Experimental Sensitivity Data for Radioactivation Analysis with 14 MeV. Neutrons

By Yuzuru KUSAKA, Haruo TSUJI and Takayoshi ADACHI

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The recent improvements, in such accelerators as the Cockcroft-Walton or Van de Graaff type in the shape, the operating methods, the available neutron fluxes and the cost of neutron generators, make them particularly useful as the neutron source in radioactivation analysis.[1-3]

In such machines 14 MeV. neutrons are produced by the $^{3}H(d, n)^{4}He$ nuclear reaction, using deuterons accelerated at 100 to 150 kV. The neutrons so produced can participate in (n, p), (n, α), (n, n) and (n, 2n) reactions for various nuclides. As the radioactivities produced by the reaction are generally of short half-lives, fast radioactivation is made possible for many elements.

Several concise summaries of theoretical sensitivity values for various elements activated by 14 MeV. neutrons have been published.[4,5] Although these data are very useful for a rough estimation of the sensitivities of each element, they are only approximate, mainly because of the inaccuracy of the present cross section value in the 14 MeV. neutron reaction.

This paper will describe experimental results concerned with the sensitivity, results which have been obtained by an experimental method with practical applications as an industrial routine analysis; it will also discuss radioactivity which has not been described in the sensitivity data so far published.

Experimental

Neutron Source. — A thick tritiated zirconium target (3 curies; The Radiochemical Centre, Amersham, Buckinghamshire, England) was bombarded with a deuteron beam of 100 to 170 KeV. and of 5 to 10 μamp. by the Cockcroft-Walton type accelerator of Konan University. The neutron strength during the irradiation was continuously measured by a proportional counter with a cesium iodide scintillator which had been set in the accelerator tube to count the associated alpha particles from the T-D reaction. The absolute neutron flux at the sample position was determined by the activation method of standard copper foil, the beta radioactivity of which was measured by a 4π gas-flow counter using Q gas.

Sample Transfer System and Neutron Irradiation.—A polyethylene "bunny" capsule (1.3 cm. i. d., 4.6 cm. long, 0.1 cm. thick) containing the sample was transferred, by means of a vacuum cleaner (500 watt), between the irradiation position, which was set to come to the sample center 3.5 cm. below the target, and the radioactivity detector through a flexible vinyl tubing (1.8 cm. i. d., 10 m. long). Irradiation was started by turning on the accelerator beam switch, and it was timed with a stop watch. At the end of 10.0 min. irradiation, the transfer system was turned on to bring the "bunny" capsule back to the well of the scintillation detector. The average travel time of the capsule from the irradiation position to the detector was 3.5 sec.

Radioactivity Measurement.—The radioactivity induced in the sample was measured by a shielded well-type sodium iodide-scintillation detector (Harshow, 4.45 cm. dia., 5.08 cm. height, well: 1.91 cm. dia., 3.81 cm. deep) optically contacted with a photomultiplier tube (RCA 6342-A). Pulses from the phototube were fed to a linear-amplifier and then to a single-channel pulse height analyzer, the output from which was fed to a scaler.

The gross gamma ray counting was done for 10.0 min. in the sensitivity determination. In another run using the same sample, the decay curve was traced for an appropriate length of time and, consequently, the produced radioactive nuclide was analyzed. In the runs the countings were started after only 5.0 sec. of irradiation. However, the oxide, carbonate, acetate and nitrate samples were started after 40 sec., it being necessary first to eliminate the influence of the ^{16}N produced by the (n, p) reaction on oxygen.

Using the mean value of neutron flux observed during the irradiation, all the net counts, which were obtained by subtracting the counts in the empty capsule from the counts with the sample, were normalized to the data in a standard neutron flux (1.0×10^{5} neutrons per cm^{2} per sec.) and were also corrected for the variations in the weight of each sample.

Irradiation Sample.—A definite amount of the sample was pressed into a pellet (1.2 cm. dia., less than 3.4 cm. long) and put in the bottom of the "bunny" capsule.

The chemical forms of the samples were as

1) E. A. Burrill and M. H. MacGregor, *Nucleonics*, **18**, No. 12, 64 (1960).
2) Y. Kusaka, *Isotopes and Radiation* (*Doitai to Hoshasen*), **4**, 79 (1961).
3) W. W. Meinke and R. W. Shideler, *Nucleonics*, **20**, No, 3, 60 (1962).
4) R. F. Coleman, *Analyst*, **86**, 39 (1961).
5) A. S. Gillespie, Jr., and W. W. Hill, *Nucleonics*, **19**, No. 11, 170 (1961).

follows: the elements of silver, aluminum, arsenic, bismuth, calcium, cerium, cobalt, chromium, copper, iron, indium, iridium, lithium, magnesium, manganese, molybdenum, nickel, phosphorus, platinum, rhodium, ruthenium, sulfur, antimony, selenium, silicon, tin, tantalum, tellurium, thallium, tungsten, zinc and zirconium; the oxides of dysprosium, gallium, germanium, mercury, lanthanum, niobium, lead, praseodymium, samarium, titanium, vanadium and yttrium; the carbonates of cesium, potassium, sodium and rubidium; the acetates of barium and strontium; fluorine as polytetrafluoroethylene; bromine as dibromobenzene; iodine as methyl iodide; nitrogen as ammonium nitrate and oxygen as oxalic acid.

Results

Radioactive Nuclide Produced by Fast Neutron Reaction. — The nuclides so produced were analyzed by the decay curves obtained in the radioactivity measurements. The quantity expressed with the counting rate of the nuclide at the end of irradiation has been plotted against the half-life of the radioactivity; it is shown in Fig. 1. In Fig. 1 the nuclide is marked with the parent isotope in the nuclear reaction, while the waved line shown under the reaction mark indicates the activation to the metastable state of an isomeric pair. The nuclear reactions shown with the black point, that is, in cerium, germanium, gold, rubidium and selenium, have not been described in previously-published sensitivity data[4,5] but, nevertheless, produce considerable amounts of radioactivity.

In Fig. 1 the ambiguous weak radioactivities of less than 100 counts per min. are not plotted.

In the other experiments, the sensitivity for the activation analysis has been estimated. In these cases the sensitivity was assumed to be the quantity of the element needed to give 500 counts in a 10-min. measurement. However, in oxygen and gold only 100 counts was adopted in the 1 min. measurement; this was because of their short-lived radioactivities. The results are shown in Table I.

TABLE I. SENSITIVITY FOR RADIOACTIVATION ANALYSIS

Element	Sensitivity mg.	Element	Sensitivity mg.
Br	18.5	Zn	260
Cu	21.2	Mn	290
Pr	24.6	O	352
Si	28.9	Zr	390
Rb	59.5	N	435
Ga	63.9	Na	490
P	65.1	Mo	497
Sb	67.4	Cd	503
Ba	76.1	Cl	505
Ag	81.6	Mg	537
Al	86.1	Ge	540
Cr	170	Sr	625
F	181	Au	685
Ce	196	Hg	690
Se	200	K	890
V	250		

More than 1 g.: As, Bi, Ca, Co, Cs, Fe, Hf, I, Ir, La, Li, Nb, Ni, Pb, Pt, Ru, S, Sm, Sn, Ta, Te, Ti, Tl, W, Y

A Consideration of Experimental Error.—The experimental error in the results described above was caused mainly by the fluctuations of the neutron flux on the sample during irradiation. A typical example of the fluctuation curve is shown in Fig. 2. In the adopted

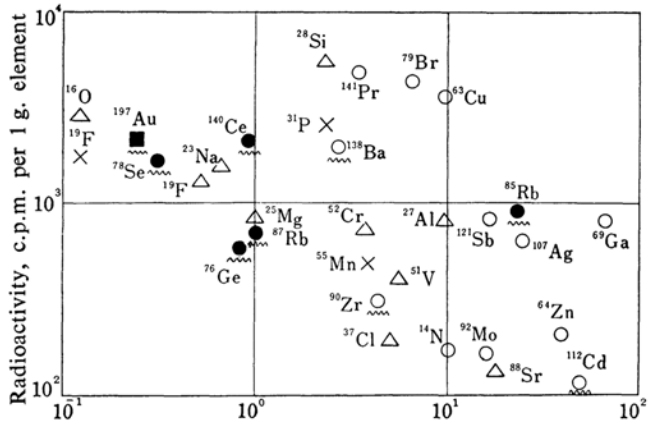

Fig. 1. Radioactive nuclide produced by 14 MeV. neutron reaction.
○: (n, 2n) reaction △: (n, p) reaction
×: (n, α) reaction □: (n, n′) reaction

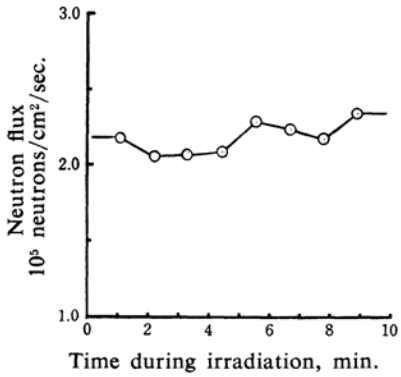

Fig. 2. Fluctuation of neutron flux during irradiation.

neutron flux itself, which was the mean value of the plots in Fig. 2, an uncertainty of 2.9% is estimated. In all the runs, an approximate 3.5% error could be assumed.

However, when relatively short-lived nuclides were to be measured, the neutron flux fluctuations near the end of irradiation become important and, therefore, the results tend to be less reliable. Although the theoretical error estimations in such nuclides are very difficult, roughly a 10% error was inferred from the experiments.

The neutron flux variation in the sample set at the irradiation position gave about a 5% error as a maximum. However, after the flux distribution had been determined by the activation method of copper monitor foils set at various positions in a polyethylene " bunny " capsule during irradiation, the corrections for sample position (sample thickness) were made in all the experimental results and, therefore, the error based on sample position became negligibly small.

Discussion

The short-lived radioactivities produced in germanium, selenium, rubidium and cerium are probably produced by (n, 2n) reactions; that is, they are due to Ge-75 m (48 sec.), Se-77 m (19 sec.), Rb-84 m (23 min.), Rb-86 m (1.02 m) and Ce-139 m (55 sec.). The radioactivity produced in gold might be due to the Au-197 m (7.2 sec.) produced by the (n, n') reaction.

It is possible improve the sensitivity by using a more sensitive gamma ray detector or higher neutron fluxes. Even in these cases, however, the relative sensitivity values are not expected to differ greatly from the data reported in the present paper. It is, therefore, believed that the present data will be useful in a general consideration of radioactivation analysis by 14 MeV. neutrons.

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Department of Chemistry
Faculty of Science
Konan University
Higashinada-ku, Kobe