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RELATIVE CROSS-SECTIONS OF (n, p) REACTIONS IN SULPHUR 32 AND PHOSPHORUS 31

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(Received for publication, February 20, 1953)

ABSTRACT. Fast neutrons from a 100 milligram ($Ra\alpha + Be$) source have been used to produce (n, p) reactions in S^{32} and P^{31}. The radioactive end products of the reactions P^{32} ($T \sim 14{\cdot}2$ days) in the case of S^{32}, and Si^{31} ($T \sim 170$ min.) in the case of P^{31}, serve as indicators of the reactions. By the principle of 'threshold detector', the initial radioactivity of the end product in each case is a measure of the average cross section of the (n, p)-reaction over a narrow band of neutron-energy between the threshold of the reaction and the upper limit of the effective neutron-energy for capture. The saturation conditions of irradiation and measurement are found out by elaborate preliminary experiments. The initial activity, measured in this way by a G.M. counter under fixed geometry, comes out to be 367 counts per 2 mins. for S^{32} and 240 counts/2 mins. for P^{31}. Various corrections for chemical or isotopic composition of the sample, self absorption of β rays, non-saturation of activity etc. are applied. The ratio of relative cross sections of (n, p) reaction for S^{32} to P^{31} is finally obtained as ~ 2 with a probable error of about $\pm 10\%$. This agrees reasonably well with the ratio $\sim 2{\cdot}57$ obtained by Cohen with much higher energy of fast neutrons. Importance of works on these lines for the interpretation of high energy excitation states of nuclei is discussed.

INTRODUCTION

Nuclear reactions with fast neutrons :

In order to explore the structure of nuclear energy levels in the region of excitation energies higher than 8 Mev, recourse has to be taken primarily to reactions with fast neutrons. The position here is, however, somewhat more complicated than in the reactions with slow neutrons. In the fast neutron reactions, the resonance capture of the neutrons, so common with the capture of slow neutrons in heavy nuclei, is comparatively rare due to the over-lapping of the highly excited states of the compound nucleus. The cross section of fast neutron reactions, therefore, would be small. But here there are chances of sufficient excitation energy being available to the nucleus for the emission of a second particle after the first particle has been emitted ; more than one excited state of the compound nucleus and more than one state of the residual nucleus are likely to contribute to the reaction yield. Thereby the cross section of the neutron reactions may be sometimes considerably enhanced. The study of this reaction is also facilitated in favourable cases if the residual nucleus is radioactive.

On account of the strong short range forces existing between the constituent particles of a nucleus, it can be treated as a condensed phase of its constituents. As in a liquid drop, or in a solid body, an excited state of such a nucleus formed

the absorption of an incident particle cannot be far removed from its original ground state, yet it may possess an excitation energy large enough for the subsequent emission of a single particle. The compound nucleus thus formed has therefore a fairly long life and is a distinct intermediate stage in the nuclear reaction. Its state and decay are independent of the initial process of its formation. This picture will, however, be strictly true when a single quantum state of the compound nucleus has been formed. When a fast incident particle is absorbed in the nucleus, a superposition of several stationary states of a highly excited compound nucleus may result ; the course of the nuclear process may then depend strongly on the relative phases of the states and may not be independent of the initial process of excitation. Bohr's method of calculation may not be strictly applicable in such a case. If, however, the density of states of the compound nucleus is very high, so that their widths overlap each other strongly, a great many states can be excited simultaneously by the incident particle. This phase relation of the states may be nearly at random and the resulting process again almost independent of the way of excitation. In such a case classical considerations may again be justified. Indeed, a statistical method assuming the existence of average values of certain magnitudes over states within not too wide interval of excitation-energy may then be applied. Weisskopf (1947) has shown that within the limits of validity of the statistical considerations discussed above, the cross section of a non-resonance process (a, b) produced by the fast particles can be taken to be of the form :

$$\sigma(a, b) = \sigma_a \eta_b ; \quad \eta_b = \Gamma_b / \Lambda \qquad \ldots\ldots\ldots \qquad (1)$$

where σ_a is the cross section for the formation of the compound nucleus by the capature of the particle a and η_b is the relative probability for the emission of the particle b from the compound nucleus. The quantity η_b is the ratio of the average (over all energy states) partial width Γ_b for the emission of b, to the average total width Λ for all kinds of particles. Based on the above model, Weisskopf has given the method of calculating the approximate cross sections of fast neutron reactions like (n, p) (n, n), (n, 2n) (n, α) etc. Confining our discussions to reactions with high energy neutrons alone as the incident particle, it is clear that

$$\sigma_a = \sigma_b \triangleq \pi r^2 \qquad \ldots\ldots\ldots \qquad (2)$$

where r is the nuclear radius. The process which may now result after the disintegration of the compound nucleus can be reactions of the type (n, p), (n, n), (n, 2n), (n, γ) and (n, α), which are likely to complete with each other in accordance with the relative probabilities of these processes.

The (n, p) reactions are endothermic in character and can take place only with fast neutrons. They are generally less probable than the (n, n) reactions because of the potential barrier preventing the proton from escaping from the nucleus as easily as the neutron. They would predominantly proceed through closely lying energy states of high excitation energy and their cross section would be of measureable value only if neutron energies of several Mev above the threshold

energy of the reaction are employed, since the outgoing proton would need that much of energy to penetrate the potential barrier. If the incident neutron energy-surplus over the threshold is large compared to the nuclear temperature T, then the processes like (n, 2n) should be the dominant feature and the cross section for the (n, 2n) reaction would nearly be equal to σ_n. The (n, α) reactions have an extremely small cross section in the heavy nuclei and in general their values will be smaller than those for the (n, p) reactions because of the higher potential barrier for the outgoing α-particle. Further, the simple capture of the neutrons leading to radiative processes like (n, γ) should also possess very small cross section at neutron energies exceeding 1 Mev, where the cross section σ(n, n) for inelastic scattering of neutrons becomes considerable. The small chance of occurrence of the (n,α) and (n,γ) reactions with fast neutrons in medium heavy nuclei can, therefore, be almost left out of account in the range of neutron energies in which the (n, p) reaction has generally a large cross section. The (n, 2n) reaction also does not compete seriously with the (n, p) reaction, when the energy of the neutrons employed is not more than a few Mev above the threshold energy of the (n, p) reaction.

Experiments have been performed by Jensen (1944), Dunlop and Little (1941), Cohen (1951) and others whose results can be used to verify the conclusions of Weisskopf's theory and to obtain approximate data on the structure of the energy levels in the high energy regions in some nuclei. The investigations are still in the initial stages. In most of these works, however, high energy neutrons produced by cyclotron-accelerated deuterons have been used. Although neutrons of more or less controlled energy can be produced by this method, the upper limit of the neutron energy spectrum generally obtained in this way is quite high (vide Cohen, 1951, figure 1). Consequently the (n, p) reactions produced by these neutrons are very often complicated by other types of reactions like (n, 2n), (n, α), (n, n) etc., taking place simultaneously. We propose, therefore, to use the fast neutrons from a 100 mgm (Raα+Be) neutron source available in this laboratory to study some (n,p) reactions with fast neutrons. The upper limit of the neutron energy spectrum from such a source is about 11 Mev (Teucher, 1949) and the spectrum has a flat maximum over the range about 2 to 4.5 Mev. With these average energies of the fast neutrons, reaction types (n, 2n), (n,α) etc., are not likely to occur with any appreciable intensities, whereas, the (n, p) reactions are expected to be the predominant feature in the medium heavy nuclei, for which the (n, p) reaction threshold generally lies between 1 and 2.5 Mev. The average cross section of (n, p) reactions in these nuclei could therefore be studied with comparatively simple technique, as explained below, when a fairly strong (Raα+Be) neutron source was employed.

METHOD OF THRESHOLD DETECTORS

In the cases where the (n, p) reactions in the nuclei lead to radioactive bodies, the relative cross section of the process can be determined by the method

as threshold detectors. For this a target material is chosen which becomes radioactive on absorbing neutrons of energies above a certain threshold value E_t and subsequently emits a γ-quantum, a proton or an α-particle. It is important that the target nuclei are given an equilibrium exposure to the neutron source.. If the neutrons from the source possess the proper threshold energy E_t and a wide spectrum extending up to the maximum energy E_m then assuming that there is no discontinuity in the capture cross section for activation in this region, the total initial activity produced can be written as

$$A = \int_{E_t}^{E_m} n_0 . \sigma(E) . F(E) . dE$$

where n_0 is the number of nuclei of the species to be activated by the neutrons in the target ; $\sigma(E) \equiv \sigma$ (n, p) denotes the cross section of the (n, p) reaction for the neutrons of energy E, to which the activity A is due, $F(E)$ is the neutron flux in the energy range E and $E+dE$. The The average cross section of the (n, p) reaction between E_t and E_m can thus be estimated by measuring the saturation activity A of the detector, if the flux $F(E)$ and the fast neutron energy spectrum of the source are known; E_t can be determined from the energy balance of the (n, p) reaction under question. If, however, the neutron flux and the energy spectrum of the source are not absolutely known but remain constant, then careful measurements of the initial saturation activity of several detectors under strictly comparable geometrical conditions are expected to give relative cross sections of the (n, p) reactions in the detector nuclei. Such studies of the variation of σ (n,p) from element to element is expected to give useful information about the density of energy levels in the region of high excitation energy for these elements.

EXPERIMENTAL DETAILS

Samples of active material were placed as coaxial cylinders just outside a β-ray G. M. counter (0.1 mm. thick copper wall). Pure reprecipitated flower of sulphur was exposed to neutrons from the 100 mgm (Raα+Be) source, filtered through about 1 mm of cadmium. For phosphorus, ammonium-dihydrogen-phosphate was used. Identical geometry of irradiation to the neutron source was employed throughout. Radio-active $_{15}P^{32}$ of mean half-life 14.2 days was obtained by the (n, p) reaction in $_{16}S^{32}$ and likewise $_{14}Si^{31}$ of half-life 170 min. was obtained from $_{25}P^{31}$. In both the cases the threshold energy of the (n, p) reaction was about 1 Mev. In the case of P^{31}, an additional short period activity seems to have developed (vide figure 2), apparently due to P^{30} produced by the (n, 2n) reaction.

For quantitative comparison of cross sections, measurements should be made such that (1) saturation mass of the substance to be activated is exposed to the neutron-source, for saturation period; if possible, (2) saturation thickness of the well-mixed active substance should be examined for initial activity.

Condition (1) will ensure that even the fastest neutrons from the source may acquire the appropriate mean free path of collision in passing through the sample nuclei. Condition (2) will ensure that maximum possible number of β-rays from the active layer is reaching the counter. These saturation conditions have been discussed by Jensen (1944). Prolonged and painstaking investigations were made in order to determine these saturation conditions and from the results obtained it was finally decided to expose 407 gm of sulphur and 494 gm of the phosphate to the fast neutron source for final measurement.

The investigation of the saturation thickness of the active layers for measurement of the saturation activity was carried out with the activated saturation mass of the substance which was thoroughly mixed up to obtain the average effect. Thin cells of the active powder were then prepared in very thin paper cylinders supported by wide-meshed wire net inside and transparent alkathene cylinder outside. A saturation thickness of 0.6 gm/cm^2 for sulphur and 0.35 gm/cm^2 for the phosphate were obtained.

For the final measurement of the initial activity produced by the (n,p) reactions in sulphur a permanent cell was constructed with thin paper and wire gauge cylinder backing inside and a strong coaxial cylinder of brass outside. The inner cylinder was 4.25 cm in diameter and the well mixed activated sulphur was closely packed into a layer 8.9 cm high and 1.04 cm thick to give the required saturation layer of density 0.6 gm/cm^2. The mass of this active layer was 90 gm. To measure the activity of this cell, the β-ray counter mentioned above was used under strictly reproducible geometry with 10 cm of lead shielding all around. The decay of the sample was followed regularly for 31 days. The semi-logarithmic plot of the nett activity is shown in figure 1, which clearly gives a half-life of 14.2 days.

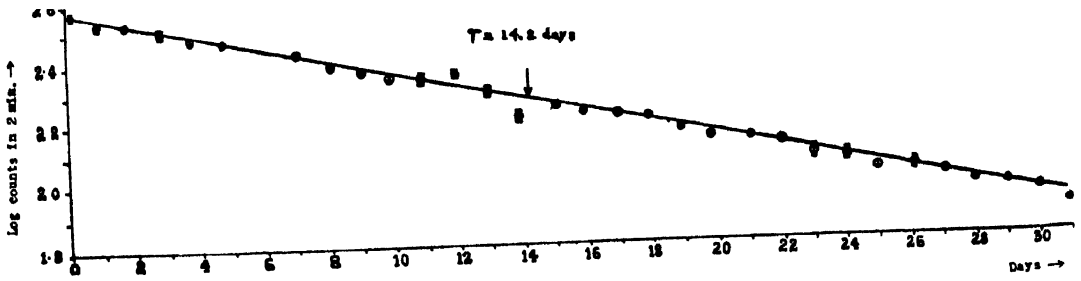

Fig. 1.

The decay curve of radio phosphorus P^{32} produced by the (n, p)-reaction in S^{32}.

Similar observations were made for a saturation mass of the activated phosphate (irradiated for 39 hrs to fast neutrons) and taken in the form of a shell similar to that of sulphur. The thickness of the shell was 0.26 cm and its weight was 42 gm, in order to give the saturation thickness of 0.35 gm/cm^2. The activity of the shell was followed for 4½ hours. The average of three such sets of observations is shown as logarithmic plot in figure 2. The half-life of the activity is clearly 170 min.

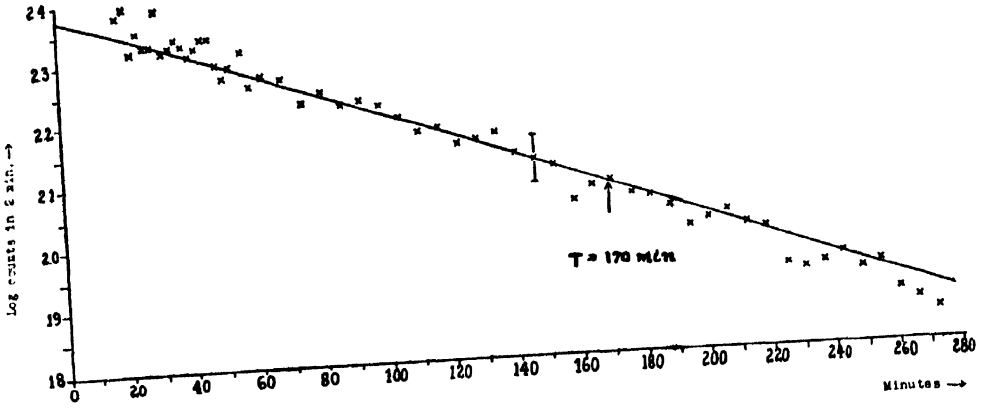

Fig. 2.

The decay curve of radio silicon Si^{31} produced by the (n, p)-reaction in P^{31}. The points near the beginning lie somewhat above the line and probably indicate the presence of a short period activity (P^{30} ~2·5 min) produced by the (n, 2n) reaction in P^{31}.

CORRECTIONS APPLIED

The initial activities obtained by extrapolating curves (1) and (2) to zero time are 367 counts per two min for sulphur and 240 counts per two min for the phosphate. The following corrections were applied to these values :

(*a*) A conversion factor of 1.05 to convert sulphur to the pure isotope S^{32}, there being 4.2% of S^{34} in the sample tested. Similarly a factor of 3.625 is necessary to convert the phosphate to the pure element.

(*b*) Since quite thick layers of the active substances were used for our measurements, the correction for self-absorption of β-rays in the layers is essential, but correction for back scattering of β-rays by the sample holder can be neglected (vide Metzer, Alder and Huber, 1948). The correction factors of 23% for sulphur and 18%for the phosphate were obtained by using the method given by the above authors. Almost identical results were obtained by another method suggested by Cohen (1951).

(*c*) In the case of sulphur, the half-life of activity being 14.2 days, the saturation activity was not attained by irradiating it for 94 hours, as we did. The correction factor on this account was calculated to be 5.7. The phosphate sample was always irradiated for saturation time and hence no correction was necessary for it on this account.

RESULT

The corrected initial activity (which is proportional to the cross section of the (n, p)-reaction) for sulphur S^{32} therefore becomes :

$$\frac{367 \text{ counts}}{2 \times 60 \text{ sec.}} \times \frac{1{\cdot}05 \times 5{\cdot}7}{0{\cdot}23} \simeq 79.$$

For phosphorus P^{31} :

$$\frac{240 \text{ counts}}{2 \times 60 \text{ sec.}} \times \frac{3{\cdot}625}{0{\cdot}18} \simeq 40.$$

The ratio of the two cross sections

$$\frac{\sigma(n, p)S^{32}}{\sigma(n, p)P^{31}} \simeq 2$$

The overall probable error of this determination is ± 10 to 15%.

REMARKS

The relative cross section for sulphur comes out to be about twice that for phosphorus. This result agrees within the limits of experimental error, with the (n, p) reaction cross section observed by Cohen (1951) who obtained 28.5×10^{-26} cm^2 for S^{32} and 12.0×10^{-26} cm^2 for P^{31}, which give a ratio 2.37. Fast neutrons of energy ranging upto $\sim$18 Mev were used for the reaction by Cohen. This energy limit being higher than the maximum neutron energy ($\sim$11 Mev, Teucher, 1949) from a (Raα+Be) source used by us, there may be difference in the state of excitation of the nuclei in the two cases leading to difference in the reaction cross sctions. Metzger, Alder and Huber (1948) have observed a resonance effect in the excitation function of the reaction P^{31} (n, p) Si^{31}, the cross section slowly increasing linearly between the neutron energies 2.3 and 3 Mev, and reaching the maximum value of 7.4×10^{-26} cm^2 at 3 Mev. Thereafter the cross section begins to fall at high neutron-energies. For S some results of the total capture cross section as a function of neutron energy have been given by Adair (1950) and others. According to these authors a large number of resonance peaks exists for neutron capture in S^{32} between 0.1 and 4 Mev neutron energies, a region in which the (Raα+Be) neutron spectrum is particularly rich. The cross section for (n,p) reaction in S^{32} is therefore likely to be higher than in P^{31}. Further work on the absolute measurement of these cross sections would be necessary to clarify the position. Measurements of the absolute cross section of the (n, p) reaction in one of the nuclei and the relative cross section for a number of other cases are in progress. General considerations regarding the density and structure of the nuclear energy levels in the region of high excitation energy can probably be attempted in the light of the theory (vide Weisskopf, 1947) when these measurements are completed.

ACKNOWLEDGMENTS

We take this opportunity to express our sincere thanks to Professor D. S. Kothari and Prof. R. C. Majumdar for constant encouragement in course of this work and to Mr. S. K. Nandi and Mr. L. Kasturi Rangan for helping us with some of the measurements. The work has been done under the financial support of the Atomic Energy Commission, Ministry of Research and Natural Resources, Government of India.

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