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Utilization of the Intense Pulsed Neutron Source (IPNS) at Argonne National Laboratory for Neutron Activation Analysis*

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UTILIZATION OF THE INTENSE PULSED NEUTRON SOURCE (IPNS) AT ARGONNE NATIONAL LABORATORY FOR NEUTRON ACTIVATION ANALYSIS*

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ABSTRACT

The Intense Pulsed Neutron Source (IPNS) neutron scattering facility (NSF) has been investigated for its applicability to neutron activation analysis. A polyethylene insert has been added to the vertical hole VT3 which enhances the thermal neutron flux by a factor of two. The neutron spectral distribution at this position has been measured by the multiple-foil technique which utilized 28 activation reactions and the STAYSL computer code. The validity of this spectral measurement was tested by two irradiations of National Bureau of Standards SRM-1571 (orchard leaves), SRM-1575 (pine needles), and SRM-1645 (river sediment). The average thermal neutron flux for these irradiations normalized to a 10 μamp proton beam is 4.0×10^{11} n/cm^2-s. Concentrations of nine trace elements in each of these SRMs have been determined by gamma-ray spectrometry. Agreement of measured values to certified values is demonstrated to be within experiment error.

INTRODUCTION

The motivation of this study was initially generated several years ago at the closing and decommissioning of Argonne's CP-5 reactor. Although this facility was primarily utilized for basic neutron research, it did provide the analytical chemist with the very sensitive and powerful capability of neutron activation analysis. Because of economic considerations, the Intense Pulsed Neutron Source (IPNS) design could not consider sample activation as a high priority item. However, several neutron beam holes which extended into the IPNS target reflector region were provided and these, although limited in sample volume and neutron flux, could conceivably be used for sample activation.

The challenge in using this high energy accelerator-based neutron source focuses upon knowing the neutron spectral distribution within the sample volume since neutrons of all energies are generated within the target. Thus, before meaningful activation analysis could be done, careful neutron dosimetry of the neutron spectrum had to be accomplished. This paper describes those efforts

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and indeed demonstrates that meaningful neutron activation analysis can be done at the IPNS facility.

INTENSE PULSED NEUTRON SOURCE (IPNS)

The IPNS is based upon a rapid cycling synchrotron (RCS) which accelerates protons to 500 MeV (ref. 1). The RCS produces 100-nm pulses of protons at repetition rates up to 30 Hz. The protons from the accelerator are transported to one of two target assemblies, the Radiation Effects Facility (REF) and the Neutron Scattering Facility (NSF), which have been optimized for radiation effects and neutron scattering, respectively. The proton beam time is divided between the two facilities such that the NSF receives the proton beam about 75% of the operating time. For this reason, two vertical irradiation thimbles were installed in the NSF. The target in each assembly is made of 238-U. Neutrons with all energies up to the incident proton energy are produced by the processes of spallation and fission, at a rate of ~20 neutrons/proton.

The REF (ref. 2) also consists of a 238-U target, two vertical irradiation thimbles, and a horizontal irradiation thimble, all surrounded by a Pb neutron reflector. The neutron reflection and the high-energy (n, 2n) reactions in Pb increases the neutron flux and also reduces the neutron gradient in a direction perpendicular to the target axis. The two vertical irradiation thimbles are located on either side of the target at positions of maximum flux. Each has a liquid He cryostat which can operate at temperatures between 4 and 1000 K. The horizontal irradiation thimble is located on an axis parallel to and directly below the target and operates at ambient temperatures. The neutron spectrum in the REF is considerably harder than in the NSF in order to maximize damage in materials which are under study.

In the NSF, the fast neutrons produced by fission and spallation reactions in the uranium target are thermalized by moderators (with a high hydrogen density) located above and below the target. Two types of moderators have been used. The first type consisted of four ambient-temperature polyethylene moderators with an inner graphite reflector. The second type consisted of four 100 K circulating liquid methane moderators with an inner 100 K beryllium reflector. Currently the polyethylene-type moderator is being used. Twelve horizontal beam tubes and one vertical neutron beam tube view selected faces of the moderators to provide neutron beams to instruments. Each horizontal beam tube contains a 36-inch thick steel beam gate. Two vertical holes (VT3 and VT4) and two horizontal holes (F-6 and H-2) may be used for sample irradiations. A polyethylene insert has been added to the vertical hole VT3 to enhance the neutron flux and has resulted in an increase of the thermal component by a factor of two.

NEUTRON SPECTRUM DETERMINATION

The multiple-foil activation technique was used to determine the neutron energy distribution in the VT-3 position with and without the polyethylene liner. Twenty-eight different neutron-activation reactions were measured at the maximum flux location. The foil activities were measured with HPGe and Ge(Li) detectors over decay periods of several half-lives for each reaction. Peak integrations and Compton-background subtractions were done by means of standard computer codes. Corrections for cover foils, neutron and gamma self-shielding, and decay during and after the irradiation were made for the activation products. The STAYSL computer code (ref. 3) was used to adjust the neutron spectrum (100 energy groups) by fitting the foil activities. This method calculates the activity for each activation reaction using ENDF/B-V (ref. 4) nuclear cross-sections and an initial input neutron spectrum calculated with the HETC (ref. 5) computer code. The spectral adjustment procedure follows a least-squares approach where uncertainties and covariances are assigned to the measured activities, cross sections, and starting flux spectrum. The output flux spectrum and covariance matrix are determined by a matrix inversion procedure which minimizes the chi-square parameter. The neutron spectrum for the VT3 polyethylene lined hole is shown in Fig. 1. Collapsed neutron spectra of both the poly and bare VT3 is presented in Table 1. Both are normalized to a 10-μamp proton beam.

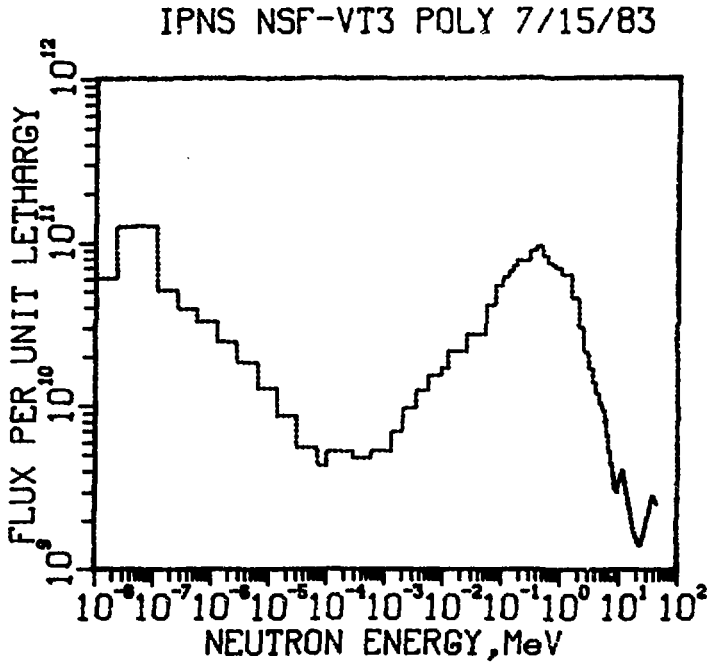

Fig. 1. Neutron spectrum IPNS NSF VT3 (poly) vertical hole.

TABLE 1

IPNS-NSF-VT3 neutron fluxes normalized to a 10-μamp proton beam current

Energy, MeV	Flux, x 10^{11} n/cm^2-s	
	Bare	Poly
Total	8.67	10.17
>0.1	3.00	3.28
>1.0	0.893	0.880
Thermal (<0.5 eV)	2.00	4.42
0.5 eV - 0.1	3.65	2.50
0.1 - 1.0	2.10	2.40
1.0 - 5.0	0.805	0.755
5.0 - 10.0	0.0418	0.0521
10.0 - 20.0	0.0243	0.0268
20.0 - 30.0	0.00811	0.00874
30.0 - 44.0	0.0119	0.0131
>44.0[a]	0.0262	0.0287

[a]Calculated HETC (ref. 5).

EXPERIMENTAL

Standard Reference Materials (SRM)

Three National Bureau of Standards Standard Reference Materials (SRM) were used for the determination of various trace element concentrations. These were SRM-1571 (orchard leaves), SRM-1575 (pine needles), and SRM-1645 (river sediment). Sample sizes ranged from 240 mg for the smallest sample to 480 mg for the largest. SRM-1571 and SRM-1575 were oven dried at 85°C for about two hours and then stored in a desiccator until weighed and encapsulated. SRM-1645 was oven dried at 50°C for about two hours and also desiccator stored. Samples were prepared for the irradiations by encapsulating in 1-cm (dia.) by 2.5-cm polyethylene vials that had been previously cleaned in distilled water and ethanol. After the vials were clean, handling during the weighing and packaging processes was done using clean polyethylene gloves.

Flux monitors of nickel and cobalt were placed in horizontal positions at the bottom, middle, and top of the irradiation package, which consisted of each of the SRMs and an empty (blank) vial. Vertical flux monitors of iron, nickel, and cobalt were strategically placed in the interior of this package so that correlations could be made with the horizontal monitors and also measure the vertical flux gradients. All flux monitors were encapsulated in polyethylene tubing to avoid sample cross-contamination.

Irradiation Conditions

Two irradiations were made in the polyethylene-lined vertical tube VT3 of the NSF. The first was a relatively short irradiation of 16.9 hours which enabled the determination of short-lived activation products. The second irradiation

spanned 12.0 days and enabled the determination of long-lived activation products. The time-averaged neutron flux of the short irradiation was calculated to be 7.47 x 10^{11} n/cm^2-s and for the long irradiation, 6.41 x 10^{11} n/cm^2-s. The measured thermal neutron fluxes for the two irradiations were 3.2 x 10^{11} and 2.8 x 10^{11} n/cm^2-s, respectively. Average proton beam currents were measured to be 8.64 μamps for the short and 6.54 μamps for the long irradiation.

Gamma-Ray Spectrometry

The counting of the SRM samples from the short irradiation started about four hours after the end of the irradiation and utilized three absolutely calibrated germanium detectors (30-45 cm^3). Each sample was counted on all detectors to reduce any calibration biases. Counting of each sample for the particular nuclide of interest was repeated at least three times within three half-life periods from the end of the irradiation. The principal gamma-ray of each nuclide was counted for sufficient length of time to provide counting statistics ranging from 0.1-5%, and the precision for replicate counts was required to be within the 5% range. Counting geometries were adjusted such that errors due to dead-time and random summing were negligible. Peak integration and identification was done by conventional computer techniques (ref. 6).

RESULTS AND DISCUSSION

The comparison of measured SRM trace element concentrations to NBS values is presented in Table 2. The excellent agreement demonstrates primarily the accuracy of the neutron dosimetry and the methodology used in determining the IPNS neutron spectrum. Uncertainties on the measured values are estimated to be ±7% which includes estimated errors in the neutron cross-sections, spectrum unfolding, detector calibration, counting statistics, and neutron flux gradient corrections. Of the nine trace elements listed, only the As, Rb, and Sb appear to be biased high. This could be attributed to the reaction cross sections used since uncertainties for these reactions are listed at about twice the magnitude as uncertainties for the reactions for the other elements.

Conclusions are that meaningful neutron activation analysis can be performed at the IPNS in the VT3 (poly) position, but this is contingent upon the fact that adequate neutron dosimetry be provided also.

TABLE 2

Comparison of measured to certified trace element concentrations

	Element Concentration, ppm unless indicated					
	SRM-1571 (Orchard Leaves)		SRM-1575 (Pine Needles)		SRM-1645 (River Sediment)	
Element	Value	This Work	Value	This Work	Value	This Work
Na	82 ± 6	82 ± 6	--	38 ± 3	5500[a]	4892 ± 342
Sc	(.06)[b]	0.06 ± 0.004	0.03[a]	0.038 ± 0.003	2[a]	1.9 ± 0.1
Cr	2.3[a]	2.5 ± 0.2	2.6 ± 0.2	2.3 ± 0.2	2.96 ± 0.28%	2.99 ± 0.21%
Mn	91 ± 4	89 ± 6	675 ± 15	654 ± 46	785 ± 97	795 ± 56
Fe	300 ± 20	290 ± 20	200 ± 10	188 ± 13	11.3 ± 1.2%	10.4 ± 0.7%
Co	0.2[a]	0.27 ± 0.02	0.1[a]	0.14 ± 0.01	8[a]	8.1 ± 0.6
As	14 ± 2	18 ± 1	0.21 ± 0.04	--	66[a]	109 ± 8
Rb	12 ± 1	17 ± 1	11.7 ± 0.1	16.5 ± 1.2	--	58.5 ± 4.1
Sb	--	5.7 ± 0.4	0.2[a]	0.4 ± 0.03	51[a]	60 ± 4

[a]Uncertified; information value only.
[b]Ref. 7.

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