Gamma-ray Spectrometry in Radioactivation Analysis with 14 MeV. Neutrons

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(Received July 25, 1964)

A commercial small-type neutron generator producing 14 MeV. neutrons is becoming popular as a neutron source for the radioactivation analysis of various elements.[1-4] For the practical application of such a neutron generator, however, it is important to determine the analytical sensitivities of various elements under definite experimental conditions. Kusaka et al. reported some experimental results on analytical sensitivities obtained by a gross gamma-ray counting method for the induced radioactivities.[5]

This paper will describe the experimental results concerning the sensitivity obtained by gamma ray spectrometry and will show the gamma ray spectra of 37 elements. All the data are normalized to definite experimental conditions so that they can be compared with each other.

Experimental

Neutron Source. — Monochromatic 14 MeV. neutrons were produced by T-d reaction in a Toshiba Neutron Generator, Model NT-20, in which a thin tritiated titanium target (4 curies; Isotope Div., Harwell, England) was bombarded with a deuteron beam of about 100 μamp. and 150 to 200 KeV. A target assembly with a sample station for neutron irradiation, similar to that described by Fujii et al.,[3] was used.

The neutron output during the irradiation of a sample was kept constant as far as possible by regulating the accelerating or extraction voltage in the generator. The output was continuously recorded by a plastic neutron scintillator which was set at a definite position below the drift tube of the generator and which was then connected to a counting-rate meter set near the operator. The absolute value of the neutron flux at the irradiation position was determined by the method of activating copper foil, the beta activity of which was measured by a 4π gas-flow counter using Q gas.

The Sample Transfer System and Neutron Irradiation. — Samples were packed tightly in cylindrical polyethylene containers (4.5 cm. long, 1.3 cm. i. d., 0.1 cm. thick). These "bunny" capsules were then transferred through a pneumatic tube system, operated by a vacuum cleaner blower (500 W.), between the irradiation position and the radioactivity detector. At the end of 10.0 min. of irradiation, the sample transfer system was turned on to bring the sample capsule back to the well of the scintillation detector. The average travel time of the capsule was 3.0 sec.

Radioactivity Measurement. — The radioactivity of the sample was measured by a shielded well-type sodium iodide scintillation detector (Harshow; 4.45

1) W. W. Meinke and R. W. Shideler, *Nucleonics*, **20**, No. 3, 60 (1962).
2) I. Fujii, H. Muto, K. Ogawa and A. Tani, *J. Atomic Energy Soc. Japan*, **5**, 455 (1963).
3) I. Fujii, H. Muto and K. Miyoshi, *Japan Analyst*, **13**, 249 (1964).
4) J. Wing, *Anal. Chem.*, **36**, 559 (1964).
5) Y. Kusaka, H. Tsuji and T. Adachi, This Bulletin, **36**, 1259 (1963).

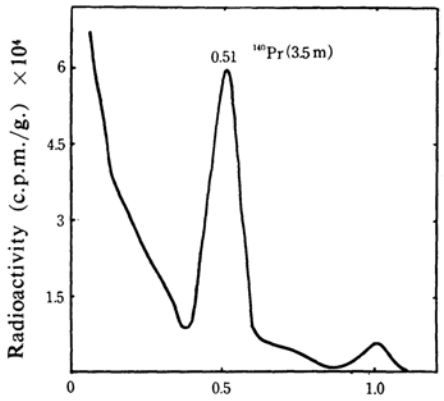


Fig. 1. Praseodymium.

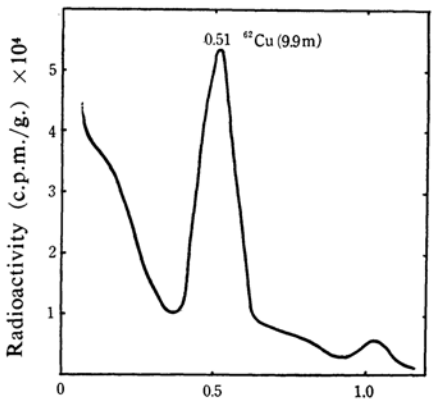


Fig. 2. Copper.

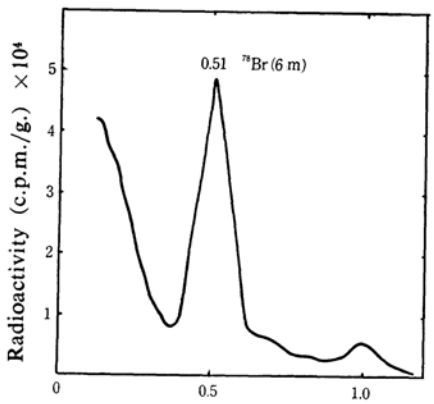


Fig. 3. Bromine.

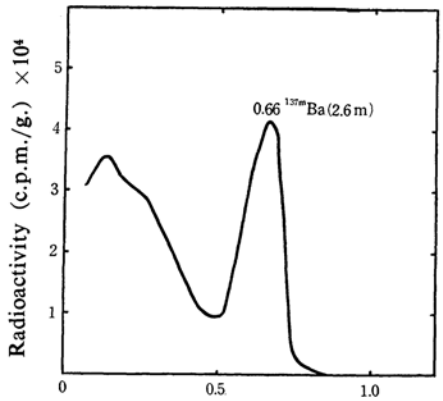


Fig. 4. Barium.

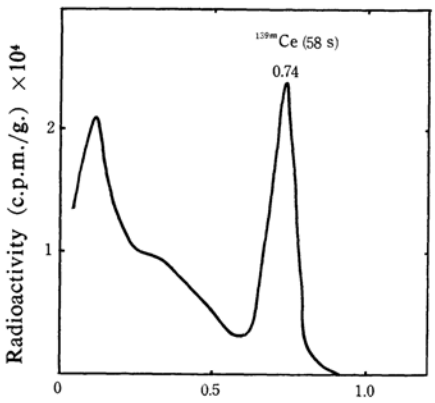


Fig. 5. Cerium.

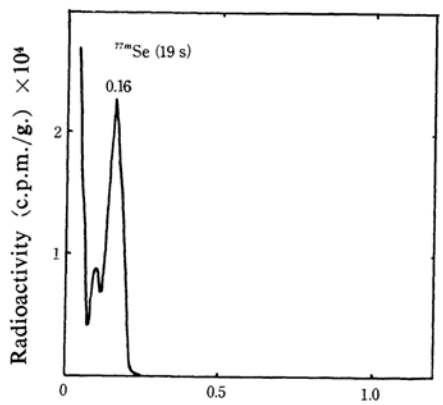


Fig. 6. Selenium.

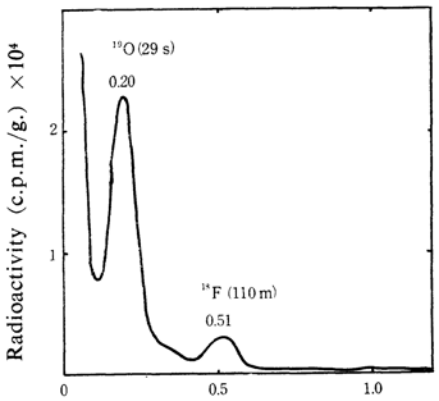


Fig. 7. Fluorine.

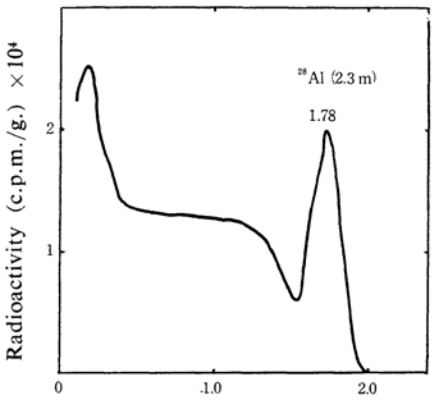


Fig. 8. Silicon.

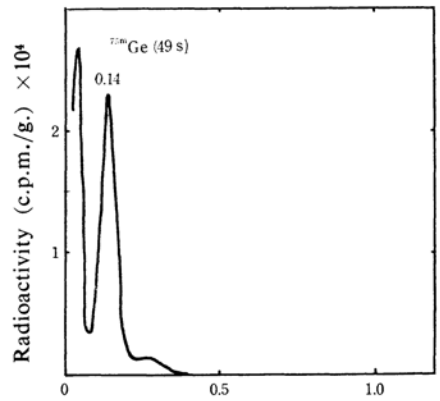


Fig. 9. Germanium.

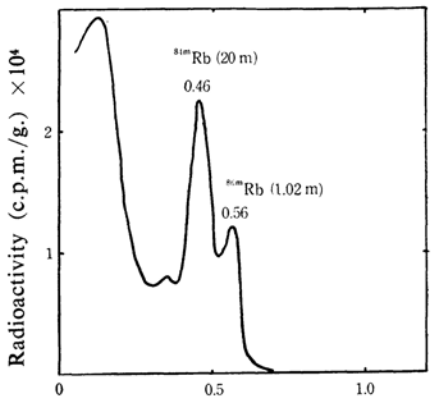


Fig. 10. Rubidium.

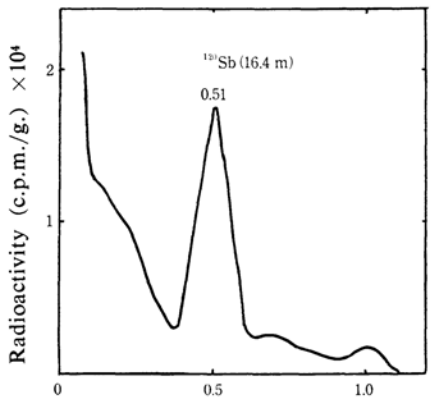


Fig. 11. Antimony.

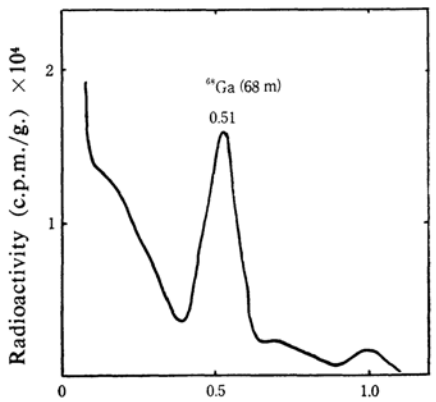


Fig. 12. Gallium.

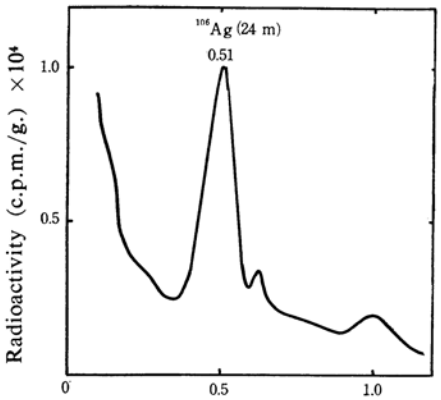


Fig. 13. Silver.

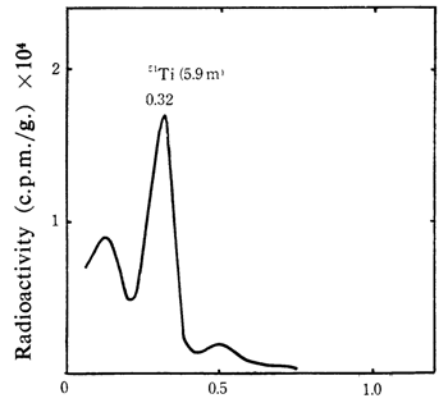


Fig. 16. Vanadium.

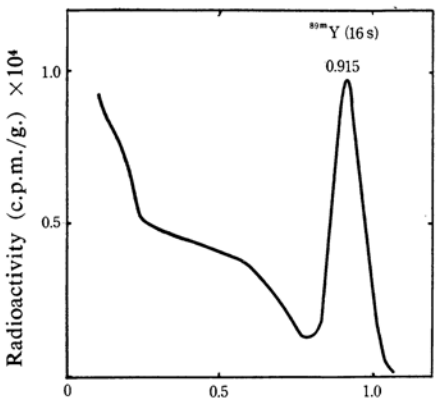


Fig. 14. Yttrium.

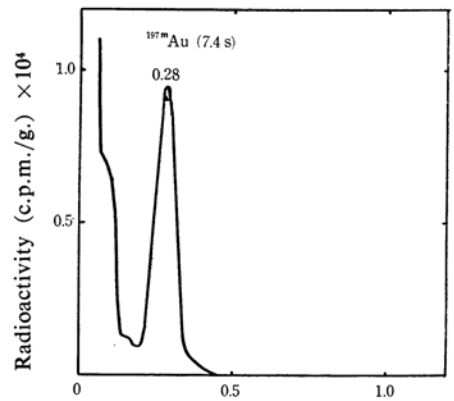


Fig. 17. Gold.

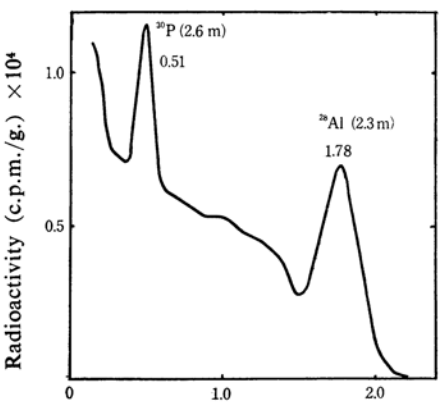


Fig. 15. Phosphorus.

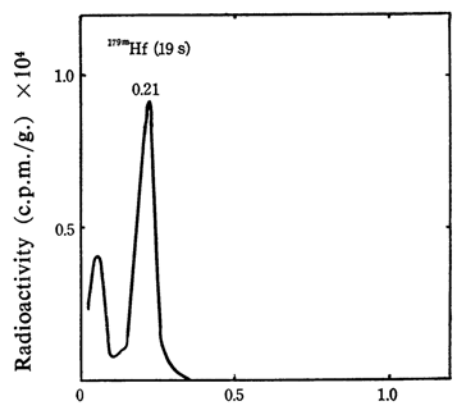


Fig. 18. Hafnium.

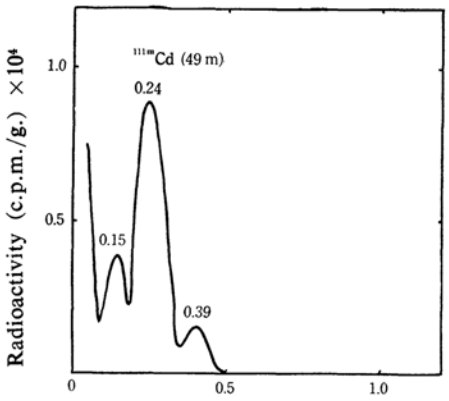


Fig. 19. Cadmium.

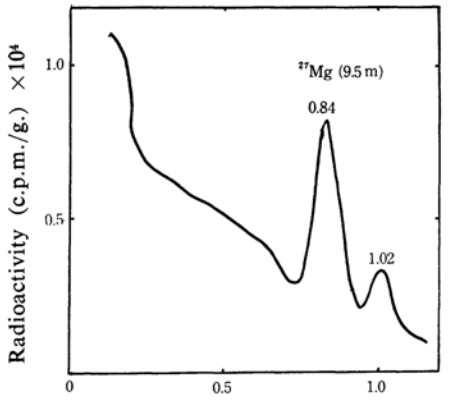


Fig. 20. Aluminum.

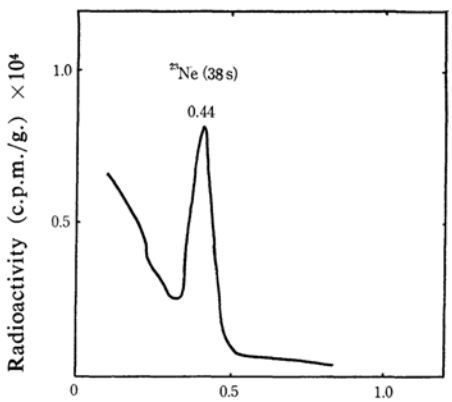


Fig. 21. Sodium.

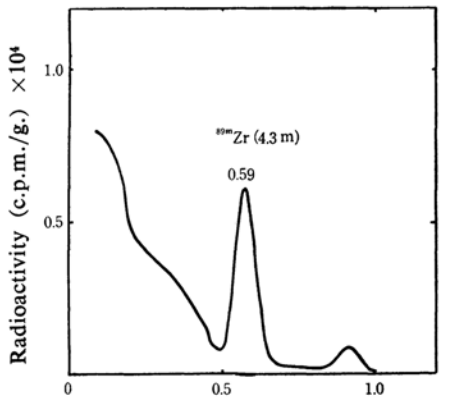


Fig. 22. Zirconium.

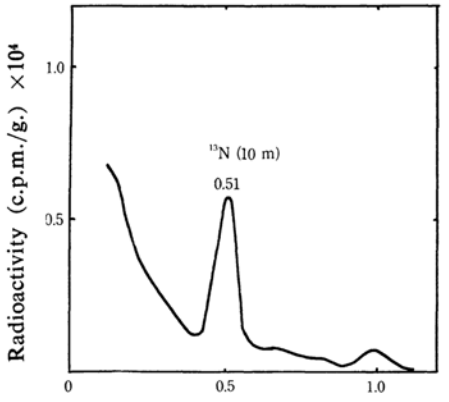


Fig. 23. Nitrogen.

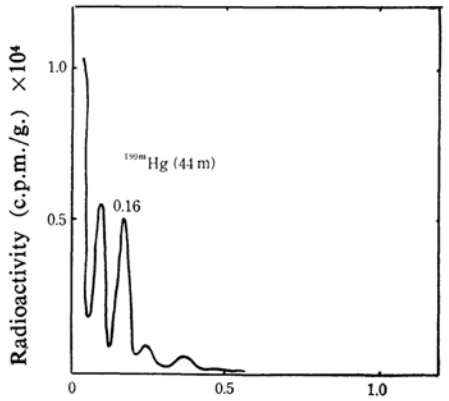


Fig. 24. Mercury

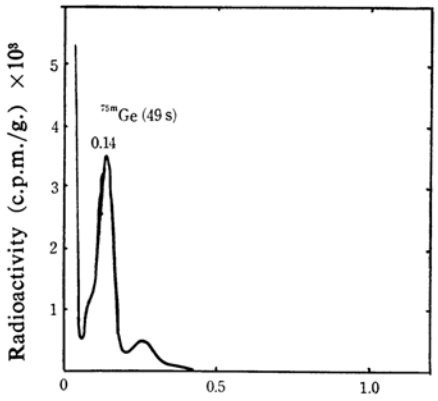


Fig. 25. Arsenic.

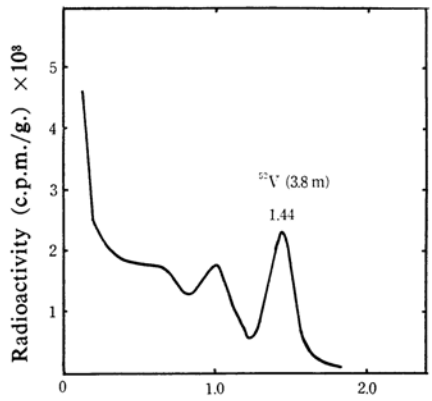


Fig. 26. Chromium.

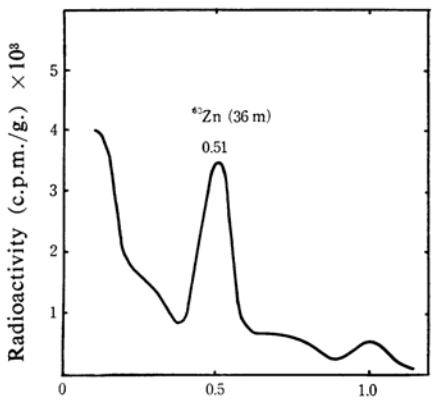


Fig. 27. Zinc.

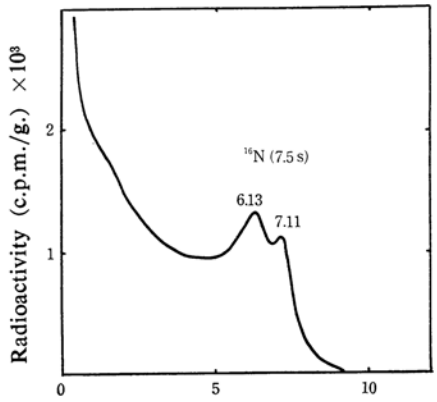


Fig. 28. Oxygen.

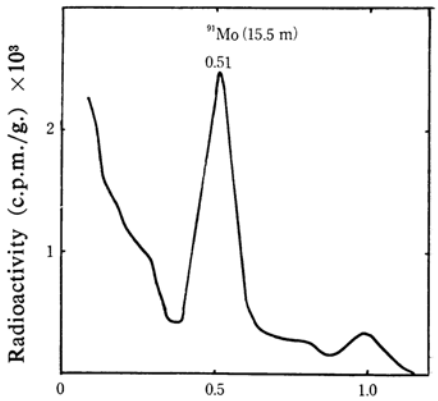


Fig. 29. Molybdenum.

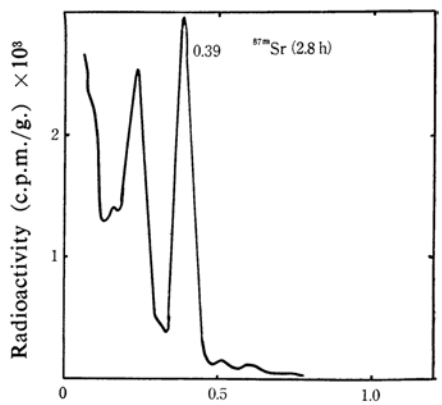
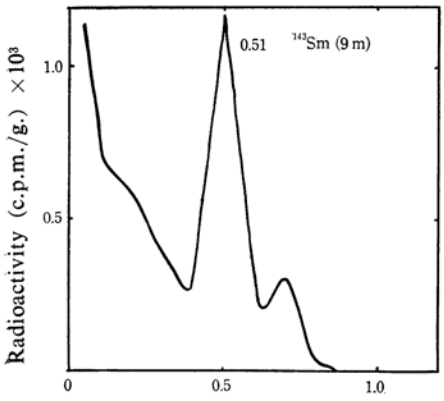
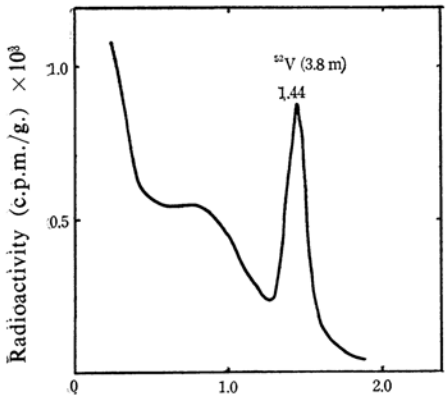


Fig. 30. Strontium.



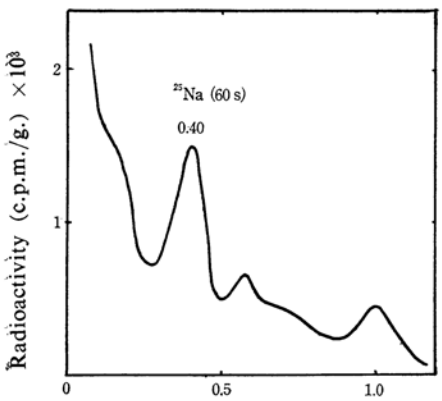
Energy, MeV.

Fig. 31. Samarium.



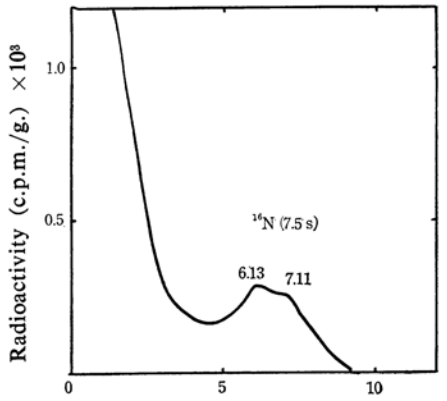
Energy, MeV.

Fig. 32. Manganese.



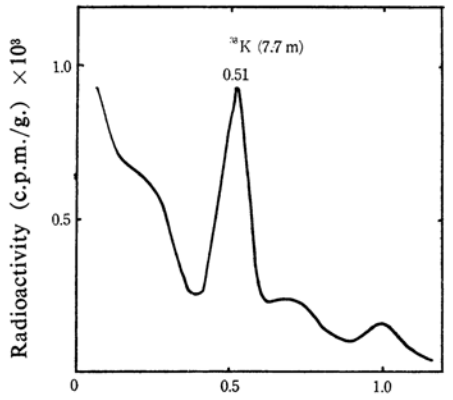
Energy, MeV.

Fig. 33. Magnesium.



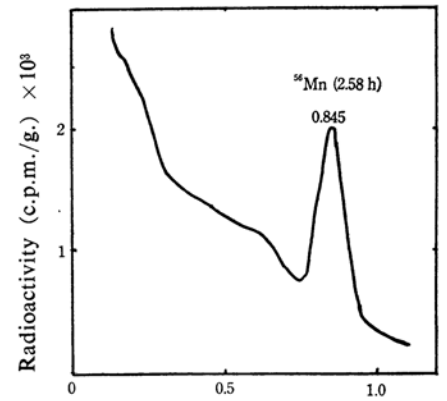
Energy, MeV.

Fig. 34. Fluorine.



Energy, MeV.

Fig. 35. Potassium.



Energy, MeV.

Fig. 36. Iron.

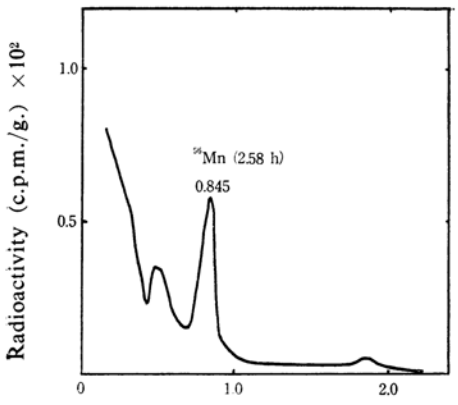


Fig. 37. Cobalt.

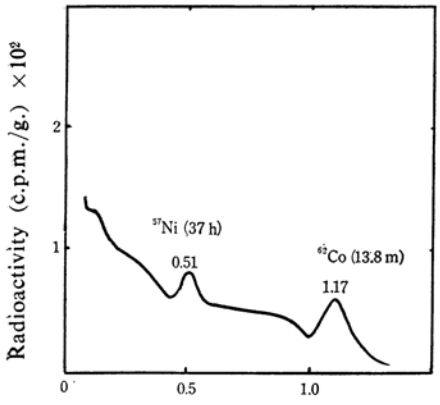


Fig. 38. Nickel.

cm. diameter, 5.08 cm. height, well: 1.91 cm. diameter, 3.81 cm. deep) mounted on a photomultiplier tube (Toshiba-7696) and connected in turn to a Toshiba 128 channel pulse-height analyzer. The resolution of the detector for 0.662 MeV. ^{137}Cs gammas was about 12%.

The countings were started 5.0 sec. after irradiation and were stored for 1.0 min. However, the samples containing oxygen were started after 1.0 min., it being necessary 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, the counting data obtained were normalized to a standard neutron flux of 1.0×10^7 neutrons per cm^2 per sec.

Irradiation Sample.—Inside the "bunny" capsule definite amounts of the sample powder mixed with pure carbon powder were packed in a definite volume (1.3 cm. diameter, 2.0 cm. long).

The chemical forms of the samples were as follows: the elements of silver, aluminum, gold, calcium, cadmium, cobalt, chromium, copper, iron, germanium, hafnium, indium, lead, magnesium, manganese, molybdenum, nickel, phosphorus, sulfur, antimony, selenium, silicon, tantalum, tin, titanium, tungsten, zinc and zirconium; the oxides of arsenic, cerium, dysprosium, gallium, gadolinium, erbium, holmium, mercury, lanthanum, praseodymium, samarium, vanadium, yttrium and ytterbium; the carbonates of potassium, sodium and rubidium; the acetates of barium and strontium; fluorine as polytetrafluoroethylene; bromine as dibromobenzene; chlorine as benzene hexachloride; nitrogen as ammonium nitrate, and oxygen as oxalic acid.

Results

The Gamma-ray Spectra of the Radioactive Nuclides Produced by 14 MeV. Neutron Reaction.—The spectra obtained for each element were compared under normalized conditions; weight of sample: 1.0 g. as element; irradiation time: 10.0 min.; neutron flux: 1.0×10^7 neutrons per cm^2 per sec.; counting time: 1.0 min. from the end of irradiation. The results are shown in Figs. 1—38, in which the photopeaks available for the activation analysis are marked with the energies and the nuclides produced. The figures are arranged in the order of the magnitude of the photopeak area, that is, of the sensitivity of the activation analysis.

In the cases of sulfur, chlorine, calcium, titanium, indium, tin, lanthanum, gadolinium, dysprosium, holmium, erbium, ytterbium, and tantalum, no photopeaks usable for the effective activation analysis appeared in the experiments.

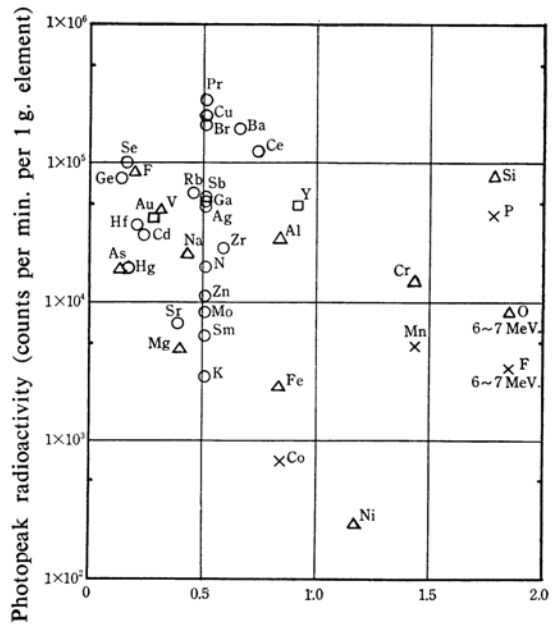


Fig. 39. Sensitivity of gamma-ray spectrometry in 14 MeV. neutron activation analysis.
○: $(n, 2n)$ reaction ×: (n, α) reaction
△: (n, p) reaction □: (n, n') reaction

The Sensitivity of Gamma-Ray Spectrometry. —The sensitivity of the analysis can be expressed with the photopeak radioactivity strength, which is obtained by drawing a straight line across the bottom of the peak in the spectrum and by summing up the counts above the line. The values for each element have been summarized in a graphic form and are shown in Fig. 39, in which the photopeak activity is marked with the target element and the kind of the nuclear reaction is plotted against the gamma-ray energy.

It may be possible to improve the sensitivities by using such things as more sensitive detectors, but these plots can give a general impression of the relative sensitivities in the 14 MeV. neutron activation analysis.

The authors wish to thank Dr. Kazunori Yuasa, Mr. Bunzaburo Saeki and Mr. Yoshinao Fujiwara of the Department of Physics, Konan University, for their kind and valuable advice through this study.

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