The Sensitivity of Radioactivation Analysis by a Low Level Neutron Source

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General discussions of the sensitivities attainable with the thermal neutron activation analysis of various elements have been undertaken in several reports.[1-7] Most of their sensitivity estimations have, however, been made for reactor irradiation, based on theoretical computation and expressed with the radioactivities at the end of the irradiation. Although these data are very useful for rough estimations of the sensitivities of each element, even in a low-level neutron flux, they often do not coincide exactly with the experimental values because of the decay effect in the short-lived nuclides to be measured, because of the differences in the kind of detector used, and so on. Moreover, in some reports long-time irradiations such as those for one week or even one month have been adopted. These durations are too long for routine analysis.

The use of a low level neutron source is suited to a " non-destructive " assay of a macro amount of elements possessing high-neutron cross section. In such cases, gamma ray measurement, for example, the use of a well-type sodium iodide scintillator, is a useful counting method. Moreover, the induced radioactivity, in general, is so weak that the counting period must be longer than in the case of a high neutron flux.

In such practical work using a low-level neutron source, therefore, the sensitivity values previously reported must be re-evaluated experimentally. This paper will describe some experimental findings on the sensitivities obtained by irradiation and measurement procedures which are designed to be suitable for a routine analysis of all the elements under consideration. The results of thermal neutron flux determination are also included.

1) G. W. Leddicotte and S. A. Reynolds, U. S. Atomic Energy Comm., AECD-3489 (1953).
2) E. N. Jenkins and A. A. Smales, *Quart. Rev.* **10**, 83 (1956).
3) H. Hamaguchi and H. Natsume, "Jikken Kagaku Koza," Ed. by Chem. Soc. Japan, Vol. 12, Maruzen Company Ltd., Tokyo (1956), p. 447.
4) U. Schindewolf, *Angew. Chem.*, **70**, No. 7, 181 (1958).
5) W. W. Meinke, *Anal. Chem.*, **31**, 792 (1959).
6) M. Okada, *J. Chem. Soc. Japan, Pure Chem. Sec. (Nippon Kagaku Zasshi)*, **80**, 1 (1959).
7) O. U. Anders, *Nucleonics*, **18**, No. 11, 178 (1960).

Experimental

Neutron Source. — The source consisted of a mixture of 84.2 mg. radium bromide (49.3 mg. as a radium element) with beryllium, which was sealed in a platinum tube (length; 40 mm.; diameter, 5 mm.; thickness, 0.5 mm.) with a polyethylene outer tube (length, 85 mm.; diameter, 16 mm.).

Neutron Irradiation.—The geometry of the paraffin moderator block used in this work was the same as that in Ref. 8. The irradiation periods for all the samples under consideration were 10.0 min., 1.0 hr. and 17.0 hr.

Radioactivity Measurement. — The radioactivity induced in the sample was measured by a well-type sodium iodide scintillation detector (Harshaw, 4.45 cm.ϕ×5.08 cm. h; hole, 1.91 cm.ϕ, 3.81 cm. deep) with a No. 6097F E.M.I. photomultiplier tube. After a 20.0 sec. interval after the end of irradiation, the measurement was started and the gross gamma ray counting was done for 3.0 min., 10.0 min. and 30.0 min.

The measurement of the absolute disintegration rates for the determination of thermal neutron flux was made with a 4π gas flow counter (Kobe Kogyo GM-917) using Q gas.

Irradiation Sample.—A definite amount of the sample was put in a polyethylene capsule (inner diameter, 16 mm.; length, 86 mm.; thickness, 1 mm.) and irradiated. The chemical forms of the samples were as follows: silver, aluminum, gold, bismuth, calcium, cadmium, cerium, cobalt, chromium, copper, iron, germanium, hafnium, iodine, indium, iridium, lithium, magnesium, manganese, molybdenum, nickel, phosphorus, platinum, rhodium, ruthenium, sulfur, antimony, selenium, silicon, tin, tantalum, tellurium, thallium, tungsten, zinc and zirconium as the elements; arsenic, dysprosium, europium, gallium, mercury, lanthanum, niobium, lead, praseodymium, samarium, titanium, vanadium and yttrium as the oxides, cesium, potassium, sodium and rubidium as the carbonates; barium and strontium as the acetates; bromine as the dibromobenzene; chlorine as the hydrogen chloride; fluorine as the calcium fluoride; nitrogen as the ammonium nitrate, and oxygen as the water. All the samples were chemically pure.

Results

Thermal Neutron Flux Determination.—Using indium and gold foils 1.3 cm. in diameter and

8) H. Tsuji, Y. Kusaka and Y. Namikawa, This Bulletin, **35**, 2045 (1962).

of various thicknesses as the neutron monitors, the thermal neutron flux in the inside bottom of the polyethylene sample capsule in the paraffin block was determined.

For the determination of the absolute disintegration rates, a conventional 4π counting method[9] was applied. In the observed counting rates, some decreasing effects based on a self-absorption effect for soft beta rays and a self-shielding effect for thermal neutrons were recognized in the relatively thick foils. To correct them, an extrapolation to zero thickness was applied, and the extrapolated value was used in the calculation of the neutron flux. The extrapolations in the case of the indium and gold foils are shown in Figs. 1 and 2.

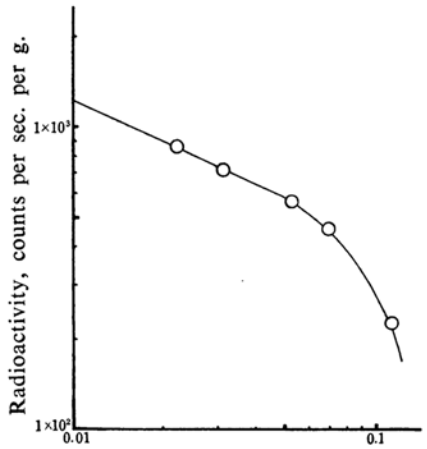

Fig. 1. 4π counting of gold foil.
(Saturation factor in irradiation is 1.0.)

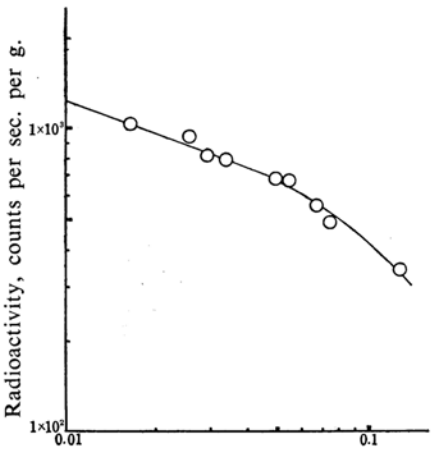

Fig. 2. 4π counting of indium foil.
(Saturation factor in irradiation is 0.5.)

9) G. Friedlander and J. W. Kennedy, "Nuclear and Radiochemistry," John Wiley & Sons, Inc., New York (1955), p. 293.

In obtaining the results, the thermal neutron fluxes were calculated as 4.1×10^3 neutrons per cm^2 per sec. in the indium foil experiment and 4.8×10^3 in the gold foil one. On the basis of these data, 4.5×10^3 was adopted as the probable neutron flux.

The Radioactivation Degree of the Elements. — All the experimental results are shown in Figs. 3, 4 and 5, in the lump. In these figures, the counting rates obtained by 3.0 min., 10.0 min. and 30.0 min. measurments for each element are plotted against the half-life of the induced radioactivity. However, when plural radioactive nuclides are produced in an element, the half-life is represented with that of the radioactivity, which is strongest at the end of the irradiation. No ambiguous radioactivities of less than 100 counts in the measurments are plotted in the figures.

TABLE I. DETECTION LIMIT IN 10 min. IRRADIATION

Detection limit	Element
1～ 10 mg.	In, Dy, Rh, V
10～ 100 mg.	Ag, Eu, Mn, I, Br, Co
100～1000 mg.	Cu, Al, Hf, Ir, Ga, Au, Sm, Cl, Si, Se, As, W
More than 1 g.	H, Li, Be, B, C, N, O, F, Na, Mg, P, S, K, Ca, Ti, Cr, Fe, Ni, Zn, Ge, Rb, Sr, Y, Zr, Nb, Mo, Ru, Cd, Sn, Sb, Te, Cs, Ba, La, Ce, Pr, Ta, Pt, Hg, Tl, Pb, Bi

TABLE II. DETECTION LIMIT IN 1 hr. IRRADIATION

Detection limit	Element
0.1～ 10 mg.	In, Dy, Eu, Rh, Mn, V
10～ 100 mg.	Ag, I, Br, Co, Ir, Au, Sm, Ga, Cu
100～1000 mg.	As, Al, Hf, W, Na, Cl, Pr, La, Cs, Te, Mo, Se, Rb, Ba, Sr, Si, Sn, Sb, Pt, Cd
More than 1 g.	H, Li, Be, B, C, O, F, Mg, P, S, K, Ca, Ti, Cr, Fe, Ni, Zn, Ge, Y, Zr, Nb, Ru, Ce, Ta, Hg, Tl, Pb, Bi

TABLE III. DETECTION LIMIT IN 17 hr. IRRADIATION

Detection limit	Element
0.1～ 1 mg.	In, Eu, Dy
1～ 10 mg.	Mn, Ir, Au, Rh, Br, V, La, I
10～ 100 mg.	As, Ga, Ag, W, Pr, Cu, Na, Co, Cs, Sm, Sb
100～1000 mg.	Rb, Al, Hf, Cl, Ru, Y, Ba, Sr, Mo, K, Te, Hg, Si, Se, Ge, Cd, Pt, Ta, P, Zn, Mg, Sn
More than 1 g.	H, Li, Be, B, C, N, O, F, S, Ca, Ti, Cr, Fe, Ni, Zr, Nb, Ce, Tl, Pb, Bi

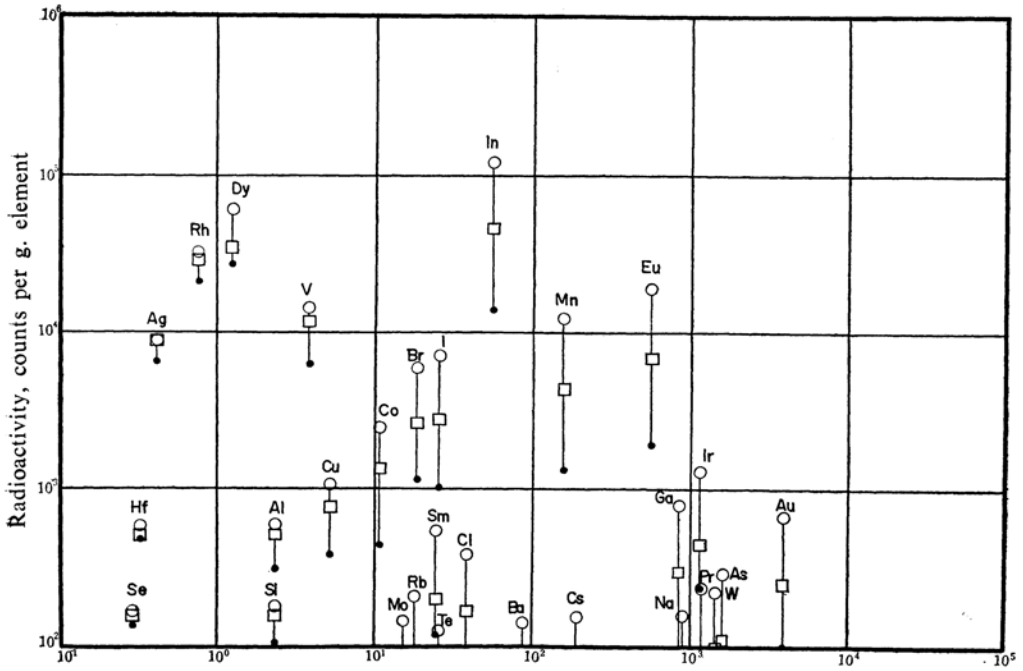

Fig. 3. Radioactivation degree in 10 min. irradiation.
• 3 min. measurement, □ 10 min. measurement, ○ 30 min. measurement

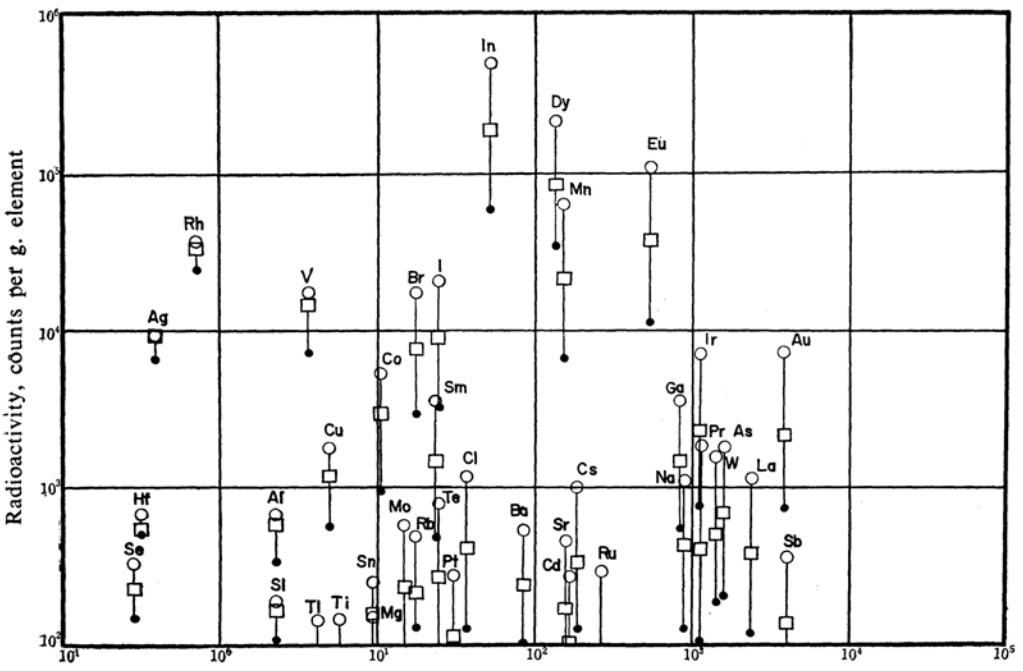

Fig. 4. Radioactivation degree in 1 hr. irradiation.
• 3 min. measurment, □ 10 min. measurment, ○ 30 min. measurement

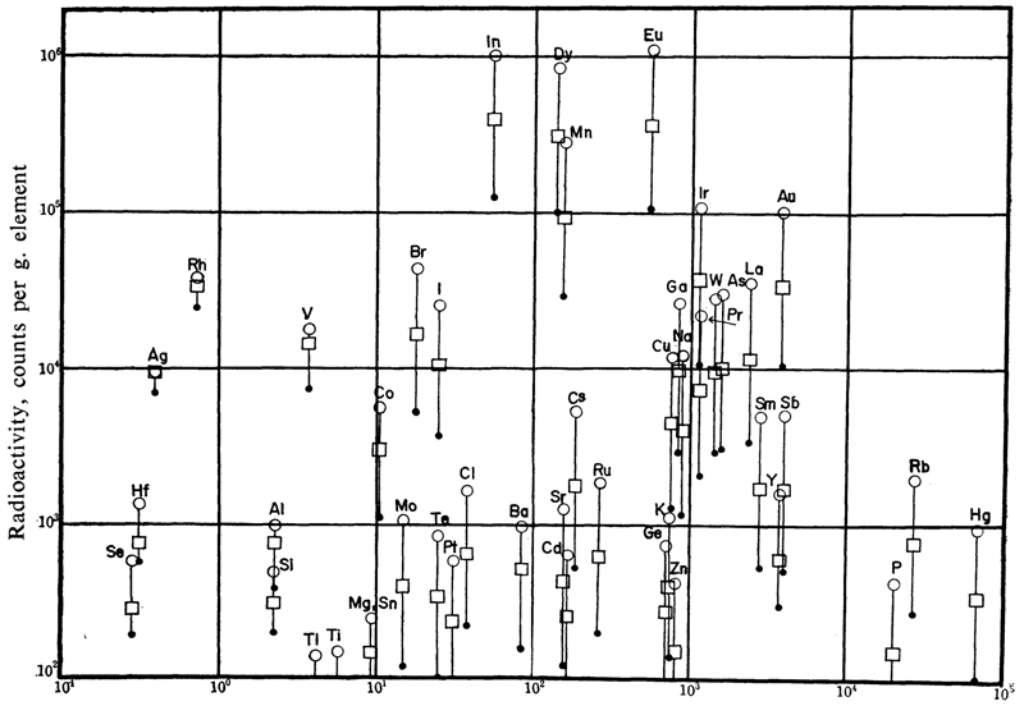

Fig. 5. Radioactivation degree in 17 hr. irradiation.
• 3 min. measurement, □ 10 min. measurement, ○ 30 min. measurement

If the detection limit in the radioactivation analysis is assumed to be the number of grams of an element required to give about 100 counts in 10 min., the detection limits summarized in Tables I, II and III can be obtained. In the tables, the elements listed in each group of detection limits are arranged in the order of decreasing sensitivity, except for "more than 1 g."

Discussion

Because of the sodium iodide scintillation-counting method, such pure beta emitters as Tl-206 and such soft gamma emitters with a high internal conversion efficiency as Co-60m and Ir-192m were measured with a low counting efficiency. For such elements a beta-ray counting method using a Geiger counter would be more suitable.

The self-shielding effect for thermal neutrons, although important in certain cases, was not considered in the treatment of the sensitivity data.

Furthermore, in a practical application, one must remember that an appropriate cooling time after the irradiation allows one to discriminate the nuclides with different half-lives; gamma ray spectrometry is also useful for the discrimination of each species.

It is possible to improve the sensitivities by using more sensitive gamma ray detectors or higher neutron fluxes. However, even then the relative sensitivity value should not change from the data reported here. Therefore, this data should be useful in general radioactivation analysis using a low-level neutron source.

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