LETTERS TO THE EDITORS

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Pile Production of Radioisotopes by Threshold Reactions

A GREAT advantage of production of radioisotopes by threshold reactions (n,p) and (n,α), relative to neutron capture (n,γ), is that the radioisotope can be separated chemically from the target material, and hence be obtained in high specific activity, in principle completely carrier-free. The rate of threshold reactions in the pile, however, is much smaller in general than that of capture reactions, not only because of the low cross-sections for neutrons in the MeV. range but also because of the low flux of neutrons of that energy in the pile. As measurement of the small yields of such reactions is often difficult, especially for long-lived radioisotopes, it is very useful to be able to predict approximate yields in advance. An example is furnished by the reaction ^{54}Fe$(n,p)^{54}$Mn, recently discussed by Stafford and Stein[1]; in this case some experiments had indicated production of the 324-day manganese-54 by pile neutrons, whereas others showed none. The most recent experiments[1], however, utilizing samples enriched in iron-54, definitely established the reaction with a cross-section for fast neutrons of 11 millibarns (10^{-27} cm.^{2}). It is the purpose of the present communication to point out that the approximate yields for threshold reactions in the pile can be predicted, and to illustrate the method for ^{54}Fe$(n,p)^{54}$Mn as an example.

A systematic survey of threshold reaction cross-sections for fission neutrons was carried out by Hughes, Spatz and Goldstein[2] in 1946. Whereas the original purpose of the measurements was to investigate the shape of the fission neutron spectrum at high energies, the present well-known spectrum[3] allows the results to be used to systematize the reaction yields. It is found[4] that the threshold reaction cross-section for fission neutrons is proportional to the integral of the fission spectrum above an 'effective energy', E_{eff}, which is a few MeV. higher than the threshold energy for the reaction, E_T. The quantity $E_{\text{eff}} - E_T$, which rises rapidly with atomic number, is simply related to the penetrability of the outgoing particle and the fission spectrum shape. Results for (n,p) reactions only are given in Fig. 1, where the cross-sections, multiplied by $10/A^{2/3}$ to correct for variation in nuclear size, are plotted against E_{eff} and compared with the integral of the fission spectrum (solid line). The average deviation of the experimental points from the curve is about a factor of two, although the cross-sections themselves cover a range of 3,000-fold. The results of Fig. 1 indicate that the cross-section for the (n,p) reaction for a neutron energy at which the penetrability becomes unity is approximately proportional to πR^2 (with R the nuclear radius), actually being about $0 \cdot 1\ \pi R^2$, as compared to $0 \cdot 2\ \pi R^2$ for (n,α) reactions[4].

For the ^{54}Fe(n,p) reaction, E_T is given by the mass difference manganese-54 – iron-54 [5] (0·71 MeV.) minus the n–p mass difference (0·78 MeV.), or $-$ 0·07 MeV. The quantity E_{eff} for this reaction is then found to be 4·2 MeV., and $\sigma 10/A^{2/3}$ is obtained from Fig. 1 as 7 mb. The cross-section itself is 9·5 mb. ($A^{2/3} = 13 \cdot 5$), in surprisingly good agreement with the 11 mb. measured by Stafford and Stein[1]. Although Fig. 1 refers to cross-sections for unmoderated fission neutrons, the cross-sections for pile neutrons as well are principally a result of unmoderated fission neutrons, for which the flux near a uranium fuel rod is approximately equal to the thermal flux[6]. It is interesting that the curve of Fig. 1 works well for the iron-54 reaction even though the experimental points used to normalize it were all for lighter elements.

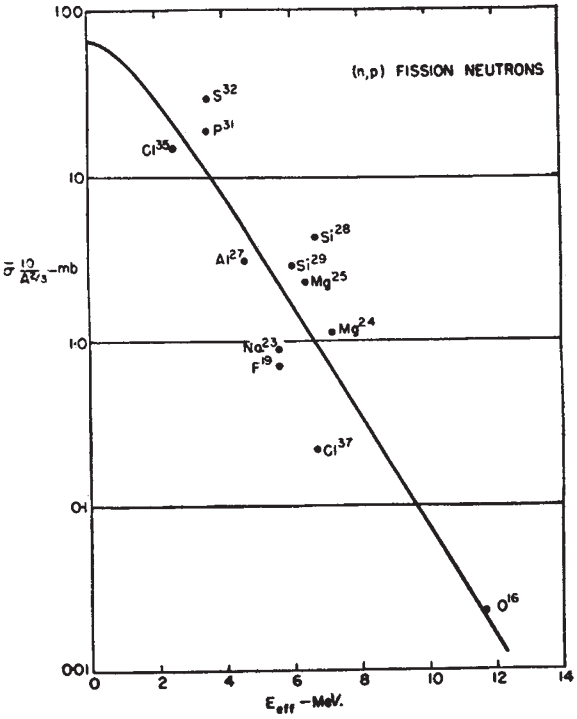

Fig. 1. Measured cross-sections of (n,p) reactions for fission neutrons as a function of effective energy compared with the integral of the fission neutron energy spectrum

The practical value of Fig. 1 and the corresponding curve for (n,α) reactions arises from the fact that approximate yield predictions can be made and thus complex and lengthy measurements avoided. The cross-sections will, of course, decrease rapidly for higher values of Z; for example, for E_T near zero, the cross-sections for (n,α) and (n,p) reactions decrease from 16 and 30 mb. respectively at $Z = 10$, to 0·01 and 1·5 mb. at $Z = 40$. Large variations in E_T occur for light elements, but these become smaller, and affect the cross-sections less, as Z increases.

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[1] Stafford, G. H., and Stein, L. H., *Nature*, 172, 1103 (1953).

[2] Hughes, Spatz and Goldstein (unpublished, see ref. 4).

[3] Bonner, Ferrel and Rinehart, *Phys. Rev.*, 87, 1032 (1952). Hill, D. L., *Phys. Rev.*, 87, 1034 (1952). Watt, B. E., *Phys. Rev.*, 87, 1037 (1952).

[4] The measurements, as well as the details of their interpretation, are described by the author in "Pile Neutron Research", 93 (Addison-Wesley, 1953).

[5] Collins, Nier and Johnson, *Phys. Rev.*, 86, 408 (1953).

[6] Ref. 4, pp. 60, 106.