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SCREENING OF IAEA ENVIRONMENTAL SAMPLES FOR FISSILE MATERIAL CONTENT

DOYLE M. HEMBREE, JR., JOEL A. CARTER, GERALD L. DEVAULT,
J. MICHAEL WHITAKER
Y12 National Security Complex, Oak Ridge, Tennessee, USA

DAVID GLASGOW,
Oak Ridge National Laboratory, Oak Ridge, Tennessee, USA

Analysis of environmental samples for the International Atomic Energy Agency (IAEA) Strengthened Safeguards Systems program requires that stringent measures be taken to control contamination. To facilitate contamination control, it is extremely useful to have some estimate of the fissile content of a given sample prior to beginning sample preparation and analysis. This is particularly true for laboratories that employ clean rooms during sample preparation. A review of the analytical results for samples submitted between January 1, 1999 and September 1, 2000 revealed that the total uranium content values ranged from 0.2 to greater than 500,000 ng/sample. Poor estimates of the uranium or plutonium content in the samples have caused some of the laboratories in the IAEA Network of Analytical Laboratories (NWAL) to experience clean laboratory contamination, sample cross contamination, and non-ideal uranium spike additions. This has led to significant increases in analysis costs (e.g., recertification of clean rooms after removing contamination, and rerunning samples) and degradation in data quality. A number of methods have been proposed for screening environmental samples for fissile material content, including gamma spectrometry, x-ray fluorescence, kinetic phosphorimetry (KPA), and inductively coupled plasma-mass spectrometry (ICP-MS). Gamma spectrometry and x-ray fluorescence are suitable for screening samples with microgram or greater quantities of uranium. ICP-MS and KPA are used successfully in some DOE NWAL laboratories to screen environmental samples.

A neutron activation analysis (NAA) method that offers numerous advantages over other screening techniques for environmental samples has recently been proposed. Fissile materials such as ^{239}Pu and ^{235}U can be made to undergo fission in the intense neutron field to which they are exposed during neutron activation analysis (NAA). Some of the fission products emit neutrons referred to as "delayed neutrons" because they are emitted after a brief decay period following irradiation. Counting these delayed neutrons provides a simple method for determining the total fissile content in the sample. Neutron activation analysis is a nuclear technique, which means that the chemical bonding environment of a fissile atom has no effect on the measurement process. Therefore, NAA is virtually immune to the "matrix" effects that complicate other methods. As a result, environmental samples such as pine needles, water, soil, sediments, or swipes can all be rapidly analyzed with little or no sample preparation. All of the sources of error in the NAA-delayed neutron counting experiment are known and can be accounted for. In addition, the method is nondestructive and does not require the use of a blank measurement.

The neutrons counted as a result of ^{239}Pu fission cannot be easily discriminated from those arising from ^{235}U. Therefore, the delayed neutron method proposed for sample screening will provide total fissile content; not the quantities of individual radionuclides. However, for most IAEA samples, it can be assumed that uranium is the predominant fissile material, and the method can be usefully employed for screening using this assumption (data collected from a large number of environmental samples supports this contention). The method is fast (~5 minutes per sample), very sensitive (3-10 pg ^{235}U), and very accurate.

An initial assessment of the NAA method was conducted using 35 swipe samples. The uranium results using thermal ionization mass spectrometry (TIMS) ranged from less than blank values to 332 ng/sample. The comparison of results between TIMS and NAA was quite favorable; the average percent difference between the two methods for the 35 samples was less than 5%. The accuracy, large dynamic range, speed, and low cost of the NAA-DN technique make the method an excellent choice for properly screening environmental samples.