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THE 870.8-keV GAMMA RAY FROM PuO_2

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ABSTRACT

A ^{252}Cf neutron source and an ^{241}Am alpha source were used with isotopically enriched water containing 43.9% ^{17}O and 43.1% ^{18}O, to study the $(n,n'\gamma)$ and $(\alpha,\alpha'\gamma)$ reactions in ^{17}O and ^{18}O. The production yields for the 870.8-keV gamma ray from ^{17}O and the 1982.2-keV gamma ray from ^{18}O were measured. In addition, the average cross sections over the ^{252}Cf fission neutron spectrum for $^{17}O(n,n'\gamma)^{17}O$ and $^{18}O(n,n'\gamma)^{18}O$ were determined.

INTRODUCTION

In the nondestructive analysis of certain radioactive materials, an 870.8-keV line appearing in the emitted gamma-ray spectrum has been used to confirm the presence of the oxide state of the material based on the fact that the first excited state of ^{17}O is 870.8 keV.

For oxide materials with multiple indigenous radiation sources, such as PuO_2 with both neutrons and alpha particles as decay radiations, the reaction mechanisms leading to excitation of the ^{17}O first excited state are not obvious. On the other hand, in the natural oxide materials, the amount of ^{18}O (0.204%) is more than that of ^{17}O (0.038%); hence, the gamma-ray decay from the excited states of ^{18}O, such as the 1982.2-keV gamma ray from the first excited state, may also be useful for identification purposes.

Experiments to determine the relative contributions from $^{17}O(n,n'\gamma)^{17}O$, $^{17}O(\alpha,\alpha'\gamma)^{17}O$, and $^{18}O(n,n'\gamma)^{18}O$, $^{18}O(\alpha,\alpha'\gamma)^{18}O$ in such cases have been performed.

EXPERIMENTAL PROCEDURE

Two series of measurements were carried out with a neutron source and alpha source, respectively. In the first series of measurements, a ^{252}Cf neutron source (1.48×10^6 n/s) was used because the neutron spectrum of ^{252}Cf resembles closely that of plutonium.

The neutron source was placed at the center of two containers of water. One container held 100 cm^3 of natural water, the other held 100 cm^3 of water enriched in the isotopes ^{17}O and ^{18}O (43.9% ^{17}O and 43.1% ^{18}O).

A high-purity germanium detector with 11.7% efficiency and 1.9-keV resolution at 1332 keV was used to detect gamma rays. The spectra were recorded using a Tracor Northern 1710 analyzer.

In the second series of measurements, ^{241}Am solutions were used because the energy of the primary alpha particle is 5.486 MeV, comparable to 5.498-MeV alpha particles from ^{238}Pu, and 5.155-MeV alpha particles from ^{239}Pu. The ^{241}Am solutions that emitted 1.205×10^8 α/s were added to each of two containers; one containing 1 cm^3 of natural water, the other containing 1 cm^3 of enriched ^{17}O, ^{18}O water (the same isotopic enrichments as above). The same detector and analyzer were used to record the emitted gamma radiation.

RESULTS AND DISCUSSION

Partial plots of obtained gamma spectra appear in Fig. 1. The dotted line is the spectrum obtained with a ^{252}Cf neutron source placed in natural water; the asterisk line is the spectrum obtained in a similar way using isotopically enriched water. Part (a) shows the region near the 870.8-keV line, where the difference of these two spectra represents the intensity of 870.8-keV gamma ray from the $^{17}O(n,n'\gamma)^{17}O$ reaction.

For the solution containers used, it was estimated that a source neutron would travel an average distance of 1.5 cm in water before escaping. A value of 0.42 ± 0.05 mb was obtained for the average cross section (^{252}Cf fission neutron spectrum) for the reaction $^{17}O(n,n'\gamma)^{17}O$ by measuring the intensity of the emitted 870.8-keV gamma ray.

Part (b) of Fig. 1 shows the region near the the 1982.2-keV line. In natural water, the observed 1982.2-keV line is seen to be very weak. This peak consists of two components, derived from both the $^{18}O(n,n'\gamma)^{18}O$ and $^{17}O(n,\gamma)^{17}O$ reactions. The ^{17}O thermal neutron capture cross section was measured by Lone and Inglis[1] to be 538 ± 65 μb; the average ^{17}O capture cross section over the fission neutron spectrum is unknown, but likely to be smaller. From the total intensity of the 1982.2-keV gamma ray, the average cross section was found (over the ^{252}Cf fission neutron spectrum) to be 2.08 ± 0.25 mb.

The gamma spectrum produced by alpha particles interacting with enriched water appears in Fig. 2. In addition to the $^{17}O(\alpha,\alpha'\gamma)^{17}O$ and $^{18}O(\alpha,\alpha'\gamma)^{18}O$ reactions, the $^{17}O(\alpha,n)^{20}Ne$ and $^{18}O(\alpha,n)^{21}Ne$

reactions can be identified. Figure 3 shows partial plots of gamma spectra obtained. The dotted line is the spectrum for ^{241}Am in natural water; the asterisk line is the spectrum acquired for ^{241}Am in enriched water.

To alpha particles, the mixed solutions are infinitely thick targets. Hence, from the observed intensities of the 870.8- and 1982.2-keV gamma rays, the production yields were found to be 1.24 ± 0.15 per 10^6 alpha particles and 0.44 ± 0.06 per 10^6 alpha particles, respectively.

The ratio between specific isotopic $(\alpha,\alpha'\gamma)$ and (α,n) reactions can also be found. From the observed intensities of the 870.8-, 1635.3-, 1982.2- and 350.7-keV gamma-ray lines, the following ratios were obtained: $^{17}O(\alpha,\alpha'\gamma)^{17}O/^{17}O(\alpha,n)^{20}Ne$ = (39 ± 2)% and $^{18}O(\alpha,\alpha'\gamma)^{18}O/^{18}O(\alpha,n)^{21}Ne$ = (5.6 ± 0.8)%.

The (α,n) reaction cross section for ^{17}O and ^{18}O gas targets for alpha particles having energies between 5 and 12 MeV had previously been studied by Hansen, et al.;[2] they reported that at 5 MeV for ^{17}O, σ_{tot} = 175 ± 15 mb; for ^{18}O, σ_{tot} = 380 ± 30 mb.

The hydrogen capture gamma ray at 2223.3 keV is a prominent peak in the obtained spectra. From the intensity of this peak, the capture cross section for the ^{252}Cf fission neutron spectrum was determined to be 0.887 ± 0.090 mb, as compared with the thermal neutron capture cross section of 332 ± 2 mb.[3]

In most PuO_2 samples (with high ^{239}Pu content), the 1982.2-keV gamma ray was not observed. The reason for this, we believe, is that the alpha particle energy from ^{239}Pu (5.155 MeV) may be too low to overcome the Coulomb barrier of ^{18}O.[4]

CONCLUSIONS

The average cross sections for the ^{252}Cf fission neutron spectrum for the following reactions have been measured: $^{17}O(n,n'\gamma)^{17}O$ (0.42 ± 0.05 mb), $^{18}O(n,n'\gamma)^{18}O$ (2.08 ± 0.25 mb), and $^{1}H(n,\gamma)^{2}H$ (0.887 ± 0.090 mb).

The $(\alpha,\alpha'\gamma)$ production yields for the 870.8-keV gamma ray of ^{17}O and 1982.2-keV gamma ray of ^{18}O were determined to be 1.24 ± 0.15 per 10^6 alpha particles, and 0.44 ± 0.06 per 10^6 alpha particles, respectively. The production yields can also be found in $(n,n'\gamma)$ reactions: they are 622 ± 30 per 10^6 neutrons and 3100 ± 150 per 10^6 neutrons, respectively. The production yields for both gamma rays are higher for neutron inelastic scattering. However, from the known alpha decay half-life and spontaneous fission half-life of plutonium isotopes, it is found that there are almost 10^8 times more alpha particles than neutrons. Therefore, the 870.8-keV gamma rays observed from PuO_2 were mainly due to the $(\alpha,\alpha'\gamma)$ reaction.

In nuclear safeguards work a very strong ^{252}Cf source, say 10^9 n/s, could be placed next to a sample to enhance the intensities of both the 870.8- and 1982.2-keV gamma rays. A search could also be made for (α,n) reaction gamma rays for confirmation of the oxide state in those cases where the ^{238}Pu and ^{240}Pu content is high.

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2. L. F. Hansen, J. D. Anderson, J. W. McClure, B. A. Pohl, M. L. Stelts, J. Wesolowski, and C. Wong, "The (γ,n) Cross Sections on ^{17}O and ^{18}O Between 5 and 12.5 MeV," Nucl. Phys. A98, p. 25 (1967).

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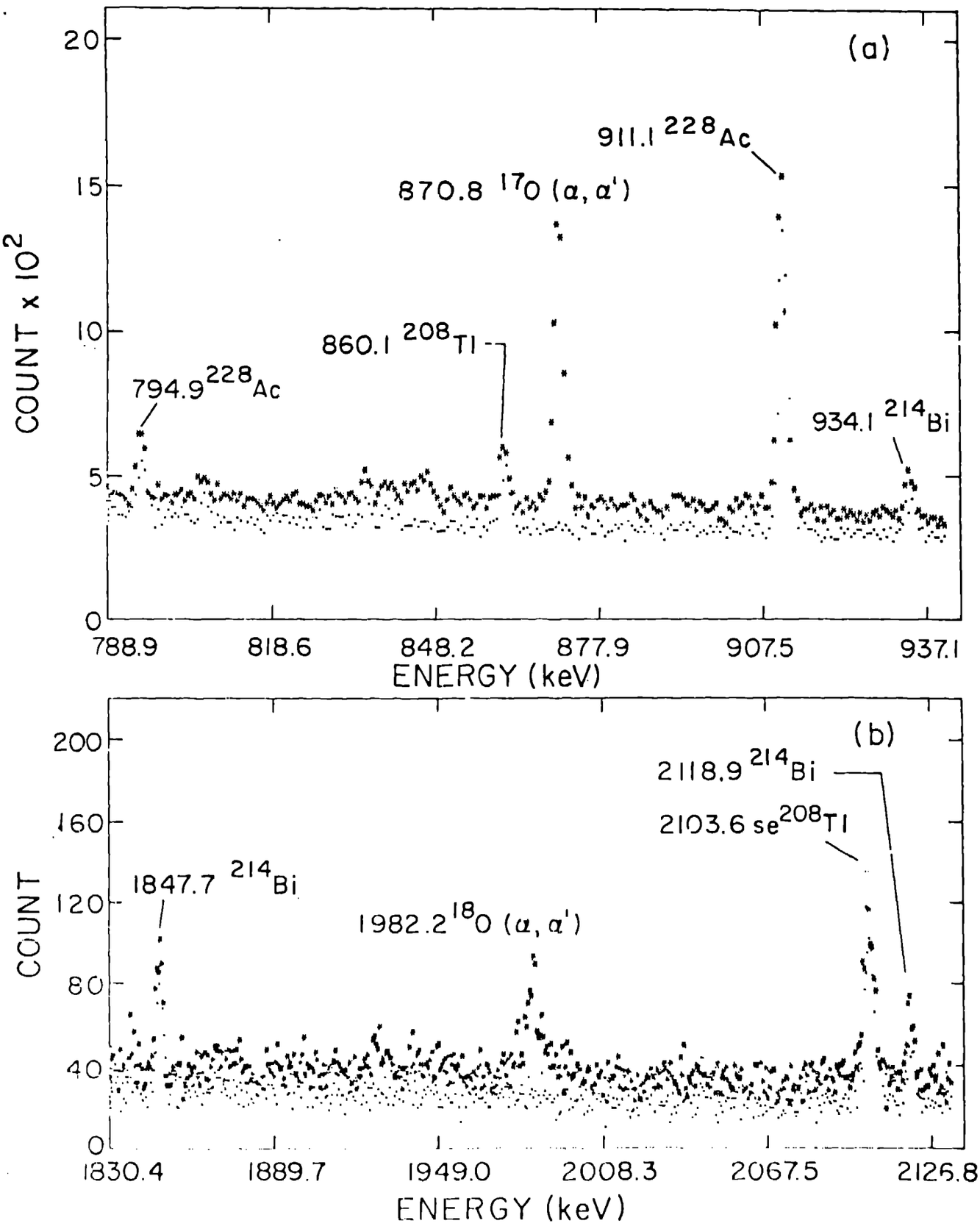
4. J. C. Peng, private communication.

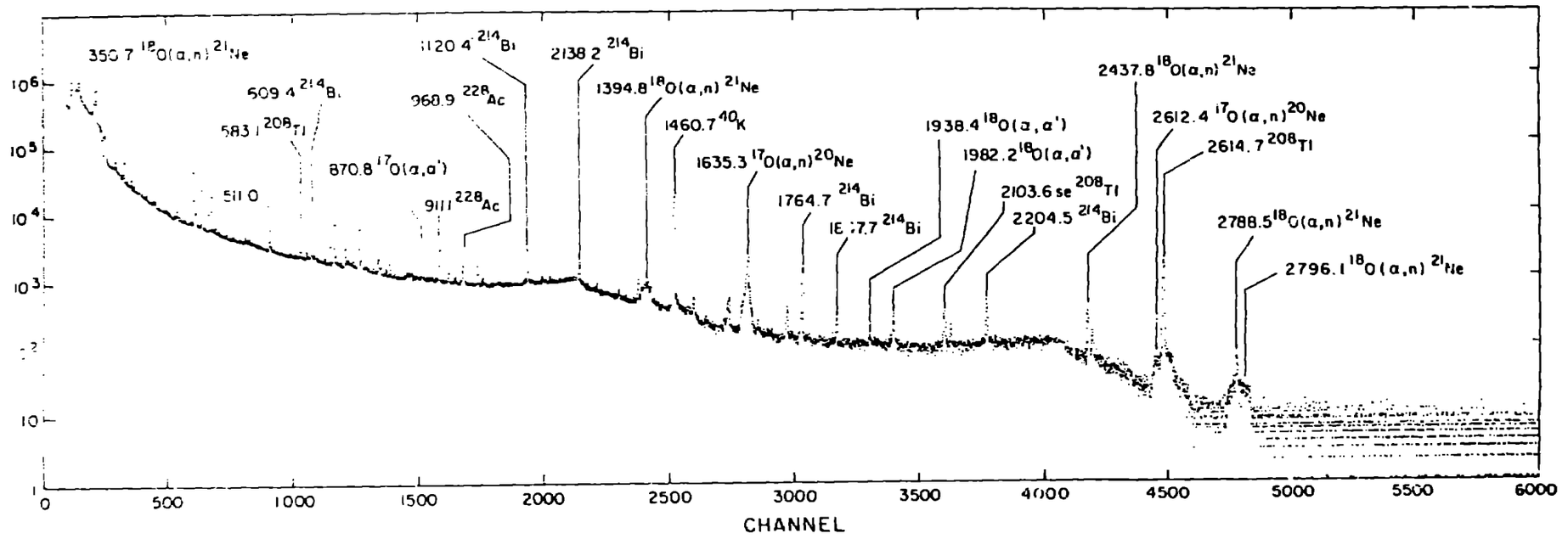
FIGURE CAPTIONS

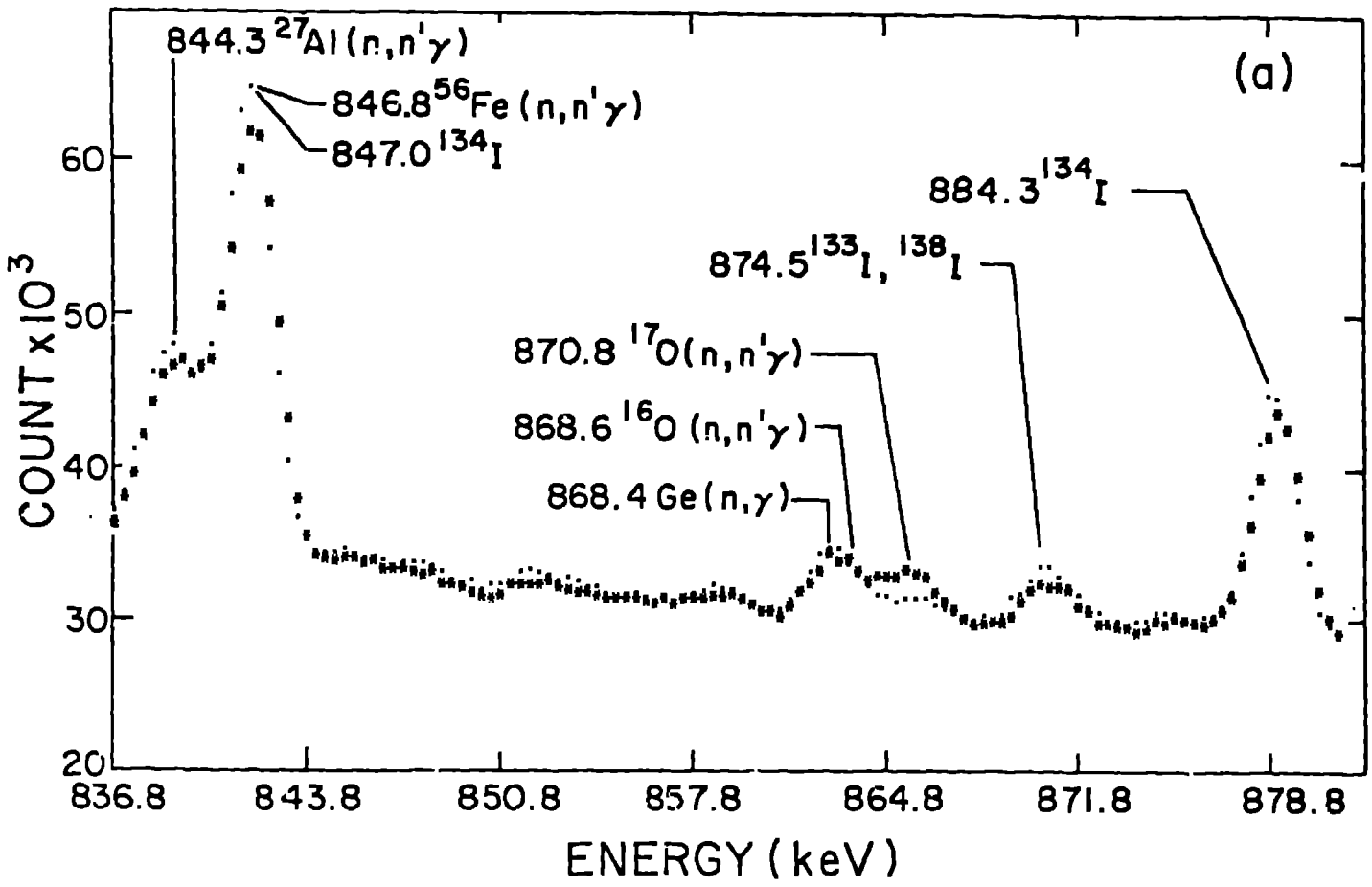
Fig. 1. Partial gamma spectra plots; the dotted line is the spectrum resulting from ^{252}Cf in natural water, the asterisk line is the spectrum resulting from ^{252}Cf in enriched water. Part (a) the 870.8-keV region; part (b) the 1982.2-keV region.

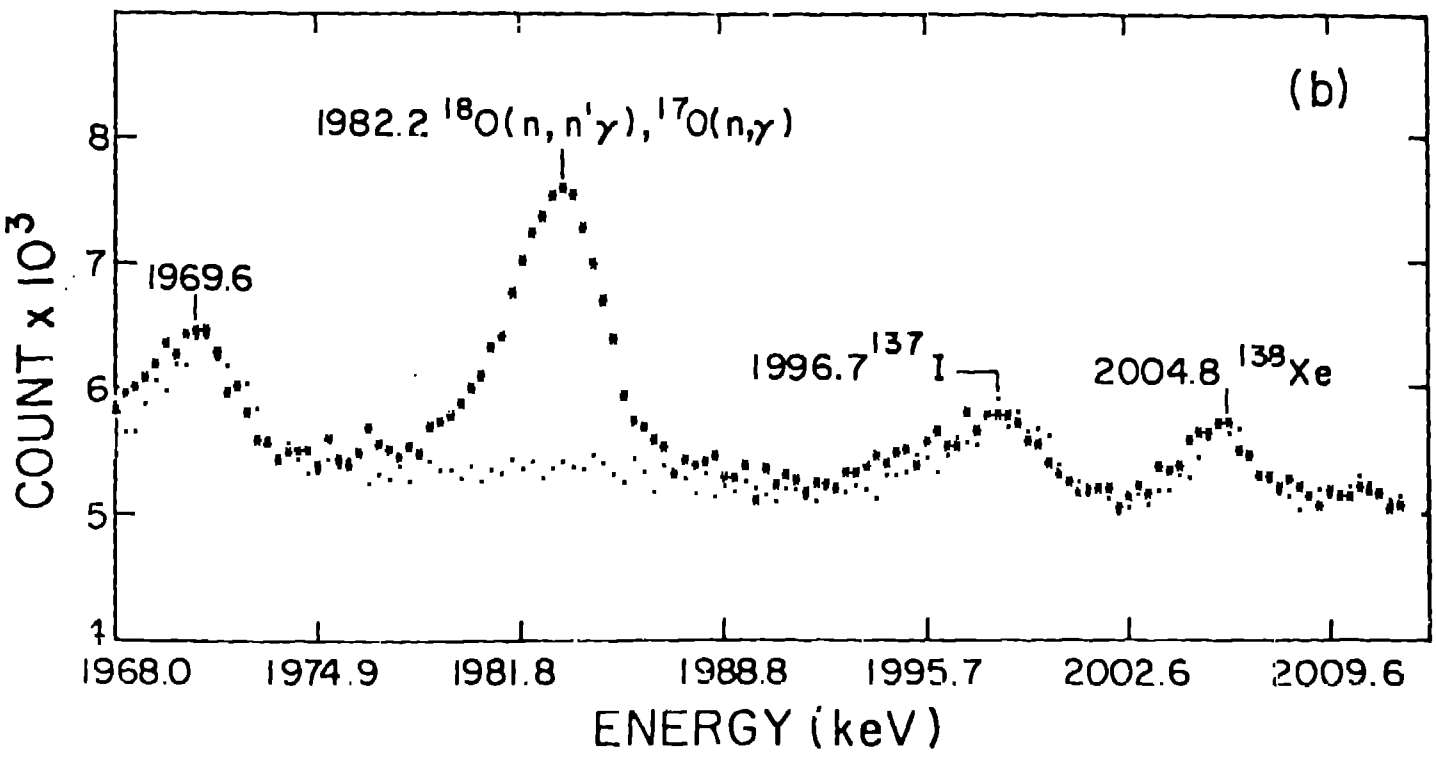
Fig. 2. Gamma spectrum from ^{241}Am and enriched water in a mixed solution.

Fig. 3. Partial gamma spectra plots; the dotted line is the spectrum of ^{241}Am in natural water, the asterisk line is the spectrum of ^{241}Am in enriched water. Part (a) the 870.8-keV region; part (b) the 1982.2-keV region.


(a)
911.1 228Ac
870.8 17O (α, α')
860.1 208Tl
794.9 228Ac
934.1 214Bi
COUNT x 10²
20
15
10
5
0
788.9
818.6
848.2
877.9
907.5
937.1
ENERGY (keV)
(b)
2118.9 214Bi
2103.6 se 208Tl
1847.7 214Bi
1982.2 18O (α, α')
COUNT
200
160
120
80
40
0
1830.4
1889.7
1949.0
2008.3
2067.5
2126.8
ENERGY (keV)


350.7 $^{18}O(\alpha,n)^{21}Ne$
509.4 ^{214}Bi
583.1 ^{208}Tl
870.8 $^{17}O(\alpha,\alpha')$
911.1 ^{228}Ac
968.9 ^{228}Ac
1120.4 ^{214}Bi
2138.2 ^{214}Bi
1394.8 $^{18}O(\alpha,n)^{21}Ne$
1460.7 ^{40}K
1635.3 $^{17}O(\alpha,n)^{20}Ne$
1764.7 ^{214}Bi
1938.4 $^{18}O(\alpha,\alpha')$
1982.2 $^{18}O(\alpha,\alpha')$
2103.6 se ^{208}Tl
2204.5 ^{214}Bi
2437.8 $^{18}O(\alpha,n)^{21}Ne$
2612.4 $^{17}O(\alpha,n)^{20}Ne$
2614.7 ^{208}Tl
2788.5 $^{18}O(\alpha,n)^{21}Ne$
2796.1 $^{18}O(\alpha,n)^{21}Ne$
10^6
10^5
10^4
10^3
10
1
0
500
1000
1500
2000
2500
3000
3500
4000
4500
5000
5500
6000
CHANNEL


844.3 ^{27}Al(n,n'γ)
846.8 ^{56}Fe(n,n'γ)
847.0 ^{134}I
884.3 ^{134}I
874.5 ^{133}I, ^{138}I
870.8 ^{17}O(n,n'γ)
868.6 ^{16}O(n,n'γ)
868.4 Ge(n,γ)
(a)
COUNT x 10^3
60
50
40
30
20
836.8
843.8
850.8
857.8
864.8
871.8
878.8
ENERGY (keV)


1982.2 ^{18}O(n,n'γ), ^{17}O(n,γ)
1969.6
1996.7 ^{137}I
2004.8 ^{138}Xe
(b)
COUNT x 10^3
8
7
6
5
4
1968.0
1974.9
1981.8
1988.8
1995.7
2002.6
2009.6
ENERGY (keV)