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Neutron-Induced Reactions in Fluorine*

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By measuring the radioactivity produced when F^{19} is bombarded with monoenergetic neutrons, the excitation functions and absolute cross sections have been obtained for the $F^{19}(n,\alpha)N^{16}$ and $F^{19}(n,p)O^{19}$ reactions from energies at which the respective activities were first observed to 8.0 Mev. The occurrence of the (n,α) reaction was first observed at a bombarding energy of 3.1 Mev; the (n,p) reaction was first observed at 4.7 Mev. Several resonances were found in the (n,α) excitation function, while the (n,p) excitation function showed a single maximum at 7.5 Mev. For neutron energies up to 8.0 Mev, the (n,α) reaction cross section is larger than that for the (n,p) reaction by a factor of 2 or greater.

INTRODUCTION

Measurements of the neutron total cross sections of most elements have been made in the neutron energy range from thermal energies to several Mev. These experiments are usually of the transmission type, and, consequently, it is not possible to deduce the process by which removal from the beam has occurred. Therefore, it is of interest to obtain the magnitudes of the individual contributions to the cross section. The total cross section is the sum of the elastic, inelastic, and reaction cross sections. For neutrons of energies less than about 20 Mev, the reaction cross section is composed of processes of the type (n,γ), (n,α), (n,p), and $(n,2n)$. Radiative capture is important only for low-energy neutrons, and $(n,2n)$ reactions are usually energetically impossible for neutrons of energies less than about 10 Mev. For neutrons in the energy range of a few Mev, (n,α) and (n,p) reactions are the most common neutron-induced reactions.

The excitation functions and absolute cross sections have been obtained[1–9] for the (n,p) reactions on N^{14}, O^{16}, Al^{27}, P^{31}, and S^{32}, and for the (n,α) reactions on Li^6, Be^9, B^{10}, N^{14}, Ne^{20}, and S^{32}. A preliminary report has been given[10] on the excitation function for the $F^{19}(n,\alpha)N^{16}$ reaction. Paul and Clarke[11] have measured the cross sections for (n,p), (n,α), and $(n,2n)$ reactions on many elements at a neutron energy of 14.5 Mev. Lillie[12] has studied the (n,p) and (n,α) reactions in oxygen and nitrogen at 14.1 Mev.

A common method of obtaining (n,p) and (n,α) reaction excitation functions is by observing, as a function of neutron energy, the pulse-height distribution in a gas counter filled with a gas that contains the nucleus to be used as a target. In some cases this is not convenient, and the reactions $O^{16}(n,p)N^{16}$, $Al^{27}(n,p)Mg^{27}$, $P^{31}(n,p)Si^{31}$, and $F^{19}(n,\alpha)N^{16}$ have been studied by observing the induced radioactivity as a function of neutron energy.

Resonances in the reaction $F^{19}(n,\alpha)N^{16}$ were first observed by Wilhelmy,[13] who bombarded an SF_6-filled ionization chamber with Po-Be and Rn-Be neutrons and observed the pulse height spectra. Peaks due to reactions on S and peaks caused by maxima in the energy distribution of the neutron sources were identified by performing the same experiment with the chamber filled with SO_2. The continuous nature of the energy spectra from these neutron sources made the interpretation of the results difficult; however, peaks were observed which, when corrected for the most recent determination of the Q-value for the $F^{19}(n,\alpha)N^{16}$ reaction,[14] can be attributed to resonances in the cross section at neutron energies of 4.4 and 4.9 Mev.

The $F^{19}(n,p)O^{19}$ reaction has been studied only to the extent that the cross section has been measured at 14.5 Mev. At this energy, Paul and Clarke[11] obtained a value of 135±50 mb.

Since the half-life of N^{16} is 7.35 sec,[2,15] the observation of the induced radioactivity as a function of neutron energy provides a convenient means of examining the excitation function for the $F^{19}(n,\alpha)N^{16}$ reaction. The Q-value for this reaction is -1.49 Mev and that for the $F^{19}(n,p)O^{19}$ reaction is -4.00 Mev.[14] Therefore, the

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(n,α) reaction can be investigated up to a neutron energy of about 4.2 Mev before the (n,p) reaction becomes possible. The half-life of O^{19} is 29 sec.[16] This is close enough to that for N^{16} so that precautions must be taken when attempting to determine the amount of N^{16} activity present at neutron energies above 4.2 Mev, and yet it is sufficiently long to make possible the separation of the two activities with little difficulty. The $(n,2n)$ reaction is not possible for neutron energies below 10.7 Mev and was not encountered in these experiments.

By observing the N^{16} and O^{19} activities produced when F^{19} is bombarded with monoenergetic neutrons from the *p-T* and *d-d* reactions, it has been possible to obtain the excitation functions and absolute cross sections for the $F^{19}(n,\alpha)N^{16}$ and $F^{19}(n,p)O^{19}$ reactions from energies at which the activities were first observed up to 8.0 Mev.

SAMPLE AND DETECTOR

In the investigation of neutron-induced reactions, it is desirable to have as large a neutron flux as possible and the greatest possible detection efficiency for measuring the effects of the reactions. If a charged-particle reaction is used as a neutron source, the size of the neutron flux is limited by the energy spread that can be tolerated and by the available charged-particle beam intensities. In order to obtain the largest possible number of target nuclei per cm^3, a solid, fluorine-containing sample was used in these experiments. The greatest possible efficiency for the detection of reactions produced in solid samples results when the sample and the detector are identical. In this manner a 4π geometry is obtained and the efficiency for the detection of decay electrons from induced radioactivity can be made almost 100% by biasing the detector to respond to all pulses greater than a certain small value. In order to investigate the neutron-induced radioactivity in F^{19}, a CaF_2 crystal was used. This crystal was found to be satisfactory both as a sample and as a detector.

The crystal was a 1 in. diam×1 in. cylinder of unactivated CaF_2 which was encased in a thin aluminum container. The inner surface of the aluminum was coated with MgO, which acted as a light reflector. Since CaF_2 fluoresces in the ultraviolet region, it was necessary to use a photomultiplier tube with a quartz window. An EMI 6255 tube with a 1 in.×1 in. photocathode surface was found to be quite satisfactory. Optical contact between the crystal and the quartz window was made with a thin layer of silicone fluid. The associated electronics were of the conventional type.

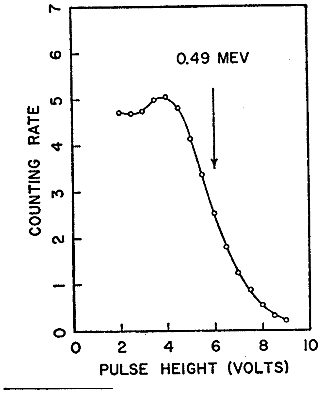

FIG. 1. Pulse-height distribution for the Cs^{137} γ ray that was obtained with the CaF_2 crystal. A channel width of 1 volt was used.

An energy calibration was obtained by observing the pulse-height distribution from the Cs^{137} gamma ray. This curve is shown in Fig. 1. Since the crystal contains elements of low Z, only a Compton scattering peak is prominent. The point at which the counting rate was one-half of the peak counting rate was taken to correspond to the maximum energy of the Compton electrons, 0.49 Mev, and is indicated by the arrow in Fig. 1. By comparing the pulse height of the Compton peaks obtained with the CaF_2 crystal and with a NaI crystal under the same conditions, the pulse height from CaF_2 was found to be smaller by a factor of 10 than that from NaI.

NEUTRON SOURCES

Neutrons of energies up to 4.9 Mev were obtained from the proton bombardment of a tritium target. This target [manufactured at the Oak Ridge National Laboratory, Oak Ridge, Tennessee] was prepared by absorbing tritium gas in a layer of zirconium metal evaporated onto a $\frac{3}{4}$ in. diam tungsten blank. The active area was $\frac{5}{8}$ in. in diameter. The target thickness was about 20 kev to 5-Mev protons. Although the target backing was cooled by an air stream, beam currents of less than 2 microamperes were used in order to prevent undue heating and possible loss of tritium. No indication of a decrease in the neutron flux was obtained over long periods of bombardment with 2 microamperes of 5-Mev protons.

With the *p-T* neutron source, an energy resolution of about 30 kev was obtained by placing the face of the CaF_2 sample-detector at a distance of 6 inches from the source. At a distance of 2.5 inches, the energy spread was increased to about 60 kev due to the decrease in neutron energy at the larger angles. Excitation functions were measured with the *p-T* source for both of these geometries.

The *d-d* reaction was used to produce neutrons of energies from 4.6 to 8.0 Mev. A gas deuterium target was used which was constructed from a $\frac{1}{2}$ in. diam ×1.5 in. platinum tube. The beam entered the target chamber through a $\frac{1}{4}$ in. hole which was covered with a 2.79 mg/cm^2 nickel foil. Deuterium pressures of about 10 inches of Hg were used. No decrease in pressure was observed over prolonged periods of bombardment with 2 microamperes of deuterons. With the face of the CaF_2 crystal placed at a distance of 2 inches from the

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[16] A. M. Feingold, Revs. Modern Phys. 23, 10 (1951).

center of the target chamber, the total spread in neutron energy, resulting from the gas thickness, straggling in the foil, and the finite solid angle, amounted to about 100 kev.

EXPERIMENTAL PROCEDURE

Since N^{16} has a relatively short half-life, the simplest procedure for measuring the relative amounts of activity produced at different bombarding energies is to count the saturation activity during bombardment. This is made possible by the occurrence of high-energy electrons in the N^{16} decay scheme; 18% of the decays go to the ground state of O^{16} with a maximum electron energy of 10.4 Mev.[15] In order to discriminate against γ rays from inelastic neutron scattering, electrons from the radioactive decay of the nuclei produced in the (n,p) and (n,α) reactions on the aluminum container, the Ca of the crystal, and the Si of the phototube, and general background, the counter bias was set so that the pulse-height analyzer would respond only to those pulses corresponding to an energy greater than about 7.5 Mev. After saturation activity was reached, the number of counts was recorded for a given amount of charge collected at the neutron-producing target. The beam current was maintained constant to within 10 percent during both the buildup and counting periods. Changes in the counting rate caused by the small variations in the beam current were essentially averaged out by recording the number of counts obtained over a period of about 20 half-lives. In this manner it was possible to obtain a relative excitation function for the $F^{19}(n,\alpha)N^{16}$ reaction up to a neutron energy of 4.9 Mev, the maximum energy obtained from the p-T source.

When using the d-d reaction as the neutron source, it was not possible to continue the excitation function in the manner described above since the deuteron bombardment of the nickel foil of the gas target chamber produced γ-ray pile-up in the crystal to a pulse height greater than that obtained from the maximum-energy electrons. It was necessary, therefore, to determine the yield of radioactivity by observing the counting rate after the beam was cut off.

At each bombarding energy the N^{16} and O^{19} activities were allowed to reach equilibrium by bombarding for at least 4 minutes. At the end of this time, the deuteron beam was cut off by a compressed air-operated tantalum cup placed at a distance of about 8 feet from the deuterium target. As soon as the beam was stopped, a counting cycle was begun. This entailed counting the activity and then recording the data in alternate 10-sec intervals. To carry out this counting procedure, the bias was set so that the pulse-height analyzer would respond to all pulses corresponding to an energy greater than about 150 kev. Thus, it was possible to count essentially all of the decay electrons from both the N^{16} and O^{19} nuclei. A typical decay curve, taken at a neutron energy of 6.30 Mev, is shown in Fig. 2. The 2.3-min activity is due to the Al^{28} produced in the (n,p) reaction on the Si^{28} content of the phototube. The 9.6-min activity from the $Al^{27}(n,p)Mg^{27}$ reaction and the longer-lived activities from the (n,p) and (n,α) reactions on calcium do not appreciably affect the decay curve over the relatively short time interval for which observations were made. Subtraction of the 2.3-min activity revealed the 29-sec activity of O^{19}, and the subtraction of both of these activities left the 7.3-sec activity of N^{16}. At all bombarding energies for which the decay curve technique was used, the N^{16} activity was sufficiently intense to allow the decay to be followed for at least 7 half-lives in spite of the other activities which were present. At 6.30 Mev, for example, 75 percent of the initial activity was due to N^{16}. The extrapolation of the individual decay curves to the time at which the beam was turned off allowed the saturation activity to be determined. The decay curves were taken at approximately 100 kev intervals over the range of neutron energies from 4.6 to 8.0 Mev. Since there was some uncertainty in extrapolating the decay curves to zero time, the relative excitation functions obtained with the d-d source are probably accurate only to 10–15 percent.

Since the excitation functions obtained with both neutron sources were taken with respect to the beam current, these curves were corrected for the p-T and d-d cross sections. The p-T yield curve used was that

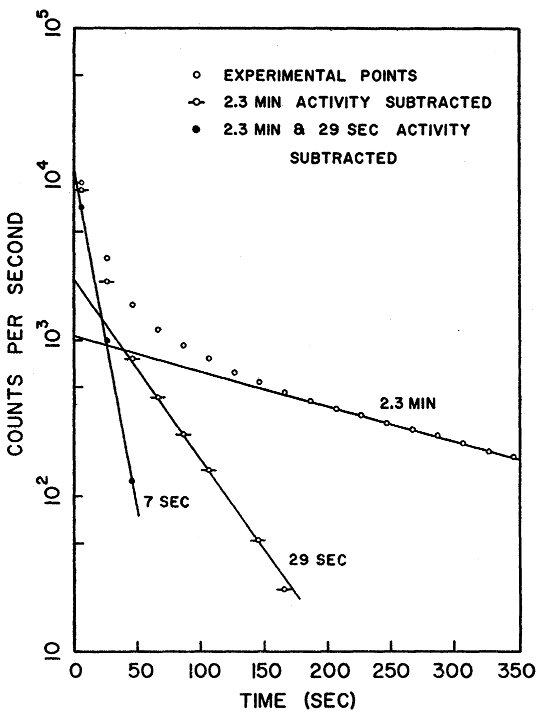

FIG. 2. Decay curve of the radioactivity induced in the CaF_2 crystal when bombarded by 6.30-Mev neutrons. Activities due to N^{16} (7 sec), O^{19} (29 sec), and Mg^{27} (2.3 min) are shown.

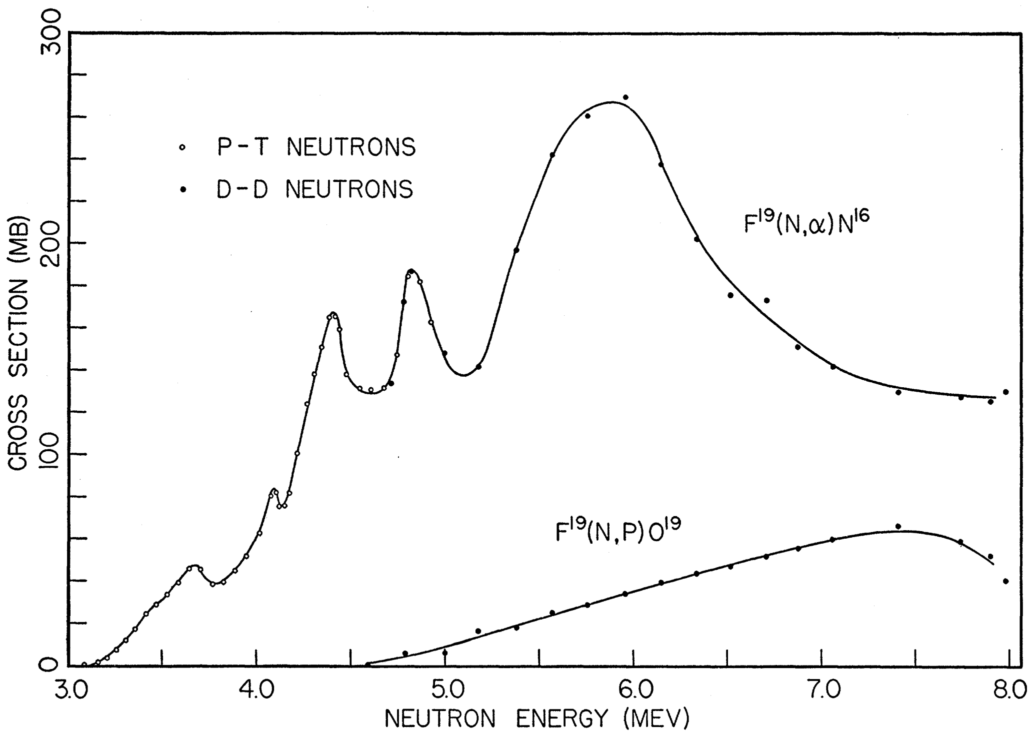

FIG. 3. Excitation functions and absolute cross sections for the reactions $F^{19}(n,\alpha)N^{16}$ and $F^{19}(n,p)O^{19}$. The energy resolution for the points obtained with the *p-T* neutron source is about 60 kev; for those obtained with the *d-d* source, the resolution is about 100 kev.

measured by Jarvis and Perry.[17] The curve for the *d-d* reaction was determined by measuring, as a function of deuteron energy, the number of neutrons per microcoulomb emitted into a forward solid angle equal to that subtended by the CaF_2 crystal at the position used to measure the relative excitation functions. Since the gas target chamber was used, the background counting rate per microcoulomb at each energy was measured by removing the gas from the chamber. The neutron detector was a long counter of the type described by Hanson and McKibben,[18] which was calibrated with a Ra-Be source of known flux. This counter has a response which is essentially constant over a wide range of neutron energies.

Since the bias was set to record essentially all of the decay electrons from both N^{16} and O^{19}, and since the neutron flux, obtained by the above method, was known, an absolute cross section was obtained for the excitation functions measured with the *d-d* neutron source. The data obtained with the *p-T* source was normalized to the (n,α) curve at a neutron energy of 4.86 Mev. Uncertainties in the measurement of the saturation activities, in the neutron flux, and in the number of counts below the bias setting of 150 kev, limit the accuracy of the absolute cross-section measurements to about 40 percent.

To insure that the effects observed with the *p-T* source by counting the saturation activity during bombardment were due entirely to the $F^{19}(n,\alpha)N^{16}$ reaction, the neutron energy region accessible with *p-T* neutrons was reinvestigated in the following manner. A 1.5 in. diam$\times\frac{1}{4}$ in. cylinder of Teflon $[(CF_2)^n]$ was mounted on an anthracene crystal of the same diameter and $\frac{7}{16}$ in. in length. The Teflon was bombarded until saturation of the N^{16} activity was reached. After turning off the beam, the amount of activity was counted for 8 sec. Accurate reproduction of the time intervals was made possible by using a mechanical timer. In order to discriminate against any activity that might be present due to the (n,p) reaction, a bias setting equal to the energy of the maximum energy electrons in the O^{19} decay (4.8 Mev) was used. The excitation function so obtained reproduced in detail that measured with the previous technique, although the statistics were somewhat poorer. Since the only materials common to the two techniques were the fluorine and the silicon of the phototube, with its induced radioactivity of low energy, it was concluded that the effects observed were due only to the $F^{19}(n,\alpha)N^{16}$ reaction.

RESULTS

The excitation functions and absolute cross sections obtained in the manner described above for the reactions $F^{19}(n,\alpha)N^{16}$ and $F^{19}(n,p)O^{19}$ are shown in

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[17] G. A. Jarvis and J. E. Perry (unpublished).

[18] A. O. Hanson and J. L. McKibben, Phys. Rev. 72, 673 (1947).

Figs. 3 and 4. The data obtained with the *p-T* source shown in Fig. 3 were taken with the geometry that produced an energy spread of about 60 kev. Resonances are indicated at 3.70, 4.11, 4.42, and 4.86 Mev. A reexamination of this energy region was made with an energy resolution of 30 kev and the results are presented in Fig. 4. This data shows the peak observed at 3.70 Mev with the 60-kev resolution decomposed into at least 3 narrower resonances. The indication of structure on the low-energy side of the 3.70-Mev resonance (Fig. 3) appeared again in the 30-kev resolution data near an energy of 3.4 Mev. No 7.3-sec activity was observed below a neutron energy of 3.1 Mev.

The measurements made with the *p-T* and *d-d* sources overlap for neutron energies from 4.6 to 4.9 Mev and both sets of data indicate the resonance at 4.86 Mev. At higher energies, an additional maximum was observed with *d-d* neutrons at 5.9 Mev. It is probable that a number of resonances observed by Bostrom *et al.*[10] near 6 Mev are included in the broad resonance at 5.9 Mev. Since the energy resolution was about 100 kev for the *d-d* neutrons, only the gross structure of the excitation function could be detected. The resonance energies and peak cross sections are summarized in Table I.

For neutron energies above 4.7 Mev, 29-sec activity corresponding to the occurrence of the $F^{19}(n,p)O^{19}$ reaction was observed. The O^{19} yield was observed to increase smoothly up to a neutron energy of 7.5 Mev. Above this energy the yield decreased up to the maximum bombarding energy used. At the 7.5-Mev peak, the cross section for the (n,p) reaction is only one-half of that for the (n,α) reaction. The (n,p) excitation function shows no indication of the broad resonance at 5.9 Mev that was observed in the (n,α) reaction.

DISCUSSION

When a target nucleus is bombarded by neutrons with an energy of several Mev, compound nucleus formation may take place with many different values of the neutron angular momentum. Therefore, it is possible to form states of the compound nucleus with a wide range of *J*-values. Since these states are highly excited, the levels will be broad and overlapping. Neutron emission from such states is highly favored over other modes of decay, and, consequently, the resonance structure of the total cross section, evident for low-energy neutrons, is not prominent for neutrons of several Mev. Although neutron emission may occur with any of the large number of incoming angular momenta, charged particle emission, which is usually of lower energy, takes place predominately with low angular momentum. Therefore, selective emission will occur from those states with *J*-values that will allow the particles to be emitted with a small angular momentum value. Since the cross sections for reactions in which charged particles are emitted are an order of magnitude smaller than the elastic-plus-inelastic scattering cross section, resonances in the reactions leading to the emission of charged particles will not appreciably affect the total cross section. Because of this effect, it is possible to observe resonance structure in the compound nucleus by measuring the excitation functions for (n,α) and (n,p) reactions, even though the resonances frequently cannot be resolved in experiments that measure the neutron total cross section. In some cases, the *J*-values of the compound nucleus will be such that resonances can be observed both in the charged particle reaction cross sections and in the total cross section.

TABLE I. Resonances in the reaction $F^{19}(n,\alpha)N^{16}$.

Neutron energy (Mev) 30-kev resolution (*p-T* source)	60-kev resolution (*p-T* source)	100-kev resolution (*d-d* source)	Other[a] measurements	Peak cross section (mb) (±40%)
3.4	3.4 (?)			
3.61±0.05				48
3.69±0.05	3.70±0.05			55
3.77±0.05				44
	4.11±0.05			90
	4.42±0.05		4.4	167
	4.86±0.05	4.9±0.1	4.9	186
		5.9±0.1		268

[a] See reference 13.

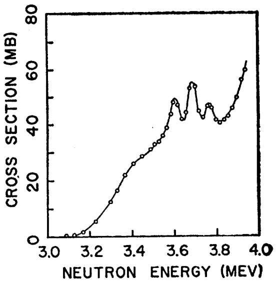

FIG. 4. Excitation function and absolute cross section for the reaction $F^{19}(n,\alpha)N^{16}$ obtained with the *p-T* neutron source. The energy resolution is approximately 30 kev.

Measurements have been made by Nereson and Darden[19] of the neutron total cross section for F^{19} from 3 to 12 Mev with an energy resolution of approximately 10 percent. In this energy range the total cross section is approximately 2 barns and is a smoothly varying function of energy except for the indication of a resonance at 4.4 Mev. It is probable that the group of resonances near this energy, observed in the $F^{19}(n,\alpha)N^{16}$ excitation function, are at least partially responsible for the total cross-section resonance.

Nereson and Darden obtained no evidence for a broad resonance at 5.9 Mev. Since a peak appeared at this energy in the (n,α) reaction excitation function but not in that for the (n,p) reaction, the *J*-values of the states of the compound nucleus must be such that α emission is favored at this energy. It is not possible to make any estimate of the spins or parities of these states on this basis, since α particles could be emitted, not only to the ground state of N^{16}, but to any of the

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[19] N. Nereson and S. Darden, Phys. Rev. **94**, 1678 (1954).

low-lying excited states,[14] the properties of which are not yet known.

The (n,α) reaction becomes energetically possible at a lower energy than does the (n,p) reaction, and, due to the difference in barrier penetration, the (n,α) reaction cross section is correspondingly higher for energies up to a few Mev above the (n,p) threshold. It is difficult, however, to explain the large difference in the cross sections at 8.0 Mev, at which energy both the α-particles and the protons may have energies greater than the barrier energy.

The cross sections obtained for the (n,α) and (n,p) reactions on F^{19} are of the same order of magnitude as the cross sections for reactions of this type on other nuclei.[1–9] The (n,α) reaction cross section measured at 3.9 Mev is 46±18 mb, which is in good agreement with the value 37±18 mb, obtained by Jelley and Paul[20] at this energy.

ACKNOWLEDGMENTS

The authors wish to express their appreciation of the continued assistance and encouragement given by Professor T. W. Bonner. Thanks are also due Dr. H. Bichsel for preparing and mounting the crystal.

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K-Capture - Positron Ratios for the First-Forbidden Transitions of Rb^{84} and the Relative Probabilities of *L*- and *K*- Electron Capture*

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The radiations from Rb^{84} have been investigated by means of scintillation coincidence spectrometer techniques in order to obtain the relative intensities of the various transitions. The energies and abundances of the radiations are 0.44-Mev β^- (5%), 0.81-Mev β^+ (10%), 1.70-Mev β^+ (9%), 0.89-Mev γ (64%), and 1.91-Mev γ (0.9%). [*Note added in proof.*—The β^- has been shown to be 0.91 Mev (2.5%).] Electron capture populates the level 1.91 Mev above the ground state, competes (54%) with the 0.81-Mev β^+ to populate the level 0.89 Mev above ground, and competes (21%) with the 1.70-Mev β^+ in effecting transitions to the ground state of Kr^{84}. The ratios of *K*-capture to positron emission for the transition to the 0.89-Mev level ($\Delta I=0$, yes) and to the Kr^{84} ground state ($\Delta I=2$, yes) are 5.15±0.38 and 2.06±0.36, respectively. These values are in qualitative agreement with theory. Nucleon configurations of the several states of Rb^{84}, Kr^{84}, and Sr^{84} are discussed. In the decay to the 0.89-Mev level an L/K-capture ratio of 0.12±0.05 was obtained by use of a value, 0.65, for the fluorescence yield of krypton. Alternatively, an experimental value for the fluorescence yield, 0.62±0.03, may be computed if the theoretical L/K-capture ratio is assumed.

THE radiations emitted in the decay of Rb^{84} have been studied by a number of investigators.[1–4] According to Huddleston and Mitchell,[4] two main positron groups are observed. The maximum energies and relative abundances were stated to be 1.63 Mev (0.39) and 0.82 Mev (0.58). A gamma ray of energy 0.89 Mev was found to be in coincidence with the lower energy positron group; the higher energy positrons were shown to represent a transition to the ground state of Kr^{84}. From the shape of the Fermi plot for the 1.63-Mev group, it was concluded that the transition is of the type $\Delta I=2$, yes. Moreover, since the spin-parity assignment for the ground state of Kr^{84}, an even-even nucleus, is 0+,[5] the ground state of Rb^{84} was given the assignment 2−. In agreement with this 2− designation and with the 2+ designation for the first excited state[5] of Kr^{84}, the Fermi plot for the lower energy positron group was found by Huddleston and Mitchell to have an "allowed shape." These authors did not observe any appreciable negative beta emission, although Beckham and Pool[2] reported that negative beta decay to Sr^{84} occurs with a probability 16 percent as great as that for positron decay. It may be noted that negative beta decay should be exoergic to the extent of ~0.5 Mev.[6] Karraker and Templeton,[3] who also observed negative electrons and positrons, reported the principal mode of decay to be electron capture; in other respects their observations are in essential agreement with those of Huddleston and Mitchell. Further, a gamma ray of energy 1.89 Mev has been observed[7] in the radiations from Rb^{84}. The best value of the half-life obtainable from the literature is 34 days.[3]

As pointed out by Huddleston and Mitchell, the

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