235}U are consistent with the isotopic abundance of natural uranium. The ^{238}U peak was well resolved from the other isotopes and was adopted for analyzing the concentration of uranium and its uncertainty in each sample.

The uranium isotopic activity for each 100 mg sample was determined by summing the activity in each dissolved fraction plus the residual material. Figures 14-17 show the percent of total uranium activity that remained undissolved from each fraction with time for each of the samples. SigmaPlotTM 11.0 (2008) was used to determine a best fit, nonlinear regression function to the data and identified that the dissolution rate is best described using the following exponential expression, which reveals an exponential decay expression

$$F(t) = Ae^{-kt} + Be^{-lt} + Ce^{-mt}$$

where values A, B, and C are fractions for each component, k , l and m are the dissolution rate constants for each component, and t is time in days. The constants in each equation represented the percentage of uranium dissolved based on the dissolution rate constants.

Chemical recovery of uranium, post anion exchange chromatography, was from measuring the known vs. measured alpha activity due to the ^{232}U tracer. Because the tracer and aliquots were homogeneously mixed through the wet-ashing procedures prior to anion exchange chromatography, the recovery of the other uranium isotopes reflects that of the tracer. If recoveries were found to be too low (less than 40%, or considerably low compared to the other samples), anion exchange chromatography and alpha spectrometry procedures were repeated for that particular sample fraction.

The major source of uncertainty was associated with measurements of alpha activity and was approximately 5%. Other contributions to uncertainty were less than 10^{-4} and were mostly associated with variations in aliquot and tracer mass. Summation of total error for a given fraction was determined through the function:

$$(\sigma_f/f) = [(\sigma_A/A)^2 + (\sigma_B/B)^2 + (\sigma_C/C)^2 \dots]^{1/2},$$

Where σ represents the standard deviation of a value, and A, B, and C reflect specific points of uncertainty. While propagating the uncertainty for the remaining undissolved uranium, any dissolution fractions contributing to the calculation also contributed to the uncertainty calculations. Since the dissolution rate is determined by how much uranium remains undissolved over time, the fusion fraction data also contributed to overall uncertainty.

5.0 Results

Figures 14-17 demonstrate that the dissolution of uranium in all the soil samples exhibits nearly identical solubility, with approximately 50% class F, 10-15% class M, and 35-40% class S. All measurements of uranium for each of the dissolution fractions were above the MDA of approximately 0.030 dpm/sample. Table 3 shows that all sample recoveries ranged from 49.4% for SLF1B to 91.8% for SLF2B.

6.0 Discussion

The addition of surfactant appears to make no difference in the dissolution rate of uranium in the samples analyzed. Results on Table 4 demonstrate that measurement uncertainties more than account for any differences in the undissolved uranium after each fraction. The heterogeneity of uranium metal contamination in the bulk soil has been well described (Glassford et al. 2013).

Furthermore, differences in uranium metal treatment used at the mill could also impact solubility. Although the feed stock at the mill was uranium metal, the contamination in the dirt floor of the mill, created by mechanically rolling hot forged uranium metal stock, remained in contact with the soil and moisture for over 50 years. Therefore, it is very likely that the elemental form of the uranium is variable. Some of the variation in solubility among all the samples may be due to the presence of UO_2 and U_3O_8 . However, the large fraction of rapidly soluble uranium suggests that a significant component of the uranium contamination is in the form of a well oxidized U^{6+} metal. In contrast, analysis of samples containing NBL CRM 129 demonstrates that pure U_3O_8 is very insoluble (Table 5). Oxidized uranium metal present in soil arising from one of several rolling methods is now likely in the form of U^{6+}O_2 which confirms the observation for the large, rapidly soluble component (FUSRAP 2010).

Dose conversion factors provided by RESRAD 6.5 (2009) were used to determine the radiation dose and risk associated with exposure to uranium contamination observed at this abandoned facility. Dose rates associated with solubility type S, M, and F natural uranium are 2.59 mrem/Bq, 0.95 mrem/Bq, and 0.158 mrem/Bq for S, M, and F classes, respectively. RESRAD also give the mortality risk for the S, M, and F classes as $1.96\text{E-}06/\text{Bq}$, $7.84\text{E-}07/\text{Bq}$, and $3.41\text{E-}08/\text{Bq}$, respectively.

7.0 Conclusions

The dissolution rates for natural uranium contamination in the dirt floor of the abandoned metal rolling mill are best described using a three component exponential function. The presence of a pulmonary surfactant in the simulated lung fluid made no observable difference in the uranium dissolution rate. After oxidation and weathering for more than 50 years, the uranium

contamination present in the soil at this abandoned rolling mill appears to exhibit a significantly soluble fraction that presents less risk to humans following inhalation exposure. Results of this study provides health physicists and engineers with quantitative technical guidance for establishing appropriate protective measures to avoid unnecessary risk for workers and the environment. Future studies should include analysis of the solubility associated with the respirable fraction of this contaminated soil and whether a greater concentration of pulmonary surfactant can alter the risk. The physical and chemical characteristics associated with the uranium contamination in the soil provide the nuclear forensics professional with a basis for interpreting what type of process generated the observed contamination.

8.0 References

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Appendix

Table 1 – Simulated Lung Fluid Composition (Moss 1979)

Chemical	Formula	Recommended Concentration (g/L)	Measured Actual (g/L)
Magnesium chloride	MgCl ₂ *6H ₂ O	0.203	0.2067
Sodium chloride	NaCl	6.019	6.0193
Potassium chloride	KCl	0.298	0.3017
Sodium phosphate, dibasic	Na ₂ HPO ₄ *7H ₂ O	0.268	0.2681
Sodium sulfate	Na ₂ SO ₄	0.071	0.0711
Calcium chloride	CaCl ₂ *2H ₂ O	0.368	0.3697
Sodium acetate	NaH ₃ C ₂ O ₂ *3H ₂ O	0.952	0.9528
Sodium bicarbonate	NaHCO ₃	2.604	2.6115
Sodium citrate	Na ₃ H ₅ C ₆ O ₇ *2H ₂ O	0.097	0.0981

Table 2 – Sample IDs, Solvent Used, and Soil Masses

Sample IDs	Solvent	Soil Mass (g)
SLFA	100% SLF	0.1020
SLFB	100% SLF	0.1064
CRMSLF	100% SLF	0.1066
BSLF	100% SLF	Blank, No Soil
SURA	98% SLF, 2% Surfactant	0.1153
SURB	98% SLF, 2% Surfactant	0.1086
CRMSUR	98% SLF, 2% Surfactant	0.1066
BSUR	98% SLF, 2% Surfactant	Blank, No Soil
PB	Tracer Blank	U Separation Only
QC	Quality Control	U Separation Only

Table 3 – Sample Fraction Data; Alpha Spectrometry Results and Recoveries

Sample ID	U-232 Activity (dpm)	%Error	U-238 Activity (dpm)	%Error	Tracer Start (dpm)	aliquot size (fraction of total volume)	Recovery
SLF1A	6.429	2.72	6.837	2.65	8.791	0.0105	0.731
SLF2A	6.960	2.63	2.227	4.40	8.519	0.0211	0.817
SLF3A	7.106	2.60	1.692	5.02	8.954	0.0210	0.794
SLF4A	6.837	2.65	1.358	5.58	9.066	0.0210	0.754
SLF5A	7.493	2.55	2.665	4.04	8.819	0.0209	0.850
SLF6A	7.228	2.59	2.880	3.90	9.145	0.0105	0.790
SLFA-F	7.707	2.44	5.176	2.89	9.289	0.00967	0.830
SLF1B	4.353	3.34	4.634	3.24	8.813	0.0105	0.494
SLF2B	8.009	2.55	3.553	3.66	8.723	0.0210	0.918
SLF3B	6.465	2.80	1.146	6.27	9.010	0.0211	0.718
SLF4B	6.368	2.82	1.465	5.57	9.098	0.0210	0.700
SLF5B	7.161	2.68	2.816	4.08	8.881	0.0209	0.806
SLF6B	6.768	2.74	2.842	4.06	9.213	0.0105	0.735
SLFB-F	7.876	2.44	7.829	2.45	9.374	0.0107	0.840
SUR1A	6.597	2.69	8.634	2.40	8.740	0.0106	0.755
SUR2A	7.923	2.49	5.373	2.94	8.856	0.0210	0.895
SUR3A	6.268	2.84	1.466	5.57	9.100	0.0211	0.689
SUR4A	6.560	2.69	2.029	4.60	9.194	0.0210	0.713
SUR5A	6.690	2.67	2.352	4.29	8.991	0.0210	0.744
SUR6A	6.468	2.80	1.847	4.98	9.447	0.0105	0.685
SURA-F	7.394	2.64	10.71	2.27	9.432	0.0110	0.784
SUR1B	6.254	2.84	6.668	2.76	8.637	0.0105	0.724
SUR2B	7.772	2.59	5.380	3.03	8.847	0.0211	0.878
SUR3B	6.439	2.800	1.931	4.88	9.166	0.0211	0.702
SUR4B	6.866	2.73	1.834	5.00	9.239	0.0212	0.743
SUR5B	6.903	2.72	4.179	3.400	9.040	0.0213	0.764
SUR6B	7.298	2.66	2.058	4.73	9.269	0.0105	0.787
SURB-F	7.090	2.53	8.967	2.300	9.426	0.00979	0.752

*number in ID represents fraction; 1=1hr, 2=4hr, 3=8hr, 4=1d, 5=7d, 6=30d, F=Fusion

***'A' represents 1st of 2 replicated experimental runs; 'B' represents 2nd of 2

***'SLF' = only simulated lung fluid used; 'SUR' = samples with surfactant added

Table 4 – Sample Fraction Data; ^{238}U Activity calculations and Uncertainties

Sample ID	Total ^{238}U Activity for Sample Fraction (dpm)	Total ^{238}U Activity (dpm)	Activity (Bq)	Total ^{238}U Activity (Bq)	Uncertainty (%)	Cumul. Uncert (%)
SLF1A	891.5		14.9		5.36	6.56
SLF2A	129.5		2.16		6.37	7.41
SLF3A	101.4		1.69		5.65	6.80
SLF4A	85.8		1.43		7.24	8.17
SLF5A	149.8		2.50		6.09	7.17
SLF6A	347.5		5.79		6.02	7.11
SLFA-F	645.0	2350	10.7	39.2	3.78	5.79
SLF1B	891.7		14.9		5.25	6.29
SLF2B	184.1		3.07		5.64	6.62
SLF3B	75.8		1.26		7.69	8.43
SLF4B	99.7		1.66		7.14	7.93
SLF5B	166.7		2.78		5.98	6.91
SLF6B	367.7		6.13		6.00	6.92
SLFB-F	871.7	2657	14.5	44.3	3.46	5.88
SUR1A	1079		18.0		5.01	6.10
SUR2A	286.1		4.77		5.19	6.25
SUR3A	100.8		1.68		7.16	7.96
SUR4A	135.3		2.26		6.37	7.26
SUR5A	150.3		2.50		6.14	7.06
SUR6A	257.7		4.30		6.69	7.54
SURA-F	1242	3252	20.7	54.2	3.48	5.72
SUR1B	879.3		14.7		5.23	6.25
SUR2B	290.3		4.84		5.25	6.27
SUR3B	130.3		2.17		6.58	7.42
SUR4B	116.6		1.94		6.64	7.47
SUR5B	257.1		4.28		5.54	6.51
SUR6B	249.6		4.16		6.41	7.27
SURB-F	1218	3141	20.3	52.4	3.42	5.58

Table 5 – CRM 129; Alpha Spectrometry Results, Recoveries, Activities and Uncertainties

Sample ID	U-232 Activity (dpm)	%Error	U-238 Activity (dpm)	%Error	Tracer Start (dpm)	aliquot size (fraction of total volume)	Recovery	Total U-238 Activity for Aliquot (dpm)
CRMSLF1	4.79	2.87	0.27	11.47	8.82	0.0106	0.54	0.49
CRMSLF2	6.87	2.46	0.31	10.65	8.74	0.0210	0.79	0.39
CRMSLF3	6.12	2.58	0.18	13.95	9.02	0.0210	0.68	0.27
CRMSLF4	5.63	2.67	0.68	7.19	9.18	0.0210	0.61	1.11
CRMSLF5	5.63	2.67	1.50	4.89	8.91	0.0210	0.63	2.38
CRMSLF6	6.28	2.55	5.87	2.63	9.24	0.0106	0.68	8.64
CRMSLF-F	6.65	2.62	11.92	2.07	9.38	0.0002	0.71	16.81
CRMSUR1	4.75	2.88	0.33	10.27	8.61	0.0106	0.55	0.60
CRMSUR2	6.18	2.57	0.39	9.48	8.99	0.0209	0.69	0.56
CRMSUR3	5.93	2.61	0.14	16.11	9.16	0.0211	0.65	0.21
CRMSUR4	6.08	2.59	0.28	11.25	9.25	0.0213	0.66	0.42
CRMSUR5	6.58	2.50	2.04	4.23	9.07	0.0210	0.73	2.81
CRMSUR6	6.87	2.46	2.78	3.66	9.32	0.0106	0.74	3.78
CRMSUR-F	6.94	2.72	12.20	2.16	9.38	0.0002	0.74	16.50
Sample ID	Total U-238 Activity for Sample Fraction (dpm)		Total U-238 Activity for Sample (dpm)		(Bq)	Total Sample (Bq)	Uncertainty (%)	Cumul. Uncert (%)
CRMSLF1	46.3				0.771		12.29	12.73
CRMSLF2	18.4				0.307		11.43	11.91
CRMSLF3	12.6				0.210		14.57	14.95
CRMSLF4	52.7				0.879		8.37	9.01
CRMSLF5	113.3				1.89		6.50	7.30
CRMSLF6	816.4				13.61		4.96	5.98
CRMSLF-F	70907		71967		1182	1199	3.34	8.78
CRMSUR1	56.5				0.942		11.22	11.74
CRMSUR2	26.9				0.448		10.42	10.98
CRMSUR3	9.9				0.165		16.69	17.04
CRMSUR4	19.7				0.329		12.06	12.55
CRMSUR5	133.5				2.22		6.02	6.95
CRMSUR6	355.7				5.93		5.61	6.60
CRMSUR-F	72822		73424		1214	1224	3.47	9.35
*CRM= Certified Reference Material 129								

Figure 1 – Contaminated Soil

Figure 2 – SURVANTA® packaging and vial

Figure 3 – Autoclave

Figure 4 – Dissolution System

Figure 5 – Centrifuge in use

Figure 6 – Surfactant suspended in solution

Figure 7 – Surfactant attached to soil

Figure 8 – LSC system (closed and open)

Figure 9 – Fusion of undissolved soil; Post-fusion cake

Figure 10 – Anion Exchange Chromatography

Figure 11 – Sample Electrodeposition

Figure 12 – Alpha Spectrometry Chamber (open and closed)

Figure 13 – Uranium Alpha Spectra

Figure 14 – Sample SLFA

Figure 15 – Sample SLFB

Figure 16 – Sample SURA

Figure 17 – Sample SURB