the higher troughs observed in the case of U^{238} and Th^{232} fission are reasonable in view of the energy of the neutrons being used. Specifically, they are not inconsistent with a constant ratio of peak to trough in the fission of all heavy nuclei at excitations at which the rate of break-up of the compound nucleus into fission fragments is comparable.

The radiochemical U^{238} fission yield data of Engelkemeir, Seiler, Steinberg, and Winsberg[50] is presented in Fig. 3 in a manner comparable to that used for our data in Fig. 1 (assuming binary fission with loss of two neutrons). The rather flat regions near symmetrical fissions in Figs. 1 and 3 suggest that perhaps the observed curves are superpositions of two yield curves, one the familiar "double-humped "curve, whose shape and absolute values are not very sensitive to neutron energy, the other a yield curve with a rather broad maximum at symmetrical fission. The peak in the latter curve would be much lower than those in the asymmetric type, but the absolute values here would increase with increasing neutron energy.

The results reported in Table I also give a little information on the charge distribution in Th^{232} fission as compared with U^{235} or Pu^{239} fission. Because the neutron-to-proton ratio in Th^{232} is higher than that in U^{235} and Pu^{239}, slightly longer fission chains are expected in Th^{232} fission. For this reason there should be no change in the relative yields of isomers close to the end of a beta-chain, since these would be formed primarily by beta-decay of a fission product with lower charge rather than directly in the fission process. Within experimental error, this is borne out in the cases of isomers in the chains of mass 77 and 115.

On the other hand, because of the higher neutron to proton ratio in Th^{232} relative to U^{235} and Pu^{239}, a lower yield of shielded isotopes is to be expected.[52] In the one case investigated (Cs^{136}) this expectation was also realized: in Th^{232} fission, the yield of Cs^{136} is apparently no greater than one quarter of that in U^{235} fission with slow neutrons. The theory of Glendenin[52] predicts for Cs^{136} a yield in Th^{232} fission about 20 times lower than in U^{235} fission.

[52] L. E. Glendenin, Ph.D. thesis, Massachusetts Institute of Technology (July 29, 1949), unpublished.

PHYSICAL REVIEW VOLUME 84, NUMBER 1 OCTOBER 1, 1951

Neutron-Capture Theory of Element Formation in an Expanding Universe*†‡

RALPH A. ALPHER AND ROBERT C. HERMAN
Applied Physics Laboratory, The Johns Hopkins University, Silver Spring, Maryland.

(Received June 11, 1951)

The neutron-capture theory of element formation by non-equilibrium processes has been extended to include explicitly the effect of the expansion of the universe, and the resulting equations have been solved on an electronic digital computing machine. Inclusion of the universal expansion is found to require an increase by a factor of five of the density of matter chosen for the start of the element-forming process over that previously found necessary to represent the observed relative abundance distribution of elements in a static universe. The following physical conditions lead to agreement in the over-all trend of theoretical with observed abundances: the element-forming process is taken to start at $\sim$140 sec after the "beginning" of the universal expansion; at this time the temperature is $\sim 1.3\times 10^{9}$°K$\cong$0.11 Mev, the neutron-proton ratio is 7.33:1, and the density of matter is $\sim 0.9\times 10^{-6}$ g/cm^3. This density value includes a correction made to account for the effect of groupiing together nuclear species in order to reduce the number of defferential equations required to describe the neutron-capture procss.

The effect of the choice of an initial neutron-proton ratio on the other physical conditions involved in representing the observed relative abundance data is considered. A neutron-proton starting ratio of 1:4, recently found by Hayashi to result from the interactions between matter and radiation in the pre-element-forming phase of the expanding universe, is shown to lead to some difficulties.

In an Appendix, Dr. T. H. Berlin, of The Johns Hopkins University, shows that the differential equations describing the element-forming process can be solved in closed form for the static case by the use of laplace transforms. However, the inclus on of the universal expansion precludes solution in closed form.

I. INTRODUCTION

THE work described in this paper serves to complete one aspect of the description of the origin and observed relative abundance distribution of the chemical elements according to a non-equilibrium neutron-capture theory. Calculations have been made of the dependence on atomic weight of the relative abundance distribution of nuclear species which results if the effect of the expansion of the universe is explicitly taken into account in the formation process. In previously reported work[1,2] the process of element formation by successive neutron captures was examined for a static universe; and it was shown that the general

* This work was supported by the U. S. Navy Bureau of Ordnance.

† Preliminary accounts of this work were presented at the New York meeting of the American Physical Society, February, 1951, Phys. Rev. 82, 296(A) (1951), and at the Washington meeting, April, 1951, Phys. Rev. 83, 236(T) (1951).

‡ This paper includes an Appendix by Dr. T. H. Berlin, Department of Physics, The Johns Hopkins University, on the exact solution of equations describing element formation in a static universe according to the neutron-capture theory.

[1] R. A. Alpher and R. C. Herman, Phys. Rev. 74, 1737 (1948).
[2] R. A. Alpher and R. C. Herman, Revs. Modern Phys. 22, 153 (1950).

trend of the observed relative abundance distribution in atomic weight, as given by Brown,[3] was theoretically reproduced over the entire atomic weight range. As will be seen in this paper, a satisfactory representation of the relative abundance data can also be obtained with a neutron-capture theory in an expanding universe. In this, as in previously reported work, the detailed features of the abundance data are not reproduced, since the theory does not contain the specific nuclear properties and processes involved. The theory of the formation process is based on the radiative capture of fast neutrons by all nuclear species, and a smoothed-out representation is taken for the well-defined dependence of fast neutron capture cross sections on atomic weight. The capture cross section data involved are those determined principally by Hughes, Spatz, and Goldstein,[4] and by Hughes and Sherman.[5]

The expansion of the universe was not explicitly included in previously reported work by the authors on the origin of the elements because of the computational labor involved. Recently, the Computation Laboratory of the National Bureau of Standards was kind enough to afford the authors the opportunity of using its new electronic computing machine, the SEAC (Bureau of Standards Eastern Automatic Computer), for the solution of this problem. The detailed coding of the problem for SEAC solution, together with the actual SEAC operation, was performed by Dr. Joseph H. Levin of the Computation Laboratory, National Bureau of Standards.

II. FORMULATION OF THE PROBLEM

While a detailed statement of the neutron-capture process of element formation in an expanding universe has already been given,[2] it seems worthwhile to describe briefly the proposed situation again here. The presently observed relative abundance distribution of nuclear species is believed to be universal, and to have been established in all essential details quite early in the history of the expanding universe, prior to the formation of galaxies and stars. In the present form of the theory, it is supposed that very shortly after the "start" of the universal expansion, the material content of the universe was neutrons. At the low densities of matter involved, neutrons were able to undergo free radioactive decay. At first the temperature was too high to allow the existence of nuclei in appreciable amounts. However, as the universe expanded and cooled, nuclear reactions yielding the various nuclear species became predominant as compared with photo and thermal dissociation processes. The first of these building-up reactions involved the neutrons and the appreciable number of protons already present from neutron decay, and yielded deuterons by radiative capture of neutrons. Heavier nuclei were built up by successive radiative capture of neutrons with intervening β-decay, the latter adjusting nuclear charge so that the resultant nuclei could continue to grow by neutron addition. Many simplifications are involved in the physical picture considered here. In particular, for species of atomic weight less than 15 or 20, reactions other than those of the (n,γ) type undoubtedly played an important role. Certainly other reactions must be introduced to carry the chain of element-building reactions past the non-existent nuclei at atomic weights 5 and 8. Moreover, the detailed abundances of heavier nuclei must have been influenced to some extent by the competition between (n,γ) reactions and β-decay, by (γ,n), $(n,2n)$ and other reactions insofar as the "shielded elements" and the abundance peak in the vicinity of iron are concerned, and by nuclear fission acting to terminate the element-forming process at high atomic weights. However, it appears that under the assumed physical conditions, (n,γ) reactions would generally be the most probable, and in predominating would determine the basic character of the relative abundance data. These and other detailed questions involved in the neutron-capture theory have been discussed elsewhere recently.[2]

The cosmological model chosen for the early stages of the expanding universe is the general nonstatic model, composed of an ideal fluid, exhibiting homogeneity and isotropy, and with no interconversion of matter and radiation. It can be shown that for the early times of interest in connection with the element-forming process, the physical state of the model can be described without recourse to any statement as to the nature of the radius of curvature of the universe at that time. Thus, it is not necessary to characterize the universe as being open, closed, or flat, questions which must be decided on the basis of information other than that gleaned from the theory of element formation.

It was shown in earlier work on the neutron-capture theory in a static medium that the density of matter must have been considerably less than the density of radiation during the formation process. In this situation, the radiation controls the expansion, and the field equations pertinent to the cosmological model described give the following time dependence for the density of matter,[2]

$$\rho_m = \rho_0 t^{-\frac{3}{2}} \text{ g/cm}^3, \tag{1}$$

and for the density of radiation,

$$\rho_r = (32\pi G/3)^{-1} t^{-2} = 4.48 \times 10^5 t^{-2} \text{ g/cm}^3, \tag{2}$$

where G is the gravitational constant, and time t is in seconds after the start of the expansion. The trace of matter is taken to be in thermal equilibrium with the radiation. If the universal expansion is supposed to involve the adiabatic expansion of black body radiation, then it follows from Eq. (2) that the temperature is given by

$$T = (c^2/a)^{\frac{1}{4}} \rho_r^{\frac{1}{4}} = 1.52 \times 10^{10} t^{-\frac{1}{2}} \,^\circ\text{K}, \tag{3}$$

[3] H. Brown, Revs. Modern Phys. **21**, 625 (1949).
[4] Hughes, Spatz, and Goldstein, Phys. Rev. **75**, 1781 (1949).
[5] D. J. Hughes and D. Sherman, Phys. Rev. **78**, 632 (1950).

where a is the Stefan-Boltzmann constant and c the velocity of light.

The equations describing the successive neutron-capture processes are derived in terms of finite volume elements, so that again the question of the radius or the total mass of the universe need not be considered. Let C_n, C_1, and C_j be the concentrations of neutrons, protons, and nuclei of atomic weight j. Let C_0 be the concentration of nucleons at the time t_0 defining the start of the element-forming process according to the simplified theoretical picture. Then one defines normalized concentrations as

$$\xi_n = C_n/C_0, \quad \xi_1 = C_1/C_0, \quad \xi_j = C_j/C_0. \tag{4}$$

Although physically it does not appear correct to select a starting time, since formation of nuclei will build up over a finite time as competing processes die out, actually it appears to be a reasonable as well as a sufficient approximation to select a time when the temperature has fallen below the deuteron binding energy. Assuming that element building starts at this time, the following equations then describe the neutron-capture process:

$$\begin{aligned} d\xi_n/d\tau &= -\xi_n - \sum_{j=1}^{J} P_j \xi_n \xi_j - 3\xi_n/(2\tau), \\ d\xi_1/d\tau &= \xi_n - P_1 \xi_n \xi_1 - 3\xi_1/(2\tau), \end{aligned} \tag{5}$$

and for the general case of species of atomic weight j,

$$d\xi_j/d\tau = P_{j-1}\xi_n\xi_{j-1} - P_j\xi_n\xi_j - 3\xi_j/(2\tau).$$

In Eqs. (5), one has

$$\tau = \lambda t, \tag{5a}$$

where λ is the decay constant of the free neutron,

$$P_j = p_j C_0/\lambda, \tag{5b}$$

and the integration is started at a $\tau_0 = \lambda t_0$. The quantities p_j are probabilities, per second, that a nucleus of atomic weight (j) will capture a neutron and become the species of atomic weight ($j+1$). Equations (5) state that the neutron concentration decreases because of neutron decay, and because of radiative capture by all other nuclear species; the proton concentration is increased by free neutron decay and decreased by the formation of deuterons; the concentration of nuclei of atomic weight (j) is increased through radiative capture of neutrons by nuclei of atomic weight ($j-1$) and is decreased through its own capture of neutrons, and, for all species the concentrations decrease as $3\xi/(2\tau)$, a term describing the dilution due to the expansion of the universe.

Qualitatively, the quantities p_j are the product of a capture cross section and a velocity. The cross sections actually used in calculations were, as already mentioned, values from a smoothed-out representation of the general dependence of fast neutron capture cross sections on atomic weight. The velocity factor was so determined as to account for the fact that at the temperature and density associated with the element-forming process the collisions among nuclei and nucleons must have obeyed the maxwell distribution law in energy. With the several factors in p_j so taken, the calculation of P_j is quite straightforward.[2]

An initial value of $\tau = \tau_0 = 0.128$ was chosen for integrating Eqs. (5). Because $\tau = \lambda t$, the corresponding initial time in seconds, measured from the "beginning" of the expansion, as well as the corresponding temperature, depend on the value used for the neutron decay constant. For a neutron half-life of 12.8 min,[6] the initial temperature and time are found to be $\sim 1.28 \times 10^9$°K (0.11 Mev) and ~ 142 seconds, respectively.[7] One must further select the initial concentration of nucleons, as well as the composition in terms of neutrons and protons. Clearly, the introduction of an initial time implies that there are no nuclear species present other than neutrons and protons to begin with. Electrical neutrality of the mixture is assumed. If the normalized concentration of neutrons, ξ_n, is taken to be unity at $\tau = 0$, and if the only process that went on until $\tau = \tau_0 = 0.128$ was the free decay of neutrons, then the initial concentrations of neutrons and protons for the formation process would have been $\xi_n(\tau_0) = 0.88$ and $\xi_1(\tau_0) = 0.12$, respectively. These values, together with $\xi_j(\tau_0) = 0$, were taken as initial conditions for the solutions of Eqs. (5). The effect of the particular choice of initial conditions is considered further in Sec. IV of this paper.

To reduce the number of Eqs. (5) to be solved simultaneously to a practical quantity,[2] the coefficients P_j were averaged for small groups of adjacent nuclear species, giving equivalent coefficients for each group as Π_j. Specifically, groups of five were taken for $5 \leqq j \leqq 94$, while for higher j twenty-element groups were used. The first five equations, applying to neutrons and species with j ranging from one to four, were included without change as part of the resulting twenty-seven equations, except that the summation in the neutron equation was taken up to $J = 4$, since it seems reasonable from the observed relative abundance data to suppose that only a small fraction of the available neutrons were used by the process in producing the quite small amounts of heavier elements. The scheme of grouping to reduce the number of equations does not seriously affect the value of the initial nucleon concentration, C_0, which yields a satisfactory representation of the relative abundance data, nor does it alter the basic appearance

[6] The value 12.8±2.5 min for the half-life of the neutron was reported in an invited paper by Dr. J. M. Robson at the New York Meeting of the American Physical Society, February, 1951, Phys. Rev. 82, 306(T) (1951).

[7] These differ from previously reported values (see reference 2) of temperature and time corresponding to $\tau_0 = 0.128$ because of the use of a different decay constant in earlier work. When $\tau_0 = 0.128$ was first chosen, the value of the neutron decay constant was taken to be such as to yield 1 g/cm³ for the radiation density at the start of the process.

of the theoretical abundance curve. The averaged or grouped values of neutron capture cross sections did not differ in essential features from the smoothed-out data on capture cross section *versus* atomic weight previously described[2]; and, therefore, it was expected that the former would again determine, without serious difference, the general relative abundance curve arising from the neutron-capture theory.

III. RESULTS

The grouping of nuclear species described earlier reduced Eqs. (5) to twenty-seven in number. These equations were set up for "single-precision" calculation on the SEAC. With this precision it was possible just to cover the spread in the observed relative abundance data with theoretical solutions.[8] The nature of the solutions obtained of Eqs. (5), plotted as the logarithm of relative abundance *versus* $\log\tau$, is illustrated in Fig. 1. In this case the initial conditions are $\xi_n(\tau_0)=0.88$, $\xi_1(\tau_0)=0.12$, and $\tau_0=0.128$, while the initial nucleon concentration was taken to be $C_0'=1.07\times10^{17}$ cm^{-3}. As is clear from Fig. 1, after a sufficiently long τ the relative abundances of all species other than neutrons decrease as $\tau^{-\frac{3}{2}}$. This means that the only further changes in abundance are those due to dilution in the continuing universal expansion. The relative abundances for large τ are therefore to be compared with the observed universal abundance data. Since the capture reactions become unimportant by large τ, the few percent of residual neutrons which will eventually decay into protons are then added to the proton concentration.

The nature of the growth curves depends quite strongly on the initial value chosen for C_0. For a C_0 larger than that used with Fig. 1, i.e., larger than $C_0'=1.07\times10^{17}$ cm^{-3}, the relative abundances rise more rapidly and go through a sharper maximum, whereas for a smaller C_0 the growth curve peaks are broadened. If C_0' is the value yielding a good representation of the general trend of the observed abundance data, then for $C_0>C_0'$ one finds too high a relative abundance of the heavier elements, while for $C_0<C_0'$ the only elements present in appreciable relative abundance are the lightest elements. These qualitative remarks, of course, imply the normalization of computed relative abundance values to the observed relative abundance of protons in all cases.

The final relative abundance values for the various solutions examined, i.e., corresponding to different values of C_0, are shown in Fig. 2, where all the solutions are joined to the data at $j=1$. Of the solutions shown, the best representation of the observed data is given by the curve marked C_0', which, as already mentioned, corresponds to a nucleon concentration at τ_0 of 1.07×10^{17} cm^{-3}. The solutions[9] shown cover a range 2.13×10^{16} cm$^{-3}\leqq C_0\leqq 2.13\times10^{18}$ cm^{-3}. The case $0.2C_0'$ runs out before reaching higher j values because the single-precision calculation did not permit greater accuracy. The dashed portion of the C_0' curve reflects the fact that for j greater than about 80 or 90, the accuracy of solution became as low as one significant figure. All curves are dashed past $j\cong185$, because by this j the general nature of the solutions permitted linear extrapolation to higher j. The data shown in Fig. 2 are those tabulated by Brown[3] as universal relative abundances, with circles and crosses distinguishing odd and even atomic weights, respectively. For $j<16$, Brown's values of elemental abundance were reduced to isobaric abundances with the aid of the tables of relative isotopic abundances given by Seaborg and Perlman.[10] For $j\geqq16$, isobaric abundances were computed directly from Brown's tables.[11]

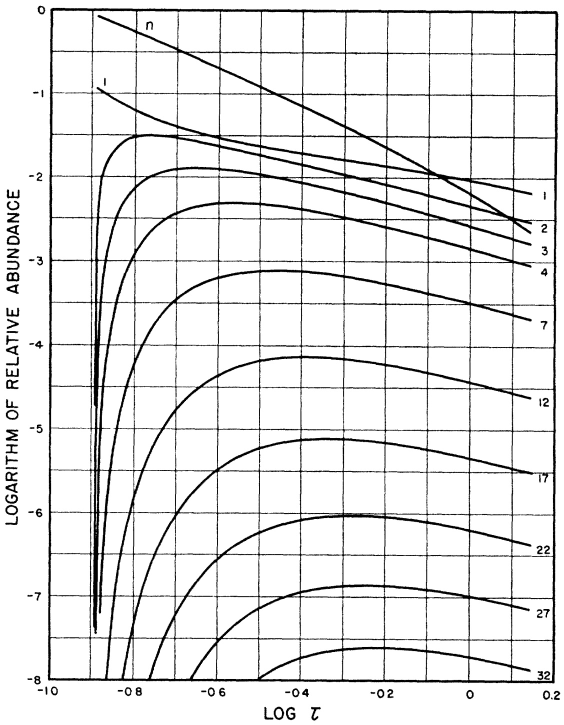

FIG. 1. Relative abundance as a function of time ($\tau=\lambda t$) according to the neutron-capture theory for an expanding universe. This is the case among those examined which best represents the observed data, namely, $C_0'=1.07\times10^{17}$ cm^{-3} at τ_0 (see Fig. 2). The curves are labeled to indicate atomic weight.

The initial nucleon concentration C_0' associated with the solution giving an adequate representation of the observed data in Fig. 2 is five times the C_0 which was

[8] A "double-precision" calculation would have yielded more accuracy for the higher atomic weights when the C_0 chosen was too small (i.e., when the theoretical solution yielded too low abundances of the heavier elements), but it was not deemed desirable because of the very considerable additional labor and machine running time required to accomplish this.

[9] The solution marked $20C_0'$ was a hand-computed solution.

[10] G. T. Seaborg and I. Perlman, Revs. Modern Phys. **20**, 585 (1948).

[11] There are two misprints in Brown's abundance tabulation, Table IV, in reference 3. Sb^{133} should read Sb^{123} and the abundance of Sn^{118} should be 0.149.

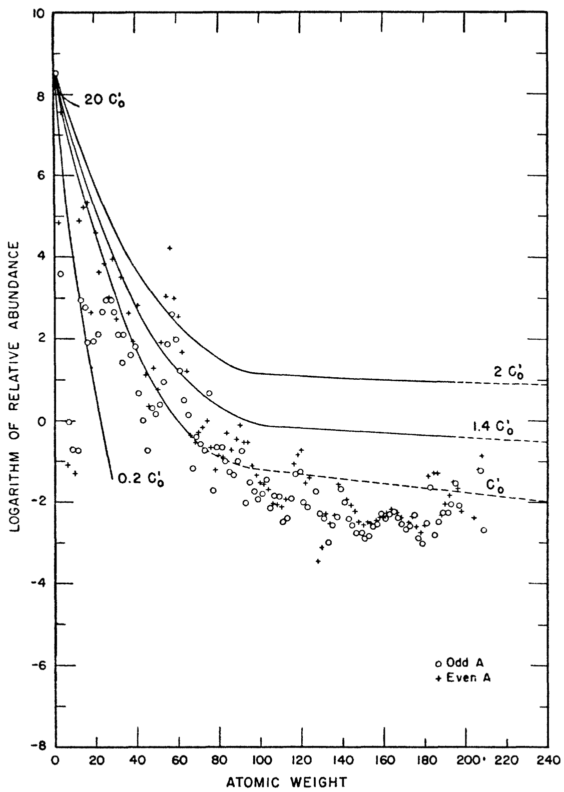

FIG. 2. Comparison of theoretical with observed relative abundances. The data are those of Brown (reference 3), while the curves are calculated according to Eqs. (5) with various values of C_0 to exhibit the effect of varying the initial nucleon concentration.

previously reported as being required in the case of element formation in a static medium.[2] The neutron-capture theory is thus seen to adequately describe the observed relative abundance data when the universal expansion is explicitly taken into account.

The effect of grouping nuclear species in order to reduce the number of Eqs. (5) was found to be small, as already mentioned. This was determined by integrating Eqs. (5) on the SEAC as in the grouped case, but with coefficients P_j corresponding to the first twenty-seven nuclear species individually, rather than with the grouped Π_j covering the entire atomic weight range by means of twenty-seven equations. The cases studied, which had initial nucleon concentrations of 1.4, 6, and 20 times the value C_0' already described as yielding an adequate solution with the grouped equations, indicated that an adequate solution would result with the ungrouped equations, provided the C_0 in the latter case was roughly five times the value in the case of grouping. Hence, a better estimate of the nucleon concentration to be assumed at $\tau=\tau_0$ in order to represent the general trend of the observed relative abundance data is $\sim 5.4\times 10^{17}$ cm^{-3}. This corresponds to a matter density of 8.9×10^{-7} g/cm^3 at $\tau_0=0.128$, or $\rho_0=1.5\times 10^{-3}$ g cm^{-3} sec$^{\frac{3}{2}}$ in Eq. (1).

It may be noted that,[2] in the nonstatic homogeneous isotropic cosmological model used in these calculations, there exists a relationship throughout the expansion between the densities of matter, ρ_m, and of radiation, ρ_r, namely, $\rho_r\rho_m^{-4/3}=$constant. Since $T=1.28\times 10^9$°K at τ_0, one obtains $\rho_r(\tau_0)=22.2$ g/cm^3. If one takes the smeared-out density of matter in the universe at the present time to be 10^{-29} g/cm^3 as suggested by Behr,[12] then according to the foregoing relationship, the present residual radiation density (not including that due to stellar radiation) is $\sim 5.5\times 10^{-30}$ g/cm^3 or $T\cong 28$°K.

IV. DISCUSSION

A. The Initial Neutron-Proton Ratio

As already mentioned, the neutron-capture theory in the approximation described does not include the variety of detailed processes and reactions which must have led in turn to the development of detailed features of the observed relative abundance data. In particular, a problem which merits more complete discussion here is the effect on the initial neutron-proton ratio of the physical conditions preceding the element-forming phase in the expansion. The choice of neutron and proton concentrations at $\tau=\tau_0$ influences the value of the initial nucleon concentration, C_0, and, therefore, of the density of matter which is required for the neutron-capture theory to represent the observed relative abundance data.

It will be recalled that the calculations described in this paper were based on initial values $\xi_n(\tau_0)=0.88$, $\xi_1(\tau_0)=0.12$. These are the neutron and proton concentrations which would result from the spontaneous decay of neutrons during the interval $0\leqq\tau\leqq 0.128$ with $\xi_n(0)=1$, and $\xi_1(0)=0$, where $\tau=0.128$ is the process starting time selected for reasons described in Sec. II. Such a formulation is a simplification of the more complex phenomena which may have occurred during the period of high temperature and density preceding the starting time selected for the element-forming process.

Hayashi[13] has recently examined in detail the many physical processes, including spontaneous neutron decay, which might have occurred during this period, the main purpose being to determine the effect of such processes on the neutron-proton ratio to be chosen as an initial condition for forming elements. Since the ratio obtained by Hayashi is quite different from that resulting from spontaneous neutron decay alone, it is desirable to discuss briefly his analysis and its effect on the neutron-capture theory.

Hayashi employed the cosmological model described in Sec. II, namely, an expanding universe of radiation with a trace of matter, in which the temperature and density of matter vary with time according to Eqs. (1) and (2). His analysis was restricted to the period of

[12] A. Behr, Astron. Nachr. **279**, 97 (1951).
[13] C. Hayashi, Prog. Theor. Phys. **5**, 224 (1950).

time after the temperature of the universe had decreased to a value less than that equivalent to the rest mass of the meson, *viz.*, $\sim 10^{12}$°K. This temperature corresponds to a time of $\sim 2\times 10^{-4}$ sec after the "beginning" of the universal expansion.

The following reactions among protons, p, neutrons, n, electrons, e^-, positrons, e^+, neutrinos, ν, antineutrinos, ν^*, and radiation, $h\nu$, were considered:

$$\begin{aligned} n+e^+ &\rightleftharpoons p+\nu^*, \\ n+\nu &\rightleftharpoons p+e^-, \\ n &\rightleftharpoons p+e^-+\nu^*, \end{aligned} \tag{6}$$

and

$$e^++e^- \rightleftharpoons h\nu.$$

The reaction rates for the processes described by Eqs. (6) were examined and, with the exception of β-processes, found to be sufficiently high to maintain certain of the concentrations at equilibrium values, even though the universal temperature decreases rather rapidly during this period.

The electron, positron, and photon concentrations were determined purely on classical statistical considerations, since the number of electrons and positrons involved in the lagging β-possesses is small compared with the number involved in the pair production—annihilation processes. Nonlinear rate equations were written to describe the β-processes, with the rate coefficients being evaluated from the Fermi theory of β-decay. There are no problems of degeneracy at the temperatures and densities considered. The rate equations were integrated numerically by Hayashi subject to the initial condition that, at $T\cong 10^{12}$°K, all particles and photons were present in equilibrium concentrations. Consequently, the initial neutron-proton ratio for the period considered by Hayashi was taken as unity. The solutions show that during the first five seconds the induced β-decay of the neutron is more important than spontaneous β-decay. Starting at unity, the neutron-proton ratio decreases to $\sim\frac{1}{3}$ by one second, to $\sim\frac{1}{4}$ by ten seconds, and is controlled by the relatively slow process of spontaneous neutron decay thereafter.[14]

Hayashi suggests that if nuclei beyond He^4 are ignored and if β-processes for the light nuclei may be neglected, then He^4 is effectively formed as $2n+2p\rightarrow He^4$, regardless of the formation route. If one neglects neutron decay and takes a starting neutron-proton ratio of 1/4 for the element-forming period, the final helium-hydrogen abundance ratio according to these ideas would be 1/6. This result is compatible with observed values. However, such an approach would appear to be an oversimplification because at low matter densities β-processes cannot be ignored in considering the details of the light element reactions and because the competition of neutron decay and universal expansion appear to be important in the element-forming process. The exact effect of a neutron-proton ratio of 1/4 should be investigated by a re-examination of the light element reactions in the manner of Fermi and Turkevich.[2]

The effect of Hayashi's neutron-proton ratio of 1/4 on the solution of Eqs. (5) has been examined by means of numerical integration on the SEAC. For this ratio two cases were studied, namely, those for $C_0=2.13\times 10^{16}$ cm^{-3} and $C_0=2.13\times 10^{18}$ cm^{-3}. In neither case did the neutron-capture process yield sufficient relative concentrations of the heavier elements. Furthermore, the growth curves for individual species in the two cases indicated that lower or higher initial values of C_0 would not improve the situation.[15] It is, of course, possible that intermediate values of C_0 might bring the theoretical curve closer to the observed data. However, it would appear to be necessary to increase the initial neutron-proton ratio to perhaps as much as unity in order to yield a fit. Unfortunately, it was not feasible to continue an examination of this problem in greater detail.

Since the neutron-capture theory is in fact quite approximate for the lightest elements and since the above mentioned SEAC calculations are not definitive, the question as to whether or not a low initial neutron-proton ratio is compatible with a satisfactory fit to the observed abundances still requires study of the light element reactions in detail.

[14] The neutron half-life used in Hayashi's calculations was 30 minutes, which is quite different from Robson's latest value of 12.8 minutes (see footnote 6). Since the neutron half-life is involved in almost all of the rate constants for Eqs. (6), a determination of the effect of changing the neutron half-life on the neutron proton ratio would appear to require a new integration.

[15] In the low density case, the neutron-capture reaction rates are so low that effectively all of the neutrons decay into protons before they can be captured, and the heavier nuclei are formed in extremely low relative abundances. In the high density case neutron-capture reaction rates are so high that spontaneous neutron decay and the effect of the universal expansion can be neglected. The result of this situation is that the few neutrons present to start with are quickly used up in forming the very lightest species, and again the heavier elements are formed in very small relative amounts. It may be of interest to note that in this case where the neutron-proton ratio is low and C_0 is very high, one may obtain an approximate analytical solution of Eqs. (5). Under these conditions the expansion terms $-3\xi_j/(2\tau)$ and the decay terms ξ_n may be ignored. In addition, one takes $P_{j-1}\xi_n\xi_{j-1}\gg P_j\xi_n\xi_j$ in the jth equation. Then the approximate solutions describing the growth curves may be written as follows:

$$\xi_n=q_0\{r_0\exp[q_0P_1(\tau-\tau_0)]-1\}^{-1},$$
$$\xi_1=r_0\xi_n\exp[q_0P_1(\tau-\tau_0)],$$

and

$$\xi_j=\frac{q_0(r_0P_1)^{1-j}}{(j-1)!}\prod_{m=1}^{j-1}P_m\sum_{s=0}^{j-1}(-1)^s\binom{j-1}{s}\exp[-sq_0P_1(\tau-\tau_0)],\quad j>1,$$

where

$$q_0=\xi_1(\tau_0)-\xi_n(\tau_0)$$

and

$$r_0=\xi_1(\tau_0)/\xi_n(\tau_0).$$

In the limit of long τ, the foregoing equations reduce to the final relative abundances in this approximation, *viz.*,

$$(\xi_1)_{\max}=q_0,\ (\xi_n)_{\max}=0,\ \text{and}\ (\xi_j)_{\max}=[q_0(r_0P_1)^{1-j}/(j-1)!]\prod_{m=1}^{j-1}P_m.$$

Calculations for the case $C_0=2.13\times 10^{18}$ cm^{-3}, with the Hayashi initial neutron-proton ratio of 1/4, check very well with solutions of the full Eqs. (5) as determined on the SEAC. The expression given for $(\xi_j)_{\max}$ is also derived in the Appendix by a different procedure.

B. Gaps at Atomic Weights 5 and 8

Hayashi has pointed out that a high proton concentration might be expected to aid materially in carrying the chain of element-forming processes across the missing nuclei at atomic weights 5 and 8.[2] In this connection, Turkevich[16] has recently suggested a reexamination of the light element reactions taking into account the high energy nonthermalized reaction products. In their previous calculations Fermi and Turkevich[2] considered all particles to have energies corresponding to the universal temperature. Because of the rapid increase with increasing energy in reaction probabilities for charged particles, the higher energy particles may be expected to provide a means of increasing the flux of nuclei crossing the gaps[17] at $A=5$ and 8.

C. Remarks Concerning Equilibrium Theories

The work described in this paper demonstrates that the general trend with atomic weight of the observed relative abundances of the elements can be represented satisfactorily by the neutron-capture theory in an expanding universe. In view of the low matter density required and the rapidity of the decrease of temperature and density with the expansion of the universe, the implication of this result in connection with equilibrium theories of element formation should again be pointed out. In these theories[18] nuclear species formed in statistical equilibrium in stellar interiors and are supposed to be distributed in space by subsequent stellar explosions. All of these stellar models contain high neutron concentrations and have high densities, $\rho \cong 10^8$ g/cm^3, and high temperatures, $T \cong 1$ Mev. Under these conditions (densities $\sim 10^{15}$ times greater than those required in the non-equilibrium theory) one would expect during an explosion a very considerable modification of the original equilibrium distribution of the elements by neutron capture and other reactions. In order to obtain the presently observed relative abundances as the end result of such a stellar explosion, it would appear necessary to carry out a non-equilibrium calculation of the type described in this paper but with the following differences. The dynamics of the explosion, and the density and temperature variation with time, as well as an initial distribution of relative abundances, would have to be known. This difficult problem amounts to finding what initial abundance distribution must be taken in order that it be modified to the observed universal distribution during the explosion.

D. Analytic Solution of the Equations of the Neutron-Capture Theory

The question has frequently been raised as to whether or not Eqs. (5) could be solved analytically. It had been suggested[2] that this could not be the case because the equations are nonlinear and because the neutron and proton rate equations contain terms of different orders. Recently, Dr. T. H. Berlin, of The Johns Hopkins University, has examined this problem and found that solutions could be obtained in closed form only if the expansion of the universe is ignored. A discussion of this solution is given in the Appendix.

We should like to express our appreciation to the National Bureau of Standards for its kindness in extending to us the use of the SEAC, and to Dr. Joseph H. Levin for his complete cooperation in coding and running the SEAC solutions. The computational assistance of Miss K. E. Pace is acknowledged with thanks. Finally, we should like to thank Dr. T. H. Berlin for his continued interest in this work.

APPENDIX

The Neutron-Capture Equations of Element Formation in a Static Universe: An Exact Solution

T. H. Berlin
Department of Physics, The Johns Hopkins University, Baltimore, Maryland

The equations describing element formation by successive neutron captures in a static universe are the set:[2]

$$\begin{aligned} d\xi_n/d\tau &= -\xi_n - \sum_{j=1}^{J} P_j \xi_n \xi_j, \\ d\xi_1/d\tau &= \xi_n - P_1 \xi_n \xi_1, \\ d\xi_j/d\tau &= P_{j-1}\xi_n\xi_{j-1} - P_j \xi_n \xi_j, \quad 2 \leqq j \leqq J. \end{aligned} \tag{A1}$$

These equations are to be solved under the initial conditions:

$$\xi_j(\tau_0)=0, \quad 2\leqq j \leqq J; \quad \xi_1(\tau_0)=\alpha; \quad \xi_n(\tau_0)=\beta,$$

where $\tau_0=0$ is obviously suitable for the static case.

Equations (A1) are linearized by changing the independent variable from τ to z so that

$$dz/d\tau = \xi_n. \tag{A2}$$

The equations become

$$\begin{aligned} dz/d\xi_n &= -1 - \sum_{j=1}^{J} P_j \xi_j \equiv -F(z), \\ d\xi_1/dz &= 1 - P_1 \xi_1, \\ d\xi_j/dz &= P_{j-1}\xi_{j-1} - P_j \xi_j, \quad 2\leqq j \leqq J. \end{aligned} \tag{A3}$$

[16] Private communication.

[17] Turkevich (private communication) has also suggested using the higher energy neutrons formed as reaction products among the light nuclei in $(n,2n)$ reactions with heavier nuclei to form shielded isobars.

[18] See Sec. III of reference 2 for a discussion and bibliography.

Equations (A3) are conveniently solved by the use of the laplace transform.[19] Setting

$$f_j(\kappa)=\int_0^\infty e^{-\kappa z}\xi_j(z)dz, \tag{A4}$$

and using the initial conditions, we have from Eq. (A3)

$$\begin{aligned} &f_1(\kappa)=(\kappa^{-1}+\alpha)/(\kappa+P_1),\\ &f_j(\kappa)=[P_{j-1}/(\kappa+P_j)]f_{j-1}(\kappa), \quad 2\leqq j\leqq J. \end{aligned} \tag{A5}$$

Then,

$$f_j(\kappa)=P_j^{-1}(\kappa^{-1}+\alpha)\prod_{k=1}^{j}P_k(\kappa+P_k)^{-1}, \quad 1\leqq j\leqq J. \tag{A6}$$

Inverting Eq. (A4), we have

$$\xi_j(z)=(1/2\pi i)\int_{\kappa_j-i\infty}^{\kappa_0+i\infty}e^{\kappa z}f_j(\kappa)d\kappa, \tag{A7}$$

where the line $\kappa=\kappa_0$ is to the right of the singularities of $f_j(\kappa)$.

If it is assumed that the P_k are all different and not equal to α^{-1}, $f_j(\kappa)$ has simple poles at $\kappa=0, -P_1, -P_2, \cdots, -P_j$. Therefore, we have

$$\xi_j(z)=P_j^{-1}+\sum_{r=1}^{j}A_{jr}e^{-P_r z}, \tag{A8}$$

where

$$A_{jr}=\lim_{\kappa\to -P_r}(\kappa+P_r)f_j(\kappa).$$

On taking the limit, we obtain

$$A_{jr}=-P_r^{-1}(1-\alpha P_r)R_{jr},$$

where

$$R_{jr}=\prod_{\substack{k=1\\k\neq r}}^{j}P_k(-P_r+P_k)^{-1}; \quad R_{11}=1. \tag{A9}$$

We now see that $F(z)$ is a known function of z. Hence, we have

$$\xi_n(z_0)-\xi_n(0)=-\int_0^{z_0}F(z)dz. \tag{A10}$$

This implies that $\tau(z)$ may be found from

$$\tau=\int_0^z ds/\xi_n(s). \tag{A11}$$

One is usually not interested in the ξ_j at a particular time but in the limiting value of ξ_j, that is, at $\tau=\infty$. This means that there is no necessity to use Eq. (A11).

From Eq. (A2), we see that $(dz/d\tau)_{\tau\to\infty}=0$, since all the neutrons are eventually captured or decay into protons, so that ξ_n approaches 0. Since $\xi_n\geqq 0$, z is a monotonically increasing function of τ and approaches a limiting value $z(\tau=\infty)=z_0$. Then $\xi_n(z_0)=0$, and from Eq. (A10) we have, because $\xi_n(0)=\beta$,

$$\beta=\int_0^{z_0}F(z)dz \tag{A12}$$

as the determining equation for z_0, which will have a unique solution for given α, β. The limiting values of the ξ_j are $\xi_j(z_0)$ obtained from Eq. (A8).

We may remark briefly on some aspects of the above solution of the problem. Let P_m, say, be zero. We then see from Eq. (A5) that $f_j(\kappa)=0$ for $j\geqq m+1$, so that $\xi_j=0$ for $j\geqq m+1$. In other words, if a species has zero (i.e., very small) capture cross section, then all heavier species will have effectively zero abundance. However, the formation chain would be expected to continue through isobars of more normal cross section.[2]

To facilitate numerical work with the approximations to the Eq. (A1), Alpher and Herman assumed that P was constant for those elements with $j\geqq 100$. This is an inconvenient assumption for the solution given. If some P's are alike, we see from Eq. (A6) that $f_j(\kappa)$ will have multiple poles. The exact inversion can easily be performed to give ξ_j but high order derivatives of $f_{100}(\kappa)$ will be involved. The resulting expression for ξ_j would be quite inconvenient for numerical work; and it would be very much simpler to assume that, instead of being constant, P_0 is a slowly increasing function of j for $j>100$ in order to maintain all P_j different. It appears from the cross-section data that this is as reasonable an assumption as taking the P_j the same.

The exact solution would be of increased interest if the dependence of the ξ_j on the initial conditions α, β and on density could be put into simple form. Since the P_j are all proportional to density, we note that R_{jr} [Eq. (A9)] is independent of density and may be computed, given the effective neutron capture cross section only.

If we set $z=y/n_0$, then from Eq. (A8), we have

$$P_j\xi_j=1-\sum_{r=1}^{j}R_{jr}\exp\left(\frac{-P_r y}{n_0}\right)+n_0\alpha\sum_{r=1}^{j}\left(\frac{P_r}{n_0}\right)R_{jr}\exp\left(\frac{-P_r y}{n_0}\right). \tag{A13}$$

The two sums over r are now density independent functions of y.

From the definition of $F(z)$, Eq. (A3), we find that Eq. (A12) may be written in the form

$$n_n(0)-n_1(0)G(y_0)=(J+1)y_0-H(y_0), \tag{A14}$$

where

$$\xi_j=n_j/n_0; \quad z_0=y_0/n_0;$$

$$G(y)=\sum_{j=1}^{J}\sum_{r=1}^{j}R_{jr}\left[1-\exp\left(\frac{-P_r y}{n_0}\right)\right];$$

$$H(y)=\sum_{j=1}^{J}\sum_{r=1}^{j}\left(\frac{n_0}{P_r}\right)R_{jr}\left[1-\exp\left(\frac{-P_r y}{n_0}\right)\right];$$

[19] H. Bateman, Proc. Camb. Phil. Soc. **15**, 423 (1910). Bateman has solved an almost identical set of equations. The principal differences are in the equations for ξ_n and ξ_1.

and $G(y)$, $H(y)$ are density independent functions that can be computed given the capture cross sections.

In Eq. (A14) we see that the initial neutron and proton densities are very simply exhibited.

We can readily evaluate ξ_j in the two limiting cases $n_n(0)$ very much greater and very much less than $n_1(0)$.

Case: $n_n(0) \ll n_1(0)$:

From the definitions of G and H,

$$G(\infty)=\sum_{j=1}^{J}\sum_{r=1}^{j}R_{jr} \quad \text{and} \quad H(\infty)=\sum_{j=1}^{J}\sum_{r=1}^{j}\left(\frac{n_0}{P_r}\right)R_{jr}.$$

Therefore, as $n_n(0)$ becomes very large, we must have

$$y_0 \cong n_n(0)/(J+1).$$

From Eq. (A13), we have $P_j\xi_j \cong 1$, so that $n_j \cong n_0/P_j$, which is finite. Thus, with increasing initial neutron density, the abundance distribution approaches the stationary distribution for which $d\xi_j/d\tau=0$.

Case: $n_n(0) \ll n_1(0)$:

In this case we can expect that the solution, y_0/n_0, of Eq. (A14) is small. Dealing with the leading terms only, then, we have

$$G(y) \cong \sum_{j=1}^{J}\sum_{r=1}^{j}R_{jr}\left(\frac{P_r y}{n_0}\right)=\left(\frac{P_1}{n_0}\right)y,$$
$$H(y) \cong \sum_{j=1}^{J}\sum_{r=1}^{j}R_{jr}\left(y-\frac{P_r y^2}{2n_0}\right)=Jy. \tag{A15}$$

These results are obtained by noting that

$$\sum_{j=1}^{J}P_j\xi_j(0)=P_1\alpha,$$

since only $\xi_1(0)\neq 0$, and by using Eq. (A13). Thus, Eq. (A14) becomes

$$n_n(0)-n_1(0)(P_1/n_0)y_0 \cong y_0,$$

so that

$$y_0/n_0 \cong [n_n(0)/n_0]/[n_1(0)(P_1/n_0)+1] \cong [n_n(0)/n_1(0)]P_1^{-1}, \tag{A16}$$

which is a small number for high density.

Now we must consider $\xi_j(z_0)$ for z_0 small, since $z_0=y_0/n_0$. This is conveniently done through Eqs. (A6) and (A7). We shall use

$$\xi_j(z_0)=\sum_{p=0}^{\infty}\left[\frac{d^p\xi_j(z)}{dz^p}\right]_{z=0}\left(\frac{z_0^p}{p!}\right). \tag{A17}$$

However, we have

$$[d^p\xi_j/dz^p]_{z=0}=(1/2\pi i)\int_{\kappa_0-i\infty}^{\kappa_0+i\infty}\kappa^p f_j(\kappa)d\kappa. \tag{A18}$$

From Eq. (A6) we see that as $\kappa\rightarrow\infty$, $f_j(\kappa)\rightarrow 1/\kappa^j$. Thus, for $p<j-1$, the integrand vanishes at infinity in the right half-plane faster than $1/\kappa$. Consequently, we have

$$[d^p\xi_j/dz^p]_{z=0}=0 \quad \text{for} \quad p<j-1. \tag{A19}$$

The first nonvanishing derivative of ξ_j at $z=0$ occurs for $p=j-1$. Transforming to the complex variable $\kappa=1/s$, we have

$$[d^{j-1}\xi_j/dz^{j-1}]_{z=0}=(1/2\pi i)\int_{\kappa_0-i\infty}^{\kappa_0+i\infty}\kappa^{j-1}f_j(\kappa)d\kappa$$
$$=(1/2\pi i)\int^{(0+)}s^{-j-1}f_j(1/s)ds, \tag{A20}$$

and

$$\left[\frac{d^{j-1}\xi_j}{dz^{j-1}}\right]_{z=0}=\alpha\prod_{k=1}^{j-1}P_k.$$

Finally, the leading term, $p=j-1$, for $\xi_j=n_j/n_0$ is

$$n_j=\left[\frac{n_1(0)}{(j-1)!}\right]\left[\prod_{k=1}^{j-1}\left(\frac{P_k}{n_0}\right)\right]\left\{\left[\frac{n_n(0)}{n_1(0)}\right]\left(\frac{n_0}{P_1}\right)\right\}^{j-1}. \tag{A21}$$

The author wishes to thank Dr. Herman and Dr. Alpher for their many pertinent suggestions and clarifying discussions.