Neutrons from C(d,n)N and Cu(d,n)Zn*

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(Received April 4, 1949)

The energy distribution among the neutrons emitted from carbon and copper bombarded by 10-Mev deuterons has been investigated by means of proton recoils in photographic emulsions. A description of the method is given. Two excited levels are observed in N^{13} at 2.29±0.12 Mev and 3.48±0.12 Mev, the latter not having been observed previously. Q for the production of N^{13} in the ground state is found to be −0.29 ±0.09 Mev. Both the reactions (d,n) and (d,$2n$) have been identified in the effects occurring when copper is bombarded by 10-Mev deuterons. Quantitative agreement with the expected energy range of the neutrons from the (d,$2n$) reaction is found. There is also evidence for some type of level structure in Zn. Discussion is given of a possible interpretation of this structure.

I. INTRODUCTION

THE energy spectrum of the neutrons from the reaction

$$Z^A+H^2\rightarrow(Z+1)^{A+1}+n \qquad (1)$$

yields information as to the energy of the state in which the nucleus $(Z+1)^{A+1}$ is formed. The special emulsions manufactured by Ilford, Ltd., and Eastman Kodak have made it possible to use the photographic plate as a detector of such neutrons. The hydrogen present in the gelatin of the emulsion (1.4 percent by weight) furnishes an adequate supply of protons to be knocked on by impinging neutrons. These protons are then automatically recorded as they render developable the silver bromide grains along their paths. Measurement of the track lengths under magnification gives a measure of the neutron energy. Application of this method to the general reaction (1) has been made by a number of workers.[1]

The energy of the neutrons given out in the reaction

$$C^{12}+H^2\rightarrow N^{13}+n \qquad (2)$$

has been studied by several investigators using deuterons of energy up to 2 Mev.[2] The results of these experiments are all in agreement in reporting a single group of monoenergetic neutrons and a Q-value of −0.27±0.02 Mev (Bennett and Richards). Hornyak and Lauritsen[3] list an excited state of N^{13} at 2.34 Mev. With deuterons of energies no greater than 2 Mev, this state could not have been excited. Using the 10-Mev deuterons available from the Washington University cyclotron, the neutrons from (2) have been studied by means of photographic plate detectors. Deuterons of this energy should be capable of forming N^{13} in several of its excited states.

The neutrons from the reactions

$$Cu^{63,65}+H^2\rightarrow Zn^{64,66}+n, \qquad (3)$$

$$Cu^{63,65}+H^2\rightarrow Zn^{63,65}+2n, \qquad (4)$$

are of interest for two reasons. First, information as to the excited states in the Zn nuclei may be obtained from a study of the energy spectrum of neutrons resulting from reaction (3). Second, neutrons from both reactions are present in profusion near a deuteron-accelerating cyclotron because of the large amount of copper in the dees and associated components of the machine. To plan proper shielding against neutrons originating in the cyclotron, it would be helpful to know their energy distribution. Reactions (3) and (4) have been studied using the 10-Mev deuterons available at this laboratory.

II. EXPERIMENTAL METHOD

A. The Deuteron Source

The fringing field of the cyclotron magnet combined with a specially designed slit system is used to isolate a monoenergetic beam of deuterons[4] for use in the investigation. The beam then passes down a long snout to a point well away from the cyclotron where experimentation can be carried out with adequate shielding. The energy of these deuterons was determined by a photographic method, in which the beam was scattered by a thin platinum foil and allowed to strike an emulsion at a glancing angle. The distribution in range of the recorded deuteron tracks was then determined and converted to an energy scale. In this way, the monoenergetic beam selected for use in these experiments was found to have an energy of 9.93±0.07 Mev with a spread of less than one percent.

B. Apparatus

Figure 1 shows schematically the experimental arrangement. The homogeneous beam of deuterons strikes a thin target, and the neutrons from the resulting reaction are detected at 90° to the incident beam by the

* Assisted by the joint program of the ONR and the AEC.

[1] H. T. Richards, Phys. Rev. **59**, 796 (1941); C. F. Powell, Proc. Roy. Soc. **181**, 344 (1943); R. A. Peck, Phys. Rev. **73**, 947 (1948).

[2] T. W. Bonner, Phys. Rev. **53**, 496 (1938); W. E. Bennett *et al.*, Phys. Rev. **59**, 781 (1941); W. E. Bennett and H. T. Richards, Phys. Rev. **71**, 565 (1947).

[3] W. F. Hornyak and T. Lauritsen, Rev. Mod. Phys. **20**, 191 (1948).

[4] E. C. Creutz and R. R. Wilson, Phys. Rev. **59**, 916 (1941).

proton recoils in the photographic plate as shown. The main beam of deuterons passes on through the thin target and emerges into air through a 1-mil aluminum window. Hence the beam dissipates most of its energy at a considerable distance from the detection plate, thus reducing background fog in the emulsion due to gamma-radiation. This is an important point since the exposures needed to obtain adequate yields of neutrons are quite long.

The plates used for detection are encased in light tight steel holders which are in turn mounted on a rigid support as shown. This support is a circular disk on which several plate holders can be mounted radially with respect to the target. The detecting plates are outside the cyclotron vacuum system.

Adequate collimation of the neutrons results from the fact that the photographic plate is almost tangential to the neutrons leaving the target. The plates are at a mean distance of 10 cm from the center of the target. Shielding from extraneous neutrons from the cyclotron chamber is achieved by the use of large paraffin blocks cast to fit into place on all sides of the photographic plates, leaving a narrow path open between plate and target for the free passage of the neutrons to be studied.

C. Emulsion Technique

The best exposure of a photographic plate to neutrons is one which yields an adequate number of recoil proton tracks with a minimum of background fog to interfere with their measurement. In order to reduce this background to a minimum, one inch of lead shielding was used on that side of the plate nearest the cyclotron. Deuteron beam currents of about 0.25 microampere were used with exposure times of approximately one hour for the target thickness employed.

The plates were developed in the usual manner employed for Ilford C-2 plates and were examined by microscope. For all scanning and measuring, a magnification of 900× was used with bright field illumination. The eyepiece micrometer employed in measurement of the tracks was a special one made by Bausch and Lomb. The divisions were ruled with a diamond point such that the rulings themselves were only two microns in width. This specification was made so that the thickness of the division markings would not interfere in estimating the last significant figure in track lengths. Calibration of the eyepiece micrometer was made with a Bausch and Lomb stage micrometer.

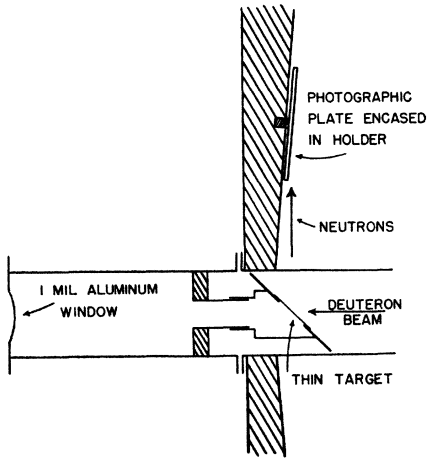

FIG. 1. Schematic arrangement for the study of neutrons from (d,n) reactions by means of photographic plates.

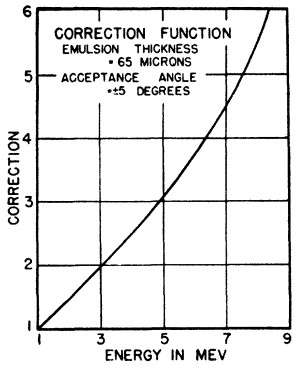

FIG. 2. Correction function consisting of geometric and cross-section corrections applied to the proton recoil spectra obtained experimentally in order to infer the correct neutron distribution.

The criteria for accepting a track for measurement were: (1) the direction of the track should be within 5° of the forward direction of the neutrons in both the horizontal and vertical planes; (2) a track should clearly begin and end within the emulsion. Tracks were measured by scanning across the plate in the direction of the incident neutrons, taking swaths at 0.2-mm intervals.

III. EXPERIMENTAL RESULTS AND CONCLUSIONS

A. Carbon

A thin carbon target (165-kev thickness for 10-Mev deuterons), was prepared by depositing soot from burning camphor on to a sheet of 0.1-mil platinum. This target was subjected to deuteron bombardment and the neutrons from reaction (2) recorded photographically. The raw data, as obtained from the measurement of recoil proton track lengths, must be altered in order to yield the neutron energy spectrum. To transform the range distribution to an energy distribution, recourse is made to a range-energy curve for the emulsion. This calibration has been made for Ilford emulsions by Lattes and his co-workers[5] and their results were used in this study.[6] The energy distribution histo-

[5] Lattes, Fowler, and Cuer, Proc. Phys. Soc. **59**, 883 (1947).

[6] Confirmation of the relative constancy in stopping power of Ilford emulsions has been demonstrated by agreement of independent workers in reporting alpha-particle ranges in the emulsions from natural radioactive elements. A batch taken at random at this laboratory yielded an alpha-particle calibration curve differing only by 0.3 percent from that obtained by the Bristol group. It has become the practice here to check the stopping power of a given batch of plates by impregnation with uranium and subsequent measurement of the UI and UII alpha-particle ranges.

TABLE I. Neutron groups from $C^{12}(d,n)N^{13}$ and the assignment of excitation energies to N^{13}.

Group	Mean energy of neutrons (Mev)*	Q (Mev)	Excitation energy of N^{13} (Mev)
A	7.53±0.07	−0.29±0.09	0.00
B	5.40±0.05	−2.58±0.08	2.29±0.12
C	4.30±0.04	−3.77±0.08	3.48±0.12

* Mean energy of neutron groups calculated following the procedure of Bethe and Livingston, Rev. Mod. Phys. **9**, 245 (1937).

gram was plotted in 200-kev intervals. In order to overcome any observer bias in measurement and to lend smoothness to the curve, the energy histogram was overlapped, i.e., the number of tracks per 200-kev interval was calculated and plotted every 100 kev. Finally, in order to infer the correct neutron spectrum, two corrections must be applied: (1) one which recognizes that the probability of recording a short track is greater than that of recording a long one, and (2) one which gives the variation of the cross section for neutron-proton scattering as a function of the neutron energy. Calculation of such correction functions have been discussed by other authors[7] and will not be repeated here. A plot of the correction used in this work is given in Fig. 2.

Figure 3 is the corrected neutron spectrum of reaction (2) representing measurement of 1500 recoil proton tracks in 2.3 cm² of plate area. Three groups are clearly resolved. To insure that none of these neutrons were extraneous, a "blank" run was made with only the platinum backing in place. The background so obtained

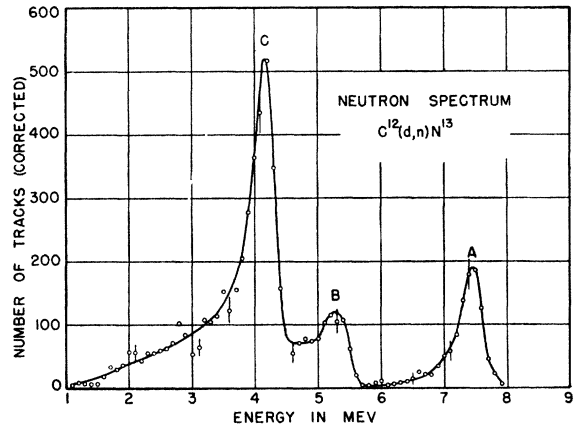

FIG. 3. Energy distribution of neutrons from $C^{12}(d,n)N^{13}$. The number of tracks in each 200-kev interval have been plotted every 100 kev and corrected according to Fig. 2. Background neutrons (see text), amounting to 100 percent of the total number of tracks measured below 1.5 Mev, ~50 percent of those in the interval 2–3 Mev, 5 percent at the peak of group C, 12 percent at group B, and zero at group A, have been subtracted prior to plotting. The standard deviation is shown at several typical points on the curve. The spectrum represents 1500 acceptable recoil proton tracks in 2.3 cm² of plate area.

[7] H. T. Richards, Phys. Rev. **59**, 796 (1941); W. M. Gibson and D. L. Livesay, Proc. Phys. Soc. **60**, 523 (1948).

was subtracted from the carbon run and the data appearing in Fig. 3 is the resulting pure carbon spectrum. Group A corresponds to the formation of N^{13} in the ground state. The Q of this transition, as calculated from the mean energy of group A, is −0.29±0.09 Mev as compared to −0.27±0.02 Mev as found by Bennett and Richards.[2] Groups B and C correspond to the formation of N^{13} in excited states. For the level associated with group B, the energy of the excited state is found to be 2.29±0.12 Mev which is in satisfactory agreement with that listed by Hornyak and Lauritsen[3] (2.34 Mev). Group C yields a level in N^{13} at 3.48±0.12 Mev, as yet not reported in other reactions involving N^{13}. Results are listed in Table I.

B. Copper

A copper foil, of thickness 0.4 mil, was bombarded by 9.93-Mev deuterons and information as to the energy of the resulting neutrons was recorded as in the case of carbon. Figures 4 and 5 represent the neutron spectrum so obtained from 1300 acceptable tracks.

At the bombarding energies used, neutrons from both the (d,n) and $(d,2n)$ reactions should be present. The threshold of the reaction $Cu^{63}(d,2n)Zn^{64}$ has been reported as 7 Mev by Livingston and Wright.[8] Meitner gives a value of 5.5 Mev as calculated from the end point of the energy spectrum of beta-particles in the decay of Zn^{63}.[9] Neutrons produced by the stripping of deuterons should also be present at this bombarding energy, as predicted by Peaslee.[10] However, a calculation of the mean energy to be expected of these neutrons at 90° to the incident beam can be made. Assuming a momentum distribution for the neutron inside the deuteron as that given by Serber[11] and a Coulomb barrier of 7 Mev for copper, a rigorous evaluation of the mean energy of stripped neutrons gives a value of ~1 Mev at 90°. Examination of Fig. 4 will indicate that these neutrons do not give a serious contribution to the observed spectrum.

A calculation of the spread in energy of the neutrons from the $(d,2n)$ reaction can be made. Following Bethe,[12] we suppose the mechanism, in the case of the most abundant (70 percent) isotope, Cu^{63}, to be

$$H^2+Cu^{63}\rightarrow Zn^{64*}+n+Q_1, \quad (5)$$

$$Zn^{64*}\rightarrow Zn^{63}+n+Q_2. \quad (6)$$

If we assume the neutron in (5) is evaporated with negligible energy, the neutron energy in (6) is found to be 3.9 Mev at 90°. The reverse situation, in which the neutron in (6) comes off with very small energy, can be arrived at if one knows the binding energy of the last neutron in Zn^{64}, and hence the minimum excitation

[8] R. S. Livingston and B. T. Wright, Phys. Rev. **58**, 656 (1940).
[9] L. Meitner, Arkiv. f. Mat. Astr. o. Fys. **33A** No. 3 (1946).
[10] D. C. Peaslee, Phys. Rev. **74**, 1001 (1948).
[11] R. Serber, Phys. Rev. **72**, 1008 (1947).
[12] H. Bethe and M. S. Livingston, Rev. Mod. Phys. **9**, 245 (1937).

energy needed for evaporation. A consideration of the masses involved yields ~13 Mev for this binding energy. Consideration of the nuclear energy surface by Feenberg,[13] predicts a value of ~12 Mev for this nucleus. Taking a mean value of 12.5 Mev for the excitation of Zn^{64}, reaction (5) will then yield a neutron of 3.5 Mev. Thus the region from zero to about 3.9 Mev in the spectrum will correspond to the production of two neutrons by deuterons on Cu^{63}. Similar calculations for the other target isotope of copper, Cu^{65}, yield the region zero to about 5.6 Mev for double neutron production. An examination of Fig. 4 reveals a Maxwellian appearance of the curve for two particle emission. An end point might well be assigned somewhere in the neighborhood of 4 Mev, in good agreement with the prediction on Cu^{63}, since a majority of the neutrons will come from that more abundant isotope. From Fig. 5 it is seen that after undergoing a slight rise, the curve falls still further to a minimum value at approximately 5.5 Mev. This is in qualitative agreement with the predicted end point of the (d,$2n$) reaction in Cu^{65}.

Above 5.5 Mev, the spectrum shows definite grouping of the neutrons. Neutrons of these energies correspond to a (d,n) reaction and hence some sort of level structure in Zn^{64} or Zn^{66} is indicated. Neutrons arising from production of these isotopes in the ground state would have energies around 16 Mev as calculated from the mass values involved. These were not detected, which is not surprising since the probability is small that Zn be formed in the ground state with the excitation energies available.[14] The probability of detection in the emulsion is also very small at these energies. The fact that a group at 8.6 Mev was the highest observed can be interpreted only by saying that neutrons above this energy are produced too infrequently to be detected by the present method in the length of exposure used.

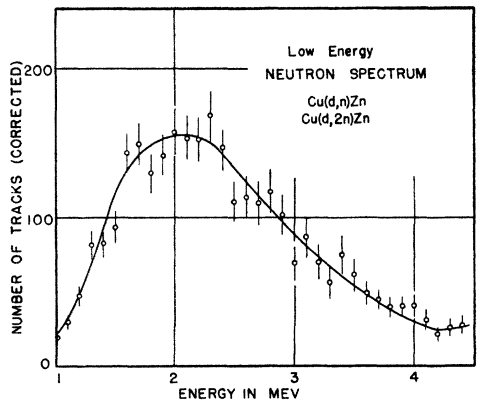

FIG. 4. Low energy distribution of neutrons from Cu(d,n)Zn and Cu(d,$2n$)Zn. The number of tracks in each 200-kev interval have been plotted every 100 kev and corrected according to Fig. 2. The spectrum represents 1000 acceptable recoil proton tracks in 2.0 cm² of plate area.

[13] E. Feenberg, Rev. Mod. Phys. **19**, 239 (1947).
[14] H. A. Bethe, Rev. Mod. Phys. **9**, 100 (1937).

TABLE II. Neutron groups from Cu(d,n)Zn and assignment of a mean energy to "level groups" in Zn^{64} or Zn^{66}. Neutron groups are numbered reading from right to left in Fig. 5. Values of the energy are given to only two significant figures, corrections affecting the third figure having not been applied.

Group	Mean energy of neutrons (Mev)	Mean excitation energy (Mev) Zn^{64}	Zn^{66}
I	8.5	6.3	9.1
II	6.4	8.5	11.3
III	5.7	9.2	12.0
IV(?)	4.6	10.3	—

Existing statistical theories of nuclei in the neighborhood of Zn predict extremely close spacing (a few hundred ev) of the levels whose energy above the ground state (~6–10 Mev) would correspond to the production of the neutrons in Fig. 5. Hence in order to account for the apparently definite grouping observed, one might suppose the compound nucleus, Zn^{65}, to be formed in states with a certain limited range of angular momenta. Transitions might then be made to an allowed range of states in Zn^{64} with the emission of neutrons whose energies are not homogeneous but spread over a finite range. Such a mechanism, with transitions of the compound nucleus to several allowed "ranges of states" in the residual nucleus, would account for the distribution in Fig. 5. Still another interpretation is possible if one assumes the density of highly excited levels in medium-heavy nuclei to vary irregularly with their energy above the ground state. Such assumptions, however, are highly speculative with the limited experimental information at hand.

The mean excitation energy of three "groups of levels" in Zn can be obtained from the mean energy of the neutron groups in Fig. 5. Corrections to these energies, which affect the third significant figure, have not been made, since the peaks in Fig. 5 presumably do not represent homogeneous groups and the usual corrections for straggling, etc., cannot be applied. Hence, only two figures are given in Table II which gives the

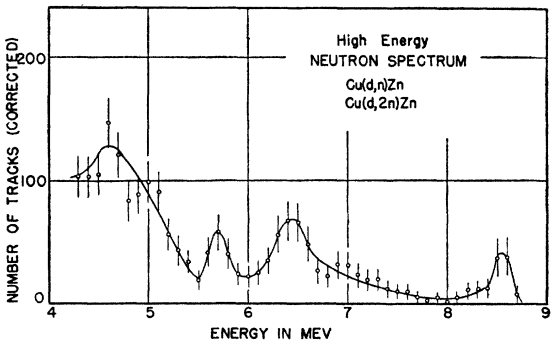

FIG. 5. High energy distribution of neutrons from Cu(d,n)Zn and Cu(d,$2n$)Zn. The number of tracks in each 200-kev interval have been plotted every 100 kev and corrected according to Fig. 2. The spectrum represents 300 acceptable recoil proton tracks in 4.1 cm² of plate area.

mean excitation energies of "level groups" according to whether they be assigned to Zn^{64} or Zn^{66}, unambiguous assignment being possible only if experiments are made with separated Cu isotopes.

The author should like to make acknowledgment to Dr. F. N. D. Kurie for his stimulating interest in the early stages of this problem; to Dr. H. Primakoff and Dr. E. Feenberg for helpful discussions, and to Miss Eileen Denison for measurement of track lengths in the copper spectrum.

PHYSICAL REVIEW VOLUME 76, NUMBER 4 AUGUST 15, 1949

Fading of Proton Tracks in Vacuum*,**

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(Received April 11, 1949)

The fading of the latent images due to protons has been compared for photographic emulsions maintained in air and in vacuum after the exposure was made. The fading in vacuum was found to be approximately 10 percent of that in air indicating a purely chemical origin for the phenomenon of fading. At the present time it is speculative whether the residual 10 percent arises from the same source in which case it could be eliminated by merely improving the vacuum, or comes from an altogether distinct process. Some suggestions are advanced concerning a possible Ag atom evaporation process.

THE fading of tracks caused by charged particles has been well established by the Bristol group *et al.* Although it is improbable that the phenomenon is confined exclusively to latent images formed by charged particles as distinct from photons, in the former case it has proved more consequential by imposing a limit on the effective exposure times which can be utilized (notably in cosmic-ray research).

A significant question arises as to whether the dissipation of the latent image has a chemical or physical origin or involves two distinct processes. There could be a reoxidation of the Ag of the latent image by the atmosphere, possibly involving the water vapor, CO_2, etc. Alternatively the Ag atoms may "evaporate" from the Ag speck[1] constituting the latent image, and rediffuse into the AgBr lattice. The former alternative seems to be favored by the following observations:

(a) The rate of fading of alpha and proton tracks was conspicuously greater in summer than in winter in St. Louis. (Summer temperature $\backsimeq$30°C and relative humidity $\backsimeq$75 percent, winter $\backsimeq$22°C and $\backsimeq$39 percent.)

(b) Under constant temperature conditions ($\backsimeq$30°C) fading was accelerated by artificially raising the humidity and retarded by lowering the humidity.[2]

(c) Elevating the temperature sufficiently, produces rather than destroys a latent image.[3] Evidence for this comes from two sources,—the spontaneous "fogging" of an emulsion which increases rapidly with temperature and also the latent image resulting from frictional contacts[4] with an emulsion, explainable by a thermal mechanism.

A logical *coup de grace* to either theory would be supplied by comparing the rate of fading of tracks in vacuum and in air under the same temperature conditions.

Kodak Nuclear Track Plates NTB and Ilford Nuclear Research Plates C2, both 50μ, were exposed to fast neutrons from the Washington University cyclotron providing abundant proton recoils in each emulsion. Some of these plates were developed immediately. The remainder were divided into two groups and were allowed to stand as follows: (1) In vacuum. The container was evacuated with a mechanical pump every few days. The pressure varied from $\simeq$0.1–10-mm Hg. These plates were exposed to atmospheric pressure only for the initial half hour after bombardment and for the few minutes necessary to occasionally remove a plate for developing. (2) In air. The experiment was performed during the winter (temperature $\simeq$22°C, relative humidity $\simeq$39 percent). Both groups of plates were at the same temperature. A developing technique was standardized to permit identical development of successive plates.

The plates were scanned for proton tracks ending in the emulsion and having a range of at least 300μ (corresponding to a proton energy $\simeq$7 Mev). Grain counts per unit length were made on each track commencing from the end of the track where the protons stopped. However, it was found that the grain density towards the end of the track was too great to permit accurate counting. Less subjective data were obtained by excluding the ends of the tracks. In the final analysis

* A more comprehensive account of fading experiments will be published in *Review of Scientific Instruments*.

** Assisted by the joint program of the ONR and AEC.

[1] Consider the latent image mechanism in this case to comprise at least two competing primary processes—thermal liberation of electrons, and evaporation of Ag atoms from sensitivity specks.

[2] Dependence on seasonal humidity has recently been verified for alpha-tracks: R. Coppens, J. de phys. et rad. **10**, 11 (1949).

[3] However this could well be the net effect of the two primary processes and does not exclude the possibility of fading by evaporation proceeding simultaneously.

[4] K. B. Mather, J. Opt. Soc. Am. **38**, 1054 (1948).