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Neutron Activation Cross Sections with Sb−Be Neutrons

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The neutron activation cross sections near 25 kev have been measured for about 50 nuclei. Gamma-ray scintillation spectrometers with known efficiency were used to compare the radioactivities produced by the several nuclei with I^{128}. This gave a significant increase in sensitivity and precision over direct beta-counting methods in many cases. Absolute standardization was obtained through scintillation-beta-counting the I^{128} in NaI(Tl) crystals irradiated with a neutron source calibrated by the manganese bath technique.

INTRODUCTION

Neutron capture in the kilovolt energy range has taken on increasing importance in cosmological theories of element formation as well as in the design of new types of fission reactors. Yet, quantitative measurement of the cross sections in this range remains a difficult technical problem. For the restricted class of nuclei in which a suitable radioactive product is formed by neutron capture, the beta or gamma activity produced offers a practicable approach to such measurements. Hummel, Hamermesh, and Kimball[1,2] have measured many activation cross sections with an antimony-beryllium source by comparison with thermal-neutron cross sections through beta activity. Long-counter extrapolations from a higher energy region form the basis for many of the other reported results.[3] The results reported here[4] were obtained with Sb-Be neutrons by determinations of individual gamma-ray intensities through the measurement of the associated peak areas.[5] Where the gamma-ray yield per disintegration was not well known, this quantity was redetermined by using pile-irradiated samples.

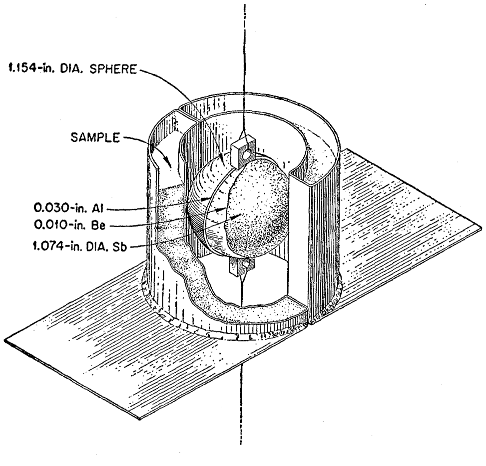

Fig. 1. Cutaway drawing of geometry in Sb−Be neutron irradiations.

EXPERIMENTAL

The determination of the iodine cross section by one of us (RLM) is recounted in Appendix I. All other cross sections were determined by comparison with iodine in split cylindrical-shell containers of about 40-ml capacity (for a pair). A spherical Sb−Be source (1.2-inch diameter) was pulled up into the approximate center of a pair of containers by a hand cord or, later, an electric motor drive, to effect an irradiation (see Fig. 1). The irradiations were done at a remote location, six feet or more from the floor of a large room, to avoid detectable activation by scattered and thermalized neutrons. The lead-shielded 3 in.×3 in. NaI(Tl) gamma-ray spectrometer was located in another building about a half-mile away. The transfer time (end of irradiation to start of count) was reduced to $2\frac{1}{4}$ minutes, which was adequate for the shortest activities measured. Samples were metal, oxide, or carbonate powders, or metal foils where possible. (See Appendix II for notes on special cases.) Samples were transferred to flat-bottomed glass beakers or folded into square packets for gamma counting. The use of flat samples facilitated corrections for gamma-ray attenuation which were generally below 25%. In a few cases of very weak radiation, essentially an infinite thickness of sample was required and the results depended directly on the absorption coefficients. In many cases the counting efficiency, including gamma-ray absorption, was checked experimentally by mixing in a known quantity of the same radioactive nuclide produced by pile irradiation.

RESULTS

Nuclei chosen for measurement were those for which the activation cross section could be expected to represent the total (n,γ) cross section (i.e., no known alternate mode of decay). The popular 54-minute In^{115} previously used as a standard[1,2] is an exception. The data are presented in Table I together with the energy of the gamma ray observed and its yield per disinte-

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[1] V. Hummel and B. Hamermesh, Phys. Rev. **82**, 67 (1951).

[2] C. Kimball and B. Hamermesh, Phys. Rev. **89**, 1306 (1953).

[3] For references, see D. J. Hughes and J. A. Harvey, *Neutron Cross Sections*, Brookhaven National Laboratory Report BNL-325 (Superintendent of Documents, U. S. Government Printing Office, Washington, D. C., 1955).

[4] See also Lazar, Lyon, and Macklin, Bull. Am. Phys. Soc. Ser. II, **2**, 15 (1957).

[5] Lazar, Davis, and Bell, Nucleonics **14**, No. 4, 52 (1956).

TABLE I. Sb−Be activation cross sections based on 820±60 mb for I^{127}.

Target isotope	E_γ (Mev)	γ/dis	σ^a (mb)	Target isotope	E_γ (Mev)	γ/dis	σ^a (mb)
Na^{23}	1.38	1.00	1.0±0.2	Ag^{107}	0.61	0.0225^a	930±180
Mg^{26}	0.83	0.70	<14	In^{115} (54 min.)	1.28	0.848	805±80
Al^{27}	1.78	1.00	1.4±0.2	Sb^{121}	0.57	0.664	950±100
Cl^{37}	2.1	0.47	1.1±0.2	Sb^{123}	0.603	1.00	456±46
K^{41}	1.52	0.20	<20	[I^{127}	0.455	0.172	820±60]
Ti^{50}	0.32	0.95	5±3	Ba^{138}	0.165	0.23^a	11.4±1.1
V^{51}	1.45	1.00	50±5	La^{139}	1.60	0.882^a	50±7
Mn^{55}	0.82	1.00	55±6	Ce^{140}	0.145	0.615	31±4
Cu^{63}	0.511	0.38	116±12	Ce^{142}	0.29	0.426^a	425±43
Cu^{65}	1.04	0.10	46±5	Pr^{141}	1.59	0.041^a	547±55
Ga^{71}	2.50	0.30	68±13	Sm^{152}	0.105	0.357	668±100
Ge^{74}	0.274	0.112	54±8	Sm^{154}	0.25	0.037^a	527±70
As^{75}	0.55	0.446^a	740±70	Gd^{158}	0.364	0.123^a	710±71
Br^{81}	1.03	0.255^a	550±55	Er^{170}	0.308+0.295	0.91	298±30
Rb^{85}	1.08	0.15	181±35	Hf^{180}	0.482	0.85	441±66
Rb^{87}	(1.85)	(0.23)	75±15	W^{186}	0.686	0.288^a	296±44
Zr^{94}	0.745	0.97	243±36	Re^{185}	0.137	0.105^a	2650±500
Zr^{96}	0.660 (Nb^{97})	1.00	22±4(?)	Re^{187}	0.155	0.162^a	970±200
Mo^{98}	0.14+0.18	0.72^a	209±21	Os^{190}	0.129	0.224	886±130
Mo^{100}	0.192	1.00	38±8				
	0.30 (Tc^{100})	1.00	45±9	Au^{197}	0.411	0.95	1120±110
Ru^{96}	0.216	0.92	321±60	Hg^{202}	0.279	0.828	57±13
Ru^{102}	0.498	0.90	386±39	Th^{232}	0.310 (Pa^{233})	0.90	500±100
Ru^{104}	0.73	1.00	211±21	U^{238}	0.210+0.270 (Np^{239})		610±61
Pd^{108}	0.088	0.0463^a	540±60				

[a] Our measurement.

gration. The half-lives used in correcting for decay were taken from Nuclear Science Abstracts.[6] A plot of the cross sections as a function of the mass number (A) is shown in Fig. 2. Qualitatively a rapid rise of cross section with $A<100$ is found, followed by a plateau, with dips near the magic neutron numbers. The two values at mass 96 differ by an order of magnitude, suggesting pronounced Z or N dependence. As an indication of the reproducibility of the measurements, the iodine gamma-activity calibration runs made during the course of the experiment are shown in Fig. 3. The departure from the half-life of the source is less than 1.3%. Comparable results were obtained in repetitions of other activations. Error estimates for the cross sections are taken as 10% except where high background or other circumstances dictated a less precise result. Agreement with the unpublished results of Linenberger[3] and others using Van de Graaff neutrons and long counters appears to be within the errors of measurement. The recent unpublished measurements of Ball *et al.* at Livermore with Sb-Be neutrons agree to better than 20%.* The results of Hummel, Hamermesh, and Kimball[1,2] agree generally within a factor of two.

DISCUSSION

Recent attempts (particularly by Cameron[7] of Chalk River) to predict (n,γ) cross sections in the kev region from nuclear models have met with fair success, giving average deviations of a factor of two or more. The precision of the present results seemed to warrant a more stringent test of some of the concepts involved. For several of the heavier nuclei measured, information on neutron widths, radiation widths, and level spacings for s-wave resonances has become available.[3,8–10] Using these data one can readily predict the average capture cross section at 25 kev for s-wave resonances.

In each case, this calculated value is approximately half the measured cross section. The difference can be attributed to resonances with higher angular momenta, particularly p-wave.[11] Using this information, Dresner[12] has derived p-wave average reduced neutron widths, using the Porter-Thomas distribution function and the following formulas:

$$\langle\sigma_{n,\gamma}\rangle=\sum_{J,l}\frac{2J+1}{2(2I+1)}\epsilon\frac{2\pi^2\bar{\lambda}^2}{D_J}\left\langle\frac{\Gamma_n(l)\Gamma_r}{\Gamma_r+\sum_{l'}\epsilon\Gamma_n(l')}\right\rangle,$$

where ϵ is the available number of channels (0, 1, or 2 for fermions) determined from I, J, l and parity conservation;

$$D_J=2\left(\frac{2I+1}{2J+1}\right)\bar{D}_{\mathrm{obs}},$$

where $\bar{D}_{\mathrm{obs}}$ is the average level spacing for *all* s-wave

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[6] Nuclear Data Group, National Research Council, Washington 25, D. C.

* *Note added in proof.*—Some results of Booth, Ball, and MacGregor are given in Bull. Am. Phys. Soc. Ser. II, **2**, 268 (1957).

[7] A. G. W. Cameron, Astrophys. J. **121**, 144 (1955).

[8] R. S. Carter *et al.*, Phys. Rev. **96**, 113 (1954).

[9] J. A. Harvey *et al.*, Phys. Rev. **99**, 10 (1975).

[10] J. S. Levin and D. J. Hughes, Phys. Rev. **101**, 1328 (1956).

[11] Recent studies of σ_{act} *vs* E_n at Duke University by E. Bilputch are also said to indicate p-wave contributions of this size.

[12] L. Dresner (unpublished).

resonances;

$$\Gamma_n = 2\gamma_n^2 \frac{(kR)^{2l+1}}{1+(kR)^{2l}} \quad (\text{for } l=0,\,1).$$

Here I and J are the spins of the target and compound nuclei.

Derived values of $\langle\gamma_n^2\rangle$ for $l=1$ are given in Table II. Total cross section data generally give a different estimate (significant upper bound) and are included in the data presented.

For a few of the lightest elements measured, where two or three known resonances should contribute most of the capture cross section near 25 kev, the measured values appear higher than those calculated from application of the Breit-Wigner isolated-level formula. This again suggests the possibility of appreciable p-wave capture from resonances not observable in total cross section measurements. This could be important in a sodium-cooled fast reactor, for example.

APPENDIX I. NEUTRON ACTIVATION OF IODINE NEAR 25 KEV[13]

Hummel, Hamermesh, and Kimball[1,2] have measured many activation cross sections with an antimony-beryllium source by comparison with thermal cross sections. Their 2.2-barn result for iodine (25-minute beta activation) seemed unexpectedly high. It seemed worthwhile to remeasure this cross section by absolute beta-counting in a NaI(Tl) crystal as done at higher energies by Martin and Taschek.[14]

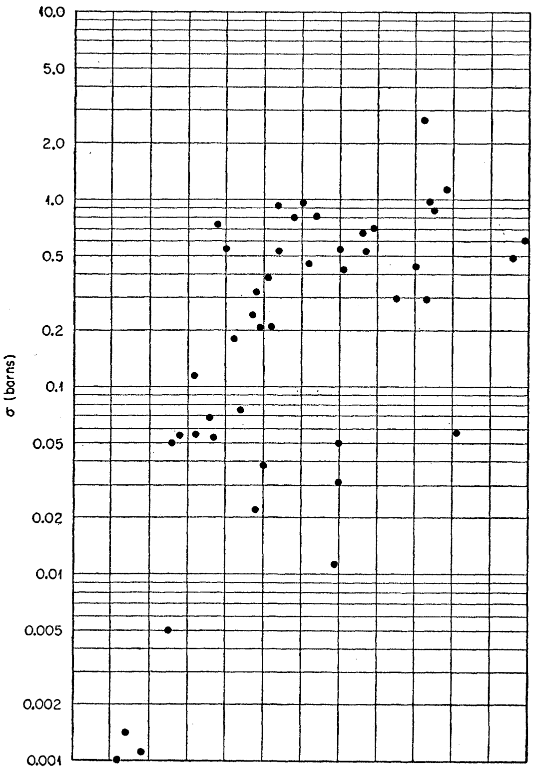

FIG. 2. σ_{act} for neutrons from Sb–Be source relative to $\sigma_{\text{act}}(\text{I}^{127}) = 0.82 \pm 0.06$ barn plotted as function of A.

A standard ORNL Sb–Be source (an Sb cylinder $\frac{3}{4}$ in. in diameter $\times\frac{3}{4}$ in. long in a hollow Be cylinder $1\frac{3}{16}$ in. in diameter $\times 1\frac{3}{16}$ in. long), calibrated at the National Bureau of Standards, was used. The source strength per unit solid angle was measured by rotating the source in front of a paraffin surrounded BF_3 counter. For the axial directions (used in the iodine irradiations) the flux was 0.915 ± 0.03 times the average. The equatorial flux, incidentally, was very near the average and constant with longitude as expected.

Several $1\frac{1}{2}$ in. $\times 1\frac{1}{2}$ in. NaI(Tl) crystals canned in aluminum foil with a thin glass window at one end were used in the experiment. Each was irradiated in an attic 25 feet above the concrete floor at 19 to 41 cm from the neutron source. The separation was carefully measured with a vertical cathetometer from a safe distance. After irradiation, the crystal was mounted on an RCA-5819 photomultiplier tube and the energy scale checked against the 661-kev Cs^{137} photopeak. The iodine beta activity was counted with an integral bias of about 140 kev. Room background at 1.8 counts/sec ranged from 8% to 33% of the beta counts. Extrapolation of

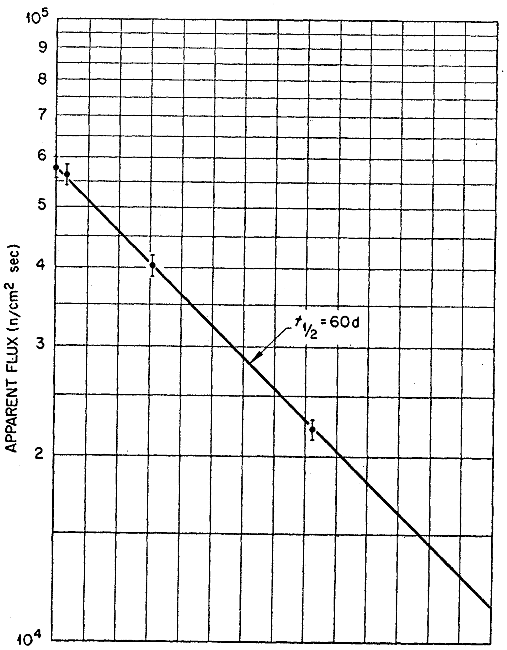

FIG. 3. Flux of Sb–Be neutron source as a function of time as determined by activation of I^{127} induced in NaIO_3 standards. (Standard deviation indicated is for counting statistics only.)

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[13] See also R. L. Macklin, Bull. Am. Phys. Soc. Ser. II, 1, 264 (1956).

[14] H. C. Martin and R. F. Taschek, Phys. Rev. 89, 1302 (1953).

TABLE II. *P*-wave reduced neutron widths calculated as outlined in text from experimental activation and total cross sections at 25 kev ($\langle\gamma_{n1}^2\rangle$ in ev). Value <0 implies *s*-wave contribution consistent with experimental data.

Nuclide	From $\sigma(n,\gamma)$			From σ(total)		
	$\langle\gamma_{n1}^2\rangle_{\text{most prob}}$	$\langle\gamma_{n1}^2\rangle_{\text{max}}$	$\langle\gamma_{n1}^2\rangle_{\text{min}}$	$\langle\gamma_{n1}^2\rangle_{\text{most prob}}$	$\langle\gamma_{n1}^2\rangle_{\text{max}}$	$\langle\gamma_{n1}^2\rangle_{\text{min}}$
Ag^{107}	1.91	12.1	0.418	1.55	4.65	0
In^{115}	3.49	17.8	1.01	1.50	2.54	0.606
Sb^{121}	20.4	∞	2.12	0.741	2.64	
Sb^{123}	12.6	∞	0.749	0.405	6.15	0
I^{127}	1.44	22.6	0.0815	<0	0	0
Re^{185}	1.94	∞	0.066			
W^{186}	4.80	∞	0.098	<0	18.3	
Re^{187}	0.065	1.21	0			
Au^{197}	5350	∞	13.7			
Th^{232}	0.448	0.940	0.0675	<0	0.573	0
U^{238}	0.694	5.26	0.202	<0	0.322	
W^{182}				<0	12.9	0
W^{183}				<0	2.57	0

the integral bias curve below 50 kev showed that the beta-counting efficiency under these conditions was 0.93 ± 0.015. The I^{128} decay has been studied recently at Columbia University[15] and a 6.9% electron-capture branch reported. About 95% of the electron-capture decay is to the ground state, and would not have been observed in the present experiment. The reproducibility of the activations corrected to saturated activity (and multiplied by the square of the separation) was not significantly different from the reproducibility calculated from the counting statistics and the cathetometer accuracy. The average of eight runs is considered reproducible to 1.6% (limit of error).

A few small effects were considered in calculating the cross section. Air scattering gave a computed 1.1% increase in neutron flux at the crystal. Effective center displacement for source and crystal led to corrections ranging from 0.2 to 1.0% at the extreme distances. The effect of multiple scattering in the crystal, calculated from the measured total cross sections of sodium[16] and iodine,[3] was +3.1%. The corrected $I^{127}(n,\gamma)I^{128}$ cross section is 0.82 ± 0.06 barn.

The primary energy of the neutrons is not well known.[17] The energy loss by collisions with beryllium has been estimated by using a homogeneous cylinder model with the primary source uniform throughout the outer beryllium-shell region of the actual source. A spherical beryllium-shell model ignoring the antimony gives comparable results. Table III shows the calculated percentage of emitted neutrons above fractions of the primary energy corresponding to maximum loss in 0, 1, 2, and 3 collisions with beryllium.

When one weights the detailed energy distribution by $(E_0)^{-\frac{1}{2}}$ corresponding to the expected $1/v$ iodine cross section dependence in the neighborhood of 20 kev (but with an upper limit of 3 for this factor), the area under the curve becomes 1.074. Thus neutron energy loss in the source increases the observed cross section by about 7.4%. Stated another way, the effective source energy is 87% of the primary energy. An effective neutron energy of about 25 kev as given by Kimball and Hamermesh[2] seems reasonable.

The early work of Linenberger and Miskel[3] at several energies gives an interpolated value of about 1 barn at 25 kev, with an uncertainty of perhaps 0.2 or 0.3 barn for the absolute value. The Hamermesh and Hummel value of 2.2 ± 0.4 barns obtained with an antimony-beryllium neutron source is certainly at variance with the present result. A few points of technique in favor of the present work, such as less ambiguity in incident flux measurement, high-efficiency beta-counting, and no reliance on a thermal cross section comparison, can hardly account for the difference.

One question that deserves a little thought in the present experiment is the possibility of self-absorption. Since the average probability of neutron absorption in the crystal was only 0.03 (i.e., 97% of the incident neutrons escape from the crystal), this at first glance seems negligible. If the absorption were due to a few narrow, isolated resonances one might still expect self-absorption to be important. Several of the iodine resonances below 100 ev have been examined.[3] Taking the gamma width (0.1 ev) and the extreme values of the reduced neutron widths (3×10^{-4} and 7×10^{-3} ev) found there as typical at 24 kev, one can show that the scattering cross section remains about ten times as large as

TABLE III. Percentage, *N*, of neutrons having energy greater than fraction, *E*, of primary energy after 0, 1, 2, and 3 collisions with beryllium.

Number of collisions	0	1	2	3
E	0.89	0.640	0.410	0.262
N(%)	67.6	93.6	98.6	99.2

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[15] N. Benczer *et al.*, Phys. Rev. **101**, 1027 (1956).

[16] H. W. Newson (private communication).

[17] Recent data on the beryllium (γ,n) threshold and the unpublished value of Tomlinson (1.692 Mev) for the antimony gamma energy give 27 kev for the primary energy. The gamma-ray momentum spreads this by ±5.5% (maximum) depending on the neutron emission angle. The presence of an additional gamma-ray at 2.1 Mev implies a second group of neutrons (<5% intensity, however) near 400 kev.

the absorption cross section at the (Doppler-broadened) peaks. The maximum peak absorption calculated is only about twice the average value found in the experiment. Since the average energy loss on scattering is about 400 ev and scattering is ten times as probable as absorption, the self-shielding at a single resonance must be completely negligible.

APPENDIX II. NOTES ON SPECIAL CASES

Mg^{26}.—The upper limit is based on failure to find a distinct gamma-ray peak and the poor statistics associated with this short-lived activity and low abundance (11.1%).

Cl^{37}.—A sample of $\mathrm{CCl_4}$ was used and a correction of a few percent was made for evaporation.

K^{41}.—The upper limit is set by K^{41} background in the gamma-ray spectrometer.

Cu^{63}.—A layer of unactivated copper was used around the sample during gamma-ray counting to absorb all of the positrons.

Br^{81}.—Lump $\mathrm{CBr_4}$ was used.

Rb^{85}.—The energetic beta-rays were also counted to increase sensitivity in this case. The efficiency was determined by mixing a known activity into inert $\mathrm{Rb_2SO_4}$ under the same conditions as in the Sb−Be irradiation.

Zr^{96}.—The cross section might be somewhat lower as the gamma-ray spectrum peak was distorted by other activities in this case.

Mo^{100}.—The Tc^{100} daughter activity was also observed, as indicated in Table I.

I^{127}.—$\mathrm{NaIO_3}$ was used.

Er^{170}.—The value of γ/dis is taken from yet unpublished work of M. E. Bunker. We would like to thank Dr. Bunker for permission to use his results before publication.

Re^{185} and Re^{187}.—$\mathrm{KReO_4}$ was used.

Hg^{202}.—$\mathrm{Hg_2Cl_2}$ was used.

Th^{232}.—The Pa^{233} daughter was extracted for the activity measurement.

U^{238}.—The Np^{239} daughter was extracted for the activity measurement.

PHYSICAL REVIEW VOLUME 107, NUMBER 2 JULY 15, 1957

B^{12}, C^{12}, and the Red Giants*

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Alpha-particle emission associated with the β decay of B^{12} has been detected and the energy spectrum investigated. These studies show that $(1.3\pm0.4)\%$ of all decays of B^{12} lead to the second excited state of C^{12} and that this state breaks up predominantly into three alpha particles with one alpha particle and the ground state of Be^8 as an intermediate stage in the process. The most probable spin and parity assignments for the state appear to be $J=0^+$, and analysis of the alpha-spectrum yields $Q(\mathrm{C}^{12*}-\mathrm{Be}^8-\mathrm{He}^4)=278\pm4$ kev, corresponding to an excitation energy in C^{12} of 7.653 ± 0.008 Mev. A new determination of the disintegration energy of Be^8 yields $Q(\mathrm{Be}^8-2\mathrm{He}^4)=93.7\pm0.9$ kev and hence $Q(\mathrm{C}^{12*}-3\mathrm{He}^4)=372\pm4$ kev. It is concluded, from the general principle of reversibility of nuclear reactions, that the second excited state of C^{12} as predicted by Hoyle is of a suitable character to act as a stellar thermal resonance in the Salpeter process, $2\mathrm{He}^4\rightleftharpoons\mathrm{Be}^8$; $\mathrm{Be}^8(\alpha,\gamma)\mathrm{C}^{12}$ under conditions expected in red giant stars.

INTRODUCTION

It has been suggested[1,2] that the fusion of alpha particles through the reactions

$$\mathrm{He}^4+\mathrm{He}^4\rightleftharpoons\mathrm{Be}^8,$$

$$\mathrm{Be}^8+\mathrm{He}^4\rightarrow\mathrm{C}^{12*}\rightarrow\mathrm{C}^{12}+\gamma,$$

or, more simply,

$$3\mathrm{He}^4\rightleftharpoons\mathrm{C}^{12*}\rightarrow\mathrm{C}^{12}+\gamma,$$

plays an important role in energy generation and element synthesis in red giant stars. These processes are believed to occur at a late stage of the red giant evolution in which the hydrogen in the central core has been largely converted into helium, and in which gravitational contraction[3] has raised the central temperature to $\sim10^8$ deg K, and the density to $\sim10^5$ g/cc. Under these conditions, as has been shown by Salpeter,[2] an equilibrium ratio of Be^8 to He^4 nuclei equal to $\sim10^{-9}$ is established. This conclusion followed from experimental measurements[4–6] which established the fact that Be^8

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* Supported in part by the joint program of the Office of Naval Research and the U. S. Atomic Energy Commission.

[1] E. J. Öpik, Proc. Roy. Irish Acad. **A54**, 49 (1951); Mem. soc. roy. sci. Liège **14**, 131 (1953).

[2] E. E. Salpeter, Astrophys. J. **115**, 326 (1952); *Annual Review of Nuclear Science* (Annual Reviews, Inc., Stanford, 1953), Vol. 2, p. 41.

[3] F. Hoyle and M. Schwarzschild, Astrophy. J. Suppl. **2**, 1 (1955).

[4] A. Hemmendinger, Phys. Rev. **73**, 806 (1948); Phys. Rev. **75**, 1267 (1949).

[5] Tollestrup, Fowler, and Lauritsen, Phys. Rev. **76**, 428 (1949).

[6] Early measurements by O. Laaff, Ann. Physik **32**, 760 (1938), and K. Fink, Ann. Physik **34**, 717 (1939) were analyzed by J. A.