Screening of Fissile Material by Delayed Neutron Activation Analysis

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1. Introduction

DNAA is a method of neutron activation analysis for detection and qualification of fissile material in various types of samples by delayed neutron counting. DNAA offers significant advantages for screening of environmental samples due to the rapid, nondestructive, sensitive and interference free analysis at relatively low cost comparing to other screening methods. [1]
Technical cooperation on the delayed neutron activation analysis (DNAA) method is being implemented pursuant to the bilateral safeguards arrangement between the United States Department of Energy and the Ministry of Science and Technology of the Republic of Korea, and the technical training was conducted at ORNL during May, 2004. The system characteristics such as reproducibility and linear range of measurement, and swipe sample screening were performed during the training. The results will be presented in this paper.

2. Experimental

2.1 DNAA system and irradiation protocol

The delayed neutron facility in the High Flux Isotope Reactor at ORNL consists of the pneumatic transfer tube, pneumatic tube programmable logic controller and user interface, neutron detector assembly and signal processing equipment. The rabbits, polyethylene sample containers, are loaded into the pneumatic tube in a fume hood loading station, irradiated for pre-selected time, and automatically moved to the delayed neutron counting assembly. The details of the transfer system are in reference [2].

Heating at the irradiation position is low and polyethylene rabbits survive irradiation in excess of 20 minutes duration. The maximum sample size is a right cylinder of a diameter of 10mm and height 18mm. The thermal neutron fluence rate was measured to be 4.32×10^{13} $n \cdot cm^{-2} \cdot s^{-1}$ and the epithermal fluence rate was approximately 1.22×10^{11} $n \cdot cm^{-2} \cdot s^{-1}$.

Typically, blank samples of filter paper are irradiated along with calibration standards, standard reference materials and calibration verification samples as appropriate. Irradiation duration is usually four minutes or less and is followed by 10-15 seconds decay and counting for 1-2 minutes. Therefore, the total time for a single analysis is about six minutes or less. The sample transfer time from the reactor to the counter is about 2.5 seconds.

2.2 Standard sample preparation

Most of the samples and standards used for calibration and surveillance of the delayed neutron counting system are prepared from diluted solutions of commercial inductively coupled plasma standards obtained from the National Institute of Standards and Technology (NIST) or from VHG Labs. The liquid materials were pipetted gravimetrically onto filter paper and dried prior to irradiation and the fly ash standard reference material was oven dried at $105^{\circ}C$ for two hours and used as received. All of the solutions were nitric acid stabilized and the aliquots prepared for analysis were taken soon after solution preparation.

2.3 Swipe sample preparation

ROK swipe samples were treated in cleanroom glove bags in order to prevent contamination. Each was too large to fit into the rabbit and was sectioned into three pieces using new disposable scissors. Each third was folded repeatedly until it fit neatly into the rabbit and the rabbit cap was installed prior to opening the glove bag.

3. Results

3.1 Reproducibility of Measurement

Two groups of ^{235}U samples, nominally 170ng and 1.68ng, were prepared from VHG uranium ICP solution. These were analyzed to establish system reproducibility. Figure 1(a) shows that the 1-sigma standard deviation in the mean is very low at the 170ng level. Counting statistics uncertainty at 1-sigma was 0.5%. At the 1.68ng level, depicted in Figure 1(b) the 1-sigma standard deviation in the mean is higher because the counting statistics were poorer at about ±2.5%. The reproducibility of the irradiation position, system timing, counting position, and sample preparation appear to be excellent and very much dependent on counting statistics variation.

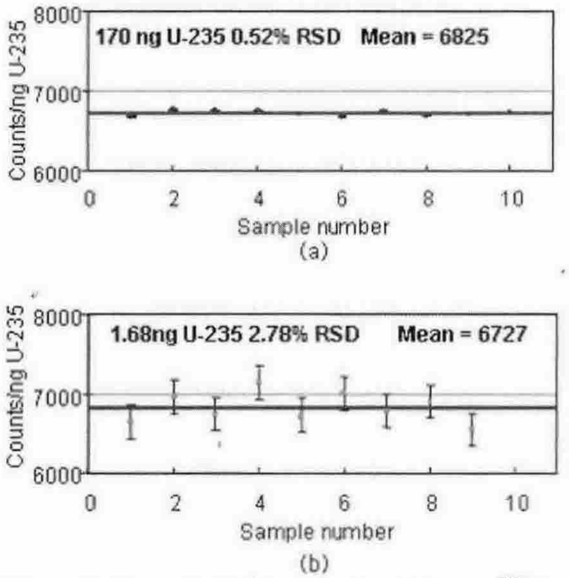

Figure 1. Reproducibility tests for different ^{235}U mass groups

3.2 Linear range of the DNAA system

In order to identify the linear range of the DNAA system, a number of samples which varied from 10^{-1} to 10^{3} nanograms of ^{235}U were prepared using NIST 3164 uranium ICP standard solution. Measured counts showed a good linearity with respect to ^{235}U contents as seen in Figure 2 and no detectable deviation from linearity was observed.

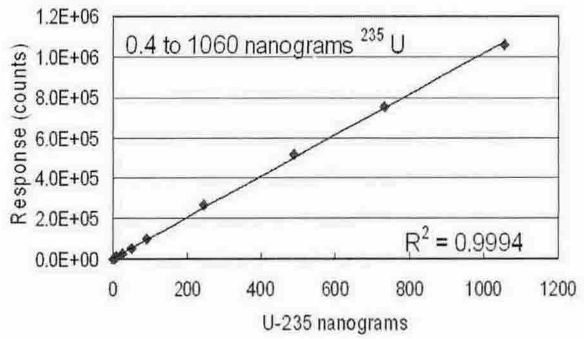

Figure 2. System linearity test from 0.4 ng to 1060 ng ^{235}U contents

3.3 Determination of ^{235}U in swipe samples

The ^{235}U contents in three sets of swipe samples taken at KAERI were determined by DNAA. One set of samples consists of two swipes, one is the sample taken at the working area and the other, named control sample, is a field blank made by cleaning the inspector's hands. The results are shown in Table 1.

Table 1. Results of ^{235}U determination ofswipe samples

	Sample #	ng ^{235}U Equivalents
1	Control sample	0.27 ± 0.02
	Swipe sample	42.5 ± 0.2
2	Control sample	0.62 ± 0.03
	Swipe sample	95.7 ± 0.3
3	Control sample	0.23 ± 0.02
	Swipe sample	4.17 ± 0.07

3. Conclusion

The irradiation and counting system of DNAA facility is confirmed to be highly reproducible and linear up to one million counts accumulated in one minute.

DNAA method has been applied for screening of IAEA swipe samples at ORNL. The wide range of fissile materials in the samples have caused of clean laboratory contamination, sample cross contamination. And all of these have led to an increase in analysis costs and a decrease in data quality. Therefore an estimate of the fissile material content of a given sample prior to beginning sample preparation for the mass-spectrometry analysis would be very useful. DNAA is excellent screening method for wide area monitoring, protection of clean room facilities and routine surveillance of nuclear facilities.

REFERENCES

[1] F. F. Dyer et al., "A comprehensive study of the neutron activation analysis of uranium by delayed neutron counting," ORNL-3342 (1962).

[2] F. F. Dyer et al., "Design and use of the ORNL HFIR pneumatic tube irradiation systems" Journal of Trace and Microprobe Techniques, Vol. 6 (2), 147-159 (1988).