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THE FERNALD ENVIRONMENTAL MANAGEMENT PROJECT

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SOLUBILITY OF AIRBORNE URANIUM COMPOUNDS AT THE FERNALD ENVIRONMENTAL MANAGEMENT PROJECT

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Abstract

The *in vitro* solubility of airborne uranium dusts collected at a former uranium processing facility now undergoing safe shutdown, decontamination and dismantling was evaluated by immersing air filters from high volume samplers in simulated lung fluid and measuring the ^{238}U in sequential dissolution fractions using specific radiochemical analysis for uranium. X rays and photons from the decay of uranium and thorium remaining on the filter after each dissolution period were also directly measured using a planar germanium detector as a means for rapidly evaluating the solubility of the uranium bearing dusts. Results of these analyses demonstrate that two distinct types of uranium bearing dusts were collected on the filters depending upon the location of the air samplers. The first material exhibited a dissolution half-time much less than one day and was most likely UO_3 . The dissolution rate of the second material, which was most likely U_3O_8 , exhibited two components. Approximately one-third of this material dissolved with a half time much less than one day. The remaining two-thirds of the material dissolved with half times between 230 ± 16 d and 1350 ± 202 d. The dissolution rates for uranium determined by radiochemical analysis and by gamma spectrometry were similar. However, gamma spectrometry analysis suggested a difference between the half times of ^{238}U and its daughter ^{234}Th which may have important implications for *in vivo* monitoring of uranium.

Introduction

The Fernald Environmental Management Project (Fernald), formerly a U.S. Department of Energy (DOE) facility for converting UO_3 and UF_6 to uranium metal, is currently undergoing safe shutdown including decontamination and dismantling (D&D) of the former production buildings. Safe shutdown describes the effort to make facilities more stable prior to potential reuse or dismantling and creates a safer environment for workers involved with D&D when demolition occurs. Where appropriate, workers are provided personal protective equipment, such as respirators, to minimize exposure to airborne uranium compounds resuspended by these activities. The requirements for personal protective equipment depend upon many factors including the concentration of airborne uranium at the work location and its solubility in the respiratory tract. Although uranium compounds at Fernald were characterized during plant operations (Fischhoff 1965), information was lacking on the characteristics of the resuspended, uranium-bearing compounds in the air during safe shutdown and D&D activities. Thus, a study was initiated to investigate the *in vitro* solubility of uranium in samples of resuspended dusts to provide additional technical support for the worker protection program at Fernald.

Prior to 1989, Fernald was a DOE uranium-metal fabrication facility producing uranium tetrafluoride (UF_4) through hydrofluorination of UO_3 or reduction of UF_6 (US DOE 1992). The UO_3 was blended with magnesium-metal granules, placed in a reduction pot and heated in a furnace to produce the intermediate uranium-metal product, called a "derby." These derbies were then remelted along with recovered uranium scrap from other stages of the process. The molten mixture was poured into graphite molds, forming ingots of varying sizes and shapes. The ingots were then machined to form billets, which were finally sent to other DOE sites. In this

process, various uranium compounds were produced as by-products including the oxides of uranium; UO_2 , UO_3 and U_3O_8 (Soldano 1998).

In lieu of site-specific information, the ICRP (1978) suggests that UO_3 is a class W compound, with a clearance half time between 10 and 100 days. Uranium compounds such as UO_2 and U_3O_8 are considered significantly less soluble (class Y), with half times in excess of 100 days. However, the solubility of uranium oxides is known to depend upon heat treatment. Soldano (1998) has indicated that uranium oxides encountered at Fernald were produced at low temperatures. Rich (1988) indicates that low-temperature uranium oxides may be considered as class W compounds. This discrepancy in the solubility classification reported for these uranium oxides suggested a need to investigate the solubility of the uranium compounds now present at Fernald.

Mercer (1967), Cooke and Holt (1974) and Chazel et al. (1998) describe results of *in vitro* dissolution studies for several different uranium compounds. Eidson et al. (1980, 1984) performed *in vitro* dissolution studies of the uranium compounds produced at several uranium mills. Eidson (1994) also conducted a comprehensive review of the *in vivo* and *in vitro* dissolution rates of uranium compounds from various industrial sources. Additionally, Metzger et al. (1996) conducted a solubility study of soluble airborne uranium compounds from a uranium processing plant.

This study evaluated the *in vitro* solubility of airborne uranium using a method in which air filters collected at Fernald were exposed to a large volume of simulated lung fluid (SLF) that was constantly being circulated and maintained at physiological pH and temperature. The large volume of SLF used in this study follows suggestions by Moss and Kanapilly (1980) and by Cusbert et al. (1994) to insure that the solution would not become saturated with uranium and

influence measurement of the dissolution rate. The filters were retained in a plastic filter assembly that could be easily removed from the system after each dissolution period for direct gamma spectrometric measurement of the uranium remaining on the filters.

Materials and Methods

Eight samples of resuspended airborne dusts, collected at four different locations in support of routine monitoring for airborne radioactive materials at Fernald, were analyzed in this study. The samples were collected using 47 mm fiberglass filters connected to a calibrated sampling pump operating at a flow rate of approximately 60 L min^{-1} . The sampling head was placed at approximately 1.5 m from the ground, facing the source of airborne radioactive materials. The total volume of air sampled through each filter was at least 10,000 L. Sample no. UC01 and the two filters consolidated in sample no. UC04 were collected from Plant 6A. Sample no. UC02 and sample no. UC03 were collected from Plant 5A. The three filters consolidated in sample no. UC05 were collected at Plant 8A and Plant 9A. Whenever more than one air filter was consolidated in a sample, the uranium bearing dusts collected on each filter were expected to be the same compounds of uranium based upon the process that was formerly conducted in the area.

Filters were cut to a diameter of approximately 38 mm using a circular die punch and mounted in a Gelman* 47 mm In-line Polycarbonate Filter Holder. This diameter was selected so that the SLF would bathe the filters rather than being forced to pass through the fiberglass filter media. The uranium-bearing surface of the filter was placed facing towards the supply of SLF. When two or more filters were combined in the assembly, they were stacked on top of each other in the same direction, with the uranium-bearing surface towards the SLF supply.

Fig. 1 illustrates the physical arrangement of the *in vitro* dissolution system. The procedure used a 750 ml volume of SLF, prepared according to the formula proposed by Moss (1979). The SLF reservoir was a 1 L plastic bottle having five holes drilled in the lid of the container. Four of the holes were fitted with two-sided hose adapters that were sealed airtight with silicone. The fifth hole was used for the pH probe. Two ports on the lid were used to supply and return SLF from the reservoir to the filter holder assembly. A short section of Tygon[†] Flexible Plastic type R-3603 Tubing was connected to the inner supply adapter and extended into the lower third of the bottle. The exterior port of this adapter was connected to a Cole-Parmer[‡] peristaltic pump using Tygon type R-1000 flexible plastic tubing. The SLF was circulated through the filter assembly at a rate of approximately 140 mL min⁻¹. This rate was determined by measuring the volume of fluid circulated in a known period of time. In this manner, the uranium-bearing filter was exposed to the entire volume of SLF which was maintained at physiological temperature (~37°C) by immersing the reservoir in a Fisher Scientific[§] model 2028S Isotemp water bath. Although the temperature of the water bath and the SLF reservoir was controlled to 37°C ± 0.01°C, the actual temperature of the filter assembly may have been slightly lower.

The two remaining hose adapters served as supply and exhaust ports for carbon dioxide gas, which was used to adjust and maintain the pH of the SLF between 7.2 and 7.4. Excess CO₂ gas was vented from the reservoir through a water bubbler to provide a visual indication of flow. The pH was continuously monitored using a Fisher Scientific Accumet Model 15 pH meter with a Fisher Scientific ACCU-pHAST pH probe. The tendency of the SLF pH was to become more basic. As the pH approached 7.4, CO₂ was added as a cover gas above the SLF in the reservoir.

The SLF would drip through the CO₂ as it was circulated back into the reservoir, reducing the pH of the SLF.

The first dissolution fraction was exposed to the uranium-bearing filter for a period of 1 hr. At the conclusion of the dissolution period, the pump was run in reverse for a fraction of a rotation to remove residual SLF from the filter holder assembly. The SLF reservoir was removed from the water bath, and the SLF was filtered by vacuum through a Gelman type GN-6 0.45 μ m membrane filter. Concentrated HNO₃ was added as a preservative to the filtered SLF and stored in a refrigerator. This procedure was repeated for each dissolution fraction.

Additional dissolution fractions were obtained at 2 hr, 4 hr, 6 hr, 24 hr, 2 d, 3 d, 7 d, 10 d, 30 d, 60 d and 100 d.

The quantity of uranium in each of the dissolution fractions was determined by specific radiochemical separation and alpha spectrometry. A ²³²U tracer was added to an aliquot of each SLF fraction that was then dried in the presence of concentrated HNO₃. The sample was wet ashed with the HNO₃ repeatedly to remove organic material and to insure the complete exchange between tracer and sample. The sample was then dissolved in 9 M HCl and the uranium separated by anion exchange chromatography (USTUR 1996). Samples were electrodeposited and counted for 75,000 seconds using a 450 mm² EG&G Ortec¹ ULTRA silicon detector. The measurement uncertainty for each sample was reported to one standard deviation.

Gamma spectrometry was used as a rapid method to characterize the solubility rate of the uranium compounds by measuring the amount of uranium activity remaining in the filter assembly after each dissolution period. Before beginning the dissolution tests, and at each dissolution fraction interval, the filter holder assembly was disconnected from the system and placed in a fixed position close to the surface of a Princeton Gamma-Tech[#] Model RG-11B/C

High Purity Planar Germanium Detector. The efficiency of the detector was determined using a standard filter prepared in the laboratory with a known amount of natural uranium solution and counted in the same geometry as the dissolution filter. The initial count of the filter mounted in the holder before the start of dissolution testing gave an estimate of the total activity initially present on the filter. Each subsequent count of the filter holder represented the activity remaining in the filter. The uncertainty reported for each gamma measurement represents counting statistics only.

Five separate energy regions of the gamma spectrum were adopted for evaluating ^{238}U and ^{234}Th . Gamma spectrometry measurements, using a personal computer-based multichannel analyzer, identified ^{238}U at 12.952 keV, 16.161 keV and 19.094 keV and ^{234}Th at 63.288 keV and 93 keV. Fig. 2 is a typical gamma spectrum obtained from measurement of a filter and shows all five peaks. Background was determined by measuring a filter holder assembly containing blank filters. The energy region associated with the 16.161 keV characteristic X-ray peak was selected to quantitate ^{238}U , while the region associated with the 63.288 keV gamma peak was selected to quantitate ^{234}Th .

After a dissolution period of 100 d, the uranium bearing materials remaining on the fiberglass filter were analyzed by dissolving the filter using the KF-pyrosulfate fusion method described by Sill et al. (1974) and the alpha spectrometry method described previously. Any residual uranium activity on the filter was added to the sum of activities measured in all previous dissolution fractions to obtain the quantity of uranium activity that was initially present on the filter. The fraction of the total activity in each dissolution fraction was then plotted against time to evaluate the dissolution rate. Finally, a least-square exponential curve fit was applied to the data to determine the dissolution half-times of the material components.

Results

The results of the radiochemical analysis for ^{238}U in each of the dissolution fractions are summarized in Table 1 and were used to calculate the dissolution rate shown in Table 2 for each of the test samples in this study. These results demonstrate that at least two distinct types of airborne uranium dusts were collected at Fernald. The filters from Plant 6A and used in UC01 and UC04, were bright yellow in color and were collected during transfer of a uranium trioxide product (UO_3). During the transfer, a hose in the vacuum line separated resulting in the atmospheric dispersion of UO_3 . This material was rapidly soluble (0.07 d and 0.05 d half times, respectively) and is representative of solubility class D.

The other type of material was collected at Plants 5A, 8A and 9A and was dark green or black in color, likely a U_3O_8 compound. Samples UC02 and UC03 were collected during the removal of ductwork in Plant 5A. During air sampling, contaminated ductwork was being reduced in size using electric saws. When Plant 5A was originally in operation, UF_4 was heated to high temperatures in the presence of magnesium fluoride to produce uranium metal. Machining of the uranium metal also occurred in this building. Thus, the likely forms of uranium contamination on the ductwork are oxides of uranium.

Sample UC05 is a composite of air filters from Plants 8A and 9A. During production, Plant 9A was used to make special uranium products through casting and machining of uranium ingots. Plant 8A was the former scrap recovery plant. Particles generated by safe shutdown and D&D activities in these areas are likely to be oxides of uranium.

The uranium dissolution rate of the material in samples UC02, UC03, and UC05 appears to have two components; approximately one-third of the material exhibited a half time between

approximately 0.01 d and 0.04 d, while the remaining two-thirds exhibited a half time between approximately 230 d and 1350 d. This material appears to exhibit characteristics of both classes D and Y solubility. The dissolution curves for each of the tested materials is shown in Fig. 3.

Table 3 provides the results of the dissolution rate analysis determined from gamma spectrometric measurements. Using the 16.161 keV X-ray peak, two distinct types of airborne uranium dusts were identified based upon their solubility characteristics. The single component, rapidly soluble material was again identified in UC01 and UC04 exhibiting a dissolution half-time of approximately 0.03 d. Likewise, the material exhibiting a two component dissolution pattern was identified in UC02, UC03 and UC05. Gamma analysis showed that approximately one-third of the material dissolved with a half time less than 2 d. The remaining two-thirds of the material on the filters dissolved with half times between approximately 35 d and 1250 d.

Gamma spectrometry also enabled the dissolution of thorium to be evaluated by measuring the change in the 63.288 keV ^{234}Th photopeak. For all samples, the activity of ^{234}Th remained relatively constant for approximately two days before a decrease was observed. An exponential curve was fit to the data to determine the clearance half time of the thorium material. Table 4 provides the results of the analysis which indicates that the half times range from approximately 3 d to 180 d. The thorium material on filter UC05 exhibited a half time of approximately 130 d, which agrees with the metabolic data for thorium reported in ICRP report no. 30 (ICRP 1978). The ICRP classifies oxides and hydroxides of thorium as class Y.

Discussion

The radiochemical analysis of uranium in each dissolution fraction provides a sensitive and reliable method for characterizing the *in vitro* solubility of the airborne uranium dusts. The rapidly soluble uranium material collected on the filters used in UC01 and UC04 is most likely UO_3 . The dissolution half time of this material was much less than one day, suggesting that it should be considered a class D compound. The material collected on those filters was reported to be in a pure, powdery form. The presumed small particle size of this aerosol may have enhanced the solubility of this material (Chazel et al. 1998). The second type of uranium material exhibited two unique solubility components and is most likely U_3O_8 . In each case, approximately one-third of this material exhibited a dissolution half-time much less than one day, while the dissolution half-time of the remaining component was much greater than 100 d. This suggests that the airborne U_3O_8 material collected at these Fernald locations may be better represented as partially a class D compound and partially a class Y compound for radiological protection purposes. The International Commission on Radiological Protection (ICRP) has published generic recommendations on the solubility of UO_3 and U_3O_8 which differ from the *in vitro* solubility results measured in this research. Since it has been demonstrated that particle size has a significant effect on solubility (Chazel et al 1998), further research into the effect of particle size on the dissolution of these uranium compounds needs to be performed.

The results of this study suggest that the characteristics of resuspended particles associated with safe shutdown and D&D activities may differ from the characteristics of particles generated during routine production operations. As a result, routine monitoring programs that were developed for conditions associated with production activities may require modification to accommodate changes in the characteristics of resuspended particles generated during clean-up operations.

Gamma spectrometry, in addition to the conventional radiochemical methods, was investigated as a rapid method to evaluate the solubility of the airborne uranium dusts. Although ^{238}U was detected by the presence of 16.161 keV Th L_β X-rays in the gamma spectrum, background variations in this region are large and lead to considerable uncertainty in the measurement results. In addition, these low energy photons undergo considerable attenuation in the filter assembly and are greatly influenced by variation in the position of the filter assembly relative to the detector. Recognizing these sources of measurement uncertainty, it was still of interest to use the planar germanium detector to measure ^{238}U and ^{234}Th remaining on the filter as a rapid, low cost method for evaluating the dissolution rate of the uranium material on the filter. Since the ^{238}U measurement results appear to confirm the results obtained using radiochemical methods, gamma spectrometry has been demonstrated to be useful in investigating material solubility and can help reduce the time, effort and cost necessary in performing the analyses.

Results obtained from the radiochemical analyses and from the direct measurement of ^{238}U were very similar. Both methods clearly identified the two different types of uranium compounds and the two component solubility of the presumed U_3O_8 compound. The rapidly soluble UO_3 in samples UC01 and UC04 exhibited an average dissolution half time of 0.06 d determined by radiochemistry and 0.03 d by gamma spectrometry. The longer component of the U_3O_8 compound in samples UC02, UC03 and UC05 varied greatly between trials, but the gamma spectrometry results showed a clear relationship to the alpha results within trials. The results obtained from the analysis of UC05 exhibited excellent agreement with the radiochemical and gamma spectrometric results, likely due to the improved performance of the germanium detector.

Fig. 4 illustrates the results of the dissolution rate analysis determined from radiochemistry and from direct gamma spectrometry for UC05. Using radiochemical methods, 31% of the material

dissolved rapidly with a half time of 0.036 d, while 68% dissolved much more slowly with a half time of 776 d. Using gamma spectrometry, 36% of the uranium material on the filters used in UC05 had a half time of 0.058 d, while the remaining 64% exhibited a half time of 1246 d. Thus, gamma spectrometry was successful in rapidly characterizing the solubility rate of the uranium compounds on the filters.

Because the uranium dissolution rates for sample UC05 obtained by radiochemical analysis were similar to those obtained by gamma spectrometric analysis, gamma spectrometry was also used to investigate the dissolution rate of ^{234}Th by measuring the 63.288 keV peak. The trend among samples UC01 through UC04 showed the thorium dissolution half time to be over three times greater than that of uranium. However, UC05 exhibited a uranium dissolution half time almost ten times greater than thorium. Fig. 5 compares the dissolution rates of the uranium and thorium compounds found on the filters used in UC05. Before approximately 2000 hr, the relative activity of ^{234}Th remaining in the solution exceeds that of the ^{238}U remaining in the solution. However, after this point, the relative activity of ^{234}Th falls below that of ^{238}U .

This observation is important for *in vivo* measurement of uranium, where photons from the decay of ^{234}Th are used to quantitate the presence of ^{238}U . Therefore, additional study is necessary to determine what impact, if any, the presence of high concentrations of phosphates in the SLF may have on the dissolution rate of thorium. However, based on the results obtained in this investigation, a whole body counter using the 63.288 keV or 93 keV ^{234}Th energy region to detect uranium *in vivo* may overestimate a uranium deposition if measured prior to 2000 hr (83 d) post-intake but would underestimate the deposition if measured after this time. Although this data is not conclusive, the trends show a clear divergence between the half times of ^{238}U and ^{234}Th for the airborne dust samples collected at the Fernald.

The measurement uncertainty due to the filter holder assembly geometry was determined to be 9.38% of the total counts in the 16.161 keV region, and 8.87% of the total counts in the 63.288 keV region. This large error is in addition to the error due to counting statistics, 0.85% and 1.20% respectively for the two energy regions. Additionally, on at least one occasion, not all of the SLF could be removed from the filter holder assembly, which apparently reduced the count rate due to increased attenuation from the SLF in the holder. This additional error, along with the error introduced by the filter geometry makes the gamma results less conclusive.

Conclusions

This research investigated the *in vitro* solubility of resuspended airborne uranium compounds arising from safe shutdown and decontamination and dismantling activities at a former uranium processing plant. Because the facility is no longer in operation, the composition of the airborne uranium materials may be difficult to predict. Results of this research suggests that site-specific analysis of the solubility of the resuspended, uranium-bearing dusts are valuable in determining whether generic classifications for particle solubility need to be revised for facilities undergoing remediation. The results of this research also demonstrate the usefulness of gamma spectrometry as a direct method to characterize the solubility rate of the uranium compounds if there is sufficient activity in the sample. Conventional radiochemical analysis can be expensive and time consuming. Gamma spectrometry has also suggested that there may be a difference in the solubility of ^{238}U and ^{234}Th that could have considerable importance in evaluating *in vivo* measurement results for uranium.

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References

- Chazel, V.; Houpert, P.; Ansoborlo, E. Effect of U_3O_8 specific surface area on in vitro dissolution, biokinetics, and dose coefficients. *Rad. Protect. Dos.* 79(1-4): 39-42; (1998).
- Cooke, N.; Holt, F. B. The solubility of some uranium compounds in simulated lung fluid. *Health Phys.* 27: 69-77; 1974.
- Cusbert, P.J.; Carter, M.W.; Woods, D.A. *In vitro* dissolution of uranium. *Radiation Protection Dosimetry* 55:39-47; 1994.
- Eidson, A.F. The effect of solubility on inhaled uranium compound clearance: a review. *Health Phys.* 67:1-14; 1994.
- Eidson, A.F.; Griffith, W.C., Jr. Techniques for yellowcake dissolution studies *in vitro* and their use in bioassay interpretation. *Health Phys.* 46:151-163; 1984.
- Eidson, A.F.; Mewhinney, J.A. *In vitro* solubility of yellowcake samples from four uranium mills and the implications for bioassay interpretation. *Health Phys.* 39:893-902; 1980.
- Fischhoff, R. L. The relationship between and importance of the dimensions of uranium particles dispersed in air and the excretion of uranium in urine. *Am. Ind. Hygiene Assn. J.* 26:26-33; 1965.
- Hall, C. Monitoring for airborne radioactive materials. RC-DPT-009; 1997.
- Hodge, V.F.; Gurney, M.E. Semi-quantitative determination of uranium, plutonium, and americium in sea water. *Analytical Chemistry* 47:1866-1868; 1975.
- International Commission on Radiological Protection. Limits for intakes of radionuclides by workers. Oxford: Pergamon Press; ICRP Publication 30, Part 1; 1978.

Kalkwarf, D.R. Dissolution rates of uranium compounds in simulated lung fluid. *The Science of the Total Environment* 28:405-414; 1983.

McKane, L.; Kandel, J. *Microbiology: essentials and applications*. Second edition. New York: McGraw-Hill, Inc.; 1996.

Mercer, T. T. On the role of particle size in the dissolution of lung burdens. *Health Phys.* 13: 1211-1221; 1967.

Metzger, R.L.; Wicher, D.; Vaselin, J.; Velasquez, P. Solubility characterization of airborne uranium from an *in situ* uranium processing plant. *Health Phys.* 72:418-422; 1997.

Moss, O.R. Simulants of lung interstitial fluid. *Health Phys.* 36:447-448; 1979.

Moss, O.R.; Kanapilly, G.M. *Generation of aerosols*. Ann Arbor, MI: Ann Arbor Science Publishers, Inc.; 1980.

Rich, B.L.; Hinnefeld, S.L.; Lagerquist, C.R.; Mansfield, W.G.; Munson, L.H.; Wagner, E.R.; Vallario, E.J. *Health physics manual of good practices for uranium facilities*. EGG-2530 UC-41; 1988.

Sill, C.W.; Purphal, K.W.; Hindman, F.D. Simultaneous determination of alpha-emitting nuclides of radium through californium in soil. *Analytical Chemistry* 46(12):1725-1737; 1974.

Soldano, M. Technical basis for internal dosimetry at the Fernald Environmental Management Project. SD-2008; 1998.

U.S. Department of Energy. *Occupational Radiation Protection*. 10 CFR 835. Washington, DC: U.S. Government Printing Office; 1993.

U.S. Department of Energy. *Fernald Site Environmental Report*. Washington, DC: U.S. Government Printing Office; Contract DE-AC05-92OR21972; 1992.

U.S. Department of Energy. Dissolution and particle size characterization of radioactive contaminants in Hanford facilities: criteria for methods of measurement. Washington, DC: U.S. Government Printing Office; Contract DE-AC06-76RLO 1830; 1990.

U.S. Nuclear Regulatory Commission. Solubility testing of actinides on breathing-zone and area air samples. Washington, DC: U.S. Government Printing Office; Technical report CR-6419: 1996.

U.S. Transuranium and Uranium Registries. Anion exchange isolation of uranium from prepared tissue solutions, revision 1. Radiochemical analysis procedures manual. Pullman, WA: Washington State University; 1996.

Figures

Fig. 1. Schematic drawing representing the dissolution system used in this experiment. The flow rate of the SLF was 140 mL min^{-1} .

Fig. 2. A one-hour gamma spectrum of the uranium-laden filter analyzed in UC05, measured prior to the start of the dissolution study. The five peaks corresponding to the regions of interest studied are each labeled.

Fig. 3. Comparison of the dissolution curves among the five experimental trials applying the results obtained using radiochemical separation and alpha spectrometry. Error was determined as one standard deviation of each measurement. The equations for each exponential curve fit through the data points are as follows:

$$\text{UC01, } y = 0.19731e^{-0.014584t} + 0.79328e^{-0.40429t}; \text{ UC02, } y = 0.72126e^{-1.2579 \times 10^{-4}t} + 0.27873e^{-2.0648t};$$

$$\text{UC03, } y = 0.61163e^{-2.1268 \times 10^{-5}t} + 0.38836e^{-1.9959t}; \text{ UC04, } y = 0.066508e^{-0.010355t} + 0.92816e^{-0.59439t};$$

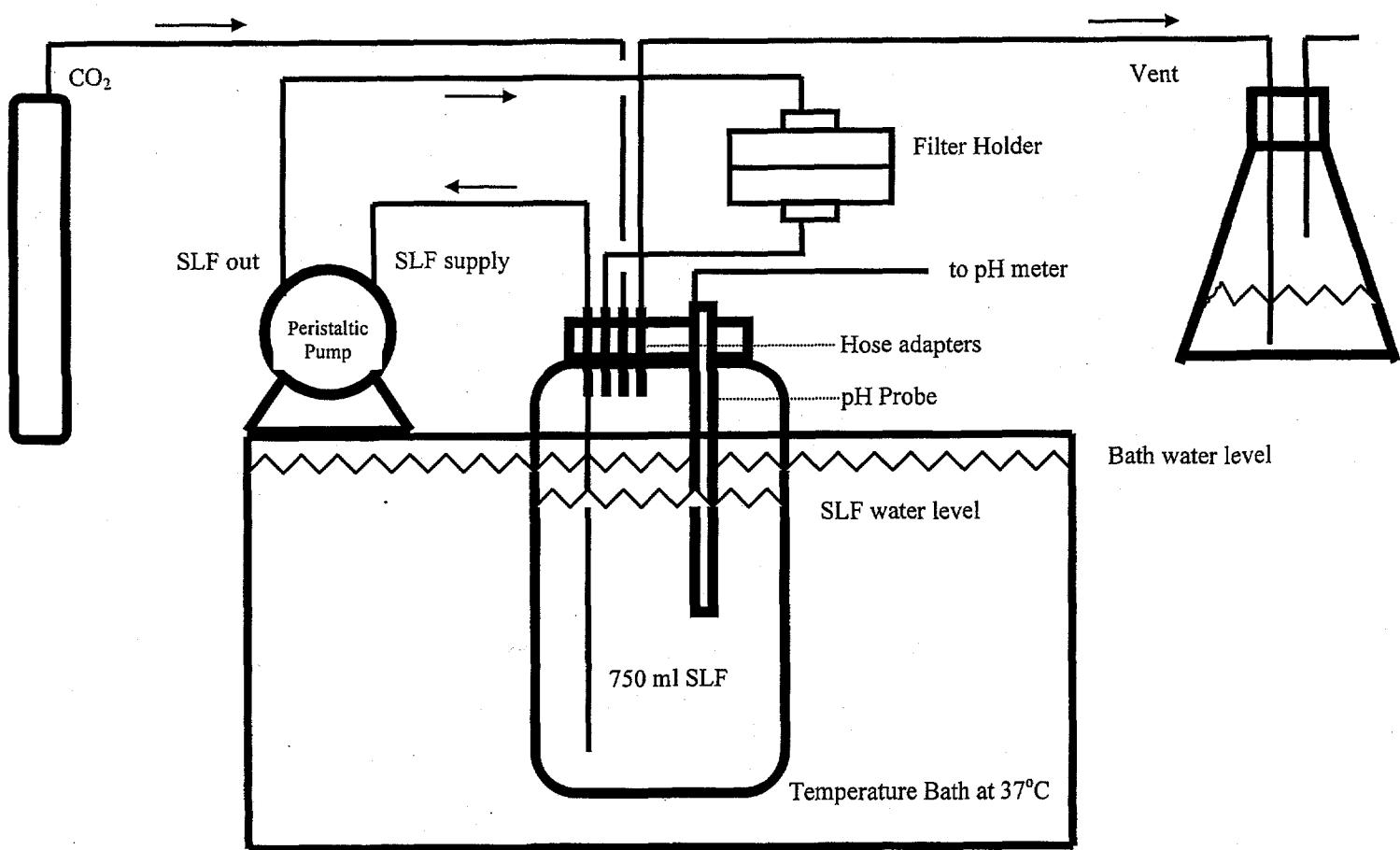
$$\text{UC05, } y = 0.68351e^{-3.7196 \times 10^{-5}t} + 0.30847e^{-0.81014t}$$

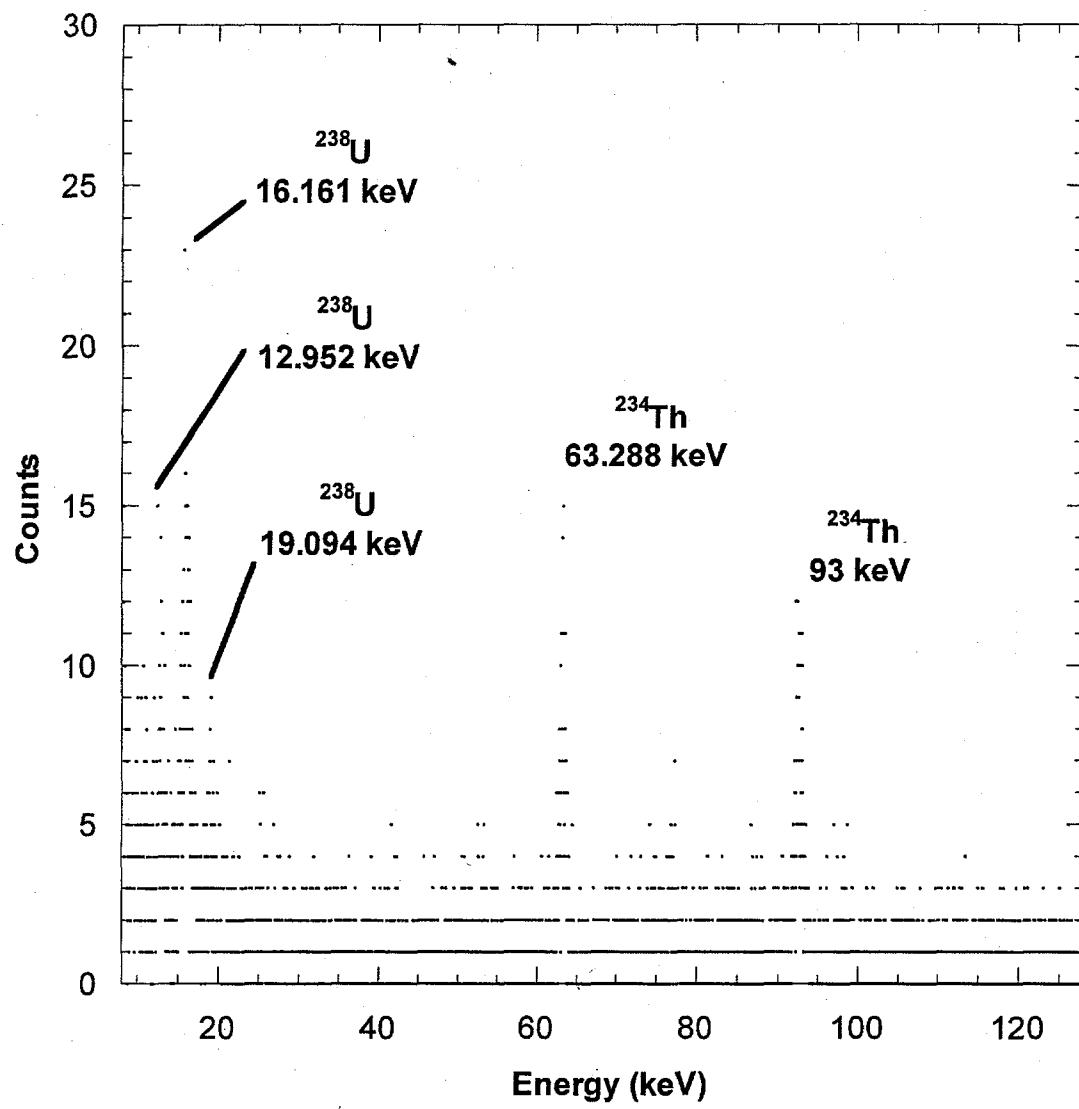
Fig. 4. Comparison of the dissolution curves for UC05 obtained using radiochemical separation and alpha spectrometry and using gamma spectrometry. The similarity between the curves is markedly apparent. For the gamma spectrometry results, error was determined using counting statistics, but the error due to detector geometry was excluded. The equation defining the alpha results is $y = 0.68351e^{-3.7196 \times 10^{-5}t} + 0.30847e^{-0.81014t}$. The equation defining the gamma results is

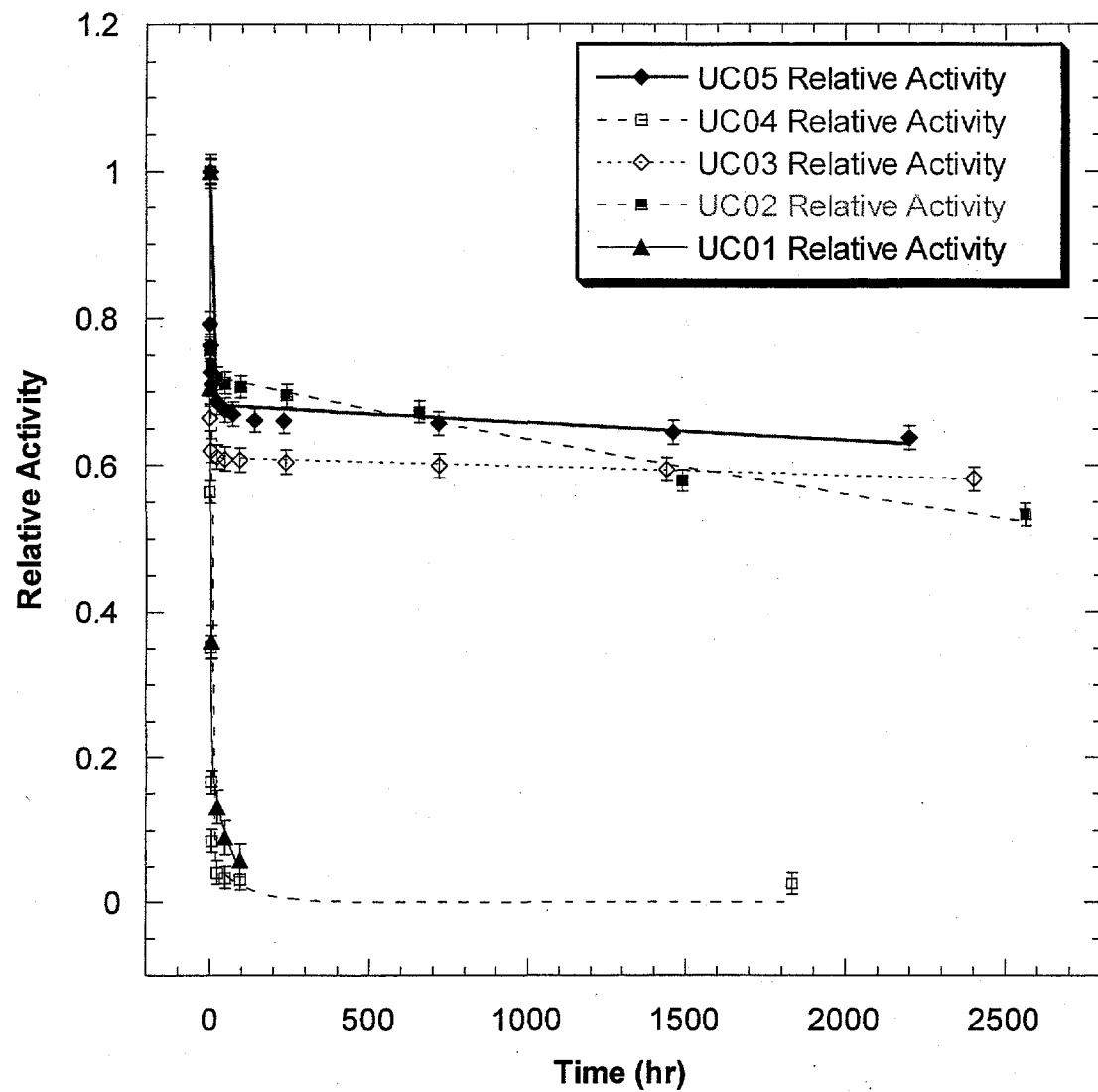
$$y = 0.63529e^{-2.3186 \times 10^{-5}t} + 0.3584e^{-0.49946t}.$$

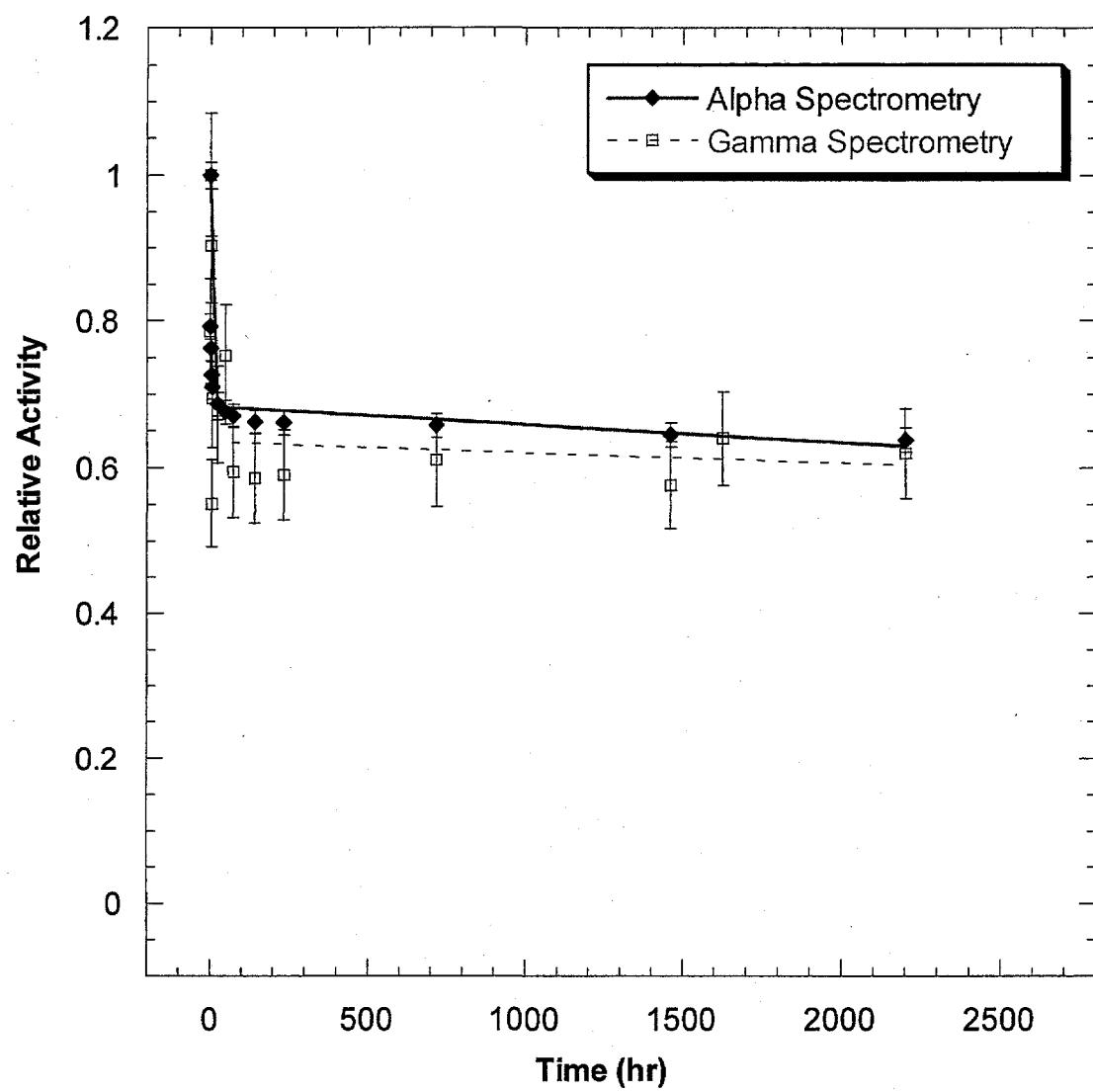
Fig. 5. Comparison between the dissolution curves for uranium using the 16.161 keV X-ray peak characteristic of ^{238}U and for thorium using the 63.288 keV gamma peak characteristic of ^{234}Th . The two-part exponential curve fit through the uranium data has the equation

$y = 0.63529e^{-2.3186*10^{-5}t} + 0.3584e^{-0.49946t}$ while the one-part exponential curve fit through the thorium data has the equation $y = 0.94541e^{-2.2374*10^{-4}t}$.









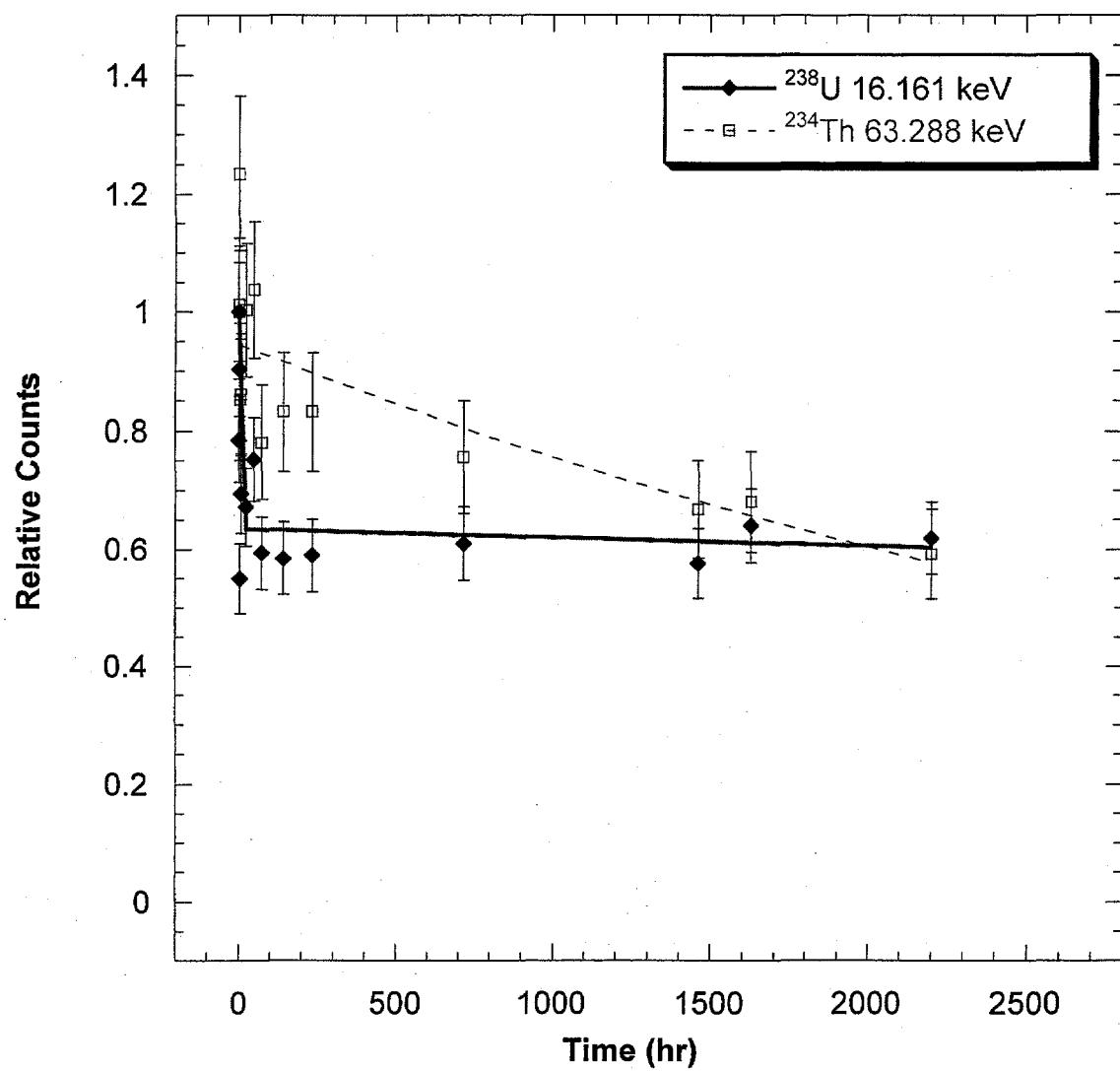


Table 1. Radiochemical results from the USTUR method performed on the filters in this experiment. The *Filter* dissolution fraction was performed last and represents the activity remaining on the fiberglass filter after the last dissolution period.

Dissolution Fraction	UC01 ^{238}U Activity (Bq)	UC02 ^{238}U Activity (Bq)	UC03 ^{238}U Activity (Bq)	UC04 ^{238}U Activity (Bq)	UC05 ^{238}U Activity (Bq)
Filter	$1.687 \times 10^{01} \pm 3.701 \times 10^{-01}$	$8.290 \times 10^{01} \pm 1.490 \times 10^{00}$	$3.084 \times 10^{01} \pm 5.449 \times 10^{-01}$	$3.247 \times 10^{00} \pm 6.633 \times 10^{-02}$	$2.129 \times 10^{01} \pm 3.904 \times 10^{-01}$
1	$8.382 \times 10^{01} \pm 2.560 \times 10^{00}$	$3.788 \times 10^{01} \pm 8.958 \times 10^{-01}$	$1.779 \times 10^{01} \pm 3.812 \times 10^{-01}$	$5.279 \times 10^{01} \pm 1.031 \times 10^{00}$	$6.951 \times 10^{00} \pm 1.616 \times 10^{-01}$
2	$9.824 \times 10^{01} \pm 2.963 \times 10^{00}$	$3.077 \times 10^{00} \pm 8.163 \times 10^{-02}$	$2.325 \times 10^{00} \pm 8.113 \times 10^{-02}$	$2.561 \times 10^{01} \pm 6.344 \times 10^{-01}$	$1.002 \times 10^{00} \pm 2.224 \times 10^{-02}$
3	$6.448 \times 10^{01} \pm 2.105 \times 10^{00}$	$2.888 \times 10^{00} \pm 6.510 \times 10^{-02}$	$4.497 \times 10^{-01} \pm 2.112 \times 10^{-02}$	$2.247 \times 10^{01} \pm 4.523 \times 10^{-01}$	$1.175 \times 10^{00} \pm 2.489 \times 10^{-02}$
4	$1.200 \times 10^{01} \pm 9.267 \times 10^{-01}$	$9.276 \times 10^{-01} \pm 2.669 \times 10^{-02}$	$1.559 \times 10^{-01} \pm 7.428 \times 10^{-03}$	$9.736 \times 10^{00} \pm 2.564 \times 10^{-01}$	$5.516 \times 10^{-01} \pm 1.482 \times 10^{-02}$
5	$8.800 \times 10^{00} \pm 3.856 \times 10^{-01}$	$8.266 \times 10^{-01} \pm 2.444 \times 10^{-02}$	$7.432 \times 10^{-02} \pm 6.859 \times 10^{-03}$	$5.255 \times 10^{00} \pm 1.474 \times 10^{-01}$	$7.919 \times 10^{-01} \pm 4.454 \times 10^{-02}$
6		$1.757 \times 10^{00} \pm 4.643 \times 10^{-02}$	$1.531 \times 10^{-01} \pm 9.873 \times 10^{-03}$	$8.975 \times 10^{-01} \pm 5.352 \times 10^{-02}$	$3.602 \times 10^{-01} \pm 1.211 \times 10^{-02}$
7		$3.428 \times 10^{00} \pm 8.642 \times 10^{-02}$	$2.729 \times 10^{-01} \pm 1.236 \times 10^{-02}$	$1.977 \times 10^{-01} \pm 1.034 \times 10^{-02}$	$1.747 \times 10^{-01} \pm 9.205 \times 10^{-03}$
8		$1.469 \times 10^{01} \pm 2.948 \times 10^{-01}$	$2.931 \times 10^{-01} \pm 1.475 \times 10^{-02}$	$7.694 \times 10^{-01} \pm 4.271 \times 10^{-02}$	$2.854 \times 10^{-01} \pm 1.326 \times 10^{-02}$
9		$7.095 \times 10^{00} \pm 1.524 \times 10^{-01}$	$6.390 \times 10^{-01} \pm 3.326 \times 10^{-02}$		$3.586 \times 10^{-02} \pm 2.219 \times 10^{-03}$
10					$9.392 \times 10^{-02} \pm 4.398 \times 10^{-03}$
11					$4.328 \times 10^{-01} \pm 1.475 \times 10^{-02}$
12					$2.395 \times 10^{-01} \pm 1.108 \times 10^{-02}$

Table 2. Summary of the results obtained from analysis of the alpha spectrometry data.

Filter Series	Exponential Curve-fit	R-value of Curve-fit	Material Fraction	Half-time (d)	Solubility Class
UC01	Two-part	0.999	79.3 \pm 4.7%	0.071 \pm 0.009	D
			19.7 \pm 4.6%	2.0 \pm 0.8	D
UC02	Two-part	0.997	27.9 \pm 1.3%	0.014 \pm 0.003	D
			72.1 \pm 0.5%	230 \pm 16	Y
UC03	Two-part	0.999	38.8 \pm 0.5%	0.015 \pm .0006	D
			61.2 \pm 0.2%	1358 \pm 202	Y
UC04	Two-part	0.999	92.8 \pm 2.3%	0.049 \pm .003	D
			6.7 \pm 1.9%	2.79 \pm 2.1	D
UC05	Two-part	0.984	30.8 \pm 2.1%	0.036 \pm .006	D
			68.4 \pm 0.8%	776 \pm 290	Y

Table 3. Summary of the uranium results obtained from analysis of the gamma spectrometry data. The 16.161 keV Th L_β X-ray peak was used in the analysis to identify ²³⁸U.

Filter Series	Exponential Curve-fit	R-value of Curve-fit	Material Fraction	Uranium Half-time (d)	Solubility Class
UC01	Two-part	0.998	51.1%	0.032	D
			48.7%	3.73	D
UC02	Two-part	0.833	56.9%	2.34	D
			37.2%	149.8	Y
UC03	Two-part	0.991	19.0%	1.76x10 ⁻⁴	D
			81.0%	34.6	W
UC04	Two-part	0.993	60.9%	0.032	D
			38.4%	1.11	D
UC05	Two-part	0.845	35.8%	0.058	D
			63.5%	1246	Y

Table 4. Summary of the thorium results obtained from analysis of the gamma spectrometry data. The 63.288 keV gamma peak from ^{234}Th was used to identify thorium.

Filter Series	Exponential Curve-fit	R-value of Curve-fit	Thorium Half-time (d)	Solubility Class
UC01	One-part	0.622	23.3	W
UC02	One-part	0.294	185.7	Y
UC03	One-part	0.939	93.4	W
UC04	One-part	0.787	3.79	D
UC05	One-part	0.775	129.1	Y

* Gelman 47 mm In-line Polycarbonate Filter Holder, Gelman Sciences, 600 South Wagner Road, Ann Arbor, MI 48106.

[†] Tygon Flexible Plastic Tubing R-1000 and R-3603, Norton Performance Plastic Corporation, P.O. Box 3660, Akron, OH 44309.

[‡] Cole-Parmer Masterflex Model 7553-70, Cole-Parmer Instrument Company, 7425 N. Oak Park Avenue, Niles, IL 60714.

[§] Fisher Scientific Isotemp 2028S, Fisher Scientific Inc., 711 Forbes Avenue, Pittsburgh, PA 15219.

[¶] EG&G Ortec ULTRA Detector, EG&G Ortec, 100 Midland Road, Oak Ridge, TN 37831.

[#] Princeton Gamma-Tech High Purity Germanium Detector Model RG-11B/C, Princeton Gamma-Tech, P.O. Box 641, Princeton, NJ 08540.