

Solubility Characterization of Airborne Uranium from a Uranium Recycling Plant

Robert Metzger, Radiation Safety Engineering, Inc., 3245 North Washington Street,
Chandler, Arizona 85225

Leslie Cole, Mountain Health Services, Inc, 151 Cecil Gray Road, Jonesborough, TN 37659

Abstract

Solubility profiles of uranium dusts in a uranium recycling plant were determined by performing *in vitro* solubility tests on breathing zone air samples conducted in all process areas of the processing plant. The recycling plant produces high density shields, closed end tubes that are punch and formed from uranium sheet metal, and high-fired uranium oxide which is used as a catalyst. The recycled uranium is cut, melted in a vacuum furnace, and part of the molten uranium is poured into molds for further processing. Air samples were taken in process areas under normal working conditions. The dissolution rate of the uranium in a simulant solution of extracellular airway lining fluid (Gamble's solution) was then determined over the next 28 d. Airborne uranium in the oxide section of the plant was found to be highly insoluble with 99% of the uranium having a dissolution half time in excess of 100 d. The solubility of the airborne uranium in other areas of the facility was only slightly more soluble with over 90% of the airborne uranium having dissolution half-times in excess of 90 days.

INTRODUCTION

Scrap uranium metal received into the plant is first cut into small pieces and is fed

into a top loading vacuum furnace where it is melted and poured into forms for shielding used in industrial radiography and medical applications. Blanks are also poured that are subsequently processed through a rolling mill and formed into hollow tubes used in the recycling of spent reactor fuel.

At the completion of the melt, the residual uranium in the crucible, called the skull, contains a high concentration of the short-lived uranium progeny ^{234}Th and $^{234\text{m}}\text{Pa}$. These metals are of lower density and tend to float to the top of the melt as well as be trapped in the crucible coating. The skull is allowed to completely oxidize by burning the uranium with air for four days to form a high fired uranium oxide powder. The uranium oxides formed from the skull are vacuumed from the crucible and are sold as a catalyst material for the plastics industry. The actual form of the uranium generated by this process is likely a mixture of uranium oxides and has not been generally determined.

THE SOLUBILITY TEST

Air samples were taken in the Shear Station, Punch Area, Forming Station, and in the Oxide Room of the plant including the crucible cleaning operation. A medium volume air sampler operating at 2 cubic feet per minute (57 liters per minute) liters of air per minute was used to collect the samples. The samples were collected using a standard 37-mm breathing zone cassette and 0.8 micron pore size Gelman¹ GN4 filter media. Total uranium activity on the filters ranged from 460 Bq to 38 kBq . Each of the filters was

¹ Gelman Sciences, Inc., 600 S. Wagner Rd., Ann Arbor, MI 48106

covered with a clean filter of the same type to form a filter-uranium-filter sandwich, which was held together using the center sections of two breathing zone cassettes. These filters were immersed in a beaker holding 90 mL of a serum lung ultrafiltrate (SUF) (Eidson 1983), which was maintained at a pH of 7.2 by keeping a layer of CO₂ over the solution. The filter sandwich was moved to a fresh beaker of solution every day for one week, and then weekly until the end of the 28 day test period. The SUF was then wet ashed with concentrated nitric acid and hydrogen peroxide to destroy the organic components in the SUF. The pH of the solution was adjusted to 2 - 3 with 6 M NaOH and it was transferred to a teflon separatory funnel. One and one-half mL of the extractive scintillator Alphaex² was added to the funnel and the uranium was directly phase transferred to the organic scintillator. Extractive scintillators contain an organophilic metal ion extractant that allows the phase transfer of a nuclide(s) into the scintillator from an aqueous solution. Alphaex contains bis(2-ethylhexyl) hydrogen phosphate, which has a high affinity for actinides (McDowell 1994).

After the phases had been allowed to separate, 1.0 mL of the organic scintillator was withdrawn and counted on a Photon-Electron Rejecting Alpha Liquid Scintillation (PERALS³) spectrometer. The PERALS spectrometer uses pulse shape discrimination to isolate the longer pulses resulting from alpha interactions in the scintillator from those

²ETRAC, Inc., 1009 Alvin Weinberg Dr., Oak Ridge, TN 37830

³ORDELA, Inc., 1009 Alvin Weinberg Dr., Oak Ridge, TN 37830

resulting from β - γ emissions. A timing circuit and gate are then used to reject the β - γ pulses and the alpha pulses are sent to a multichannel analyzer for display of the alpha energy spectrum. The counter typically has a background of 0.03 counts per minute under an alpha peak, and an alpha counting efficiency of >99 percent. Energy resolution is about 220 keV at an alpha energy of 5 MeV. The lower limit of detection for a one hour count ranged from 0.002 to 0.003 Bq depending on the specific spectrometer used. This sensitivity makes it possible to develop solubility profiles from breathing zone air samples which contain trace quantities of radionuclides.

After the leach test period was complete, the filter sandwich containing undissolved uranium was placed in a beaker with concentrated nitric acid. The Gelman GN4 filter media readily dissolved in nitric acid. The beaker was placed on a hotplate and additions of nitric acid and hydrogen peroxide were made until the organics had all been driven off and a clear solution remained. After a pH adjustment, the uranium was extracted into the Alphaex extractive scintillator, which was counted on the PERALS spectrometer.

After the experiment was complete, the total activity and the fraction undissolved at each time period were determined and fitted to a three exponential model (Equation 1) using a standard Marquardt fitting algorithm (Kuester 1973). This fitted model was then used to find the dissolution half-times for the uranium on the air sample. This data then can be used with caution, in the ICRP 30 (ICRP 1979) based internal dosimetry codes, or

in the ICRP 66 (ICRP 1994 & 2003) and later codes. This test method has been previously validated against other *in vitro* solubility test methods (Metzger 1996) and *in vivo* bioassay data (Metzger 1997).

$$\% \text{ undissolved} = A_1 e^{-\lambda_1 t} + A_2 e^{-\lambda_2 t} + A_3 e^{-\lambda_3 t} \quad (\text{Equation 1})$$

RESULTS

Solubility profiles of the airborne uranium in the processing plant indicate that dusts are very insoluble. In the oxide section of the plant, only 1 percent of the uranium dissolved with a 0.6 day half-time, while the remaining 99 percent dissolved with a half-time of more than 100 days. Uranium dusts in the other areas of the plant were only slightly more soluble, with over 90% of the uranium having dissolution half times in excess of 90 days. Complete results of the study are shown in Table 1 and Figure 1 below. Particle size analysis performed under separate work indicated that the activity median aerodynamic diameter for the dusts was a relatively large 20+ microns.

References

Eidson, A.F.; Mewhinney, J.A. *In Vitro* Dissolution of Respirable Aerosols of Industrial Uranium and Plutonium Mixed-Oxide Nuclear Fuels. *Health Physics*. 45:1023-1037; 1983.

International Commission on Radiological Protection. Limits for Intakes of Radionuclides by Workers. Oxford: Pergamon Press; ICRP Publication 30, Parts 1,2 & 4; 1979.

International Commission on Radiological Protection. Human Respiratory Tract Model for Radiological Protection. Oxford Pergamon Press; ICRP Publication 66, 1994.

International Commission on Radiological Protection. Guide for the Practical Application of the ICRP Human Respiratory Tract Model. Oxford Pergamon Press; ICRP Supporting Guidance 3, 2003.

Kuester, J.L.; Mize, J.H. Optimization Techniques with Fortran. New York: McGraw-Hill; 1973.

McDowell, W.J.; McDowell, B.L. Liquid Scintillation Alpha Spectrometry. Boca Raton,

Florida: CRC Press Inc.; 1994.

Metzger, R.L., Jessop, B.H., & McDowell, B.L., Solubility testing of actinides on breathing zone and area air samples, NUREG/CR-6419, U.S. Nuclear Regulatory Commission, Washington, D.C., February, 1996.

Metzger, R.L., Wichers, D, Vasein, J., & Velasquez, Solubility characterization of airborne uranium from an in situ uranium processing plant, *Health Phys*, Volume 22, No. 3, March, 1997.

Table 1. Fitted parameters for Equation 1 in each process area. The table shows the fraction and dissolution half-time (days) for each component.

Process Area	Fraction	Half-Time	Fraction	Half-Time	Fraction	Half-Time
Oxide	0.01	0.63	0.14	108	0.85	>3000
Punch	0.05	0.8	0.12	90	0.83	166
Forming	0.02	0.19	0.05	2.9	0.93	236
Shear	0.02	0.71	0.04	15	0.94	>3000
Crucible	0.05	1	0.09	23	0.86	138

Figure 1. Solubility Profiles of Airborne Uranium in Serum Lung Ultrafiltrate from a Uranium Recycling Process

