Application of Combined Alpha Spectrometry and Neutron Activation Analysis for the Isotopic Determination of Thorium in Aged Thorium Dioxide

S. P. Lamont, A. Maddison, J. T. Elliston, S. E. Glover

(Washington State University) invited

Thorium is a naturally occurring element that has a number of applications, including industrial (lantern mantles, welding operations, and preparation of refractories) and medical (thorotrast imaging agent), and has been proposed as a source of fissile nuclear energy via the production of ^{233}U from ^{232}Th.

The solubility of thorium compounds and associated progeny in the human lung is very important for determination of dosimetric consequences. Current models used for worker protection in the United States (ICRP 30) place stringent limitations on the inhalation of ThO_2. These limits are currently below those listed for ^{239}Pu, often referred to as "the deadliest substance on earth." Many assumptions are made regarding the fate of progeny that may dramatically affect the dosimetry of thorium incorporated via inhalation. This work discusses some of the results from the evaluation of the isotopic solubility of thorium and associated progeny from aged and recently fired thorium dioxide in simulated lung fluid.

Certified Reference Material (CRM) 66 (New Brunswick Laboratory) was used as the source of aged ThO_2. This material had been fired to 900°C in 1958 and had reached near equilibrium with its progeny. Newly fired material was prepared by dissolving the CRM, precipitating thorium as an oxalate, and refiring to 900°C. The material was sandwiched between two filters and enclosed in a filter holder. This filter holder was then placed in simulated lung fluid at 37°C with a pH range of 7.3 to 7.4 for some specified length of time. Upon completion of the time, the filter was removed from the solution and placed in fresh simulated lung fluid for the next time interval.

Radiochemical analysis of the isotopic thorium is conducted as described by Glover et al.[1] Briefly, and aliquot is taken from each fraction, ^{229Th} tracer is added, then taken to dryness, and ashed several times with HNO_3 and H_2O_2 to destroy any organic material and to ensure complete isotopic mixing of the tracer and ThO_2 The aliquot is redissolved in 8 *M* HNO_3, and the thorium is isolated using anion exchange methods. The resulting eluent containing the thorium is then electroplated onto vanadium planchets using the methods described by Glover et al.[2] for thorium determination by alpha spectrometry and preconcentration neutron activation analysis. Radiochemical yield for the PCNAA technique is determined during the alpha spectrometry measurement. Detection limits for this method are 3.3×10^{-4} Bq/sample for both ^{228}Th and ^{230}Th and 5×10^{-6} Bq/sample for ^{232}Th with a precision of ~3 to 5% fat the limit of quantification. These detection limits will allow characterization of the solubility even in the early fractions for the thorium isotopes.

The determination of ^{228}Ra in these fractions is also very important. A recovery corrected radium determination method was developed specifically for these measurements to improve the detection limits compared to direct counting of the solution. This method is based on extraction of the radium onto Empore Radium Rad Disks. Chemical yield was determined using ^{223}Ra as a tracer immediately following separation of the radium from the simulated lung fluid by gamma-ray spectrometry. The disk is then allowed to decay for at least ten half-lives of ^{228}Ac ($t_{1/2}$=6.15 h) so that ^{228}Ra and ^{228}Ac reach equilibrium. The disk is counted on a geometry-calibrated gamma detector using the 911-keV gamma ray of ^{228}Ac for the determination of ^{228}Ra.

USTUR-0152-00