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A Neutron-Capture Theory of the Formation and Relative Abundance of the Elements*,**,***

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There exists an inverse relationship between the relative abundance of nuclear species in the universe and the neutron-capture cross sections of these nuclei. Abundant nuclei have small capture cross sections, and *vice versa*. On the basis of this correlation, a non-equilibrium theory of the formation of the elements is developed in which the elements are built up by a process of successive neutron captures. The coefficients in the equations of this theory involve the neutron-capture cross sections of the elements, and for this reason relationships between capture cross sections, atomic weights, and neutron energies are obtained from available data.

According to this theory, the primordial material was a gas of neutrons only. As the universe expanded, neutrons decayed to protons and electrons; the capture of neutrons by protons then led to deuterons. These nuclei in turn captured neutrons, and progressively heavier nuclei were formed. The neutron content in these nuclei was controlled by beta-decay between successive neutron captures.

The physical conditions which are indicated for the period of element formation are inconsistent with a cosmological model of the early stages of the universe based on matter only. It appears that the early stage was probably a universe of radiation with a trace of matter present. According to this picture, the element-building process began some 200 to 300 seconds after the start of the expansion, at which time the temperature was of the order of 10^9°K, and the density of matter was of the order of 10^{-3} g/cm^3. Because of the expansion of the universe, and because of the decay of neutrons, the production of elements must have been essentially complete in a time of the order of magnitude of the neutron decay lifetime. Preliminary calculations based on this theory successfully predict the observed relative abundance data.

I. INTRODUCTION

It is the purpose of this paper to describe a neutron-capture theory of the formation and relative abundance of the elements. That a neutron-capture process was responsible for the formation of the elements is suggested by the relative abundance data themselves. In fact, it will be shown that there exists an inverse relationship between the relative abundance of nuclear species and their cross sections for fast neutron capture, i.e., abundant nuclei have small capture cross sections.

In developing the neutron-capture theory, it is accepted that the elements were formed, and their relative abundances determined, in a pre-stellar stage of the universe. Bethe[1] has shown in his work on the energy production in stars that the stars must have been formed with essentially their present composition, except for H and He. Since most stars apparently acquired their present form quite early in the history of the universe, the elements must have been produced near the beginning of the universal expansion, some two or three billion years ago. Further evidence on this point is provided by the present existence of naturally radioactive isotopes. If the assumption is made that the stable and radioactive isotopes of a given element were of equal abundance when formed, one may compute from known decay constants and present relative abundance data when formation occured. In all cases the age of the elements is calculated to be of the order of several billion years. It is to be expected that the physical conditions prevailing in the early stages of the expanding universe played an important role in the formation of the elements.

It was suggested first by Gamow[2] that the present relative abundance of elements is the result of a non-equilibrium process. An apparently significant correlation between relative abundance and nuclear binding energies—abundant nuclei exhibiting large binding energies, and *vice versa*—suggested to a number of other investi-

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*** A preliminary report of this work was given by R. A. Alpher, H. A. Bethe, and G. Gamow, Phys. Rev. **73**, 803 (1948).

[1] H. A. Bethe, Phys. Rev. **55**, 434 (1939).

[2] G. Gamow, Phys. Rev. **70**, 572 (1946).

gators that the observed relative abundance of elements corresponds to thermodynamic equilibrium between nuclei at some high temperature and density in prestellar matter or in certain types of stars.[3] However, equilibrium theories have failed to give a reasonably simple explanation of the origin of light and heavy elements.

The exposition of the neutron-capture theory is developed according to the following plan. The relative abundance data are briefly described in Section II. The correlation of large abundance with small neutron-capture cross sections, and *vice versa*, is discussed in Section III, and a general expression relating capture cross sections, atomic weights, and neutron energies is obtained for use in the mathematical formulation of a neutron-capture theory. In Section IV, a theory of successive neutron captures is formulated, and preliminary calculations described which indicate that this theory successfully represents the observed relative abundance data. The cosmological implications of the neutron-capture theory are discussed in Section V. In addition, details of the abundance data not directly explained by this theory are discussed, and some of the difficulties of the theory are indicated.

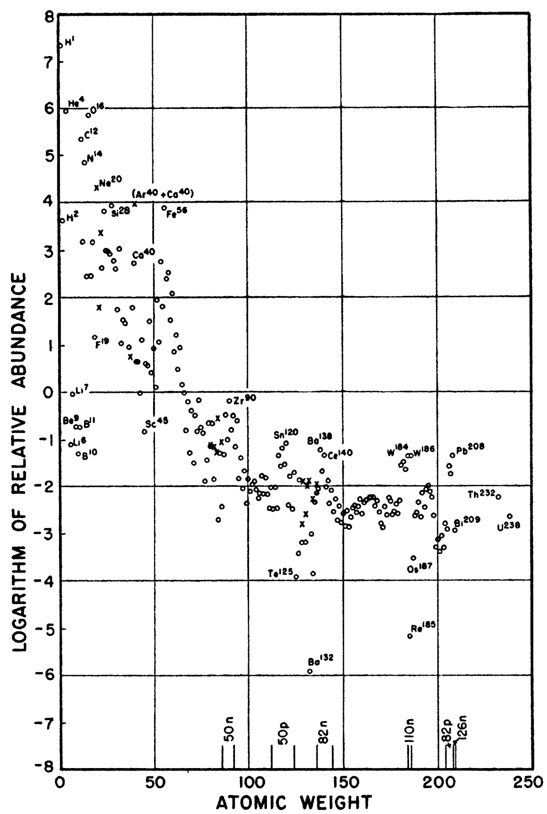

FIG. 1. Relative abundance of the elements in the universe, according to Goldschmidt (1938). Short-lived naturally radioactive elements are not plotted. Crosses indicate interpolated abundances which include the noble gases Ne, Ar, Kr, and Xe. Atomic weight ranges of "magic number" nuclei are shown on the abscissa.

II. DESCRIPTION OF RELATIVE ABUNDANCE DATA

The most recent, published tabulation of the observed relative abundance of the elements is due to Goldschmidt.[4] Recently, Brown[5] announced a revision of Goldschmidt's tabulation based in part on a new procedure for analyzing the composition of meteorites. It is reported that this as yet unpublished work does not alter the major features of the data as given by Goldschmidt.

Goldschmidt's data are reproduced in Fig. 1. The logarithms of relative abundance (number of atoms of a given atomic weight per 10,000 atoms of Si) are plotted against atomic weight. Isobaric abundances have been added in preparing the plot. According to Goldschmidt, the abundances may be reasonably interpreted as applying to the universe as a whole since the composition of the universe is generally considered to be homogeneous on a large scale. While the abundances of the noble gases are not well determined in the universe, they are fairly well determined on earth. On the basis of these terrestrial abundances, Goldschmidt has interpolated for the universal abundances. On Fig. 1, crosses are used to denote relative abundance values which include these interpolated noble gas data.

The many members of the radioactive families

[3] The following is a partial list of papers on the equilibrium theory: T. E. Sterne, Mon. Not. R. Astr. Soc. **73**, 736, 767, 770 (1938); C. von Weizsäcker, Physik. Zeits. **39**, 633 (1938); S. Chandrasekhar and L. R. Henrich, Astrophys. J. **95**, 288 (1942); G. Wataghin and P. S. de Toledo, Phys. Rev. **73**, 79 (1948); G. B. van Albada, Bull. Astr. Inst. Neth. Vol. **X**, No. 374, Sept. 12, 1946. See also Astrophys. J. **105**, 393 (1947); O. Klein, G. Beskow and L. Treffenberg, Ark. Mat. Astr. Fys. **33A**, No. 1 (1946); G. Beskow and L. Treffenberg, Ark. Mat. Astr. Fys. Pt. I, **34A**, No. 13 (1947); Pt. II, **34A**, No. 17 (1947); J. Géhéniau, I. Prigogine, and M. Demeurs, Physica **13**, 429 (1947); F. Hoyle, Mon. Not. R. Astr. Soc. **106**, 343 (1946).

[4] V. M. Goldschmidt, "Geochemische Verteilungsgesetze der Elemente. IX. Die Mengenverhältnisse der Elemente und der Atom-Arten," I. Matematisk-Naturvidenskapelig klasse, 1937, No. 4. (Oslo, 1938.)

[5] H. S. Brown, Bull. Am. Phys. Soc. **23**, 10 (1948).

are not plotted in Fig. 1. The observed abundances of the daughter elements derived by successive disintegrations from parent elements of the series are in excellent agreement with the predictions of established statistical laws of radioactive disintegration. Only the observed abundance of the long-lived parent elements and perhaps the abundance of the daughter elements, when originally formed by processes other than the decay of parent elements, require explanation.

It should be noted that the elements Li, Be, and B are quite scarce compared to other elements in their region of atomic weight. As will be explained later, this scarcity probably arises from thermonuclear reactions, different than those by which the elements were formed, and subsequent to the time of element formation. The scarcity of Ba^{132} and Re^{185} is not verified by Brown's tabulation, and is therefore open to question.

Ranges of atomic weights corresponding to the "magic number" nuclei have been indicated on the abscissa. The "magic number" nuclei, containing 50 and 82 protons, or 50, 82 and 110 neutrons, exhibit a completed "shell" structure which is not yet understood on a theoretical basis.[6] Nuclei containing 126 neutrons, essentially coincident with 82 proton nuclei, have also been classed as "magic number" nuclei.[7] As will be discussed below, these nuclei, which are quite abundant according to Fig. 1, are found to possess small neutron-capture cross sections.

The principal difference between terrestrial and universal abundances is that H and He are much less abundant on earth than in the universe. The reason for this does not appear to lie in the theory of the formation of the elements, but in the mode of formation of the planets. For example, in Weizsäcker's theory of the formation of the planetary system,[8] H and He would have been captured by the condensing planets only with difficulty. Since these elements probably constituted more than 90 percent of the planet-forming material, this conveniently accounts for the angular momentum problem in planetary theories.

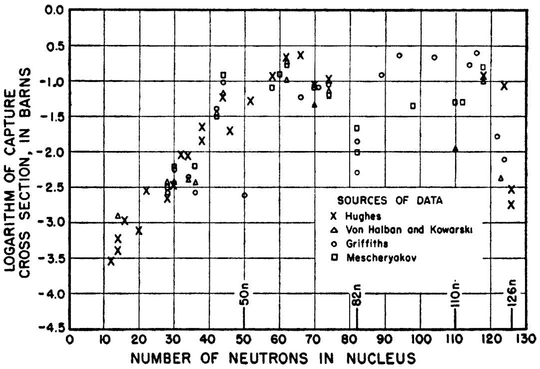

FIG. 2. Illustration of small capture cross sections associated with "magic number" nuclei. Data of von Halban and Kowarski, Griffiths and Mescheryakov are fitted to Hughes' data at 1 Mev for Ag^{107}.

III. NEUTRON-CAPTURE CROSS SECTIONS

When the relative abundances of nuclear species are plotted *versus* atomic weight A, as in Fig. 1, it is found on the average that elements of even A are about ten times more abundant than elements of odd A.[9] There are not sufficient experimental data to demonstrate that nuclei of odd A have larger capture cross sections then those of even A. It is reasonable, however, to expect that this is so. Because of the well-known even-odd variation of nuclear binding energies, nuclei will more readily capture neutrons if the product nucleus is even. Feenberg[10] in his recent work on the semi-empirical theory of the nuclear energy surface, has arrived at the rule that odd A nuclei with even charge are favored to have relatively larger capture cross sections, which agrees with the observation that such nuclei are less abundant.

Wigner and Way[11] have pointed out that the "magic number" nuclei should exhibit small capture cross sections, because these nuclei exhibit some kind of completed shell structure on an isotopic number plot. In Fig. 2 are given some experimental data on capture cross sections *versus* the number of neutrons in the capturing nucleus, illustrating the small cross sections for such

[6] H. A. Bethe, Rev. Mod. Phys. **8**, 82 (1936); G. Gamow and C. L. Critchfield, *Theory of the Atomic Nucleus and Nuclear Energy Sources* (Clarendon Press, Oxford, in print).

[7] M. G. Mayer, Phys. Rev. **74**, 235 (1948). *Note added in proof:* Since our paper was written, the work of Mayer indicates that nuclei with 110 neutrons are probably not "magic number" nuclei but rather that nuclei with 126 neutrons are to be so considered.

[8] C. von Weizsäcker, Zeits. f. Astrophys. **22**, 319 (1944). Reviewed by G. Gamow and J. A. Hynek, Astrophys. J. **101**, 249 (1945), and S. Chandrasekhar, Rev. Mod. Phys. **18**, 94 (1946).

[9] W. D. Harkins, Phys. Rev. **38**, 1270 (1931).

[10] E. Feenberg, Rev. Mod. Phys. **19**, 239 (1947).

[11] E. P. Wigner and K. Way, private communication.

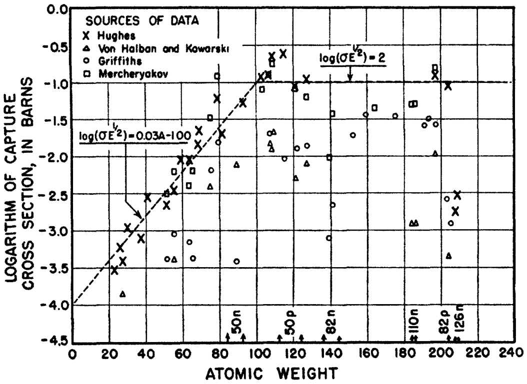

FIG. 3. Capture cross sections measured as a function of atomic weight. Data of von Halban and Kowarski and of Griffiths are extrapolated to 1 Mev, the energy at which Hughes' data are measured, by the $1/v$ law, from their energies of measurement. Mescheryakov's data are fitted to Hughes' data at Ag^{107}. Dotted lines are from cross section formulae fitted to Hughes' results and used in calculations of neutron-capture theory.

nuclei. The data are those of von Halban and Kowarski,[12] Griffiths,[13] Mescheryakov,[14] and Hughes.[15] In plotting Fig. 2, the first three sets of data were adjusted to fit Hughes' data at Ag^{107}, his data being considered the most accurate in absolute value.

An outstanding feature of the relative abundance data in Fig. 1 is the approximately exponential decrease in abundance with increasing A up to $A\cong 100$, with relative constancy for higher values of A. It is expected then that the neutron capture cross sections of the elements should increase rather rapidly with increasing A up to $A\cong 100$, and become essentially constant for higher A. The four sets of data described above are replotted *versus* atomic weight in Fig. 3, and it may be seen that the capture cross sections behave in the manner suggested. In Fig. 3, Mescheryakov's data are fitted to Hughes' value for Ag^{107}, while the other data were extrapolated to 1 Mev from the energies at which they were measured by means of the $1/v$ law. Figure 3, and cross section data in general, indicate the validity of the $1/v$ law for the capture cross sections of the elements for medium fast neutrons. We shall be interested principally in medium fast neutrons, and will accept the $1/v$ law, therefore, in combining cross section data. The correlation between capture cross sections and relative abundance is further illustrated by Fig. 4, where the capture cross sections measured by Hughes at 1 Mev are plotted against the relative abundance of the particular nuclei as given by Goldschmidt.

It appears from Fig. 3, and from Hughes' data in particular, that the cross sections of the elements may be represented approximately by[16]

$$\log_{10}(\sigma E^{\frac{1}{2}}) = -30.886+0.03A, \quad \text{for } A<100, \quad (1a)$$

and

$$\log_{10}(\sigma E^{\frac{1}{2}}) = -27.886, \quad \text{for } A>100, \quad (1b)$$

if σ is in cm^2 and E is in ergs. The inclusion of E in Eqs. (1) implies the $1/v$ law, and the validity of the variation of σ with atomic weight for all energies of interest here. The cross section data available substantially verify this latter point over the range of medium fast neutrons. Fluctuations from Eqs. (1) in Fig. 3 are due for the most part to the "magic number" nuclei, to the lack of monochromatic neutrons in most of the experiments, and to the difficulty of determining the absolute neutron flux.

In the theory to be developed, Eqs. (1) are used to describe the neutron-capture cross sections of the elements. For purposes of preliminary calculations, therefore, we have used a "smoothed" fit to the cross section *versus* atomic weight data, and have in fact ignored as detailed variations the even-odd dependence of cross sections, the small cross sections associated with the "magic number" nuclei, and the cross sections which are known for many elements.

One further assumption concerning the capture cross sections of the elements is implied. The nuclei built up in the process to be described must have been formed initially with a neutron excess greater than those observed, say, in the known Fermi-elements. In order to use Eqs. (1),

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[12] H. von Halban and L. Kowarski, Nature **142**, 392 (1938). Used 220-kev photo-neutrons produced by ThC″ γ-rays in heavy water.

[13] J. H. E. Griffiths, Proc. Roy. Soc. **170**, 513 (1939). Used 40-kev photo-neutrons produced by Ra γ-rays on Be.

[14] M. G. Mescheryakov, C. R. Acad. Sci. URSS **48**, 555 (1945). Used the 1- to 1.5-Mev neutrons from $D-D$ bombardment.

[15] D. J. Hughes, Phys. Rev. **70**, 106A (1946). See also MDDC **27**, Apr. 29, 1946. Used 1-Mev pile neutrons.

[16] If σ is given in barns, and E in ev, then Eqs. (1) may be written

$$\log_{10}(\sigma E^{\frac{1}{2}}) = 0.03A-1.00, \quad \text{for } A<100,$$

and

$$\log_{10}(\sigma E^{\frac{1}{2}}) = 2, \quad \text{for } A>100.$$

it is necessary to assume that the instability of nuclei with respect to β-decay does not materially affect their capture cross sections.

IV. EQUATIONS OF THE NEUTRON-CAPTURE PROCESS

We have seen that there is good evidence that the neutron-capture cross sections of the elements played an important role in the process by which the elements were formed. We have also seen that the element-forming process must have gone on in the prestellar state of the universe. The process of element formation which is suggested is therefore the following. Very shortly after the beginning of the universal expansion, the ylem[17] was a gas of neutrons only. These neutrons began to decay into protons and electrons, the density being sufficiently low to allow free neutron decay, but the temperature being sufficiently high that the mean thermal energy per neutron was higher than the mean binding energy per nucleon in nuclei, so that nuclei as such could not be formed. When the temperature decreased sufficiently in the expansion, the capture of neutrons by protons began, yielding deuterons. These nuclei in turn captured neutrons, and successively heavier nuclei were built up. The nuclei formed in this manner must have had large neutron excesses, and would therefore have undergone successive β-disintegrations into stable forms during and after the process of element formation. The process must have been terminated by the decrease in capture reaction rates resulting from the density decrease in the expansion, and by the decrease in the number of available neutrons as a result of their radioactive decay.

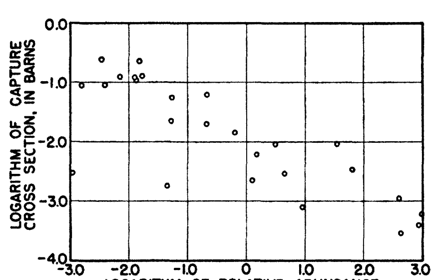

FIG. 4. Correlation of neutron-capture cross sections with relative abundance of nuclear species. The cross section data are due to Hughes, at 1 Mev, while the relative abundance data for the particular nuclei are due to Goldschmidt.

[17] According to Webster's New International Dictionary, 2nd Ed., the word "ylem" is an obsolete noun meaning "The primordial substance from which the elements were formed." It seems highly desirable that a word of so appropriate a meaning be resurrected.

We shall assume that temperature was sufficiently high and density sufficiently low so that the ylem can be treated as an ideal gas. We shall further suppose that the collision energies between neutrons and nuclei, and, in fact, the capture reaction rates, were sufficiently high that one may treat the neutron capture reactions as an equilibrium process at any instant of time. Then, from the kinetic theory of gases, one may write for the number of neutrons captured per second per unit volume by nuclei of atomic weight A, with the collision energy in the range dE at E,

$$dN_A = Bm_n^{\frac{1}{2}}n_n n_A \times[(m_n+m_A)/(m_n m_A)]^{\frac{1}{2}}\sigma_{n,A}e^{-E/kT}EdE, \quad (2)$$

where

$$B=(8/\pi m_n)^{\frac{1}{2}}(kT)^{-\frac{3}{2}}.$$

Here n_n and n_A are the concentrations of neutrons and nuclei of atomic weight A, respectively, m_n and m_A are the corresponding masses, and $\sigma_{n,A}$ is the neutron-capture cross section of the nuclei of atomic weight A. The concentrations n_n and n_A are functions of time, both because of the expansion, and because of changes resulting from the building-up process. As we have seen, $\sigma_{n,A}$ is a function of the energy E and of the atomic weight A, so that the total number of transmutations of nuclei from atomic weight A to atomic weight $A+1$, per unit volume and per unit time, is given by

$$N_A = Bn_n n_A[(1+A)/A]^{\frac{1}{2}}\int_0^\infty \sigma_{n,A}e^{-E/kT}EdE, \quad (3)$$

where the mass of the nucleus of atomic weight A is taken as an integer A times the mass of the neutron. From Eq. (3) we see that the probability per second that a nucleus of atomic weight A will capture a neutron is given by

$$p_A n_n = Bn_n[(1+A)/A]^{\frac{1}{2}}\int_0^\infty \sigma_{n,A}e^{-E/kT}EdE. \quad (4)$$

Substitution of Eqs. (1) for the approximate capture cross sections of the elements gives

$$p_A n_n = B n_n [(1+A)/A]^{\frac{1}{2}} \times 1.3 \times 10^{-31+0.03A} \times \int_0^\infty e^{-E/kT} E^{\frac{1}{2}} dE \text{ sec.}^{-1}, \quad \text{for } A<100, \tag{5a}$$

and

$$p_A n_n = B n_n [(1+A)/A]^{\frac{1}{2}} \times 1.3 \times 10^{-28} \times \int_0^\infty e^{-E/kT} E^{\frac{1}{2}} dE \text{ sec.}^{-1}, \quad \text{for } A>100. \tag{5b}$$

For $A>100$, one may replace $[(1+A)/A]^{\frac{1}{2}}$ by unity, with sufficient accuracy. Carrying out the integrations indicated in Eqs. (5), we find for the probability that a nucleus of atomic weight A will capture a neutron

$$p_A n_n = 1.4 \times 10^{-19+0.03A} \times [(1+A)/A]^{\frac{1}{2}} n_n \text{ sec.}^{-1}, \quad \text{for } A<100, \tag{6a}$$

and

$$p_A n_n = 1.4 \times 10^{-16} n_n \text{ sec.}^{-1}, \quad \text{for } A>100. \tag{6b}$$

It should be noted that as a result of the integration over all collision energies, temperature has cancelled out as a factor in the determination of the rate at which the capture processes go on. This results from the assumption that all the processes follow the $1/v$ law. Any assumption other than the $1/v$ law would have required the specification of a temperature. It should be noted that thermal dissociation of nuclei as a result of high energies on the "tail" of the Boltzmann distribution has been neglected. Although it is not necessary to specify the temperature, there are several facts which indicate what the temperature must have been during the period of element formation. First, as already mentioned, no particularly small abundances are observed which would correspond to those nuclear species known to possess very large resonance capture cross sections for thermal neutrons. It seems reasonable, therefore, that the temperatures were well above the resonance levels, i.e., above 10^3 ev. On the other hand, at a temperature of 1 Mev or higher (about 10^{10}°K), the energies of many of the neutrons would be in excess of the binding energy per nucleon in the nucleus. A temperature of the order of 10^5 ev ($\sim 10^9$°K) seems, therefore, to be indicated as the correct one.

The element-forming process suggested is one of successive neutron captures. In such a process the rate of increase of concentration of nuclei of atomic weight A must be equal to the difference between the rate at which nuclei of atomic weight $A-1$ capture neutrons and become nuclei of atomic weight A, and the rate at which nuclei of atomic weight A in turn capture neutrons and become nuclei of atomic weight $A+1$. The differential equations for such a process may be written as

$$dN_A/dt = p_{A-1} n_n n_{A-1} - p_A n_n n_A. \tag{7}$$

There is one such equation for each atomic weight A among the nuclear species. The quantities n_A are the concentrations of nuclei of atomic weight A, and $p_A n_n$ the probability per second that a nucleus of atomic weight A will capture a neutron. In an exact theory Eq. (7) must take into account the fact that the concentration of nuclei will change with time because of the expansion of the universe, entirely aside from the formation process, and, of course, the reaction rates will be affected by the expansion. The concentration of neutrons will change with time because of the expansion, and because the neutrons are used up in forming the other elements, including protons by β-decay.

The first step in the element-building process is the formation of deuterons by means of capture of neutrons by protons. Now the probable number of neutrons captured per second per unit volume by protons is given, according to Eq. (4), by

$$p_A n_n n_p = [4 n_n n_p / \pi^{\frac{1}{2}} (kT)^{\frac{3}{2}}] \times \int_0^\infty \sigma_{n,p} e^{-E/kT} E dE, \tag{8}$$

where $\sigma_{n,p}$ is the capture cross section of protons for neutrons.[18] This particular capture cross section is known not to follow the $1/v$ law for medium high energies, so that temperature does not cancel out completely as a parameter. On the other hand, if the universal expansion was

[18] See H. A. Bethe, *Elementary Nuclear Physics* (John Wiley & Sons, Inc., New York, 1947).

adiabatic, as indicated by relativistic cosmology,[19] then temperature changes during the time of the process were small.[20] Therefore the probable number of capture processes forming deuterons varied during the time of the process essentially only as the product $n_n n_p$.

The concentration of neutrons at a time t after the start of the process, $n_n(t)$, was related to the initial concentration, $n_n(0)$, except for the effect of the expansion, and except for the number used in forming other elements, as

$$n_n(t)=n_n(0)e^{-\lambda t}, \tag{9}$$

where λ is the decay constant of the neutron. Since protons were formed from the β-decay of neutrons, the product $n_n n_p$ is given by

$$n_n(t)n_p(t)=[n_n(0)]^2 e^{-\lambda t}(1-e^{-\lambda t}). \tag{10}$$

This is a slowly varying function of time, t, if t is not long as compared to the neutron decay lifetime. Because of the exponential decrease of neutrons available for building up nuclei, as a result of their decay, the period of element formation must have been of the order of a neutron decay lifetime. With this short time to be considered, the range of densities encountered as a result of the universal expansion must not have been very large, say, about one or two orders of magnitude at most. Furthermore, although neutrons and protons were used in making the other elements, the total amount of other elements is now observed to be small as compared to the great abundance of hydrogen. As a consequence, we shall assume that the rate of deuteron formation may be taken as constant, unaffected by the expansion and by the number of neutrons and protons used in making the other elements.

One further assumption is required, namely, that $p_A n_n$, the probability per second that a nucleus of atomic weight A will capture a neutron, was constant for the process. This would be approximately true if the density decrease caused by the expansion were small, and if the decrease in number of available neutrons were small.

As a result of the foregoing discussion, we write Eq. (7) approximately as

$$\left.\begin{aligned}
&p_p n_n n_p=\text{const.},\\
&dn_2/dt=p_p n_n n_p-p_2 n_n n_2,\\
&dn_3/dt=p_2 n_n n_2-p_3 n_n n_3,\\
&\qquad\vdots\\
&dn_A/dt=p_{A-1} n_n n_{A-1}-p_A n_n n_A.
\end{aligned}\right\} \tag{11}$$

Of Eqs. (11), the first states that the number of deuterons formed per second by neutron-proton capture is constant, whereas the second states that the rate of increase of concentration of deuterons equals the difference between the probable number of deuterons formed per second and the probable number of nuclei of atomic weight three formed per second.

Solutions of Eqs. (11) satisfying the conditions that except for neutrons and protons, the concentrations of all nuclei are zero at the start, are[21]

$$\begin{aligned}
n_2/n_p&=(p_p n_n/p_2 n_n)[1-(\exp -p_2 n_n t)]\\
&=(p_p/p_2)[1-(\exp -p_2 n_n t)],\\
n_3/n_p&=(p_p n_n/p_3 n_n)\{1-p_2 p_3 n_n^2\\
&\quad\times[((\exp -p_2 n_n t)/p_2 n_n)\\
&\quad\times(p_3 n_n-p_2 n_n)^{-1}\\
&\quad+((\exp -p_3 n_n t)/p_3 n_n)\\
&\qquad\times(p_2 n_n-p_3 n_n)^{-1}]\},\\
&=(p_p/p_3)\{1-p_2 p_3[((\exp -p_2 n_n t)/p_2)\\
&\quad\times(p_3-p_2)^{-1}+((\exp -p_3 n_n t)/p_3)\\
&\qquad\times(p_2-p_3)^{-1}]\},\\
n_4/n_p&=(p_p/p_4)\{1-p_2 p_3 p_4\\
&\quad\times[((\exp -p_2 n_n t)/p_2)\\
&\quad\times(p_3-p_2)^{-1}(p_4-p_2)^{-1}\\
&\quad+((\exp -p_3 n_n t)/p_3)\\
&\quad\times(p_2-p_3)^{-1}(p_4-p_3)^{-1}\\
&\quad+((\exp -p_4 n_n t)/p_4)\\
&\qquad\times(p_2-p_4)^{-1}(p_3-p_4)^{-1}]\},\ \text{etc.,}
\end{aligned} \tag{12}$$

[19] R. C. Tolman, *Relativity, Thermodynamics, and Cosmology* (Clarendon Press, Oxford, 1934).

[20] The period of element formation must have lasted only for a time of the order of the decay lifetime of the neutron.

[21] Differential equations of the type given in Eq. (7), with factors $p_A n_n$ constant, have been studied by H. Bateman, Proc. Camb. Phil. Soc. **15**, 423 (1910). In the case where the coefficients $p_A n_n$ are replaced by decay constants, Eq. (7) and the solutions, Eqs. (12), govern the growth of daughter elements in a radioactive decay series. See E. Rutherford, J. Chadwick, and C. D. Ellis, *Radiations from Radioactive Substances* (Cambridge University Press, Teddington, 1930).

TABLE I.

Range of atomic weights	$\bar{p}_j$ (sec.$^{-1}$)
1	$p_1=\bar{p}_1=0.525\times10^{-21}$
1 through 20	$\bar{p}_2=0.165\times10^{-19}$
21 through 40	$\bar{p}_3=0.628\times10^{-19}$
41 through 60	$\bar{p}_4=0.249\times10^{-18}$
61 through 80	$\bar{p}_5=0.984\times10^{-18}$
81 through 100	$\bar{p}_6=0.392\times10^{-17}$
101 through 120	$\bar{p}_7=$
121 through 140	$\bar{p}_8=$
141 through 160	$\bar{p}_9=$
161 through 180	$\bar{p}_{10}=$ 0.695×10^{-17}
181 through 200	$\bar{p}_{11}=$
201 through 220	$\bar{p}_{12}=$
221 through 240	$\bar{p}_{13}=$

where there is obviously the additional restriction that no two p's may be equal. As indicated in writing Eqs. (12), each quantity p_A appears with n_n as a factor, so that except in the exponents, n_n cancels out on the right-hand side. Evidently the ratios n_A/n_p may be computed with n_nt as the independent variable. We shall be interested only in relative concentrations, and since n_p is uniquely determined by n_n in this approximation, the computed ratios may as well be called n_A/n_n.

When numerical values were assigned to the p's, Eqs. (12) proved to be finite series of terms of large and nearly equal magnitude, with alternating signs. Equations (12) were evaluated to atomic weight 4 or 5, showing general agreement with the observational data. Since the computed abundances were very small differences between very large numbers, evaluation of Eqs. (12) became increasingly tedious and inaccurate. To circumvent this difficulty, it was found necessary to reduce the number of equations to be solved by calculating only certain ones of the n_A/n_n, which we denote by $\bar{n}_j/n_n$. Separation by twenty atomic weight units was selected as giving a reasonable number of equations to be solved. The particular $\bar{n}_j/n_n$ evaluated were assigned a $\bar{p}_j$ representing 1/20th of the mean p_A for the group of twenty species (according to atomic weight) they replaced. This follows from the fact that the transition from selected element j to selected element $j+1$, where j is interpreted as an index of the group of twenty represented, involves the successive capture of twenty neutrons. The effect of this simplification was to require solutions of 12 equations only, of the form

$$d\bar{n}_j/dt=\bar{p}_{j-1}n_n\bar{n}_{j-1}-\bar{p}_jn_n\bar{n}_j. \qquad (13)$$

The quantities $\bar{p}_j$ used in the calculation are given in Table I. Even with this simplification, solution of Eq. (13) in the form of Eqs. (12) could not be carried out for more than the first few values of j, because of the rapid exponential decrease of relative abundance with increasing j. Consequently Eq. (13) was integrated numerically.

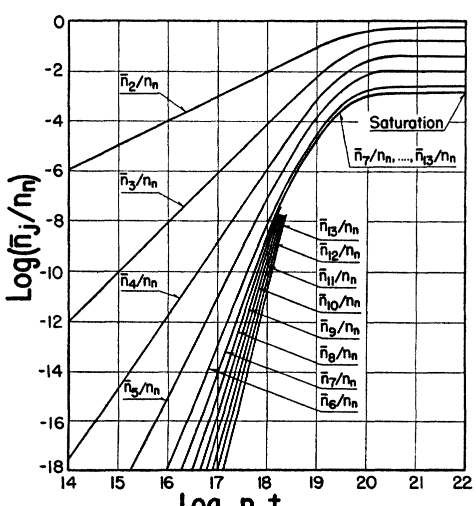

FIG. 5. Relative abundances computed by neutron-capture theory as a function of time. The quantity n_n is the neutron concentration prevailing during the period of element formation.

Solutions thus obtained are given in Fig. 5, in the form $\log(\bar{n}_j/n_n)$ *versus* $\log(n_nt)$. The quantities $\bar{n}_j/n_n$ correspond to the desired relative abundances, except, of course, for an arbitrary additive constant. The set of computed relative abundances corresponding to $\log(n_nt)=17.91$, i.e., $n_nt=0.81\times10^{18}$ sec./cm^3, was selected as giving the best representation of the observational data. This set of solutions is compared in Fig. 6 with the observed relative abundance data. A constant was added to the computed values to adjust theory and data at H^1. Although each computed $\bar{n}_j/n_n$ value represents a range of twenty in atomic weight, a smooth curve has been drawn through the points as indicative of the result of a step-by-step solution for each atomic weight.

For all practical purposes, there are no further relative changes between successive $\bar{n}_j/n_n$ with increasing n_nt after n_nt reaches a value of about 1.5×10^{20} sec./cm^3. Certainly by $n_nt\cong5\times10^{21}$ sec./cm^3 there is no further change in relative abundance, and the solutions as set up in this preliminary form have attained saturation.

Determination of the best fit of computed relative abundances is very sensitive to the choice of n_nt, as is evident from Fig. 5. To illustrate this sensitivity, sets of solutions for $n_nt=0.51\times10^{18}$ and $n_nt=1.3\times10^{18}$ are shown on Fig. 6. These values are clearly well above and well below the best fit given by $n_nt=0.81\times10^{18}$ sec./cm^3. Relative abundances corresponding to saturation, according to the present computations, are also indicated on Fig. 6. In a more exact treatment of the element-forming process, one should take into account completely the effect of the universal expansion and the diminution resulting from radioactive decay and from capture by nuclei, of the number of neutrons available. The result of this may be reasonably expected to be that the best fit to the observational data will correspond to the saturation of the neutron-capture process.

The dependence of the fit of theory to observed abundance data on the probabilities $\bar{p}_j$ may be illustrated in two ways. First, let us assume that all quantities $\bar{p}_j$ were too small by a factor of 10. It may be seen from the form of Eqs. (12) that any factor common to all $\bar{p}_j$ would cancel everywhere except in the exponents of the $(\exp-\bar{p}_jn_nt)$ terms. In the exponents, on the other hand, if one compensates an error in all $\bar{p}_j$ by multiplying them by 10, the value of the exponent can be maintained unchanged by multiplying n_nt by $\frac{1}{10}$. The net effect on the theory is to change the n_nt value for best agreement of the theory by a factor of 10, which is not a serious effect in this sort of preliminary examination of the problem. The effect of an error in a particular $\bar{p}_j$ may be more noticeable. One may estimate such effects by comparing the theoretical abundance curve with the detailed features of the observational data. For example, Pb^{208} is about ten times more abundant than the computed value. However, in Fig. 3, we see that at 1 Mev the capture cross section of Pb^{208} is about 100 times less than the average heavy element value used. Hence an error of a factor of 100 in the capture cross sections appears to lead to an error by a factor of the order of 10 in the computed abundances.

V. INTERPRETATION AND DISCUSSION

Before discussing the results of the neutron-capture process, it is pertinent to examine the early state of the expanding universe by means of a currently accepted cosmological model. It can be shown[2,19] for an expanding universe in which the material is considered to be a perfect fluid, and in which radiation is neglected, that the time rate of change of any linear dimension, l, is related to the mean density of matter, ρ, in the universe, by the equation

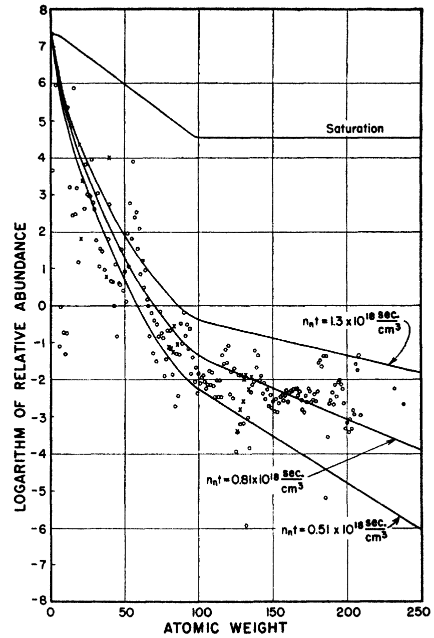

FIG. 6. Comparison of relative abundances computed by the neutron-capture theory with Goldschmidt's relative abundance data. The best fit is for $n_nt=0.81\times10^{18}$ sec./cm^3. The other curves illustrate the sensitivity of the best fit to the value of n_nt. The "saturation" curve represents the relative abundances had the process as formulated continued for an indefinite period.

$$d(l/l_0)/dt=\left[(8\pi G\rho l^2/3l_0^2)-(c^2/R_0^2)\right]^{\frac{1}{2}}\text{ sec.}^{-1},\quad(14)$$

where c is the velocity of light, G is the constant of gravitation, and l_0 and R_0 are constants of dimension length whose values are fixed according to the currently observed features of the universe. If we use Hubble's universal expansion rate,[22] $k=1.8\times10^{-17}$ cm/sec./cm, the present mean density of matter in the universe, $\rho\cong10^{-30}$ g/cm^3, and set $l=l_0=10^{10}$ cm, i.e., the side of a cube currently containing one gram of matter, then we find from Eq. (14) that $R_0=1.67\times10^{27}(-1)^{\frac{1}{2}}$ cm. The quantity R_0 is interpreted as the radius of curvature of the space. Thus the

[22] E. Hubble, *The Observational Approach to Cosmology* (Clarendon Press, Teddington, 1937).

cosmological model is an open one, a model which expands monotonically to infinity.

If we replace ρ by $1/l^3$, Eq. (14) can be integrated; if it is assumed that linear dimension $l=0$ when $t=0$, then one obtains

$$t=(-ab^2)^{\frac{1}{2}}[(\rho^{-\frac{2}{3}}-a\rho^{-\frac{1}{3}})^{\frac{1}{2}} + a\sinh^{-1}(-a^{-1}\rho^{-\frac{1}{3}})^{\frac{1}{2}}], \quad (15)$$

where

$$a=(8\pi GR_0^2)/(3c^2l_0^2), \quad \text{and} \quad b=(8\pi G/3)^{-\frac{1}{2}}.$$

Expanding the two terms in Eq. (15) in series, and combining terms, one obtains

$$t=b[(2/3\rho^{\frac{1}{2}})+(1/5a\rho^{5/6}) + (3/28a^2\rho^{7/6})+\cdots]. \quad (16)$$

The expansions are valid for

$$(-1/a\rho^{\frac{1}{3}})^2<1. \quad (17)$$

Inserting numerical values for R_0 and l_0, we find that Eq. (16) becomes

$$t=8.9\times10^2\rho^{-\frac{1}{2}}-1.54\times10^{-5}\rho^{-5/6}+\cdots, \quad (18)$$

where the condition for validity of the expansion is that

$$\rho>1.94\times10^{-22} \text{ g/cm}^3.$$

For densities greater than 10^{-18} g/cm^3 we can write, with sufficient accuracy

$$t=8.9\times10^2\rho^{-\frac{1}{2}} \text{ sec.}, \quad (19)$$

where ρ is in g/cm^3. It is interesting to note that Eq. (18) corresponds to the first term in Eq. (16), which term does not contain the radius of curvature R_0.

With the aid of the foregoing, it is possible to obtain some estimates of the physical conditions of the ylem, and of the matter in which the elements were formed. In making these preliminary calculations, it was assumed that the neutron concentration was constant, and, in fact, we found that the theory best represented the observed relative abundance data with a value $n_nt=0.81\times10^{18}$ sec./cm^3. This quantity admits of a rough interpretation, namely, as the product of the arithmetic average neutron concentration during the process times the time duration of the process. Actually the neutron concentration must have decreased at least exponentially with time during the process and an arithmetic average is therefore rough. We write

$$n_nt=(n_n)_{\mathrm{Av}}\Delta t=\int_{t_0}^{t_1} n_n dt, \quad (20)$$

thereby defining the average n_n. In Eq. (20) t_0 and t_1 are the starting and ending time of the process. Considered either by weight or by relative number of nuclei, hydrogen is now observed to be predominantly abundant in the universe. The number of neutrons and protons used in making the heavier elements must therefore have been small. To obtain an estimate of the initial density and starting time for the process, we neglect, in first approximation, anything other than neutrons and protons. According to the law of radioactive disintegration, the total number of neutrons in the universe at any time t is given by

$$N_n(t)=N_n(0)e^{-\lambda t}, \quad (21)$$

where $N_n(0)$ is the number of neutrons at time $t=0$ and λ is the neutron decay constant. Correspondingly the number of protons is

$$N_p(t)=N_n(0)-N_n(t). \quad (22)$$

The density of matter at time t is

$$\rho(t)=m[N_n(0)/V(t)], \quad (23)$$

with $V(t)$ the volume of the universe at this time and m the mass of a nucleon. The neutron concentration at time t is given by

$$n_n=N_n(t)/V=[N_n(0)/V]e^{-\lambda t}=(\rho/m)e^{-\lambda t}. \quad (24)$$

We find that

$$\int_{t_0}^{t_1}(\rho/m)e^{-\lambda t}dt=(n_n)_{\mathrm{Av}}\Delta t =0.81\times10^{18} \text{ sec./cm}^3. \quad (25)$$

The particular cosmological model we have chosen leads to Eq. (19) relating universal density and time. Substituting Eq. (19) into (25), we have

$$\int_{t_0}^{t_1}(e^{-\lambda t}/t^2)dt=1.68\times10^{-12}. \quad (26)$$

This can be integrated to give

$$(e^{-\tau_0}/\tau_0)-(e^{-\tau_1}/\tau_1)+E_i(-\tau_0) -E_i(-\tau_1)=1.68\times10^{-12}, \quad (27)$$

where $\tau=\lambda t$, λ being the neutron decay constant.

If we assume that the process of element formation took a time of the order of one neutron lifetime (latest estimate about 1800 sec.) then Eq. (27) gives for the starting time of the process on the time scale of the particular cosmological model used[23]

$$\tau_0 \cong 13.8, \quad \text{or} \quad t_0 \cong 2.5 \times 10^4 \text{ seconds.} \qquad (28)$$

According to Eq. (19) the density of matter at this time was

$$\rho_0 \cong 1.3 \times 10^{-3} \text{ g/cm}^3. \qquad (29)$$

With this density, and a temperature of the order of 10^9°K (10^5 ev), it is clear that our treatment of the ylem as an ideal gas was justified. In addition, at this temperature one does not need to consider relativistic effects for neutrons, protons, and nuclei.

Although the starting time and initial density given above are only order of magnitude quantities, they are nevertheless in seeming contradiction with the qualitative description of events prior to and during the process. Equation (19) clearly indicates that with this cosmological model, the density of matter should have dropped sufficiently in a time of the order of seconds after the beginning of the expansion to permit the radioactive decay of neutrons to begin. Then, at a time of the order of 10^4 seconds later, there would have been very few neutrons left to initiate and carry on the neutron-capture process. If the cosmological model leading to Eq. (19) is correct, the starting time of the process must have been several orders of magnitude earlier than 10^4 seconds.

In addition, we have seen that the temperature during the element-forming process should have been of the order of 10^5 ev, or about 10^9°K. At this temperature, the density of black body radiation would have been

$$\rho_{\text{radiation}} = 0.841 \times 10^{-35} T^4 \cong 10 \text{ g/cm}^3. \qquad (30)$$

Thus if radiation were present, then the radiation density exceeded the density of matter by many orders of magnitude. It would therefore appear that radiation was dominant in determining the behavior of the universe in the early stages of its expansion, and the cosmological model which has been introduced is probably incorrect.

Preliminary calculations of a cosmological model involving black body radiation only[24] (the effect of matter on the behavior of the model being negligible in the early stages because of the great difference between radiation and matter density) indicate that at some 200 to 300 seconds after the expansion began the temperature would have dropped to about 10^9°K, at which time the neutron-capture process could have begun. In such a model the density of radiation varies as $1/t^2$, whereas the density of the small quantity of matter present varies as $1/t^{\frac{3}{2}}$.

Whatever the correct cosmological model may be, it seems clear that the temperature change during the relatively short time taken by the process must have been small. This is particularly significant in view of the relative scarcity in the universe of the isotopes of Li, Be and B (see Fig. 1). This scarcity applies to terrestrial material as well as to the universe as a whole. If these elements were plentiful on earth, then their scarcity in stellar material would be readily explained. In stellar interiors, these elements in particular[1] have very short lifetimes for thermonuclear reactions with protons. Consequently they should be scarce in stars. But the fact that they are scarce on earth means that if the primordial matter involved in forming the planets did not have an intervening existence in a stellar configuration, then these elements emerged from the prestellar state with their present characteristic scarcity.

Relativistic cosmology indicates that the expansion of a general non-static model must be considered as adiabatic. Then, for a universe of matter only, temperature varies with time as $T_{\text{matter}} \propto t^{-4/3}$ and with a radiation universe $T_{\text{radiation}} \propto t^{-\frac{1}{2}}$. Starting, in both cases, with $T = 10^9$°K, and with $t_0 \cong 2.5 \times 10^4$ seconds in the first case, and with $t_0 \cong 250$ seconds in the second case, we find that for a process time of the order of the neutron half-life, the temperature will have dropped only by a factor of about ten. Now at 10^8°K, thermonuclear reactions of the various isotopes of Li, Be, and B with protons could go on at an appreciable rate. It is therefore sug-

[23] It should be noted that values of the function $-Ei(-x)$ in the range $15 \leq x \leq 20$ in steps of 0.1 are tabulated by R. C. Herman and C. F. Meyer, J. App. Phys. **17**, 258 (1946).

[24] R. C. Tolman, Phys. Rev. **37**, 1639 (1931).

gested that the scarcity of these elements is a result of a period of thermonuclear reactions following the neutron-capture process, and prior to the formation of stellar configurations.

We have already pointed out that the nuclei formed during the neutron-capture process must have had a considerable excess of neutrons, and consequently must have undergone β-disintegrations during and after the process. For a given atomic weight, one would expect the successive disintegrations of such nuclei to have stopped when a nucleus of the lowest possible charge consistent with stability was reached. It is pertinent to note, therefore, in the relative abundance data, that as a rule the most abundant of a group of isobars is generally the one with lowest charge.[25] Thus in Goldschmidt's tabulation, excluding the noble gases, there are 41 sets of stable isobars heavier than atomic weight 64, of which in 33 cases the nuclei of lowest charge are predominantly abundant. These data in themselves suggest that the abundance distribution with respect to atomic number was established in final form by β-decay processes, and, in fact, that the elements were formed by a neutron-capture process.

Critchfield and Smart,[26] in examining the problem of β-disintegrations associated with the process of successive neutron-captures, have concluded that in general nuclei can consist of no more than about 70 percent neutrons; the next neutron added to the nucleus would not be bound. At this limit of neutron content, they estimate that β-decay lifetimes would be of the order of a 0.1 second. On the other hand, the time lapse between successive neutron captures in the element-forming process which has been described is given by the reciprocal of $p_A n_n$, the probability per second that a nucleus of atomic weight A will capture a neutron. For a density of 10^{-3} g/cm^3, this time between captures is larger than 0.1 second, so that the process could go on with nuclei well below the limit of neutron content.

According to the approximate theory presented, there is no reason why elements of larger and larger atomic weights might not have been formed by successive neutron captures. However, the increase of spontaneous and induced fission cross sections with increasing atomic weight among the heaviest nuclei effectively precludes this. Subjecting elements heavier than uranium to a flux of neutrons at 10^5 ev would certainly have prevented any reasonable natural abundance for these elements. However, there is still the possibility that heavier elements were formed, to exist for a short time, and then undergo fission. The material which went into the "tail" of the abundance plot, for atomic weights greater than about 240, should then be found redistributed at lower atomic weights according to the fission yields of these elements. Published fission yield data[27] show very little dependence on the atomic weight of the parent element. For elements of the order of 230 to 240 in atomic weight, mass yield curves have definite maxima in the neighborhood of atomic weights 93 and 139. The peaks are broad, about 10 atomic mass units wide at 80 percent to 90 percent of the peak yield. Examining the abundance data on Fig. 1, we find in the neighborhood of these atomic weights abundance peaks which were previously interpreted as arising from the small capture cross sections of the nuclei there. It is possible, then, that the large abundances for these atomic weights are the superimposed result of the two different processes. Because of the expected low capture cross sections at atomic weights of the order of 93 and 139, fission products arriving in these regions would tend to accumulate there, rather than to capture neutrons and be redistributed over the higher atomic weights. Those of the transuranic elements formed which do not undergo neutron-induced fission would nevertheless have been unstable with respect to spontaneous fission or α-decay. In the latter case, abundances in the region of uranium would be enriched by disintegration products from the "tail" of the abundance plot.

Several difficulties which arise with the neutron-capture process have not yet been described. The first of these is that it appears to be impossible to build past atomic weights 5, 8 and 11 by a

[25] This has also been pointed out by F. C. Frank, Proc. Phys. Soc. London **60**, 212 (1948).

[26] C. L. Critchfield and J. S. Smart, private communication.

[27] C. Goodman (Ed.), *The Science and Engineering of Nuclear Power* (Addison-Wesley Press, Inc., Cambridge, 1947).

process of successive neutron captures alone, because of the absence of stable nuclei at these atomic weights. However, because of the high temperature, reactions with deuterons or with tritons may be reasonably introduced for the low Z nuclei, and thereby make it possible to bridge these gaps. A similar problem arises with the radioactive species in the gap between lead and thorium and uranium. Some of the decay products from uranium, thorium, etc., in this region have extremely short α-decay lifetimes, i.e., short compared with the time between successive neutron captures. In order to have nuclei of these atomic weights present to permit neutron captures successively to uranium, it is necessary to assume that these short-lived α-emitters were formed with neutron excess, and that several β-decay steps separated them from the α-emitting state. If the β-decay rates were sufficiently long, then such nuclei could have been present during the process in sufficient quantity.

There is in addition the problem of the so-called "shielded" isotopes. The neutron-capture process built up elements according to an atomic weight scheme. The distribution with respect to atomic number was presumably established later by β-decay. However, with nuclei formed with a neutron excess, β-decay would stop at the stable nucleus of lowest Z. The theory which has been described does not yet explain, then, why one finds sets of stable isobars, since transitions from a nucleus ${}_ZX^A$ to a nucleus ${}_{Z+1}X^A$ are not allowed if ${}_ZX^A$ is stable.

Finally, the correlation of abundance peaks with small capture cross sections may appear rather too exact, in view of the fact that the nuclei formed in the process must have had some neutron excess. Consequently, a "magic number" nucleus with 50 neutrons, for example, may have been β-unstable, and one should therefore correlate abundance peaks with nuclei now containing, say, 47 neutrons. This may not be a serious problem, however, since both the abundance peaks and the range of atomic weights in which "magic number" nuclei lie are broad, and a displacement by several units in atomic weight could be tolerated. This would imply that when the nucleus formed differed from a stable configuration by more than several successive β-disintegrations, the decay rates were fast compared to the time between successive neutron captures. A similar criticism applies to the correlation of the even-odd periodicity in abundance with an even-odd variation in capture cross sections, since proton-neutron ratios were different during the period of successive neutron captures. For application of the even-odd correlation, it appears to be necessary to assume that as a general rule the number of successive β-disintegrations undergone by nuclei was even.

Further studies are now being carried on of the cosmological model required by the neutron-capture process. In addition, equations for the process are now being integrated in which neutron decay and the universal expansion are explicitly included.

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