Neutron Absorption Cross Sections of Pa^{231} and Pa^{232}*

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Activation studies of Pa^{231} in the graphite region of the Materials Testing Reactor have resulted in a value of 200±15 barns for the thermal neutron absorption cross section of Pa^{231}. Resonance neutron effects were eliminated by the method of cadmium differences and by conducting the irradiations in a region where the ratio of resonance to thermal neutrons is small.

Measurements of the Pa^{232} and Pa^{233} activities produced in a highly intense neutron irradiation of Pa^{231} have resulted in a value of 760±100 barns for the absorption cross section of Pa^{232}.

I. INTRODUCTION

ADDITIONAL measurements of the thermal neutron cross section of Pa^{231} have been conducted in an effort to resolve the large discrepancies existing among previously reported values. At least six independent measurements, conducted over the past ten years, and giving values ranging from 107 to 290 barns, have been reported.[1–4] A rigorous intercomparison of these results is prevented by the widely varying conditions under which the measurements were conducted. Some measurements were based on determinations of the induced Pa^{232} activity, while others were based on alpha assays of the U^{232} daughter. The use of a wide variety of flux-monitoring techniques introduces additional uncertainties. In some instances effective flux values were computed from power-flux calibrations; in other instances fluxes were measured with gold foils calibrated in terms of a known neutron source strength; while in a third series of measurements the flux was calculated from the Pu^{238} content of Np^{237} monitors irradiated with the samples. An additional ambiguity, perhaps the most serious, concerns possible extreme differences in the energy distribution of neutrons in the various reactors used for the measurements.

The values reported here were based on absolute measurements of the Pa^{232} activity produced in a highly thermalized neutron irradiation of Pa^{231}. Resonance neutron effects were eliminated by conducting the irradiations in a region where the resonance neutron population was relatively low, and by the method of cadmium differences. Additional irradiations conducted in an extremely high neutron flux were used to evaluate the absorption cross section of Pa^{232}.

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[1] Q. Van Winkle, Argonne National Laboratory Report, ANL-4281 (unpublished).

[2] Seller, John, and Elson, Argonne National Laboratory Report, ANL-4282 (unpublished).

[3] A. H. Jaffey and Q. Van Winkle, Argonne National Laboratory Report, ANL-4283 (unpublished).

[4] Chalk River, Canada Progress Report, PR-CRR-24, 1954 (unpublished).

II. EXPERIMENTAL

A. Absorption Cross Section of Pa^{231}

About 25 microliters of a nitric acid solution of Pa^{231}, having a specific activity of 4×10^4 disintegrations/min ml, were sealed in quartz ampules and were irradiated for periods of up to 80 hours in a thermal neutron facility of the Materials Testing Reactor (MTR). In their irradiation positions, the ampules were located approximately on the vertical center line of the reactor and were separated from the lattice by approximately one foot of beryllium and four feet of graphite.[5] Moderation in the beryllium and graphite is sufficient to give a cadmium ratio of about 10^3 for gold.

Following the irradiations the samples were diluted with $6N$ HNO_3 and the protactinium was purified by a series of extractions with di-isopropylketone (DIPK) and thenoyltrifloroacetone (TTA), $0.05M$ in benzene. Finally, the protactinium was stripped from the TTA with a series of $0.1N$ HNO_3 washes and was concentrated by evaporation. The activities were prepared for absolute beta counting by vacuum evaporation from a tungsten filament onto aluminum foils having a weight area ratio of 120 micrograms/cm^2. In this manner uniform and extremely thin specimens of the activity were obtained. Beta-particle absorption in the sample itself may be regarded as negligible. A small fraction of the weaker particles are, however, absorbed in the backing material. Absorption studies made with foils equivalent in thickness to the backing showed the effect to be of the order of two percent. Accordingly, the activity measurements were corrected to this extent. Although corrections made in this manner are not exact, disintegration rate measurements involving such corrections are expected to be more reliable than those conducted on solvent evaporated deposits where sample localization effects, hence large self-absorption effects, are almost certain to occur.

The decay of the Pa^{232} was followed over a period of 10 half-lives with a windowless 4π beta counter. The presence of alpha emitters (the parent Pa^{231}) under an operating voltage corresponding to the midpoint of the beta plateau is noted by an increase in the background

[5] J. R. Huffman, Nucleonics **12**, No. 4, 20 (1954).

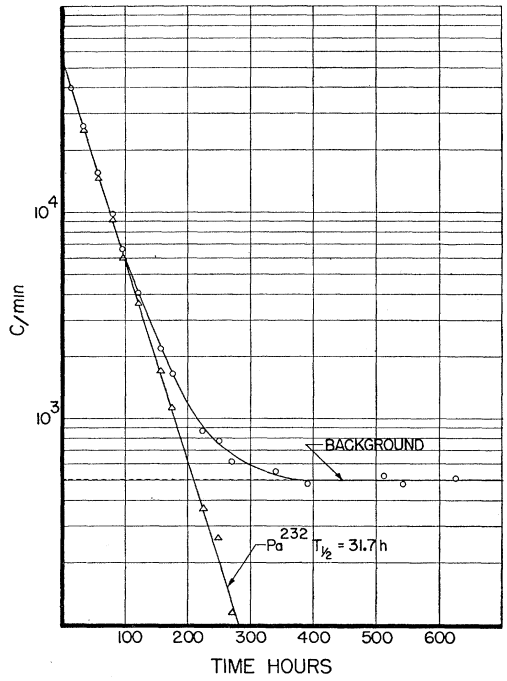

FIG. 1. Decay of Pa^{232} produced in a thermal neutron irradiation of Pa^{231}.

counting rate over the usual room conditions. This increase, however, presents no problem since the increased background may be established without ambiguity after the 1.32-day Pa^{232} has disappeared. A typical decay curve is given in Fig. 1. Coincidence loss corrections, amounting to 0.4 percent per 10^5 counts/min have been applied to the data shown. Subtraction of the background from the gross decay gives the decay of a single activity having a half-life of 1.32 days in agreement with the value reported by Jaffey and Hyde.[6]

Assuming the Pa^{231}/Pa^{232} ratio to be unchanged in the post-irradiation chemistry and in the process of foil preparation, the Pa^{232} produced per unit amount of Pa^{231} may be established from the 4π beta measurements and from direct observations of the alpha activity on a given foil. Measurements of the alpha activities were conducted with a 4π windowless counter under a voltage corresponding to the midpoint of the alpha plateau. Since the range of Pa^{231} alphas in aluminum is approximately fifty-fold greater than the backing thickness, practically all particles emitted under the 2π solid angle in the direction of the backing are recorded. From the range of Pa^{231} alphas in aluminum and the backing thickness, it may be shown that particles emerging at angles less than 1.3° in the direction of the backing are absorbed. Geometry considerations lead to a correction factor of 1.0 percent which may be applied to the activity measurements.

Particle counting under these conditions was shown to be quantitative by repeating the activity measurements in a conventional 2π alpha chamber. The $2\pi/4\pi$ counting rate ratio, averaged for several samples, was found to be 0.51, somewhat lower than the 0.52 geometry commonly assumed for alpha counting from infinitely thick backings. Because the exact backscattering factor for the 2π measurements was not known, the 4π measurements were considered more reliable and were, accordingly, used in the cross-section calculations.

The results of five runs conducted over a period of thirty days are summarized in Table I. A cadmium-covered irradiation, run No. 5, was used to establish the magnitude of the resonance neutron absorption effect. Column 2 lists the disintegration rate $A_{12}{}^0$ of Pa^{232} extrapolated to zero time and corrected to irradiation saturation. A correction amounting to two percent has been applied to account for beta absorption in the backing. Column 3 gives the Pa^{231} disintegration rate $A_{11}{}^0$ determined from 4π counting. A one percent correction has been applied to correct for absorption effects in the backing. An additional correction has been applied to correct for the growth of the alpha emitting U^{232} following the post-irradiation purification of protactinium. Column 4 lists the ratios of the Pa^{232} and Pa^{231} activities, $A_{12}{}^0/A_{11}{}^0$. Subtraction of the value of $A_{12}{}^0/A_{11}{}^0$ for run No. 5 (cadmium-covered) from the respective values for runs 1–4 gives the Pa^{232}/Pa^{231} activity ratios corrected for resonance activation, column 5. Column 6 lists values for the thermal neutron flux ϕ_{th} and column 7 gives the values of the thermal neutron absorption cross section σ_{th} found from the relation

$$\sigma_{th}=\frac{\lambda_{11}(A_{12}{}^0/A_{11}{}^0)_{th}}{\phi_{th}}. \qquad (1)$$

A simple average of the results gives as a final value $\sigma_{th}=200\pm15$ barns for the thermal neutron absorption (activation) cross section. The precision limits assigned to this value are based on estimated uncertainties of $+15$ and ±10 percent in the values used for $(A_{12}{}^0/A_{11}{}^0)_{th}$ and ϕ_{th}, respectively.

The thermal neutron flux values were established from measurements of the Co^{60} activity produced in Co flux monitors irradiated with the samples. Measurements of the specific ionization were carried out in a high pressure ionization chamber calibrated with National Bureau of Standards samples of Co^{60}. Conver-

TABLE I. Activation analysis of Pa^{231}.

Run No.	$A_{12}{}^0\times10^3$ dis/min	$A_{11}{}^0$ dis/min	$A_{12}{}^0/A_{11}{}^0$	$(A_{12}{}^0/A_{11}{}^0)_{th}$	$\phi_{th}\times10^{12}$ n/cm^2 sec	σ_{th} barns
1	14.2	53.3	266	263	0.831	205
2	66.4	232	286	283	0.852	215
3	18.1	71.2	254	251	0.886	176
4	73.3	252	291	288	0.914	203
5	0.95	351	3			

[6] A. H. Jaffey and E. K. Hyde, Phys. Rev. 79, 280 (1950).

sion of specific ionization values to thermal flux involved the use of the following constants: $T_{\frac{1}{2}}(Co^{60})=5.30$ years, and $\sigma_{th}(Co^{59})=37.0$ barns. Corrections for self-protection and flux depression were applied to the thermal flux values in the manner outlined by Hughes.[7]

B. Absorption Cross Section of Pa^{232}

A measurement of the pile neutron absorption cross section of Pa^{232} was made from determinations of the Pa^{232} and Pa^{233} activities produced in a prolonged high-flux irradiation of Pa^{231}. In this instance the sample was irradiated for 386 hours in a lattice position of the MTR in an average thermal flux of 1.56×10^{14} n/cm^2 sec. Following the irradiation the protactinium was purified as previously described with a series of DIPK and TTA extractions. Since Pa^{232} has a large thermal neutron fission cross section substantial quantities (activity-wise) of fission products relative to Pa^{233} are formed. Accordingly, special precautions were taken to eliminate those fission products whose activities would interfere with the determination of the 27.4-day Pa^{233}. Three DIPK extraction cycles followed by three with TTA were found adequate for this purpose. Zr^{95} which follows protactinium chemistry was eliminated by the addition of Zr holdback carrier prior to the solvent extraction cycles.

The decay of the sample, given in Fig. 2, is separable into two components, one corresponding to the 1.32-day Pa^{232}, and the other corresponding to the 27.4-day Pa^{233}. Corrections for resolving time losses and for room and counter background have been applied to the data shown. The identity of the 27.4-day activity as Pa^{233} was further verified by pulse-height analysis of the sample with a NaI(Tl) spectrometer. The absence of gamma active impurities, particularly Zr^{95}, was also demonstrated by the pulse height data.

The zero time counting rate intercepts of the separated activities, obtained from Fig. 2, may be used to evaluate the neutron absorption cross section of Pa^{232}. The production rates of Pa^{232} and Pa^{233} in a neutron flux of ϕ neutrons/cm^2 sec are given by

$$dN_1/dt=\phi\sigma_0N_0-(\lambda_1+\phi\sigma_1)N_1, \quad (2)$$

$$dN_2/dt=\phi\sigma_1N_1-(\lambda_2+\phi\sigma_2)N_2, \quad (3)$$

where the subscripts 0, 1, and 2 refer to Pa^{231}, Pa^{232}, and Pa^{233}, respectively, and where N, σ, and λ have their conventional meanings. The flux destruction terms $\phi\sigma_1$ and $\phi\sigma_2$ have been included since for fluxes of the order of 10^{14} n/cm^2 sec the product $\phi\sigma$ may be comparable in magnitude to λ.

Dividing the solution of Eq. (2) by that of Eq. (1) and multiplying the result by λ_2/λ_1 gives a lengthy but straight-forward expression for the Pa^{233}/Pa^{232} activity ratio in terms of ϕ, t, σ_1, σ_2, and the decay constants.

[7] D. J. Hughes, *Pile Neutron Research* (Addison-Wesley Press, Cambridge, 1953).

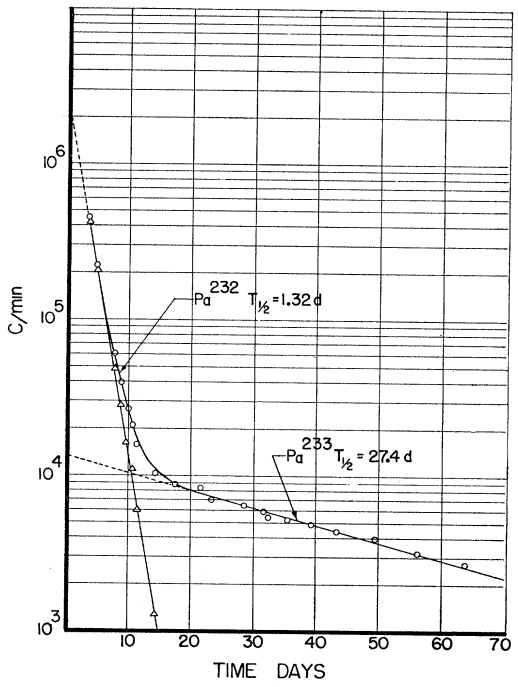

FIG. 2. Decay of Pa^{232} and Pa^{233} produced in a high-flux irradiation of Pa^{231}.

Substitution of values for these terms and a value of 5.82×10^{-3} (corrected for absorption in the backing) for the Pa^{233}/Pa^{232} zero time activity ratio (from Fig. 2) into this expression results in a value of 760 ± 100 barns for the absorption cross section of Pa^{232}.

The values used for the decay constants were those corresponding to half-lives of 1.32 and 27.4 days; the value used for ϕ was 1.56×10^{14} n/cm^2 sec; and t was taken as 386 hours. The value used for σ_2, i.e., the cross section of Pa^{233}, was 130 barns, an average of the 152-barn value reported by Halperin *et al.*[8] and the 107-barn value reported by Smith *et al.*[9]

III. DISCUSSION

Within the approximation that Co^{59} and Pa^{231} are $1/v$ absorbers in the thermal region, and that resonance absorption effects are eliminated by the method of cadmium differences, the cross section of 200 ± 15 barns for Pa^{231} corresponds to an energy for which the cobalt cross section was measured. Since the value used for Co was the thermal value of 37.0 barns, the cross section of Pa^{231} also corresponds to thermal energy, or 0.025 ev.

Since cadmium-covered irradiations were not conducted in the high-flux irradiations of Pa^{231} the value of 760 ± 100 barns for the Pa^{232} cross section must be

[8] Halperin, Stoughton, Ellison, and Ferguson, Atomic Energy Commission Report TID-2010, Vol. 3, No. 3, September, 1953 (unpublished).

[9] Smith, Passell, Reeder, Alley, and Heath, Atomic Energy Commission Report IDO-16226 (to be published).

regarded as that corresponding to a pile neutron distribution. This value is seen to be in considerable disagreement with the value of 40^{+40}_{-20} barns reported by Elson and co-workers.[10] No explanation for this large discrepancy is immediately apparent. However, the 760-barn value is consistent with that expected from cross-section systematics in this mass region.[11] Since Pa^{232} consists of an odd number of protons and an odd number of neutrons, the binding energy for an additional neutron is high; hence, an absorption cross section considerably higher than those of Pa^{231} and Pa^{233} is to be expected.

[10] Elson, Sellers, and John, Phys. Rev. **90**, 102 (1953).

[11] J. E. Evans, Phys. Rev. **96**, 845 (1954).

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$N^{14}(\alpha,n)F^{17}$ and $Na^{23}(\alpha,n)Al^{26}$ Reactions*†

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The $N^{14}(\alpha,n)F^{17}$ and $Na^{23}(\alpha,n)Al^{26}$ reactions have been studied by two methods, a proton recoil telescope and a slow neutron threshold detector. An energy level in F^{17} at 0.54±0.04 Mev has been observed and a ground state Q value of −4.76±0.07 Mev was determined. Energy levels in Al^{26} were found at 0.3, 1.0, 1.4, 1.8, 2.5, and 2.9±0.2 Mev and a ground-state Q value of −2.9±0.2 Mev was determined. In the latter reaction an excitation curve of the seven-second beta activity in Al^{26} has been observed with a threshold Q of −3.2±0.2 Mev, indicating the presence of an isomeric state in Al^{26} at about 300 kev.

I. INTRODUCTION

RECENT interest in the hypothesis of charge independence of nuclear forces and conservation of isotopic spin has made desirable the studying of nuclear level structure by several reaction routes. As mentioned in a previous paper,[1] (α,n) reactions have been neglected in the past. In the present work the reactions $N^{14}(\alpha,n)F^{17}$ and $Na^{23}(\alpha,n)Al^{26}$ are investigated.

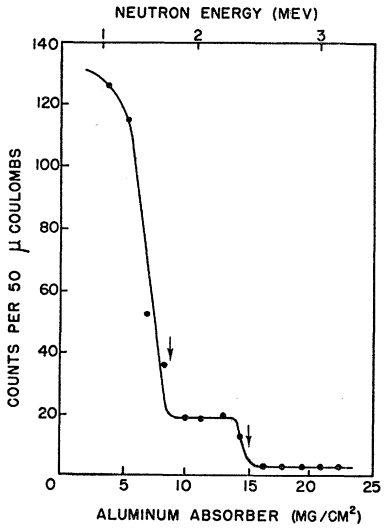

FIG. 1. Integral range curve of recoil protons from $N^{14}(\alpha,n)F^{17}$ reaction. Observation at 0°.

II. METHODS

The energies of the neutron groups were determined using a proton recoil telescope. The recoil protons from a hydrogenous radiator were absorbed in aluminum giving integral range curves, the geometry being determined by coincidences from two unpeaked proportional counters.

Threshold reaction energies for different levels were determined using an enriched BF_3 counter as a slow-neutron detector and varying the energy of the alpha-particle beam from the cyclotron in a helium range cell. A detailed description of these methods has been given in an earlier paper.[1]

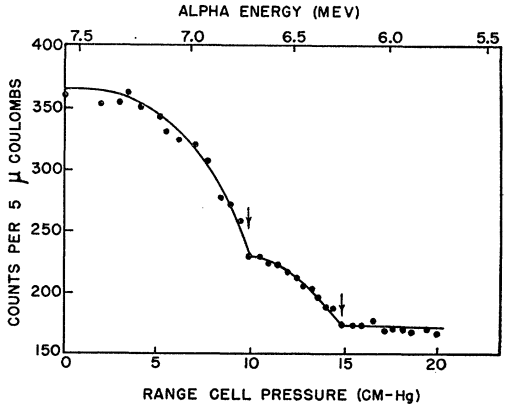

FIG. 2. Slow-neutron threshold curve for $N^{14}(\alpha,n)F^{17}$ reaction.

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[1] A. R. Quinton and W. T. Doyle, Phys. Rev. **101**, 669 (1956).