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A COMPUTER-AUTOMATED NEUTRON ACTIVATION ANALYSIS SYSTEM

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ABSTRACT

An automated delayed neutron counting and instrumental neutron activation analysis system has been developed at Los Alamos National Laboratory's Omega West Reactor (OWR) to analyze samples for uranium and 31 additional elements with a maximum throughput of 400 samples per day.

INTRODUCTION

An automated neutron activation analysis system has been developed to analyze large numbers of geological and environmental samples. Approximately 180,000 stream sediment samples have been analyzed for uranium concentrations using delayed neutron counting (DNC) and for 31 additional elements using instrumental neutron activation analysis (INAA) at maximum system throughput. At lower throughput, additional elements can be detected and excellent sensitivity for most elements is obtained.

The system utilizes two independent pneumatic transfer systems each having a high-efficiency delayed-neutron detector and four lithium-drifted germanium (Ge(Li)) gamma-ray detectors under the control of a single data acquisition computer. Samples are irradiated in the graphite thermal column of the OWR in a flux of $\sim 6 \times 10^{12}$ n/cm^2/sec and are counted immediately for delay-neutron emission to obtain the uranium assay. The samples are then routed to a delay device where the samples are allowed to decay for a preset time (typically 20 min) before they are counted for short-

lived gamma activities. The samples are then given a second irradiation and stored for later counting of the longer-lived activities at night when the reactor is shut down. System parameters are summarized in greater detail elsewhere.[1]

Gamma-ray spectra along with timing and flux data and DNC data are collected and dumped to magnetic tape for subsequent data reduction by a second on-line minicomputer. Automatic analysis of data is performed by a code which identifies gamma-ray peaks and calculates elemental concentrations.

SYSTEM DESCRIPTION

A. Hardware

1. Pneumatic Sample Handling Facilities. Samples to be entered into the automated system are loaded into 4 cm^3 irradiation vials (rabbits) 1.2-cm dia. by 6.0-cm long. The pneumatic system is designed to automatically load a sample and rapidly move the sample into the thermal column of the OWR, from there to the neutron detector, and later to a shielded location in front of a Ge(Li) gamma-ray detector for subsequent radioactive decay counting.

The individual samples are weighed and loaded into loader clips. The automatic loader for each system can hold four clips and can sequentially load a maximum of 200 samples into the pneumatic system on command from the control computer. The samples are irradiated in a thermal neutron flux in a special irradiation port placed in the graphite thermal column of the OWR. A fission ion chamber is inserted in the graphite stringer adjacent to the end of the port to monitor the relative neutron flux at the irradiation position.

A delay loader was designed for use in the system to allow the irradiated samples to decay for set periods after irradiation before they are gamma-ray counted. A number of samples are allowed to stack up on the input side of the delay loader and then are removed one at a time from the bottom of the stack, at the sample rate at which samples are being added to the top of the stack.

In order to route samples between the loader and reactor and the various counting stations in the system, two four-way pneumatic diverters are used in the system. After samples have been irradiated a second time, the samples are blown to a storage unloader where the samples can be stored for any length of time in the same order in which they were irradiated. The unloader is

mounted at the top of a cased hole which extends underground for shielding.

2. Control and Data Acquisition. The control and data acquisition system is normally controlled by a PDP-11/34 minicomputer. A second computer, a PDP-11/60, is used for on-line data analysis. All hardware in the system are interfaced to the control and data acquisition computer through CAMAC. Photo detectors are installed in the pneumatic system to detect the passage of samples at eleven locations to detect any malfunctions.

The neutron detector used in the system is described in detail elsewhere.[2] Delayed neutron counts are recorded in a CAMAC scaler that can be cleared and read by the computer. Each multielement analysis system utilizes four Ge(Li) gamma-ray detectors employing standard electronics and conventional nuclear ADCs set for 4096-channel conversion gain. Spectra are stored in 4096-word CAMAC memory modules. The memory modules can be cleared, read out, and enabled or disabled to accept digital conversions on command from the computer. Since the samples counted vary greatly in gamma-ray source strength, real times and live times are accumulated with each gamma-ray count.

B. Software

1. Control and Data Acquisition. Two basic programs exist to acquire 1) Multielement data from short half-life isotopes, and 2) Multielement data from long half-life isotopes. The multielement analysis program for short half-life activities, SHORTS, in addition to the delayed neutron counting cycle, includes a decay period, a gamma-ray count period, and an additional second irradiation for each sample. Each sample cycle takes a minimum of 32 minutes to complete; thus there can be as many as 15 samples in each system at any given time. The program automatically backs up accumulated gamma-ray spectra to magnetic tape and stores all pertinent data including irradiation times and fluxes required by the long half-life analysis program.

The multielement analysis program for intermediate and long half-life activities, LONGS, includes only a gamma-ray counting cycle for each sample. The LONGS program loads and counts samples that have normally decayed for 4 or more days after being processed by the SHORTS program. The LONGS program is usually run at night or on weekends while the reactor is shut down.

2. Gamma-Ray Data Reduction. The computer program for gamma-ray data reduction, RAYGUN, is a variant of GAMANAL[3] tailored to the PDP-11. Data to be analyzed are shipped to the Data Analysis Computer as a 4096 channel spectrum in which the

data of the first 30 channels have been replaced by a header block of constants required for data reduction.

The RAYGUN analysis code requires approximately 24K words of PDP-11 memory in which to run. A single spectrum can be fit, gamma-ray peaks identified, and elemental concentrations calculated in approximately 17 seconds.

SYSTEM PERFORMANCE

The analytical detection of uranium using DNC has been employed for many years.[4] The standard DNC measurements in the system are calibrated for fixed irradiation (20 s), decay (11 s), and count (30 s) times using samples of known natural uranium concentration. The counting scheme is less than optimal but results in a high specific sensitivity for uranium in geological samples (e.g., a four-gram sample with 2 ppm uranium will yield 5000 net neutron counts with a statistical accuracy of 1.4%).

The INAA techniques employed are novel only in respect to the high degree of automation and analysis volume. Sample irradiation times are selected on the basis of desired precision and sample size. Short and long sample irradiations of 20 and 96 seconds induce sufficient activity for sample masses greater than 1 gram and provide good counting statistics for the majority of elements of interest. For higher precision, irradiation times of 20 and 220 seconds are typically used.

The samples are counted directly in their irradiation vials. The vials are sufficiently free of trace elements that analyses are made with no blank subtraction. Counting-room background subtractions are not required for the short-lived activities. Minor contributions to the gamma-ray peaks associated with ^{46}Sc, ^{59}Fe, ^{60}Co, and ^{65}Zn are present when counting long-lived activities. Provision for discrete background subtraction is incorporated in the analysis code.

At the maximum sample throughput rate of 400 samples per day, the samples are only counted twice (at 20 min and 14 days) and concentrations of 32 elements are typically determined. At a throughput rate of 100 samples/day the automated system yields elemental assays for 15 elements from the "shorts" analysis (See Table 1.); 14 elements from the "intermediate" analysis; and 18 elements from the "longs" analysis in typical geological samples. Table 1 gives detection limits in geological samples for specified irradiation, decay, and count times. The table was compiled from data on a large number of geological samples and standards. Uncertainties in the measured trace-element concentrations are usually less than 10% at concentration values one order of magnitude above the lower detection limits.

Table 1. Elemental Lower Limits of Detection[1]

Shorts Analysis		Intermediates Analysis		Longs Analysis	
Element	Detection Limit[2] (ppm)	Element	Detection Limit[3] (ppm)	Element	Detection Limit[4] (ppm)
Na	1000	Na	300	Sc	0.04
Mg	2700	K	4500	Cr	2.5
Al	3200	Ga	45	Fe	300
Cl	120	As	3.0	Co	0.20
K	4500	Br	4.0	Zn	15
Ca	1500	Sb	1.0	Se	5
Ti	750	Ba	800	Rb	20
V	8.0	La	7.0	Sb	0.50
Mn	60	Sm	4.0	Cs	0.60
Cu	350	Yb	1.0	Ba	300
Sr	400	Lu	0.2	Ce	4.0
I	40	W	5.0	Eu	0.09
Ba	200	Au	0.04	Tb	0.15
Dy	0.9	U	10	Yb	0.50
U	.05			Lu	0.06
				Hf	0.35
				Ta	0.45
				Th	0.35

[1]Lower Limit values were calculated on the basis of a nominal 4 gram sample.
[2]Based on a 20 sec irradiation; 1275 sec decay; 480 sec gamma ray count.
[3]Based on a 240 sec irradiation; 5 day decay; 1800 sec gamma ray count.
[4]Based on a 240 sec irradiation; 21 day decay; 2 hour gamma ray count.

DISCUSSION

The automated activation analysis system has thus far been used mostly for high volume analysis of samples. The new software currently employed with the system allows much more flexibility in the choice of irradiation, decay, and count times. These changes allow a much greater variety of sample size and matrix to be analyzed with increased sensitivity and precision and with the advantages and reproducibility[5] of a fully automated INAA system.

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