Production of P³³ with Thermal Neutrons

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(Received June 23, 1952)

A soft beta-activity was found in old P³² samples from the Harwell reactor. It was established that it definitely is due to a P isotope, probably P³³, thus confirming the findings by Jensen and Sheline. The amount of P³³ and P³² samples from a 14-day irradiation of sulfur has been found to 1.4 ± 0.2 percent. The following data were found: half-life 25 ± 2 days, maximum beta-energy 246 ± 5 kev, number of gamma-rays at most 3.5 percent of the betas, cross section for the presumed S³³(n,p)P³³ reaction 0.03 barn. From energy considerations the latter reaction was found to be exothermic, whereas the reaction leading to P³² is endothermic, and this was supported by getting a P activity as rich as 30 percent in P³³ by means of a purer thermal neutron flux. The cross section for the S³³(n,p)P³³ reaction for thermal neutrons was found to be 2.3×10^{-3} barn.

R ECENTLY Jensen *et al.*¹ and Sheline *et al.*² announced the discovery of the radionuclide P⁸³. Similar work was at that time in progress in this laboratory, and later the work was extended and has yielded the following results: (1) confirmation of the physical data of P³³ and its production with reactor neutrons; (2) a method of its production from sulfur in a purer state by means of thermal neutrons only; (3) a chemical proof that the activity is due to a phosphorus isotope. The most significant facts will be stated briefly here.

The half-life of a strong P³² sample (Harwell) showed (in 1950) too large a value, namely, 14.68 ± 0.08 days (14.3 days was the accepted value³); but when aged for 15 months a soft beta-component, present as a negative ion inseparable from P³², remained. This was taken as evidence for a P isotope, which was fully confirmed when the American publications appeared.^{1,2} Our data confirmed the results of these authors quite well: The half-life was preliminarily found to be 25 ± 2 davs (Sheline²: 25 ± 2 davs; Jensen¹: 24.8 ± 0.5 davs). The maximum beta-energy was found to be very close to that of Ca⁴⁵. An absorption comparison, however, using only the first 2/5 of the practical range, was made with S³⁵ and Ca⁴⁵, yielding an energy of 246±5 kev for P³³; but it should be noted that this result presumes that the spectra are of very similar form and that the upper energy limit of Ca45 is 250 kev.3 (Jensen's figure for P33 is 0.26 ± 0.02 Mev; Sheline's is 0.26 ± 0.01 Mev.) The activity found in aged reactor-produced P32 (Harwell) gave a figure of 1.4 ± 0.2 percent P³³ activity after a 14-day irradiation of the sulfur sample in the usual, central position of the reactor.⁴ Taking the cross section for the $S^{32}(n,p)P^{32}$ reaction as 0.012 barn,³ a cross section of 0.03 barn was found for the presumed reaction $S^{33}(n,p)P^{33}$ for this neutron energy distribution. The P^{33} content of old P^{32} samples (0–250 days) was found to increase with age according to the rate expected using the above-mentioned figures. The P^{33}/P^{32} ratio was determined using an absorption method (see Fig. 1).

No gamma-rays were detected in P³³, the experimental limit being that less than 3.5 percent gammas were present. (The figure was based on a calibration of the counter with Co^{60} 1.2-Mev gamma-rays.)

It was of interest to look for a method of producing pure P³³ directly, instead of by aging P³² samples. As the Q-values of an (n,p) reaction is simply 750 kev minus the beta-energy of the product nuclide, the S³³(n,p)P³³ reaction is exothermic by about 500 kev. This suggested that P³³ might be produced by means of thermal neutrons, and since the S³²(n,p)P³² reaction is endothermic by about 900 kev the activity should be purer the better the fast neutrons are cut off. A sulfur sample was therefore irradiated for 14 days near the reflector in the Harwell reactor, where the thermal/fast ratio should be very high (the total flux being 5×10¹⁰ neutrons/cm² sec). The purified P-activity showed about 30 percent P³³ (see Fig. 1), the rest being P³²;

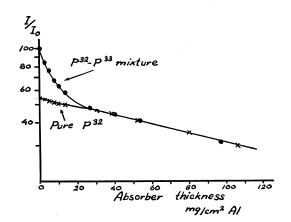


FIG. 1. Absorption curves taken with a 1.4-mg/cm² mica endwindow counter, showing the presence of about 50 percent P³³ (upper curve) in a sample obtained after irradiating sulfur with thermal neutrons. (The curve was taken 6 weeks after irradiation.) The lower curve represents almost pure P³².

¹ Jensen, Nichols, Clement, and Pohm, Phys. Rev. 85, 112 (1952).

² Sheline, Holtzman, and Fan, Phys. Rev. 83, 919 (1951). ³ Nuclear Data, National Bureau of Standards Circular 499 (1950).

⁴L. B. Emlet, Chem. Eng. Progress 46, 591 (1950); W. J. Arrol, AERE-I/R-748 (Harwell) (unpublished).

this is much higher than the 1.4 percent found in the center and supports the view that the reaction is exothermic. (The presence of P^{32} shows that a fast neutron flux is still present, of order 10^8 neutrons/cm² sec.) The specific activity calculated for sulfur is $0.1 \ \mu\text{C/gram}$, giving a thermal cross section of only 2.3×10^{-3} barn for the $S^{33}(n,p)P^{33}$ reaction. This is about 7 percent of the cross section found for the neutrons in the center. It seems safe to assume that the fast neutron flux might be simply reduced by at least a factor of ten by means of moderating material, thus directly making P^{33} at least 90 percent pure. It is possible, moreover, to use a much higher neutron flux (e.g., in Chalk River), where the order of one millicurie P^{33} per kg sulfur might be produced.

The chemical identity proof for P^{33} involved dissolving of the sulfur in CS_2 , filtering through a pile of filter papers (more than 60 percent of the P-activity was found on the first paper, whereas S^{35} went through), treating the paper with HNO₃ and a 0.1-mg phosphate carrier, lanthanum precipitation,⁴ dissolving and cationexchange processing,⁴ and finally a molybdate precipitation. Absorption curves for samples from the filter paper, lanthanum precipitation, and the molybdate all showed the same P^{33}/P^{32} ratio within experimental error. The chemical steps should eliminate all cations and most of the anions, except possibly arsenate and silicate. Even if the latter is true, some separation would likely have been observed and, moreover, Si or As isotopes are not likely to be produced from sulfur, for physical reasons.

I thank Mr. J. L. Putman and the AERE, Harwell, for their kindness in performing the irradiation in the reactor.

PHYSICAL REVIEW

VOLUME 88, NUMBER 3

NOVEMBER 1, 1952

Magnetic Analysis of the Proton Groups from the $Na^{23}(d,p)Na^{24}$ Reaction*†

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(Received July 10, 1952)

Thin targets of sodium iodide evaporated onto platinum and thin nickel backings have been bombarded with deuterons accelerated to energies ranging from 1.5 to 2.2 Mev by the large M.I.T. air-insulated electrostatic generator. Proton groups emitted at 90 degrees to the incident deuterons were analyzed with a 180-degree magnetic spectrograph. Twenty of the observed proton groups are assigned to the Na²³(d, p)Na²⁴, Na²⁴ * reaction and correspond with energy levels in Na²⁴ up to 4.5 Mev. Several of the groups are closely spaced, and level spacings in Na²⁴ as low as 17.0 \pm 3 kev have been observed.

I. INTRODUCTION

THE element sodium, atomic number 11, exists in nature as a single isotope of mass number 23. Thus, the protons emitted from the interaction of deuterons on sodium are associated with the formation of Na²⁴ in various states of excitation according to the reaction Na²³(d,p)Na²⁴. Na²⁴ subsequently decays with a half-life of 14.9 hours to Mg²⁴ by emission of 1.390-Mev beta-particle, accompanied by two cascade gamma-rays of 2.758 and 1.380 Mev.¹

At the present time, the determination of the lower energy levels in Na²⁴ can best be made by either of two methods. The gamma-ray spectra observed from slow neutron capture in Na²³ can give information regarding radiative transitions between the levels of the residual nucleus, according to the reaction Na²³ (n,γ) Na²⁴. Although not uniquely fixing the positions of the excited levels, such gamma-ray measurements do provide confirmatory evidence for levels assigned from other nuclear reactions. Some recent measurements of gammatransitions in Na²⁴ by Kinsey and his collaborators are discussed later in this paper and are compared with the results obtained here. Measurement of the proton energies from the Na²³(d,p)Na²⁴ reaction, as done in the present work, yields more directly the energy levels of the residual nucleus.

A number of proton groups from the deuteron bombardment of sodium have been reported by other investigators. Lawrence, ^{2,3} using deuterons of 2.15 Mev, first observed two proton groups with Q-values of 4.92 and 1.72 Mev. Murrell and Smith,⁴ using deuterons of 0.85 Mev, found four groups they attributed to Na²³(d,p)Na²⁴ with Q-values of 4.76, 4.58, 3.50, and 1.38 Mev. Recently, Whitehead and Heydenburg⁵ have made a more

^{*} A portion of this work was reported at the New York meeting of the American Physical Society, Phys. Rev. 82, 304 (1951). † This work has been supported by the joint program of the ONR and AEC.

¹ Nuclear Data, National Bureau of Standards Circular 499, 19 (1950).

² E. O. Lawrence, Phys. Rev. 47, 17 (1935).

³ M. S. Livingston and H. A. Bethe, Revs. Modern Phys. 9, 245 (19437). ⁴ E. B. Murrell and C. L. Smith, Proc. Roy. Soc. (London)

A173, 410 (1939).

⁶ W. D. Whitehead and N. P. Heydenburg, Phys. Rev. 79, 99 (1950).