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Soil Sample Preparation Using Microwave Digestion For Uranium Analysis

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Abstract

A new sample preparation procedure has been developed for digestion of soil samples for uranium analysis. The technique employs a microwave oven digestion system to digest the sample and to prepare it for separation chemistry and analysis. The method significantly reduces the volume of acids used, eliminates a large fraction of acid vapor emissions, and speeds up the analysis time. The samples are analyzed by four separate techniques: Gamma Spectrometry, Alpha Spectroscopy using the open digestion method, Kinetic Phosphorescence Analysis (KPA) using open digestion, and KPA by Microwave digestion technique. The results for various analytical methods are compared and used to confirm the validity of the new procedure. The details of the preparation technique along with its benefits are discussed.

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Analytical Methods and Results

As part of an initiative to reduce the use of hazardous liquids and emission of acid vapors, a study was conducted to test the use of Microwave oven for digesting soil samples. The traditional sample preparation techniques use large amounts of acids for open digestion in beakers. The use of a Microwave digestion vessel reduces the acid

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usage significantly and practically eliminates any emissions during the sample preparation phase of the analysis.

A soil sample (about 1 kg) was taken from an area containing background levels of natural Uranium. The soil was prepared by drying, high temperature dry-ashing and sieving to remove moisture, organics and large pieces to get a homogeneous sample. An aliquot of 771 grams was placed in a 500 mL Marinelli beaker and analyzed by gamma spectrometry. The analysis provided an activity of 0.68 ± 0.14 pCi/g at the 95% confidence level for U-238. Three sets of samples each containing five 20-gram fractions of the soil sample were prepared and were analyzed by three different analytical methods.

The first analytical method was performed by Alpha Spectroscopy. Five separate soil samples (1.0 gram aliquots) were digested using concentrated nitric acid (HNO_3) and hydrofluoric acid (HF) in Teflon beakers. After neutralization of HF by boric acid (H_3BO_3), samples were dissolved in 8N HNO_3 and Sodium Nitrite ($NaNO_2$). Using preconditioned Bio-Rad anion resin, samples were introduced to the column and Uranium was separated and collected. After samples were dried over a hot plate, they were dissolved in 9N HCl, and a second column containing preconditioned Bio-Rad anion resin was applied to elute Uranium, using 0.1N HCl. Finally, Uranium was isolated by co-precipitation with neodymium fluoride and mounted on polypropylene filter over carbon substrate. The samples were counted using an alpha spectroscopy instrument. The results are presented in Table 1 where the uncertainty for the average is the standard deviation of the individual points.

Sample Fraction	U-238 (pCi/g)	U-235 (pCi/g)	U-234 (pCi/g)
1	0.57	0.025	0.62
2	0.60	0.039	0.58
3	0.70	0.044	0.68
4	0.71	0.041	0.74
5	0.58	0.044	0.53
Average	0.63	0.039	0.63
Uncertainty (2-sigma)	0.14	0.016	0.16
Total U (μg/g)	1.84 ± 0.41		

Table 1 - Alpha Spectroscopy Results

For the second preparation method, an established open digestion method was used. Five soil aliquots of 1 gram each were prepared by transferring to Teflon beakers, and wet ashed twice using 50mL of HNO_3 and 150mL of (HF). The samples were digested twice more with 200 mL of HNO_3. The samples were volumed up to 100 mL with 0.1M HNO_3, 1mL of the solution transferred to a glass cuvette with 1.5 mL of the Uraplex complexant, and analyzed by KPA. The results are presented in table 2 where the uncertainty for the average is the total propagated uncertainty.

Sample Fraction	Total U (µg/g)	Uncertainty (2-sigma)
1	1.22	0.18
2	1.37	0.12
3	1.43	0.13
4	1.35	0.12
5	1.31	0.15
Average	1.34	0.21

Table 2 - KPA Results using open digestion

For the final test, five soil aliquots of 0.1 grams were prepared by placing the soil in a microwave digestion vessel with 5 mL HNO_3, 5 mL H_2O, and 4 mL of HF. The vessels were sealed, placed in the microwave oven, and digested for 15 minutes at 150 pSI and 175 °C. The final solution was clear and colorless, indicating a good dissolution. The samples were wet ashed twice using 20 mL of HNO_3, volumed up to 10 mL using 0.1 M HNO_3, transferred 1 mL to a glass cuvette with 1.5 mL of the Uraplex complexant, and analyzed by KPA. The results are presented in table 3 where the uncertainty for the average is the total propagated uncertainty.

Sample Fraction	Total U (μg/g)	Uncertainty (2-sigma)
1	1.54	0.12
2	1.33	0.11
3	1.34	0.14
4	1.33	0.12
5	1.29	0.12
Average	1.37	0.29

Table 3 - KPA Results using microwave digestion

In addition, five sets of samples were analyzed by the ICP-MS using the EPA 3050B method. However, this method does not use HF acid during the digestion process and the effect is that the sample is leached instead of being totally dissolved. As a consequence, the results from the ICP-MS analysis was lower than the KPA results by a factor of two. The results from this set of samples were not used for this study. The final results for total Uranium in the soil sample are summarized in table 4.

Analytical Method	Final Result	Uncertainty (2-sigma)
Gamma Spectrometry	1.99	0.41
Alpha Spectrometry - Open Digestion	1.84	0.41
KPA - Open Digestion	1.34	0.21
KPA - Microwave Digestion	1.37	0.29

Table 4 - Summary of results by different methods

Conclusions

The results show that the new sample preparation method for KPA produces values that are statistically similar to the established open sample digestion. The Gamma Spectrometry and Alpha Spectrometry results are consistent with each other and higher than the KPA results by about 38%. This bias is likely due a number of factors such as that for U-238 the KPA detection limit is better than the Alpha Spectroscopy, Iron interfering with the KPA complexant, and possible dissolution problems. Any dissolution problems with alpha spectroscopy would have been corrected by the addition of the U-232 tracer during the sample preparation. The exact cause of the bias will be determined in a follow-up study.

The sample preparation by microwave significantly reduced the usage of acids and other reagents, hazardous emissions, and the waste generated during sample preparation. In addition, the sample preparation time is reduced from an average of 3 days to a few hours. The savings in labor, material, and regulatory compliance strongly favor the microwave digestion as the preferred method for sample preparation.

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