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Pulse-Height Measurements of Recoils from $B^{10}(n,\alpha)Li^7$

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Thin films of boron deposited over a small area by vacuum evaporation have been exposed to the National Bureau of Standards thermal-neutron flux in pulse ionization chambers. Pulse ionization measurements in the past have yielded He^4-to-Li^{7*} pulse ratios considerably higher than the energy ratio of 1.753 from momentum conservation. Under suitable operating conditions we have obtained ratios as low as 1.778. Columnar recombination appears to cause most of this variation. The probability of the reaction $B^{10}(n,\alpha)Li^7$ going to the ground state was determined to be 6.52±0.05 percent.

I. INTRODUCTION

SEVERAL groups have conducted measurements of the average energy expended to produce an ion pair in argon as a function of energy and type of ionizing particle. The groups may be divided into two categories: one employing total ion collection and the other electron collection in pulse ion chambers or proportional counters. Jesse's[1] group, which has made extensive measurements of the total ionization in argon, found the ionization proportional to energy, independent of the particle type. On the other hand, various experimentalists employing electron collection have found a marked variation in the average energy expended per ion pair created. In general their results indicated that w increases as the ionizing density increases, either by the decreasing velocity of a given type of a particle or by the variation of specific ionization with z. For alpha particles in argon, Cranshaw and Harvey[2] have proposed the following formula:

$$w=27.5+1.9E^{-\frac{1}{2}},$$

where E is the initial alpha energy in Mev, and w is in electron volts.

The $B^{10}(n,\alpha)Li^{7*}$ reaction has permitted measurements of the ionization of particles at initial energies considerably less than alpha-particle energies from radioactive emitters, since the alpha particle and Li^{7*} ion have reaction energies of only 1.473 and 0.841 Mev, respectively. From conservation of momentum, the energy ratio $E_\alpha/E_{Li^{7*}}$ is precisely determined from the atomic masses and is 1.753. Using pulse-ionization techniques, the following ionization ratios have been reported. Stebler, Huber, and Bichsel:[3] 1.89±0.02; Hanna:[4] 1.841±0.025; and Rhodes, Franzen, and Stephens:[5] 1.878±0.014. These values exhibit a considerable departure from the ratio of particle energies, in conflict with Jesse's results. Our laboratory has investigated this reaction under a wide variety of experimental conditions with pulse-ionization chambers.

II. EXPERIMENTAL METHOD

The pulse-height distributions of recoils from thin films of boron exposed to a thermal neutron flux have been obtained in pulse-ionization chambers with argon-carbon dioxide mixtures.

A. Boron Films

Our laboratory has employed boron films (deposited by vacuum evaporation of natural boron from carbon crucibles) in pulse-ionization chambers for measurements in connection with the absolute calibration of the National Bureau of Standards standard neutron source. The films are deposited over an area 2.0×3.4 cm^2 on thin 15-mil quartz wafers which were initially coated with platinum to render them conductive.

The films are clamped to the inside of the cover plates of small pulse-ionization chambers.

B. Description of Chambers

Two similar pulse-ionization chambers were built, one of brass and one of aluminum, the latter being more suitable for absolute neutron flux measurements because of the low Al absorption cross section. The chambers have a pillbox design, $2\frac{5}{8}$ in. in internal diameter and $1\frac{1}{8}$ in. deep. Myvaseal gaskets were used for the vacuum seal between the cover plate and the chamber flange. A parallel plate mounted on teflon supports at a distance of 1.4 cm from the boron film was used for electron collection within the aluminum chamber. A carbon steel wire 1.14 mm in diameter, mounted with its axis 1.7 cm from the boron film, was the collecting electrode in the brass chamber.

After thorough cleaning with versene, water, and acetone, the chambers were connected to a copper vacuum system and evacuated to a pressure of 10^{-5} mm Hg. Normally the chambers were heated to a temperature of about 50°C for several hours with an infrared lamp until thoroughly degassed. To purify the gas the filling was made through an 850-cm length of copper tubing coiled in a trap containing butyl acetate and dry ice. A mixture of 5 percent carbon dioxide, 95 percent

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* Now with Picatinny Arsenal, assigned to Brookhaven Nationa Laboratory, Upton, New York.

[1] Jesse, Forstat, and Sadauskis, Phys. Rev. **77**, 782 (1950).

[2] T. E. Cranshaw and J. A. Harvey, Can. J. Research **26**, 243 (1948).

[3] Stebler, Huber, and Bichsel, Helv. Phys. Acta **22**, 362 (1949).

[4] G. C. Hanna, Phys. Rev. **80**, 530 (1950).

[5] Rhodes, Franzen, and Stephens, Phys. Rev. **87**, 141 (1952).

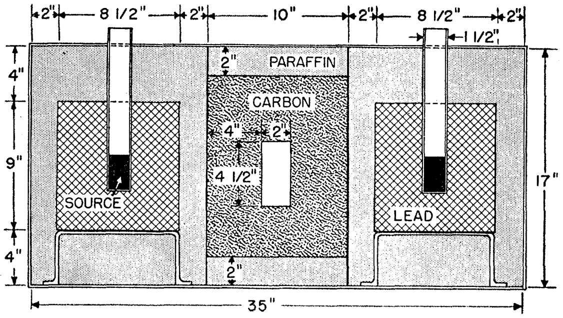

FIG. 1. NBS thermal neutron flux.

argon was used as the filling gas for most of the measurements.

C. Electronic Circuits

The chamber was connected by a cathode follower to a Bell-Jordan[6] preamplifier and amplifier system. When parallel plate collection was used, the first two amplifying tubes of the preamplifier were triode-connected to increase the signal-to-noise ratio. In terms of the particle energies measured, the amplifier rms noise for the collecting wire arrangement was 15 kev for the 20-μsec differentiating-time constant and 18 kev for the 2.0-μsec time constant. Amplifier noise was measured by feeding pulses through 1.35-$\mu\mu$f condenser to the collection electrode. A gain of roughly 200 000 was employed. Spurious counts were almost entirely eliminated by running the filament of the cathode follower attached to the chamber on dc. By heating the high coupling voltage and filter capacitors during the evacuation, counts from high-voltage breakdown were practically eliminated.

Pulses from the amplifier were fed into a three-channel discriminator built at the National Bureau of Standards. Periodic tests of the stability, linearity, and calibration of the system were made with a Model-100 pulser. Both amplifier and analyzer were connected to the line voltage through Sola constant-voltage harmonic transformers. Normally the analyzer was left on continuously, and drifts were small after the amplifier and chamber had warmed up for two hours or longer.

D. Description of Geometry

Exposures were made in the National Bureau of Standards standard slow-neutron flux (Fig. 1) which consists of a $2\frac{1}{8}$ in.$\times 4\frac{5}{8}$ in. slot in a moderating geometry between two equally-spaced one-curie Ra$-$Be(α,n) neutron sources which are imbedded in lead cylinders to reduce the gamma flux in the exposure region. Two inches of paraffin and four inches of carbon are between the cylinders and the slot. An aluminum holder, which slides into the slot, contains the pulse chamber and cathode follower. The holder exterior has been milled for the snug fit of a 0.020-in. Cd cover.

III. ANALYSIS OF DATA

Measurements were made with and without Cd, and the Cd difference counting rate was determined as a function of discriminator setting. This procedure almost entirely eliminates spurious counts from the data and the Cd difference counting rate is confined to the pulses resulting from the capture of slow neutrons (less than 0.3 ev) by boron. The Cd counting rate was about 2 percent of the difference counting rate. The pulse-height analyzer can be used as a two-channel differential analyzer or as a triple-integral discriminator unit. For the purpose of locating the leading edges of the He^4 and Li^{7*} pulses from thin films we found it more convenient to use the analyzer in the latter fashion. Assuming the energy of a particle emerging from a plane film is given

$$E=E_0\left(1-\frac{x}{R\cos\theta}\right),$$

where x is the distance from the origin of the particle to the top of the film, θ the angle of the particle direction with respect to the normal to the film, and R the range of the particle in the film material, the energy distribution of N_0 particles emitted per second in the upper hemisphere from a film of thickness L may be calculated to be

$$\frac{dN}{dE}=\frac{N_0R}{2E_0L}\quad\text{for}\quad E_0>E>E_0\left(1-\frac{L}{R}\right).$$

Half of the emerging particles are included in this interval. If discriminator readings are taken in this energy interval, the integral plot of counting rate *versus* discriminator setting permits a linear extrapolation to the leading edge of the pulses. Of course the amplifier noise (and emission of a gamma by the Li* in flight) causes a deviation of the observed counting rate from the extrapolated value near the maximum energy E_0

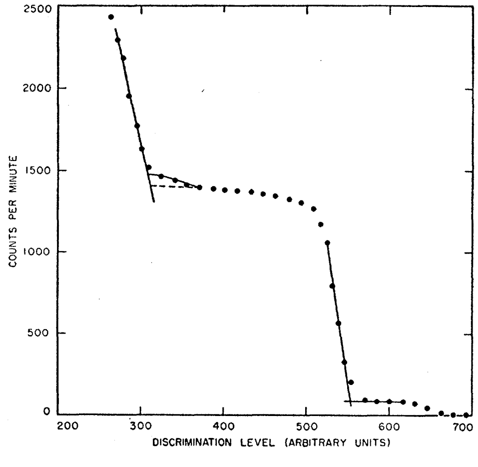

FIG. 2. Pulse-height distribution from B^{10} film, parallel plate collection, 1500 volts, 4 atmos. (99 percent A, 1 percent CO_2).

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[6] W. H. Jordan and P. R. Bell, Rev. Sci. Instr. 18, 703 (1947).

and points that fall within two rms amplifier noise values are excluded. Typical examples of the measurements for the two geometries are shown in Figs. 2 and 3. No attempt has been made to ascertain the leacing edges of the pulses from the reaction going directly to the ground state. The extrapolation of the Li* leading edge must be terminated at the top of the Li (ground-state) plateau. For relatively thick films this is somewhat difficult since the pulse spread due to noise tends to obscure the level where the extrapolation should be terminated; however, this level may be calculated from the counting rate on the alpha level since

$$\frac{dN}{dE}=\frac{N_0L}{2E_0R[1-(E/E_0)]^2}\quad\text{for}\quad E<E_0\left(1-\frac{L}{R}\right).$$

The path of the ionizing particle in the chamber gas, of course, alters the pulse height perceived at the collecting electrode since the pulse height is proportional to the mean-voltage drop of the ionization electrons divided by the total voltage drop between electrodes. For parallel plate collection the energy distribution of monoergic alphas from a film of infinitesimal thickness is

$$\frac{dN}{dE}=\frac{N_0D}{E_0R_m}\quad\text{for}\quad E>E_0\left(1-\frac{R_m}{D}\right),$$

where D is the plate separation and R_m the mean distance of the ionization electrons from the track origin measured along the track (electric center). Films employed were thin enough so that a linear extrapolation of the leading edges of the integral distribution to determine energy intercepts was valid for both geometries.

Usually a standard alpha emitter such as polonium (5.298-Mev alpha particle) is used to check chamber characteristics and the collecting voltage is varied until a plateau region is found in which the maximum pulse height appears constant. If particles whose average specific ionization is greater than the standard are being investigated, the most heavily ionizing particle should be used to check saturation. In the present case both the 1.473-Mev alpha particle and the 0.841-Mev Li* ion have been investigated for pulse saturation.

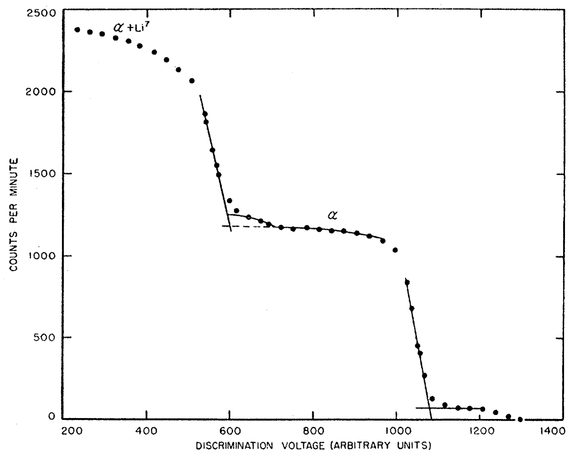

FIG. 3. Pulse-height distribution from B^{10} film, wire collection, 1200 volts, $1\frac{1}{2}$ atmos. (95 percent A, 5 percent CO$_2$).

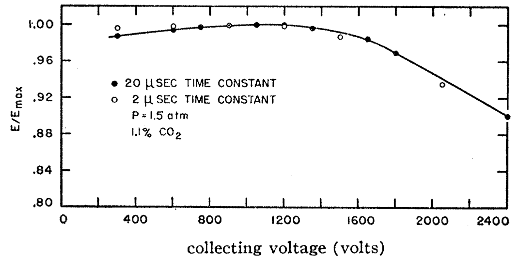

FIG. 4. Variation of pulse height of 1.473-Mev alpha ionization with collecting voltage, parallel plate chamber.

Carbon dioxide or methane (customarily 5 to 10 percent) is usually added to the argon in pulse ion chambers to reduce the random electron velocity by inelastic collisions to the region where electron scattering by argon atoms is minimal because of the Ramsauer effect and a higher drift velocity is thus obtained than with pure argon. When a mixture containing 1 percent of carbon dioxide was used at a pressure of $1\frac{1}{2}$ atmospheres, the alpha-particle pulse heights showed a maximum at 1050 v for parallel plate collection (Fig. 4) and a maximum and 300 v for the collecting wire geometry (Fig. 5). At this partial pressure of carbon dioxide, the mean free path of electrons between collisions with carbon dioxide molecules was great enough so that the electron velocity attained when the field was more than about 800 v/cm was high enough to cause appreciable scattering by the argon atoms and the resulting increased collection time caused loss in pulse height after amplifier differentiation. The steep gradient around the wire causes the optimum voltage to be reached much sooner than with the parallel plate geometry where the field is constant. For a filling with 5 percent carbon dioxide at $1\frac{1}{2}$ atmospheres, the optimum voltage for the collecting wire was discernible at 1200 v. Measurements were always made at voltages below the proportional region. At all other conditions tried (except the last listed), the pulse heights increased or remained constant with increasing high voltage. Some of the results are listed in Table I. The errors listed include an estimated calibration error of 0.5 percent.

IV. BRANCHING RATIO

If neutron capture by B^{10} gave monoergic alpha particles, the number of alphas escaping the film with energies greater than E would be given by

$$N_E=N_0\left[1-\frac{L}{2R(1-E/E_0)}\right]\quad\text{for}\quad E<E_0\left(1-\frac{L}{R}\right).$$

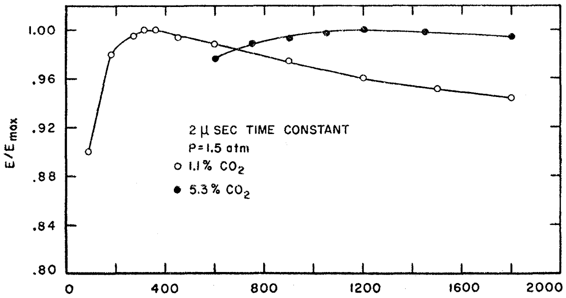

FIG. 5. Variation of pulse height of 1.473-Mev alpha ionization with collecting voltage, 1.14-mm diameter wire.

Since the alphas from the ground state have 20.7 percent more energy than those from the excited state, the range will be increased by 1.207, and to have the same thickness correction for both alpha groups it follows that

$$1-\frac{L}{2R(1-E^*/E_0^*)}=1-\frac{L}{2R(1-E/E_0)\times 1.207},$$

where the asterisks refer to the reaction going to the excited state. This expression reduces to

$$\frac{E^*}{E_0^*}=1.207\frac{E}{E_0}-0.207.$$

When the counting rates are measured on the two alpha plateaus at energy ratios determined by the above equation, the ratio of the counting rates will be independent of film thickness. For the conditions shown in Fig. 3 the ratio was determined at $E/E_0=0.875$ and $E^*/E_0^*=0.849$. (The ground-state alpha counting rate should be calculated at the point where the excited-state counting rate is determined to get the true difference. This refinement is unnecessary for thin films.)

Repeated measurement cycles were taken and the probability of the reaction going to the ground state, for thermal neutrons, was found to be 6.52±0.05 percent (the error quoted is the statistical probable error). This result compares with 5.8±0.1 percent obtained by Hanna,[4] 6.5±0.7 percent by Bichsel *et al.*,[7] and 5.9±0.9 percent by Rhodes *et al.*[5]

TABLE I. Pulse ionization ratios.

Chamber	Pressure (atmos)	Percentage of gas mixed with argon	Collecting voltage (volts)	$E_\alpha/E(\mathrm{Li})^*$ [a]
Parallel plate	4	1.1 CO_2	1500	1.795±0.015
Parallel plate	4	1.1 CO_2	3000	1.794±0.015
Parallel plate	4	5.3 CO_2	1900	1.82 ±0.02
Parallel plate	1.5	5.3 CO_2	1500	1.779±0.015
Parallel plate	1.5	1.1 CO_2	1050	1.777±0.015
Collecting wire	3	10 CH_4	1500	1.87 ±0.02
Collecting wire	1.5	5.3 CO_2	1200	1.824±0.015
Collecting wire	1.5	1.1 CO_2	310	1.834±0.015
Collecting wire	1.1	10 CH_4	1200	1.84 ±0.02
Collecting wire	1.1	5.3 CO_2	1500	1.80 ±0.02

[a] The large errors shown in the last column were for natural boron films.

V. DISCUSSION

As the pressure of the 5 percent carbon dioxide, 95 percent argon mixture was decreased from 4 atmospheres, the ratio of the He^4 to Li^{7*} pulse heights decreased from a value above 1.8 to a value approaching the conservation figure of 1.753. Columnar recombination has been reported high in carbon dioxide[8] and the decrease in pulse height with increasing pressure has been noted earlier.[9] When the carbon dioxide proportion was decreased to 1 percent and the pressure to $1\frac{1}{2}$ atmospheres, the optimum voltages for the maximum pulse heights were much lower and in the collecting wire geometry so low that considerable recombination occurred, especially for the Li^{7*} ionization electrons. At $1\frac{1}{2}$ atmospheres pressure, parallel plate measurements with both the 1 percent and 5 percent carbon dioxide mixtures yielded a He^4:Li^{7*} pulse ratio of 1.778 (av), which is 1.4 percent higher than the ratio of particle energies. Assuming the 0.841-Mev Li^{7*} ion has the same value of w as an alpha particle of the same velocity, the Cranshaw-Harvey formula gives a value of 1.823 for the ionization ratio. Our measurements indicate a considerably smaller variation in w and it is quite possible that some recombination was still occuring at our most favorable conditions, but the small size of our chambers precluded measurements at lower pressures. Unfortunately the variation in pulse heights with changing voltage due to the variation in conditions for minimum argon scattering tends to mask the recombination effect.

In conclusion, the lack of proportionality between pulse heights and particle energy observed in argon-filled pulse-ionization chambers seems largely due to columnar recombination. Since the maximum recombination occurs in the region where ionization density is greatest, alpha-particle ionization would be decreased by recombination most severely near the end of the range and w would appear to vary in the manner observed by Cranshaw and Harvey. Our measurements tend to corroborate Jesse's results made with total ion collection in pure argon in which the alpha-particle ionization was found proportional to particle energy. From our experience, a partial pressure of 0.04 atmosphere carbon dioxide is about optimum for parallel plate collection of the electronic component and should give less recombination than the usual 5 percent carbon dioxide, 95 percent argon in high-pressure applications.

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[7] Bichsel, Hälg, Huber, and Stebler, Helv. Phys. Acta **25**, 119 (1952).

[8] M. Moulin, thesis, Paris, 1910 (unpublished); Haeberli, Huber, and Baldinger, Helv. Phys. Acta **26**, 145 (1953).

[9] Bridge, Hazen, Rossi, and Williams, Phys. Rev. **74**, 1083 (1948).

For wire collection a greater partial pressure will be required.

It is possible to analyze the boron content of these films by quantitative chemical techniques and films of this type have been used to calibrate the National Bureau of Standards thermal-neutron flux to an error of less than 2 percent.

ACKNOWLEDGMENTS

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Influence of Chemical State on Stopping of Fast Charged Particles*†

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The suggestion is made to use the characteristic electron dispersion frequencies or corresponding wavelengths (λ_0) from molar refraction data for the prediction of influences of the chemical state on the stopping power of fast charged particles. Changes in λ_0 for atoms in different chemical states are transformed into changes in mean excitation potentials in the Bethe-Bloch theory and to changes in stopping power. Mass absorption data for Bi^{210} beta rays by Fournier are corrected for the anomalous Z/A-factor for hydrogen and then show the predicted effects for different groups of hydrocarbons. The very accurate data by T. J. Thompson on fast protons are compared with those from the "λ_0 approach" for a number of cases. Qualitative agreement is present for most of them but quantitatively the situation is less satisfactory. Certain favorable cases of large anomalies, not yet investigated, are predicted.

THE influence of the chemical state on the stopping power of fast charged particles was suggested by Bothe[1] to be caused by changes in the dispersion frequencies present in the Bohr stopping theory.[2] A more detailed and modern discussion[3] involves the oscillator strengths at different frequencies and its influence on the mean excitation potential in the Bethe-Bloch theory.

The problem of estimating changes in excitation potentials, without waiting for detailed information on the complete valence electron absorption spectrum of light, might be approached by evaluating the dispersion frequencies from *refractive index data*. This was done by Bohr[2] for H_2 and He, but the procedure should be generalized to polyatomic substances in any state of aggregation (except conductors). The characteristic frequency, ν_0 (or wavelength λ_0), is obtained for a variety of substances from molar refraction

$$R=\frac{n^2-1}{n^2+2}\frac{M}{d}=\text{const}\frac{1}{\nu_0^2-\nu^2}. \tag{1}$$

The value of λ_0 is a sort of mean value, the oscillator strengths of the various transition possibilities being taken into account.[4] As about the same excitation phenomena occur with light and with charged particles,[5] one should be allowed to attribute changes in λ_0 to corresponding changes in stopping power. The use of the ionization potentials may sometimes be of value but would not be general enough for our purpose.

Evaluation of λ_0 for the lightest elements in different chemical states by means of atomic[6] and molar refraction data gives a picture rather analogous to that of chemical refractometry; i.e., those compounds which show increments in refraction (due to absorption in the near ultraviolet[6]) also show increments in stopping power compared with some standard. Aromatic hydrocarbons are, for instance, expected to show higher stopping power than aliphatic ones.

In a study of the mass absorption coefficients of Bi^{210} beta rays for 17 hydrocarbons, Fournier[7] reported large increments (as much as 12 percent) but apparently neglected the anomalous Z/A-factor for hydrogen. After appropriate corrections, by use of a reasonable figure for the hydrogen mass absorption coefficient, the values for the increments are reduced to 3 percent or less. Grouping the data belonging to different hydrocarbon classes together, we find interesting differences as shown in Fig. 1. Actually the same sequence is expected from the λ_0-approach, although the experi-

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[1] W. Bothe, Jahrb. Radioakt. u. Elektronik **20**, 46 (1923); Handbuch der Physik (J. Springer, Berlin, 1933), Vol. 22, No. 2.

[2] N. Bohr, Phil. Mag. **25**, 10 (1913); **30**, 581 (1915).

[3] R. Platzman, *Symposium on Radiobiology*, Oberlin College, edited by J. Nickson (John Wiley and Sons, Inc., New York, 1952), Chap. 9.

[4] K. L. Wolf and K. F. Herzfeld, *Handbuch der Physik* (J. Springer, Berlin, 1928), Vol. 20.

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