ACTIVATION ANALYSIS OF TRACES WITH VERY HIGH
NEUTRON FLUX AND WITH 140-MeV ELECTRON BREMSSTRAHLUNG*

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Enzo Ricci

Analytical Chemistry Division
Oak Ridge National Laboratory
Oak Ridge, Tennessee 37830
USA

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ABSTRACT

Two unique "rabbit" facilities have recently been installed at ORNL; the neutron activation analysis system at the High Flux Isotope Reactor (HFIR), and the photon (and fast-neutron) activation analysis facility at the Oak Ridge Electron Linear Accelerator (ORELA). A salient feature of the HFIR irradiation site is a thermal neutron flux of $5 \times 10^{14} \text{ cm}^{-2} \text{ sec}^{-1}$. In turn, the ORELA facility provides for simultaneous bombardment of samples and standards with bremsstrahlung photons or fast neutrons from a 140 MeV, 30 Kw electron beam. Results obtained at these facilities are discussed while their main features are described.

NEW TRENDS AND FACILITIES

Present trends in biomedical and environmental science demand analytical determinations for many trace elements in a variety of samples. Thus, great efforts are currently being made to develop methods for multielement analysis at reasonable cost.¹ At the same time, high-quality analysis of specific samples is becoming increasingly necessary.

Present ORNL analytical policy aims, on one hand, at simultaneous analysis (preferably instrumental) for numerous trace elements, to attain a low cost-per-element. On the other, we hope to reduce the price of quality serial analyses for specific elements by non-destructive methods. Two facilities for activation analysis were recently completed at Oak Ridge National Laboratory, that can meet these demands. Both are pneumatic transfer systems, one installed

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at the High Flux Isotope Reactor (HFIR) and the other at the Oak Ridge Electron Linear Accelerator (ORELA). The HFIR facility is for neutron activation analysis at a very high neutron flux -- ideally suited for multielement analysis. The ORELA facility is mainly for photon-activation analysis with 140 MeV electron bremsstrahlung, and allows specific determinations of elements -- particularly of light atomic weight -- for which neutron activation is insensitive.

Figure 1 shows the HFIR "rabbit" facility. Aside from features common to other pneumatic systems, because of the high flux it is provided with an intermediate "hold up" station to allow decay of very hot samples, and with master-slave manipulators (not shown) for sample unloading. The reactor vessel contains the core, surrounded by a removable and a permanent beryllium reflector. The cylindrical "rabbit" capsule (2 cm^3 capacity) is positioned vertically at the irradiation site, in the permanent beryllium sheath. At this site the thermal neutron flux is $4.8 \times 10^{14} \text{ cm}^{-2} \text{ sec}^{-1}$ and the resonance flux, $1.1 \times 10^{13} \text{ cm}^{-2} \text{ sec}^{-1}$. The large flux ratio (~ 44) diminishes the significance of resonance activation; thus, absolute activation analysis can be performed with accuracy. The thermal flux varies within the "rabbit," at this site, by about 1% longitudinally and by 3-4% across it. As the capsule may be spun during bombardment, the latter effect can be minimized. Plastic and magnesium "rabbits" are used for short irradiations, and graphite capsules for long ones. A Ge(Li) detector system with 12% relative efficiency (75 cm^3) and a Nuclear Data 50/50, 4096 channel analyzer, plus a PDP-15 computer are assigned to the HFIR facility for obtaining and processing data.

The ORELA produces an electron beam of 120-160 MeV with power of up to 30 Kw (i.e., about 220 μA current); this beam impinges on a thick, cooled tantalum target. Though this design is aimed at increasing the fast-neutron/gamma-ray output ratio -- for physics experiments -- it still results in a satisfactory bremsstrahlung photon beam. Our bombarding facility is carefully positioned to place the "rabbit" in this photon beam. The design of the "rabbit" is original and has deserved a U.S. Patent²; it is shown in Figure 2. Four 2 cm^3 aluminum capsules are carried by an aluminum sphere which, during irradiation, spins and precesses on an upstream of air in a cylindrical bombardment chamber. The air stream thus provides not only for support and cooling of the sphere, but also for homogeneous bombardment of the capsules inside it.

RESULTS AND DISCUSSION

Because of the newness of these facilities, only examples of results obtained at them can be mentioned at this writing. Furthermore, these are preliminary results on presently available samples, containing relatively large elemental concentrations. More complete data will be given at the time of the Colloquium.

At the HFIR we have begun a program to measure concentrations of air particulates (multielement analysis) both in pristine environments and near pollution sources, particularly energy plants. The first experiments were necessarily aimed at establishing levels of impurities in air-filtering materials, because these are going to determine the analytical detection limits of our instrumental analyses. Table I exemplifies results for four elements found in samples of Whatman No. 42 filter paper; they correspond to a 10-minute irradiation followed by 3-4 hours decay.

Absolute activation analysis was used in these determinations; the necessary thermal and resonance flux data were obtained by simultaneous bombardment of aluminum diluted manganese ($\sigma_0 = 13.3$ b, $I_0 = 15.6$ b) and gold ($\sigma_0 = 98.8$ b, $I_0 = 1558$ b) monitors. The solution of the equations resulting from the flux measurements, and the absolute calculations were carried out at the PDP-15 computer. Table I also shows satisfactory results of an accuracy test of the absolute activation analysis subroutine, for which known concentrations of manganese and gold in aluminum (monitors) were redetermined. Clearly, the detection limits listed can be easily improved (see chlorine data) and the number of elements determined increased, by adequate combination of irradiation lengths and decay times; this will be the next step in our development work.

Table II lists results of a survey of photon-activation, interference-free sensitivities carried out at the ORELA facility. It shows, as expected, that the best sensitivities correspond to the light elements. An unexpected result, however, is that these sensitivities -- which correspond to 140-MeV bremsstrahlung photons -- as well as the types of operating nuclear reactions are very much the same as the ones reported for experiments at much lower electron energies^{3,4}. Thus, neither unusual sensitivities nor excessive interferences were found to distinguish ORELA's setup from other photon-activation facilities. We believe

this effect to be due to photon absorption and degradation by the thick tantalum target required for neutron production--the main objective of the ORELA. Examples of analyses are given in Table III. Though they correspond to relatively large carbon concentrations, they represent two important areas of application of the ORELA facility -- specific determinations in pure metals and in environmental (pollution) samples. Errors in results of Tables II and III are generally of about 5-10%, but a few data have higher uncertainties because they were obtained while the performance of the facility was being adjusted. To enhance sensitivity and accuracy in specific light-element determinations, coincidence equipment -- for annihilation gamma counting -- will be installed in this facility, and Cherenkov counting will also be attempted to discriminate against low-energy positron emitters.

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FIGURES

Figure 1. Neutron Activation Analysis Pneumatic Transfer System at the High Flux Isotope Reactor.

Figure 2. Bombarding Chamber and Sphere "Rabbit" for Photon Activation Analysis at the Oak Ridge Electron Linear Accelerator.

TABLE I

EXAMPLE OF RESULTS OBTAINED AT
THE HFIR FACILITY

Element	Measured Product		Analysis of Whatman No. 42 Air Filter Paper		Test of Absolute Activation Analysis Method		Error	Corresponding Detection Limits ^c
	Nuclide	Half-Life (hr.)	Sample 1 (p.p.m.)	Sample 2 (p.p.m.)	Found (%)	Known (%)	(%)	(p.p.m.)
Na	²⁴ Na	15.0	16.8	16.8			0.7 - 0.6	0.09
Cl	³⁸ Cl	0.62	291	282			1.2 - 1.7	3.1
Mn	⁵⁶ Mn	2.58	0.27	0.26	0.0867	0.087	0.9 - 0.8 ^a -0.3 ^b	0.002
Br	⁸² Br	35.3	0.24	0.22			4.2 - 5.6	0.009
Au	¹⁹⁸ Au	64.8			0.109	0.109	0	

^aWhatman filter Analysis. ^bTest of absolute A.A. subroutine. ^cOnly for conditions of 10-min. irradiation and 3-4 hr. decay.

TABLE II

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SENSITIVITIES FOR PHOTON-ACTIVATION ANALYSIS AT THE ORELA^a

Element (Target)	Reaction	Product (T _{1/2})	Gamma-Ray Measured (MeV)	Sensitivity ^b (cpm/ μ g)	Absolute Specific Activity (dpm/ μ g)
O(PbO)	γ, n	¹⁵ O(2.1m)	0.511	1,120	4,750
C	γ, n	¹¹ C(20.5m)	0.511	2,600 ^c	
F(NaBF ₃ OH)	γ, n	¹⁸ F(110m)	0.511	5,900	25,930
Mg	$\gamma, p; \gamma, pn$	²⁴ Na(15.0h)	1.369 2.754	7.05 3.37	151
P(red)	γ, n	³⁰ P(2.5m)	0.511	1,940	8,250
	$\gamma, 2p$	²⁹ Al(6.6m)	1.28	6.43	137
	$\gamma, 2pn$	²⁸ Al(2.3m)	1.780	6.47	171
Ni	γ, n	⁵⁷ Ni(36.0h)	0.511 1.37 1.89 + sum	48.8 18.8 1.87	452
Cd	γ, n	¹⁰⁵ Cd(55.0m)	0.511	37.9	161
	γ, n	^{111m} Cd(48.6m)	0.150 + 0.247	465	1,720
	γ, n	¹¹⁵ Cd(53.5h)	0.49 + 0.53	5.37	
	daughter	^{115m} In(equil.)	0.335	9.19	119
	γ, p	¹¹² Ag(3.1h)	0.617	2.74	68.2
Hg(HgS)	γ, n	¹⁹⁷ Hg(65h)	0.191	100 ^c	
Pb(PbO)	γ, γ'	^{204m} Pb(67m)	0.912 + 0.899	4.87	37.5
	γ, n	²⁰³ Pb(52h)	0.279 + 0.401	127	777

^aEnd of bombardment (one T_{1/2}, maximum 2h) at 30kW power.^bCounted on top of 3 in. X 3 in. NaI (TI) through 1.24 cm lucite absorber.^cApproximate determination.

TABLE III
EXAMPLES OF CARBON ANALYSES
AT THE ORELA FACILITY

Sample	Carbon Content %
Ta Crystal No. 1	0.0134
Ta Crystal No. 2	0.0296
Bottom Ash (Allen Steam Plant)	1.26

FIGURE I

ORNL DWG 70-1260

LEGEND

- ① REACTOR VESSEL
- ② GRATING
- ③ PERMANENT BERYLLIUM
- ④ IN REACTOR STATION "Z"
- ⑤ HOLE V-7 IN PERMANENT BERYLLIUM
- ⑥ FLOW RESTRICTOR
- ⑦ ACCESS HOLE VH-4
- ⑧ INTERMEDIATE HOLD-UP STATION "Y"
- ⑨ FLIGHT TUBE
- ⑩ PENETRATION NO. 116
- ⑪ OVERHEAD SHIELDING
- ⑫ LOADING STATION "X"
- ⑬ SAFETY VALVE ASSEMBLY
- ⑭ HOT CELL
- ⑮ EXPERIMENTAL AREA ELEV. 833'-0"

FIGURE II

ORNL-DWG. 69-128

Top View

Side View

ALUMINUM SPHERE

SPINNING CHAMBER