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NUCLEOSYNTHESIS IN THE FARLY HISTORY OF THE SOLAR SYSTEM

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ABSTRACT

This paper revises the model of Fowler, Greenstein, and Hoyle (FGH) for the nucleosynthesis of D,Li,Be, and B by high energy particles from the sun during the early history of the solar system. In this model these nuclei are produced by spallation reactions, mainly on 0^{16} , in metric-sized planetesimals. Large numbers of neutrons are also produced. A fraction of these are thermalized and react by $B^{10}(n,\alpha)Li^7$, $Li^6(n,\alpha)H^3$, to produce the terrestrial Li and B isotopic ratios. Additional D is produced by $H^1(n,\gamma)$ on hydrogen retained in the planetesimals as H_20 . The total energy required in high energy particles is about 2×10^{44} ergs.

The nuclear calculations have been generalized in an approximate manner to include a dependence on the duration of the irradiation caused by the long lifetime of Be¹⁰. The first stage of the calculation yields the required spallation yields and time-integrated neutron flux to produce the terrestrial Li, Be, and B abundances and isotopic ratios. The required flux is 4×10^{21} n/cm², identical with that obtained by FGH, and does not depend significantly on the choice of irradiation time.

The predicted spallation yields depend more strongly on the irradiation time. These are compared with cloud chamber data for 0^{16} + 300 MeV neutrons. The predicted low spallation yield for Be⁹, which merely reflects its low

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relative abundance, is consistent with the cloud chamber data. Good agreement is not obtained for the B/Li spallation ratio; however, for large values of the irradiation time, this could be due to uncertainties in the parameters. This does not seem possible for short irradiation times, however. The nuclear processes are less reasonable if C is present in the planetesimals or if higher energy particles are assumed in order to get appreciable amounts of LiBeB from spallation of SiFe as well as from 0^{16} .

The second stage of the calculation yields the required hydrogen concentration, $H/Si \approx 1$, and "dilution factor," $F_d \approx 20$ (approximately the ratio of unirradiated to irradiated material) to give the terrestrial D/H ratio. FGH calculated $F_d = 10$ and H/Si = 8. The amount of water is considerably reduced in the present calculations. Although a loss of 0 during the formation of the earth still must be postulated, the amount to be lost is much less than in the FGH case.

We point out that the FGH model is compatible with suggestions that the moon has a high water content and that biotic material has formed in the carbonaceous chondrites and on the lunar surface.

The nucleosynthesis of C^{13} in its present terrestrial abundance appears quite feasible; however, in this case the solar C^{13}/C^{12} ratio will be much less than that observed terrestrially. Conflicting experimental results exist on this point at the present time. Observed C^{13}/C^{12} variations in meteorites appear to be due to chemical fractionation. The fact that the isotopic composition of Li, Gd, and K in stone meteorites is identical with that found terrestrially requires that both terrestrial and meteoritic material were subjected to the same particle flux and had the same fraction of material irradiated. This implies that the earth and the meteorites had a common initial history if the basic features of this model are to be retained. A lunar origin for stone meteorites could very well provide the required astrophysical situation to meet this requirement, whereas an asteroidal origin presents many more difficulties.

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I. INTRODUCTION

Deuterium and the isotopes of lithium, beryllium, and boron (DLiBeB)[†] are important exceptions to the general rule that the chemical elements can be synthesized by chains of thermonuclear reactions occurring in stellar interiors as, for example, described by <u>Burbidge</u>, <u>Burbidge</u>, <u>Fowler</u>, <u>and Hoyle</u> [1957] (B²FH). At stellar temperatures and densities DLiBeB are destroyed rather than synthesized by thermonuclear reactions.

The fact that particles may be accelerated to high energies by magnetic activity on the surfaces of typical stars such as the sun led to suggestions that it may be possible to produce LiBeB by means of spallation reactions on the CNO nuclei present [BFH, 1957; Burbidge, Burbidge, and Fowler, 1958; Bashkin and Peaslee, 1961]. However, Fowler, Greenstein, and Hoyle [1960] (FGH) pointed out that it was unreasonable to assume that simple spallation would produce the observed $\text{Li}^7/\text{Li}^6 \approx 12.5$ and $\text{B}^{11}/\text{B}^{10} \approx 4$ ratios; moreover, one would expect to get more B than Li from spallation of CNO; whereas the meteoritic abundance ratio is $Li/B \sim 5$. FGH showed that these difficulties could be overcome if the spallation were assumed to occur in solid bodies rather than in a gaseous medium. Neutrons will also be produced in the spallation reactions and a fraction of these will become thermalized and be captured in the bodies. As the concentration of LiBeB increases, the thermal neutrons will begin to react with the Li⁶ and B¹⁰ by Li⁶(n, α)H³ and B¹⁰(n, α)Li⁷. The (n, α) reactions on Li⁷, Be⁹, and B¹¹ are endothermic and will not occur for thermal neutrons. This increases the B^{11}/B^{10} and Li^7/Li^6 ratios; furthermore, the

In referring to a series of nuclei we will generally omit commas and the word "and". Following the notation used by cosmic-ray physicists, we will frequently refer to LiBeB as "the description"

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conversion of B^{10} into Li⁷ changes the Li/B ratio in the direction of the present-day meteoritic abundance ratio. Neutrons are not effective in the case of nucleosynthesis on stellar surfaces because most of the neutrons will be captured by the $H^1(n,\gamma)D^2$ reaction and few will be available for reaction with Li⁶ and B^{10} ; furthermore, at the low densities in stellar atmospheres many of the neutrons would beta-decay before undergoing any reaction (see FGH Section III, 10).

FGH placed the site of nucleosynthesis in solid bodies called planetesimals which were assumed to have formed at low temperatures in the solar nebula during the hearly history of the solar system. The source of high energy particles was the early sun which was assumed to be magnetically very active [<u>Hoyle</u>, 1960]. Further discussion of the astrophysical conditions is given in Section II.

The approach used in this paper will be similar to that of FGH. A revision is appropriate because there has been considerable improvement in the experimental geochemical and nuclear data which was used by FGH. These changes will be considered in more detail at the appropriate places in the following discussion.

Section II outlines the astrophysical conditions assumed in the present calculations. These have not changed significantly from those assumed by FGH; so this section could be omitted without loss of continuity by those familiar with the FGH paper. Section III contains the nuclear calculations. As well as changes due to experimental work since the publication of FGH, the other parameters of the model have been thoroughly reviewed and in some cases revised. The basic idea behind this re-examination was that - even if we could not improve on the FGH values - we should determine the extent of their uncertainty and the effect of these uncertainties on the conclusions. Much of the

discussion of the choice of, and the uncertainties in these parameters has been placed in appendices in order to maintain continuity for readers not interested in the details of the calculations. Section IV is a discussion of the conclusions and consequences of the nuclear calculations. We have tried to be selective rather than comprehensive in that we have not attempted to re-examine all the topics discussed by FGH. Rather we have concentrated on the interpretation of recent experimental work which sheds light on the conclusions drawn from these calculations. An exception is that an interpretation of the rather large amount of data on the isotopic composition of xenon and other rare gases in meteorites has not been attempted although the production of these isotopic anomolies was - and still is - an important consequence of the FGH calculation.

II. ASTROPHYSICAL CONSIDERATIONS

As in FGH, the present calculations are based on a model for the early solar system proposed by <u>Hoyle</u> [1960]. The reader is referred to the work of Hoyle and to FGH for the details of this model. This paper will attempt no modification or significant elaboration of these discussions.

The early solar system according to Hoyle is outlined below and presented schematically in Figure 1.

(a) Rotational instability developed in the condensation of the Sun at a time when its radius was about 3×10^{12} cm ≈ 40 R_o which is slightly inside the present orbit of Mercury. This leads to the formation of an equatorial disk of mass $\approx 1\%$ of the solar mass. The Sun was spinning rapidly at this early stage.

(b) A magnetic torque coupling developed between the Sun and the disk which opposed and slowed the rotation of the Sun. Solar rotation led to a twisting

of the field lines and a transfer of angular momentum to the disk causing it to separate and move out from the Sun. Eventually the bulk of the material moved to distances corresponding to the major planets leaving the region of the terrestrial planets gas-free.

(c) In the region of the terrestrial planets any bodies greater than metric dimensions would be left behind in the outflowing gas. These are the "planetesimals" referred to earlier. The planetesimals were composed of the non-volatile portion of solar material, mainly MgSiFe in oxidized form, plus hydrogen bound as H₂O. The hydrogen plays an important role in the moderation of the neutrons. We assume that the volatile constituents $\rm H_{\odot},~CH_{h},~NH_{3},$ and the rare gases were not incorporated into the planetesimals in any appreciable amounts. Further, it must be assumed that the He³ produced by the spallation reactions can escape rapidly. This is necessary because He³ acts as a "neutron sink" cue to its large neutron capture cross-section (5500b). More discussion of this assumption is included in Section IIID and Appendix D. (d) The rotational energy ($\sim 5 \times 10^{45}$ erg) of the solar condensation was stored in the sun as magnetic energy and was dissipated by means of solar flares. About 2×10^{44} erg appeared in the form of high energy particles, mostly protons. Following FGH we adopt the proton energy spectrum derived theoretically by Parker [1957] which agreed with the experimental spectrum observed by Meyer, Parker, and Simpson [1956]. The mean energy of this spectrum is about 500 MeV; however, neither this nor the exact form of the spectrum enter the calculations directly. The accelerated particles travelled along magnetic field lines and irradiated the planetesimals. In order that the attenuation of the charged particles be small, the residual gas density in the region of the terrestrial planets must be of the order 10^{-11} - 10^{-12} gm/cc.

At these densities the fraction of the hydrogen ionized by the particle flux is about 10^{-5} - 10^{-4} , which is considerably greater than the 10^{-7} set by <u>Hoyle</u> [1960] as the minimum for which the coupling of the magnetic field to the disk can be maintained.

III. NUCLEAR CALCULATIONS

The nuclear process with which we shall be concerned is the irradiation of the planetesimals by high-energy protons. For each 10^6 Si atoms p protons react and in turn produce n neutrons and L_s nuclei of LiBeB by spallation reactions. Of the n neutrons produced, n_r reach thermal energies and are captured. The effect of the capture process on the L nuclei is to deplete Li^6 and B^{10} and enhance Li^7 by the reactions $Li^6(n,\alpha)H^3$ and $B^{10}(n,\alpha)Li^7$. Neutron capture on the hydrogen retained in the planetesimals as H_20 as well as deuterons from the spallation processes produce deuterium. (We use the term "spallation" to include all nuclear reaction mechanisms by which L nuclei may be produced in high energy reactions.)

A. Production of LiBeB

The equations for the production rate of L nuclei will be written in terms of the number of neutrons, n, as the independent variable. Writing the equations in this way, <u>i.e.</u>, using the number of L nuclei produced per n neutrons, has the effect of averaging over much of the complicated time and space dependence of the general problem. Also there is no necessity to make any specific assumptions about the planetesimal sizes except that they must be large enough for a secondary neutron flux to be developed. According to recent calculations by <u>Mitler</u> [1964], this implies that the planetesimals must have radii greater than about 20 cm.

Thus, for the production of Li⁶ we can write (following FGH)

$$\frac{dLi^{6}}{dn} = \alpha_{6} - \frac{f_{r}\sigma_{6}Li^{6}}{\sum_{A}\sigma_{A}N_{A}} \qquad (1)$$

The symbol Li⁶ indicates the abundance of Li⁶ at some stage during the irradiation process. The quantity α_6 is the ratio of the production rate of Li⁶ by spallation to the neutron production rate. It also includes the production of He⁶ by spallation which beta-decays to Li⁶ with a 0.8 sec half-life. Strictly speaking, α_6 represents the ratio of the cross-sections averaged over the differential proton energy spectrum <u>within</u> the planetesimal, averaged over depth within the planetesimal, averaged over the chemical composition of the planetesimals, and averaged over the planetesimal size distribution. <u>Mitler</u> [1964], in a re-evaluation of the FGH calculation, attempts to calculate these spallation rates directly. However, at the present time we feel too little is known even of the cross-sections themselves (let alone the other required distributions) to attempt such a direct evaluation; thus α_6 is retained as a parameter. The other LiBeB spallation rates will be treated similarly as parameters. We assume only that these production rates are independent of n, <u>i.e.</u>, that the shape of the proton spectrum does not change with time.

The second term in Equation (1) is the rate at which Li⁶ disappears due to the (n, α) reaction (proportional to σ_6 Li⁶) relative to the rate at which neutrons are being formed. The cross-section for Li⁶(n, α)H³ is σ_6 . The rate at which neutrons are being formed is equal to the rate at which they are captured (proportional to $\Sigma N_A \sigma_A$) divided by the fraction of the neutrons produced which react, f_r , where the neutron capture cross-sections and relative abundances of the nuclear species in the planetesimals are σ_A and N_A respectively.

The neutron capture cross-sections depend on the temperature. This was

estimated by FGH to be 130-200° K although somewhat higher temperatures $(200-250^{\circ})$ would be more in accord with recent solar evolution calculations [Hayashi, 1961; Iben, 1965]. For this range of temperatures the capture cross-sections are expected to vary as 1/v or as $T^{-\frac{1}{2}}$ to a fairly good approximation, since the planetesimals are composed predominantly of light elements. Therefore, since only the ratio of cross-sections appears in the equations, values measured for 2200 m/s (about 293° K) can be used.

As is shown in Appendix D, no nucleus which makes an appreciable contribution to $\sum_{A} N_{A} \sigma_{A}$ has its abundance significantly changed by the irradiation process; thus this sum is taken to be constant and is designated by the symbol \sum for convenience.

Equation (1) gives

$$\operatorname{Li}^{6} = \alpha_{6} n \left(\frac{1}{\psi_{n} \sigma_{6}} \right) \left[1 - \exp(-\sigma_{6} \psi_{n}) \right]$$
(2)

$$= \operatorname{Li}_{s}^{6} \left(\frac{1}{\psi_{n} \sigma_{6}} \right) \quad \text{for } \psi_{n} \sigma_{6} >> 1 , \qquad (2')$$

where Ψ_n is defined as $nf_r/\Sigma = n_r/\Sigma$, n_r being the number of neutrons which react. Defined in this way, Ψ_n is the value of the time-integrated neutron flux (neutrons/cm²) which reacts in the planetesimal. For the cases of interest $\sigma_6 \Psi_n \approx 3-5$, so Equation (2') may be used. The amount of Li⁶ made by spallation is defined as $Li_s^6 = \alpha_6 n$.

The equations for the production of Be^9 and B^{11} are simpler since these nuclei are not affected by the neutrons and produced only by spallation.

$$\frac{dBe^9}{dn} = \alpha_9 \tag{3}$$

$$\frac{dB^{11}}{dn} = \alpha_{11}$$

The quantity α_9 includes only direct formation of Be⁹ since B⁹ is particle unstable and Ld⁹, although itself particle stable, decays to a particle unstable state in Be⁹. The quantity α_{11} includes the contributions of C¹¹ $(t_{\frac{1}{2}} = 20.5 \text{ min})$ and Be¹¹ $(t_{\frac{1}{2}} = 13.6 \text{ sec})$ as well as the direct production of B¹¹.

The production of B¹⁰ is complicated by the existence of long-lived Be¹⁰ with a mean life (half-life/0.693) $\tau = 3.9 \times 10^6$ yr for beta-decay to B¹⁰. Thus, if the time of irradiation is not long compared to the mean life of Be¹⁰, a significant fraction of the present-day B¹⁰ may survive the irradiation as Be¹⁰. Only the Be¹⁰(n, γ)Be¹¹ reaction is energetically possible at thermal energies, and the cross-section for this is expected to be very small since the neutron binding energy in Be¹¹ is only about 0.5 MeV.

Astrophysically, the time of irradiation corresponds to the time required to dissipate the magnetic energy stored in the early Sun. <u>Hoyle</u> [1960] estimated this to be comparable to the contraction time of the early Sun. This was taken to be roughly 10^7 yr in FGH and considered to be very long compared to the mean life of Be¹⁰. Since this estimate is only order of magnitude, we have generalized the equation for the production of B¹⁰ to hold for cases where the irradiation time is short or comparable to the lifetime of Be¹⁰. As our best estimate of the contraction time, we will take the value of roughly 3×10^7 yr obtained by <u>Tben</u> [1965] in recent calculations on solar evolution.

In terms of time the production rates of B^{10} and Be^{10} are:

$$\frac{dB^{10}}{dt} = r_{10} + \frac{Be^{10}}{\tau} - \frac{\Psi_n}{T} B^{10} \sigma_{10}$$
(4)

$$\frac{dBe^{10}}{dt} = r_{10}' - \frac{Be^{10}}{\tau}$$
(5)

where r_{10} and r'_{10} represent the spallation rates of B¹⁰ plus C¹⁰ ($\tau_{\frac{1}{2}} = 19 \text{ sec}$) and Be¹⁰ respectively. For this calculation we have assumed explicitly that any variations in the proton flux occur in times small compared to τ . Thus r_{10} and r'_{10} will be taken as constant, and the neutron flux (neutrons/cm² sec) will also be steady in time and given by Ψ_n/T where T is the duration of the irradiation. The term Be¹⁰/ τ represents the beta-decay rate of Be¹⁰, and the third term in Equation (4) represents the depletion of B¹⁰ by the (n, α) reaction with cross-section σ_{10} .

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Equations (4) and (5) may be solved to give the present-day B¹⁰ abundance

$$B^{10} = Be^{10}(T) + B^{10}(T)$$

$$= Be_{s}^{10} \left[\frac{1 - \exp(-T/\tau)}{T/\tau} + \frac{1 - \exp(-\psi_{n}\sigma_{10})}{\psi_{n}\sigma_{10}} - \frac{\exp(-T/\tau) - \exp(-\psi_{n}\sigma_{10})}{\psi_{n}\sigma_{10} - T/\tau} \right] + \frac{B_{s}^{10}}{\psi_{n}\sigma_{10}} \left[1 - \exp(-\psi_{n}\sigma_{10}) \right]$$
(6)

$$= B_{s}^{10} \left\{ \frac{1}{\psi_{n}\sigma_{10}} \left[1 + f_{10} - f_{10} \exp(-T/\tau) \right] + f_{10} \left[1 - \exp(-T/\tau) \right] \right\}$$
 (6')
for $\psi_{n}\sigma_{10} >> 1$ and $\psi_{n}\sigma_{10} >> T/\tau$

where $B_s^{10} = r_{10} T$ and $Be_s^{10} = r_{10}' T$ are the amounts of B^{10} and Be^{10} produced in spallation, and $f_{10} = Be_s^{10}/B_s^{10}$. For the special case when $T/\tau >> 1$, Equation (6) reduces to:

$$B^{10} = (Be_{s}^{10} + B_{s}^{10}) \frac{[1 - exp(-\psi_{n \ 10})]}{\psi_{n}\sigma_{10}}$$
(6")

which is identical to Equation (14) used by FGH if we note that their symbol, B_s^{10} , includes the Be¹⁰ produced by spallation. For the case where $T/\tau \ll 1$

and Ψ_n large, Equation (6) reduces to $B^{10} = Be_s^{10}$ which, physically, means that all the B_s^{10} produced by spallation will be "burned up" by the neutron flux and the present-day B^{10} is just the Be_s^{10} produced by spallation which did not decay in the short irradiation interval. If we do not assume Ψ_n large, then for $T/\tau \ll 1$ Equation (6) reduces to that given by FGH in Appendix 1, Equation (A1). For most cases to be considered $\Psi_n \sigma_{10} \approx 15$, so Equation (6') will be valid for values of $T \lesssim 10^7$ yr.

Li⁷ is produced by direct spallation and as a product of the (n,α) reaction on B¹⁰. Since the amount of the B¹⁰ burned is Be¹⁰_s + B¹⁰_s - B¹⁰,

$$\text{Li}^{7} = \text{Li}_{s}^{7} + \text{Be}_{s}^{10} + \text{B}_{s}^{10} - \text{B}^{10}$$
 (7)

where Li_{s}^{7} includes the contribution of Be^{7} ($t_{\frac{1}{2}} = 53$ days) as well as Li^{7} produced directly.

B. Calculation of the Neutron Flux

As noted earlier in the discussion of Li⁶ production, we feel that experimental data on spallation and neutron cross-sections, and our understanding of the development of secondary fluxes in solid bodies are insufficient to make a direct calculation of the relative abundances from nuclear data. We will thus turn the calculation around and use the observed abundances to estimate the neutron flux and the spallation yields. Rearranging Equation (7) with the help of Equations (6') and (2'), an equation for Ψ_n may be derived:

$$\Psi_{n} = \frac{Li^{7} + B^{10}}{Li^{6} \sigma_{6} \frac{\alpha_{7}}{\alpha_{6}} + B^{10} \sigma_{10} g}$$
(8)
$$= \frac{I_{7} + I_{10} B/Li}{I_{6} \sigma_{6} \frac{\alpha_{7}}{\alpha_{6}} + I_{10} \frac{B}{Li} \sigma_{10} g}$$
(8)

where the irradiation time dependence is contained in g:

$$g = \frac{1 + f_{10}}{1 + f_{10} [1 - \exp(T/\tau)] + \frac{f_{10} \psi_n \sigma_{10}}{T/\tau} [1 - \exp(-T/\tau)]}$$

For $T/\tau \gg 1$, g = 1, Equation (8) reduces to Equation (21) in FGH.

We have made the following choice for the other parameters in Equation (8'):

$$I_{6} = Li^{6}/Li = 0.0742 \text{ terrestrial value}$$

$$I_{7} = Li^{7}/Li = 0.926 \qquad " \qquad "$$

$$I_{10} = B^{10}/B = 0.196 \qquad " \qquad "$$

$$\sigma_{6} = 945 \text{ b at } 2200 \text{ m/s from Hughes and Schwartz [1958]}$$

$$\sigma_{10} = 3813 \text{ b under same conditions}$$

$$B/Li = 0.21 \text{ from the meteoritic abundances of Shima and Honda [1963], Krankowsky and Mueller [1964], and Shima [1962]
$$\alpha_{7}/\alpha_{6} = Li_{8}^{7}/Li_{8}^{6} = 1.9 \text{ based on the work of Gradsztajn, Epherre, and Bernas [1963].$$$$

 $f_{10} = 0.25$

A detailed discussion of the abundance and nuclear data used to obtain these numbers is given in Appendix A along with a discussion of the effect of uncertainties in the various parameters. The principal change from the parameters used by FGH is in α_7/α_6 which FGH estimated as 1. FGH used a value of B/Li = 0.24 which differs only slightly from that used here.

Equation (8') is quadratic in Ψ_n and can be solved for various choices of T/τ . The results of this calculation are presented in Table 1. Note that for irradiation times ranging from 0 to ∞ , Ψ_n varies only by a factor of two, <u>i.e.</u>, the neutron flux required to produce the observed light element abundance is not especially sensitive to the time of irradiation. (This will not be

true of the predicted spallation yields discussed in the next section.) In accordance with the above discussion we have chosen 3×10^7 yr or $T/\tau \cong 7.5$. Corresponding to this time, we adopt for the remainder of this paper a value for the neutron flux of $\Psi_n \cong 4 \times 10^{21}$ neutrons/cm², identical to the FGH value.

C. Prediction of Spallation Yields

Using Equations (2'), (3), (6') and the definitions of α_7/α_6 and f_{10} , it is possible to use the meteoritic abundances (Appendix A) to predict the relative yields of L nuclei in spallation. These yields are shown in Table 2 for $T/\tau \approx 7.5$ and compared with the meteoritic abundances. The Be¹⁰ produced in spallation is included in the B¹⁰ yield. The effects of the thermal neutrons are immediately apparent from this comparison.

The calculated relative spallation yields may be compared with the cloud chamber data of <u>Fuller</u> [1954] for 0^{16} + 300 MeV neutrons which is discussed in Appendix A. Neutrons cannot be detected in a cloud chamber; thus only the spallation product charge distribution is measured. The data given in Appendix A are the prompt yields of the spallation process itself and a correction must be made for the extent to which subsequent beta decay has modified the distribution. (This has been done using the cross-sections given in Appendix A plus an estimate of 3 mb for He⁶.) It is possible to make this correction fairly well; however, there are no reliable nuclear considerations for separating the Be⁹ and Be¹⁰ contributions to the Be yield. Consequently, in both the calculated and observed yields shown in Table 3, Be¹⁰ has been included with Be rather than B. The LiBeB spallation yields are expressed as fractions of the total L_s yield. The symbol (B/Li)_s refers to the B to Li yield ratio in spallation and should be distinguished from B/Li which is the meteoritic

abundance ratio. The yields as calculated by FGH are also shown to illustrate the overall effect of the changes in the parameters.

Note that the results in Table 3 are more sensitive to the irradiation time than was the neutron flux. This, as well as the trend of the results, can be understood by considering the effect of Be¹⁰. For small T, the present day B¹⁰ abundance consists almost entirely of the Be¹⁰_s [see Equation (6')]; thus the irradiation must have stopped when Be¹⁰_s \approx B¹⁰. However, since B¹⁰ is quite small (1.5) and the B¹⁰_s/Be¹⁰_s ratio (1/f₁₀) is fixed in the model at 4, the amount of B¹⁰_s will be relatively small (6). Since the abundance of Li⁷ is large (33), almost all of it must therefore come from spallation; hence (B/Li)_s has to be small and cannot differ much from B/Li. On the other hand, for large T the present-day B¹⁰ corresponds to that part of the (B¹⁰_s + Be¹⁰_s) which survived the neutron irradiation. Since the depletion factor due to thermal neutrons is large (~12.5 for T/ $\tau = \infty$), B¹⁰_s is relatively large (12.5 × 1.5 ≈ 19), and the B¹⁰_s contribution to the present day Li⁷ is sizable and less Li⁷_s is required. The required (B/Li)_s is then considerably larger.

Two separate comparisons between the calculated and experimental data can be made.

(1) The Be Yield. The low calculated Be_s is a direct consequence of the low Be^9 meteoritic abundance which has been reduced by a factor of 30 from the Suess-Urey value used by FGH as a result of the measurements of <u>Sill and</u> <u>Willis</u> [1962]. As Table 3 shows, the experimental data do indicate that $Be^9 + Be^{10}$ is low; further the crude breakdown of the 0^{16} cloud chamber data into isotopic cross-sections made in Appendix A gave a low value for $\sigma(Be^9) = 2.5$ mb. A low Be^9 yield is not unreasonable from the point of view of nuclear physics because (a) the other mass 9 isobars do not contribute to the yield and (b) Be^9 is only bound by 1.7 MeV and has no particle-stable excited states.

(2) The $(B/Li)_s$ Ratio. The calculated and experimental values disagree for all T. The statistical error in the experimental ratio is about ± 0.4 so that $(B/Li)_s \ge 1$ seems certain. This is also expected on general grounds since spallation yields typically decrease as the mass difference between target and product increases.

From the physical arguments given above it can be seen that $(B/Li)_s$ is fairly sensitive to f_{10} for small T/τ , and also to B/Li and α_7/α_6 especially at the larger values of T/τ . For example, at $T/\tau = 3$, changing B/Li to 0.3 and f_{10} to 0.1 will increase $(B/Li)_s$ by roughly a factor of 2. The discrepancy at large T/τ may not be significant because of the uncertainty in these parameters; however, for small values of T/τ , the changes in these three parameters that would be required to bring $(B/Li)_s$ up to 1.0 seem unreasonably large. This argues for a large value of T/τ .

For comparison Table 3 also includes the cloud chamber data of <u>Kellogg</u> [1953] for C¹² + 90 MeV neutrons. Consider a variation of the model in which amounts of C comparable to the concentrations observed in the carbonaceous chondritic meteorites (5 per cent by weight) may have been present in the planetesimals. This variation is interesting because in many respects the carbonaceous chondrites appear to be a good approximation to primordial solar system material [<u>Ringwood</u>, 1962; <u>Urey</u>, 1964; <u>Anders</u>, 1963]. The presence of C would require that a weighted average of the C and O values for α_7/α_6 and for f₁₀ be used rather than that for O¹⁶ alone. However, on the assumption that α_7/α_6 and f₁₀ are not appreciably different for C than O, the calculated spallation yields would be unchanged. Table 3 shows that the experimental (B/Li)_s is larger, as expected, for C; thus, if C is assumed present in the planetesimals, the discrepancy between the calculated and observed (B/Li)_s is worse. From this point of view the original assumption

that C was highly depleted in the formation of the planetesimals seems preferable. More discussion of this point will be given in Section IVB and in Appendix D.

For the type of primary flux ($\overline{E} \sim 500$ MeV) we have assumed up to now, LiBeB are produced almost entirely by the spallation of O¹⁶ (and perhaps of C¹²). However, we can also consider a situation in which a much "harder" incident flux is assumed. At high proton energies the cross-sections for producing L nuclei from SiFe targets are probably comparable to those for 0¹⁶ [based on Be⁷; see, for example, <u>Perfilov</u> (1960)]. Below about 1.5 GeV the SiFe cross-sections drop off rapidly; whereas the 0¹⁶ cross-sections remain constant down to about 100 MeV. The nuclear reaction mechanism by which the L nuclei are produced is probably different in the two cases. For an O¹⁶ target L nuclei are the residues left behind following the emission of protons, neutrons, alphas, etc., from the original nucleus. For a SiFe target L nuclei are produced by the so-called "fragmentation" mechanism in which they are themselves emitted whole from the initial nucleus. Although the exact nature of the fragmentation process is the subject of considerable debate among nuclear chemists at the present time, it is expected on general grounds that the yield of an L nucleus is a steeply decreasing function of its nuclear charge dropping off something like exp(-Z) [Perfilov. 1960]. Thus contributions from SiFe spallation would tend to lower the experimental (B/Li) ratio with which the calculated ratio should be compared. However, the SiFe contributions must be quite large to make the calculated and experimental ratios agree. At $T/\tau = \infty$, for example, roughly 50 per cent of the Li must come from SiFe. If the material bombarded contained comparable numbers of 0 and SiFe nuclei and were only exposed to particles of \sim 1 GeV or greater, then agreement would be obtained. The first requirement is

realized in the planetesimals; however, the second is more difficult to realize. From a natural source one usually finds a particle spectrum in which the number of particles increases strongly with decreasing energy. The L yield will then be dominated by the contributions from 0^{16} spallation by the more numerous particles of less than 1 GeV. If one arbitrarily assumes an incident flux of only high energy particles, this still does not solve the problem because in a solid body the high energy particles will generate large numbers of secondary particles in the 0.1-1 GeV range. Consequently, inside the body there is again a sharply decreasing flux with increasing energy and the difficulty remains. If one postulated that the bodies were small enough to avoid such secondary production, then the number of neutrons thermalized would be too small to significantly affect the Li⁶ and B¹⁰ abundances. All in all, assuming a "harder" flux will not explain the discrepancy in the calculated and experimental (B/Li)_c ratios.

D. Deuteronomy, The Synthesis of Deuterium;

The Concentration of Hydrogen in the Planetesimals

Using the neutron flux calculated in Section IIIB, we can now write the equation for the production of deuterium by $H^{1}(n,\gamma)D^{2}$ and from deuterons emitted in spallation.

$$\frac{dD}{dn} = \alpha_2 + \frac{f_r \sigma_1 H}{\Sigma}$$
(9)

where α_2 is the ratio of the deuteron to neutron production rates and σ_1 is the thermal neutron capture cross-section for hydrogen (0.332 b). In order to calculate D/H from Equation (9) H/Si must be known. Again the problem must be turned around to find the value of H/Si required to give the terrestrial D/H ratio (1.5 × 10⁻⁴).

. The planetesimals probably had radii that were large compared to the depth of penetration of the cascade produced by the primary particles; thus only the outer shells were irradiated. The terrestrial D/H is presumably obtained after a thorough mixing of the planetesimal material, <u>i.e.</u>, the terrestrial D/H is the D/H in the irradiated material multiplied by the fraction of the material which was irradiated. FGH defined this as $1/F_d$. With this definition F_d is a "dilution factor". Equation (9) gives

$$\frac{D}{H} = \frac{\Psi_n \sigma_1}{F_d} \left[1 + \frac{\alpha_2 \Sigma}{f_r \sigma_1 H} \right]$$
(10)

Let us rewrite Σ/Si in the form

$$\frac{\Sigma}{\mathrm{Si}} = \sum_{A} \sigma_{A} N_{N} = \sigma_{1} \left(\frac{\mathrm{H}}{\mathrm{Si}}\right) + \Sigma' \qquad (11)$$

where Σ' is a calculable constant for a given chemical composition. Assuming that the other parameters can be estimated, Equation (10) has two unknowns, F_d and H/Si. A second equation may be obtained from the fact that the LiBeB abundances relative to Si will be diluted by a similar factor. (Consideration of how a nuclear cascade develops in a solid body shows that the dilution factors for D and for LiBeB are not identical; however, we shall neglect the difference in the following discussion.)

$$L_{s} = \frac{\alpha_{L} n}{F_{d}} = \frac{\alpha_{L} n_{r}}{F_{d} f_{r}} = \frac{\alpha_{L} \psi_{n} \Sigma}{F_{d} f_{r}}$$
(12)

where L_s is the total amount of LiBeB (including the Li $_s^6$ lost) produced in spallation and α_L is the ratio of light element to neutron production rates. Equations (10), (11), and (12) can be solved for F_d and H/Si if the parameters α_2 , f_r , Σ' , and α_L can be estimated. The details of the estimation of these parameters have been included in Appendices B through D. It is important, however, to include some discussion of these estimates in the main text because the uncertainties in them introduce a rather large spread in the possible values of H/Si and F_d .

We have chosen a value of $\alpha_2 = 0.1$ based on the yield of primary deuterons relative to that for primary and secondary neutrons. The uncertainty in this parameter is not expected to introduce appreciable error. (See Appendix A.)

The calculation is more sensitive to α_L ; thus the uncertainty in the estimates of the number of secondary neutrons is more serious. (See Appendix B.) (a) An upper limit to α_L is set by the primary cross-sections. On this basis FGH estimated $\alpha_L = 0.1$. At present the best estimates of primary cross-sections give $\alpha_L = 0.075$ but with enough uncertainty that $\alpha_L \approx 0.1$ is a reasonable upper limit.

(b) Estimates of secondary neutrons produced by cosmic rays ($\overline{E} \approx 4$ GeV) give a lower limit: $\alpha_{L} \approx 0.02$.

(c) By following through a cascade on the average for a 500 MeV incident proton, values in the range 0.03-0.04 are obtained. These values appear low relative to the semi-empirical cosmic-ray values; thus $\alpha_L \approx 0.05$ has been adopted as our best estimate.

There are two reasons why f_r may be different from unity, namely (a) non-thermal capture and (b) surface leakage. (See Appendix C.) It turns out that the amount of H in the planetesimals is always large (H/Si \geq 1); thus non-thermal capture will not be important and only approximately 10 per cent of the neutrons will be lost this way. For large bodies (\geq 100 cm) mechanism (b) probably amounts to 10-20 per cent based on estimates of neutron leakage from the earth and the moon by Lingenfelter, Canfield, and Hess [1961].

Thus, values of f_r between 0.7-0.8 seem indicated. We take $f_r \sim 0.75$ as our best estimate.

There are two choices for Σ' depending on whether a meteoritic or solar composition is chosen for the planetesimals. (See Appendix D.) Σ' is dominated by the contribution from Fe (high abundance and high σ_A). Because the Fe abundance is higher in meteorites than in the sun, the meteoritic value of Σ' is higher than that based on solar abundances. In several respects Type I carbonaceous chondrites appear to be a good approximation to primordial matter [<u>Ringwood</u>, 1962]. Without entering into the controversy of how valid this approximation is [<u>Urey</u>, 1964; <u>Anders</u>, 1963], we only want to note that because this class of meteorites is very rich in iron, they give a value of Σ' that can be considered an upper limit. On the other hand, the solar abundances being low in iron - will give the lower limit to Σ' . The values are

> $\Sigma' = 3.4$ (carbonaceous chondrites) $\Sigma' = 1.2$ (solar)

As H. C. Urey has pointed out for many years [see, for example, Urey, 1964] the difference in the Fe/Si ratio between the sun and the material of the inner solar system is probably real and represents an iron-silicate fractionation in the formation of the latter. Our choice for the chemical composition (and hence Σ ') of the planetesimals depends on whether the fractionation occurred before or after the irradiation. We have placed the FGH irradiation after the loss of gas from the region of the terrestrial planets; and, in the absence of gas, we can think of no mechanism by which large quantities of silicate material could be removed from the inner solar system. The iron-silicate fractionation then must have occurred during the formation of the planetesimals prior to their irradiation. Consequently, the meteoritic composition giving $\Sigma' = 3.4$ is most appropriate for the present calculation.

Considering Σ' to be a constant assumes that the concentration of He³ ($\sigma_A = 5500$ b) remains small during the irradiation. This requires not only that it be highly depleted during the formation of the planetesimals but also that the He³ produced in the planetesimals by spallation (both directly and as H³) diffuse out in a time short compared to the mean lifetime for the He³(n,p)H³ reaction (~6 × 10⁴ yr). The conclusion drawn by FGH (see their Appendix 3) that no solution is possible if spallation-produced He³ is retained in the planetesimals is still valid even though some of the numbers entering the calculation have been revised in this paper. The reader is referred to FGH, Appendix 3 for further details. The question of whether He³ can diffuse from the planetesimals was considered by <u>Mitler</u> [1963] who found, using the FGH parameters, that the He³ would escape rapidly enough to justify neglecting it in the calculation of Σ' .

The way in which the calculated values of F_d and H/Si depend on the parameters discussed above is illustrated in Table 4. Table 4 lists some solutions for (F_d , H/Si) for various choices of Σ ', f_r , and α_L . All of the above results are for $\psi_n = 4 \times 10^{21} \text{ n/cm}^2$ and $\alpha_2 = 0.1$.

Following the above discussion, our "best" estimates, corresponding to $\alpha_L = 0.05$, $\Sigma' = 3.4$, and $f_r = 0.75$, are $F_d = 20$ and H/Si = 1.2. The corresponding solutions obtained by FGH were $F_d = 10$ and H/Si = 8.0. It should be emphasized that the uncertainties in the parameters, particularly α_L , cause considerable uncertainty in F_d and H/Si. As can be seen from Table 4, the range $0.02 \leq \alpha_L \leq 0.1$ and $1.2 \leq \Sigma' \leq 3.4$ corresponds to $10 \leq F_d \leq 37$ and $0.5 \leq H/Si \leq 11$. The present calculation tends to indicate a higher F_d value and a lower H/Si value than the results obtained by FGH although much uncertainty still remains.

We conclude this section with a brief discussion of the total energy required to produce the terrestrial and meteoritic DLiBeB. The method of calculation is very similar to that given by FGH (Section III.9). For the carbonaceous chondritic composition the cross-sections given in Appendix B indicate that there is roughly one L nucleus produced per six proton reactions. For a mean incident proton energy of 500 MeV, about 2/3 of the protons will undergo a nuclear interaction; i.e., on the average about 9 incident protons of average energy 500 MeV are required to produce one L nucleus; thus 4.5 GeV of energy must be dissipated in spallation reactions for each L nucleus produced. Section IIIC showed that roughly 50 L nuclei per 10^6 Si atoms were produced by spallation. If we assume that this figure holds for all the matter in the inner solar system, and use the FGH estimate of 4.6 × 10^{49} Si nuclei in the terrestrial planets, we obtain 50 × 10^{-6} × 4.6 $\times 10^{49} \times 4.5 \times 1.6 \times 10^{-3} = 1.7 \times 10^{43}$ ergs as the total amount of energy required. If we assume, following FGH, that ~ 10 per cent of the accelerated particles interacted with the planetesimals, then about 2×10^{44} ergs in high energy particles are required. Using Hoyle's estimate of 5×10^{45} as the total amount of rotational energy to be dissipated, we see that about 4 per cent is required to appear as high energy particles.

IV. DISCUSSION

A. The Water Content of the Planetesimals

Even though it can be argued that H/Si can be calculated only to within an order of magnitude ($1 \le H/Si \le 10$), the important conclusion, as emphasized by FGH, that the hydrogen (water) content of the planetesimals is intermediate between the solar ($H/Si = 3 \times 10^4$, 0/Si = 30) and terrestrial ($H_20/Si \sim 4 \times 10^{-3}$)

values can still be drawn. It is interesting to note that our best estimate of H/Si = 1.2 based on carbonaceous chondrite abundances for non-volatile elements, is close to that measured in this class of meteorites. However, as discussed in Section IIIC, the fact that the calculated $(B/Li)_{s}$ ratio is considerably less than would be expected from the observed C concentration of the carbonaceous chondrites removes any significance from this observation.

In Appendix D we show that $0/Si = 3.8 + \frac{1}{2}$ H/Si for the carbonaceous chondrite composition. For H/Si = 1.2 this gives 0/Si = 4.4. There is no reliable way to estimate 0/Si for the earth as a whole. However, if the earth has 0/Si = 3-4 like the silicate phase of the meteorites, then we must postulate, as did FGH, that some oxygen as well as hydrogen was lost in the time between the irradiation of the planetesimals and the formation of the earth. However, in the present calculation the amount of 0 which must be lost $[(0/Si)_{lost} = 0.4-1.4]$ is considerably less than for FGH (3.2-4.2). Loss of 0 is necessary in any model for the formation of the earth and the meteorites which assumes that the first bodies to form in the early solar system were something resembling carbonaceous chondrites. Such models must also assume that the excess C would be lost as well as the 0.

If loss of 0 during the formation of the earth is really necessary, it presents serious difficulties - as has been emphasized to us by H. C. Urey because the 0 must be lost under conditions such that other volatile elements such as mercury were retained.

A further chemical difficulty in the present calculations has been mentioned to us by both Urey and E. Anders. For nuclear reasons in Section IIIC we have assumed that the C concentrations of the planetesimals was low, and the irradiation was assumed to occur after the escape of H_2 from the inner solar system. Assuming that the iron in the planetesimals is oxidized

(which would be true if the planetesimal constituents were in a state of chemical equilibrium), there is thus no reducing agent available to convert the iron to the metal to form the core of the earth.

The purpose of the present section is to define necessary chemical and physical conditions such as these for which the nuclear calculations of Section III would be valid. We have not attempted to develop detailed models by which the planets and the meteorites can form from planetesimals; moreover, we have not discussed the difficult question of how the planetesimals themselves formed. If reasonable models cannot be formulated which remove the above difficulties, serious modifications in the nuclear calculations will be necessary.

<u>Urey</u> [1965] discusses the possibility that certain lunar surface features indicate the presence of liquid water. Urey considers this to be terrestrial water transported to the lunar surface following the collision of the earth with another lunar-sized body. Another solution considered but not favored by Urey was that the moon escaped from the earth. However, a simpler solution would be found in the FGH model, namely that the moon, unlike the earth, retained a large amount of the water of the planetesimals. <u>Gold</u> [1964] and <u>Kopal</u> [1963] have previously suggested that water is escaping from the moon's interior. <u>Urey</u> [1963] argues that the water was only on the surface because a high concentration of water would lower the melting point of silicates causing the interior of the moon to be completely molten. This would cause extensive lava flows on the moon's surface. He feels that there is no evidence for extensive volcanic activity.

Urey also suggests that most stone meteorites come from the moon. In particular the organic compounds present in the carbonaceous chondrites arise from the contamination of the moon with primitive terrestrial biotic material. In the alternative we have proposed the carbonaceous chondrites would

represent material knocked from the moon prior to losing its water and other volatile material. The possible presence of organic and biotic constituents can be understood on a qualitative basis. FGH (page 212) have already pointed out that "conditions during the formation of the planetesimals were highly favorable to the building of biologically interesting molecules. The possibility is suggested that pre-biotic or even biotic materials could have been formed in the planetesimals before the formation of the Earth." On the present point of view additional organic and biotic activity might well have taken place on the moon itself. This suggests that the great care being taken in Russian and American investigations of the lunar surface to avoid terrestrial contamination is to be highly commended.

We emphasize that we are not claiming on the basis of our calculations that the moon has a high water content or that biotic activity has occurred in some meteorites and on the moon. We are only pointing out that, if independent evidence reveals either of these to be true, then the present model provides a means by which they could occur.

B. Effects of the Planetesimal Irradiation on

the Abundances of Other Nuclei

(1) C^{13}/C^{12} . FGH found that the amounts of C^{13} produced by spallation of 0^{16} in the planetesimals were comparable to the C¹³ abundance in the earth's crust. This conclusion also holds for the present calculations. From the cloud chamber data of Fuller [1954] for 0¹⁶ + 300 MeV n we have estimated that the ratio of the C¹³ to the L spallation yield is about 3/8. From Table 2 the L spallation yield is roughly 50 per 10⁶ Si which is the abundance obtained after the mixing of the irradiated and unirradiated material. The C¹³ abundance produced by spallation is thus 50 \times 3/8 \approx 19 per 10⁶ Si with an uncertainty of about 50 per cent. In our discussion up to now we have considered two possibilities for the planetesimal carbon abundance: (a) C was depleted in the formation of the planetesimals to the present crustal abundance; or (b) C was present in the concentrations observed in the carbonaceous chondrites. We have favored alternative (a) in the discussion thus far. For alternative (a) we have $C = 1.7 \times 10^3$ (based on the crustal abundance given by Taylor [1964]. The terrestrial C^{13}/C^{12} ratio is 1/90 which gives $C^{13} = 19$ which, considering the uncertainties, is fortuitous agreement. Assuming that C is 5 per cent by weight, alternative (b) gives $C^{12} \approx 5 \times 10^5$ which is 2.5 $\times 10^4$ times the amount of C¹³ produced by spallation rather than the observed factor of 90; thus the nucleosynthesis of C¹³ in the present process would not be possible if alternative (b) is adopted. Since the neutron capture cross-section for C^{12} is small (4 mb), the amount of C^{13} formed by $C^{12}(n,\gamma)$ will be negligible regardless of the initial C concentration $(C^{13}/C^{12} \cong 4 \times 10^{-27} \times 4 \times 10^{21} = 1.6$ $\times 10^{-5}$).

Considerable effort has been made to measure the solar C^{13}/C^{12} ratio. <u>Greenstein, Richardson and Schwarzschild</u> [1950] were only able to set an

upper limit of $C^{13}/C^{12} < 1/36$ based on the absence of a $C^{13}N^{14}$ isotopic doublet. Recently <u>Wyller and Greenstein</u> [1964] have reviewed these data and set $C^{13}/C^{12} < 1/200$. <u>Righini</u> [1956] found a weak spectral feature which he identified as C^{13} and allowed him to set $C^{13}/C^{12} \approx 1/4000$; however, the latest work by <u>Righini</u> [1963] places this ratio still lower at $C^{13}/C^{12} \approx 1/10,000$. On the other hand Delbouille [1964] has detected C^{13} in a different $C^{13}N^{14}$ band from that used by the above workers. He quotes a ratio of $C^{13}/C^{12} \approx 1/100$ which would be identical, within experimental error, to the terrestrial ratio.

If the terrestrial and solar ratios are identical, as indicated by the work of Delbouille, then the nucleosynthesis of C^{13} must have occurred prior to the formation of the solar system. This is because the amount of C^{13} produced by spallation on the sun's surface is small compared to the high C^{12} abundance $(C^{12} = 1.7 \times 10^7 \text{ on the Si} = 10^6 \text{ scale})$. This would rule out alternative (a) above. Conversely, if C^{13}/C^{12} is much smaller on the sun, as indicated by the observations of Righini, then alternative (a) is indicated strongly.

<u>Stawikowski and Greenstein</u> [1964] measured the C^{13}/C^{12} ratio in comet <u>Ikeya 1963a</u>, a near-parabolic comet. They found $C^{13}/C^{12} \sim 1/70$ but equal to the terrestrial 1/90 within experimental error. Comets are thought to represent the outer parts of the solar system, and it is difficult to see how they could have been heavily irradiated. If we assume that such comets have always been in a long-period orbit; then, although they could come quite close to the sun in a given orbit, their total exposure (flux times time) would be considerably less than a planetesimal which remained in the inner parts of the solar system. Further, the large C abundance in comets would seem to rule out obtaining the terrestrial C^{13}/C^{12} ratio by spallation even if we assume that comets represent "undiluted" material. These observations can best be explained by assuming nucleosynthesis of C^{13} prior to the formation of the solar system

and are consistent in this respect with Delbouille's measurement of the solar C^{13}/C^{12} ratio. If one chooses to adopt Righini's measurements, then it is necessary to conclude that comets were made from the material of the inner planets. This is contrary to prevailing opinion on the origin of the comets.

The interpretation of measured meteoritic C^{13}/C^{12} ratios is complicated by the fact that chemical processes can produce measurable changes in the isotopic composition of light elements. Boato [1954] measured the C^{13}/C^{12} ratio in the reduced and organic C from the carbonaceous chondrites and found that the ratios obtained were within the range observed in terrestrial igneous and sedimentary rocks. However, Clayton [1963] found that C^{12}/C^{12} in the carbonate minerals from the Orgueil and Ivuna Type I carbonaceous chondrites was about 6 per cent larger than the ratio in any terrestrial carbon. Chemical processes which would give a fractionation of this magnitude are unknown of the earth: but, according to Clayton, cannot be completely ruled out. If the difference were nuclear in origin, it could be readily explained by the FGH model by assuming, for example, a 6 per cent difference in dilution factor between terrestrial material and that of the Orgueil and Ivuna meteorites. However, in Section IV3 arguments against a nuclear origin will be given. (2) Li⁷/Li⁶. The Li⁷/Li⁶ ratio is sensitive to the magnitude of the neutron flux. Thus, variations of the time-integrated neutron flux in various parts of the early solar system would give variations in the Li⁷/Li⁶ ratio. (The neutron flux and proton flux are proportional, so a variation in the neutron flux reflects a variation in the proton flux.) Variations produced in this manner would be independent of any mixing of irradiated and unirradiated material subsequent to the irradiation. The sensitivity can be understood

by considering how the Li^6 and Li^7 abundances vary during the course of the irradiation as measured by the number of neutrons reacted per 10^6 Si, n.

For small n_r the (n,α) reactions will be unimportant and the Li⁶ and Li⁷. abundances will rise linearly with n_r with a slope determined by the spallation rates. As the (n,α) reactions become more and more important for larger n_r the Li⁶ abundance will reach an equilibrium value; however, the Li⁷ abundance will still increase at a rate even faster than the spallation rate because of the contribution from B¹⁰ (n,α) Li⁷. Thus, the Li⁷/Li⁶ will be increasing at a rate greater than linear; consequently a given percentage variation in n_r will produce a larger percentage variation in Li⁷/Li⁶.

If the meteorites come from the asteroid belt and if the meteorites were formed from planetesimals that occupied this region of the solar system (l.5-5 astronomical units with the maximum concentration at about 2.5 AU), then we might expect some difference in the meteoritic and terrestrial ${\rm Li}^7/{\rm Li}^6$ ratios.

<u>Shima and Honda</u> [1963] reported Li⁷/Li⁶ ratios of 10.5 in three chondrites (Bruderheim, Harleton, and Ehole) compared with the terrestrial ratio of 12.5. This would be in the direction predicted by the above analysis because the meteorites should represent planetesimals which were, on the average, further from the sun during the bombardment and, hence, have a smaller Li⁷/Li⁶ ratio. However, <u>Krankowsky and Müller</u> [1964] have repeated these experiments including two of the three meteorites studied by Shima and Honda and can detect no difference in the meteoritic and terrestrial ratios to within 2 per cent. Work by <u>Ordzhonikidze</u> [1960] also revealed no difference. <u>Poschenrieder</u>, <u>Herzog</u>, and Barrington [1965] studied the Li⁷/Li⁶ ratio in various portions of the Holbrook chondrite using an ion-microprobe mass-spectrometer. Their preliminary results indicate large variations (from 9.5 to 27.5) in the Li⁷/Li⁶ ratio.

Although there are experimental questions which remain to be settled, the work of Krankowsky and Müller appears to be most definitive at the present

time. Thus, we shall assume that there is no difference between the meteoritic and terrestrial Li⁷/Li⁶ ratios to within 2 per cent. This implies that the meteoritic and terrestrial planetesimals saw neutron fluxes identical to less than 2 per cent.

Whether or not this could occur astrophysically depends on when, where, and how the earth and the meteorites formed. The requirement that the fluxes be identical has such far-reaching implications for the history of the solar system that it is not proper to assume it from these rather narrow nuclear considerations. However, without claiming their validity, we can discuss the required astrophysical conditions. However, before discussing these conditions, we want to consider the point that, despite the requirement of constant flux, isotopic variations in certain elements are still possible due to variations in F_d . One possibility, for example, would be that meteorites formed from planetesimals which were larger or smaller than those from which the earth formed. In discussing these "dilution anomolies" we assume that meteorites are second generation objects resulting from mixing the irradiated and unirradiated material of the planetesimals. Thus three examples will be considered involving nuclei which are strongly affected by the irradiation so that variations in isotopic composition could remain even after mixing.

(3) Dilution Anomalies

<u>A. Gd, Sm, Eu</u>. These elements have isotopes with very large thermal neutron capture cross-sections: Gd^{155} (58,000 b), Gd^{157} (240,000 b), Sm^{149} (41,500 b), Sm^{152} (220 b), Eu^{151} (8700 b), Eu^{153} (320 b). For these cases the fractional changes in abundance, $\Delta Na/Na$, is given by

$$\frac{\Delta Na}{Na} = -\frac{1}{F_d} \left[1 - \exp(-\psi_n \sigma_A) \right] \cong -\frac{1}{F_d} \quad \text{for } \psi_n \sigma_A \gg 1$$

For $\psi_n = 4 \times 10^{21} \text{ n/cm}^2$, the $1/F_d$ approximation will hold for all the cases considered here. Physically, this equation corresponds to the situation when the nuclei in question have been completely depleted in the irradiated material; thus the maximum difference in the abundance of these nuclei between material which has been irradiated and mixed and material which has never been irradiated is just the fraction of the former material which has been irradiated. For our best estimate of $F_d = 20$, this maximum fractional change is 0.05; thus, measurements of the isotopic composition of these elements, if done with sufficient precision, can be used to measure differences in the fraction of material irradiated between various samples of solar system material. Note that such measurements, although of nuclei with large neutron capture cross-sections, give no information about the magnitude or variations in the neutron flux. Murthy and Schmitt [1963] were unable to find any difference between the terrestrial isotopic composition of these elements and that of four meteoritic samples: 2 chondrites (Holbrook, Forksville), 1 carbonaceous chondrite (Murray), and 1 achondrite (Pasamonte). They estimated that the uncertainty in the measurement of the isotopic ratios was one per cent.

For simplicity in notation let $q = 1/F_d$, the fraction of material irradiated. From the above discussion, we conclude that this experiment shows that

$$|q_{terr} - q_{met}| \leq 0.01$$

Since the calculated value of q = 0.05 is based on the terrestrial D/H ratio, it should be identified with q_{terr} . Thus q_{met} can differ from q_{terr} by as much as 0.01/0.05 or 20 per cent. If q were smaller (F_d larger), then there is room for still larger differences between q_{terr} and q_{met} . What the above discussion means physically is that as q gets smaller, it becomes increasingly harder to see the effect of the admixture of irradiated material on the overall

isotopic composition, and one is only measuring the unirradiated material. The principal point of this discussion is that it is not necessary to assume that the terrestrial and meteoritic q-values are exactly identical in order to explain the experimental results of Murthy and Schmitt; and the required 6 per cent variation in q to explain the variations in C^{13}/C^{12} observed by Clayton is consistent with the work of Murthy and Schmitt.

It is possible to decide experimentally whether the above permissible variations in q do exist or whether we must assume that $q_{terr} = q_{met}$. This is because the abundances of nuclei which are <u>produced</u> in amounts not negligible compared to the amount present before the bombardment are more sensitive to q than those <u>depleted</u> during the planetesimal bombardment. To illustrate this we will consider two examples: K^{40} and V^{50} . <u>B.</u> K^{40} . K^{40} can be produced by (a) $K^{39}(n,\gamma)K^{40}$ and (b) spallation of Fe. (a) In the irradiated material $K^{40*}/K^{39} = \psi_n \sigma_{39} = 4 \times 10^{21} \times 2.2 \times 10^{-24} \approx 9 \times 10^{-3}$ using K^{40*} to denote the K^{40} produced in the bombardment. Dilution with unirradiated K lowers this by a factor of $F_d = 20$ for the material which eventually formed the earth. Thus $(K^{40*}/K^{39}) = 4.5 \times 10^{-4}$ from neutron capture.

(b) After irradiation and dilution 50 L nuclei per 10^6 Si were produced by spallation. From the cross-sections given in Appendices A and B, there is about 1 L nucleus produced per 6 proton reactions or a total of 300 protons reacting per 10^6 Si atoms. Based on the inelastic cross-sections of Appendix B, about 17 per cent of the protons react with Fe group nuclei. <u>Honda and Lal</u> [1964] measured a cross section of 8 ± 1 mb for K⁴⁰ produced by the spallation of Fe with 730 MeV protons. This energy is somewhat high for the type of primary spectrum we have assumed, thus a lower cross section is appropriate for the present calculation. Consequently, we estimate that about one per cent

of the spallation reactions on Fe (corresponding to a cross-section of 5.5 mb) will produce a K^{40} nucleus and take the planetesimal K^{39} abundance to be 3.6×10^3 on the Si = 10^6 scale. Thus

$$\left(\frac{K^{40*}}{K^{39}}\right)_{s} = \frac{300 \times 0.17 \times 0.01}{3.6 \times 10^{3}} = 1.4 \times 10^{-4}$$

The total (K^{40}/K^{39}) produced by planetesimal bombardment of terrestrial material is roughly 6×10^{-4} . The present K^{40}/K^{39} ratio is 1.2×10^{-4} . The mean life for K^{40} is 1.88×10^9 yr; thus, at 4.5×10^9 yr ago $K^{40}/K^{39} = 1.2 \times 10^{-4} \cdot e^{2.4} =$ 13×10^{-4} of which 6×10^{-4} or about one-half was produced in the planetesimal bombardment. If q_{met} can differ from q_{terr} by up to 20 per cent, then $(K^{40}/K^{39})_{met}$ could differ from the terrestrial value by one-half of the 20 per cent or up to 10 per cent. The effect is much larger than in the GdSmEu case because K^{40*}/K^{39} was not negligible compared to the primordial ratio even after dilution. The above calculation is uncertain to about a factor of 3. Thus it is conceivable that all of the present K^{40} could have been made in this process; or, conversely, only a fraction may have been produced. C. V^{50} . Here production occurs only by spallation of Fe-group nuclei and the calculation will be similar to that for K^{40} . We estimate that the V^{51} abundance is 190; thus

$$\frac{v^{50*}}{v^{51}} = \frac{300 \times 0.17 \times 0.04}{190} \cong 10^{-2}$$

This ratio is what would be obtained after the mixing of irradiated and unirradiated material. The observed V^{50}/V^{51} ratio is 2.4 × 10⁻³, thus all of the terrestrial V^{50} can be easily produced by planetesimal bombardment. Such a suggestion has previously been made by Shima and Honda [1963]. The factor

of 4 excess in the above calculation is probably not significant. If the v^{50} is produced only in the irradiated material and then diluted with the v^{51} in the unirradiated material, the v^{50}/v^{51} ratio will be proportional to q, and the up to 20 per cent variation in q permitted by the GdSmEu results would give up to 20 per cent variations in v^{50}/v^{51} .

<u>Burnett, Lippolt and Wasserburg</u> [1965] have measured the K⁴⁰ isotopic abundance in nine stone meteorites, covering all the major classes. With the exception of the Norton County achondrite, any differences in isotopic composition between the meteorites and terrestrial samples are less than 1%. Various Norton County samples show enrichments of up to 1.5%; however these could have been produced by cosmic rays. Based on the above discussion, this means that the terrestrial and meteoritic q values differ by less than 2%. The calculated value for q could be high by as much as a factor of 3, but this would only raise the limit set by the K⁴⁰ experiment to 4%. The straight-forward interpretation of the Gd and K experiments is that $q_{met} = q_{terr}$, i.e., the meteorites must have been formed from an identical sampling of the planetesimals as terrestrial material if one wishes to retain the model discussed in this paper.

The Orgueil carbonaceous chondrite was among the meteorites analyzed for K^{40} . This means that the excess C^{13} observed by <u>Clayton</u> [1963] probably has a geochemical origin.

V. CONCLUSION

The present analysis shows that the experimental work carried out since the publication of FGH has rather sharply defined the astrophysical conditions under which the model would be valid. In particular the lack of isotopic anomolies in Li, Gd, and K requires a common initial history for meteoritic and terrestrial material in that both types of material must see the same particle flux and have the same fraction of material irradiated. Whether the

FGH model can be accepted then depends on the answer to the questions of where do meteorites originate and how were they formed.

It has been proposed many times [most recently by Lovering, 1958 and <u>Ringwood</u>, 1959] that the meteorites are fragments of a small planet which once occupied the asteroid belt and has since broken up. <u>Anders (see [1965]</u> for example) has developed the idea initially proposed by <u>Fish</u>, <u>Goles and</u> <u>Anders [1960]</u> that the meteorites could be formed in large asteroids. In both of these models, meteorites represent material from the asteroid belt and were formed in that region of space. With such models for the origin of meteorites, it is difficult to meet the requirements of the FGH model. There are three possibilities, all of which are unlikely but not impossible.

(a) The particle flux and planetesimal size distribution were constant over a range of 1-2.5 AU. The latter condition is reasonable but, we see no good reason that the former should be true. Taking an oversimplified view of Hoyle's model for the early solar system, we can predict a 1/r dependence for the proton flux within the nebular disk: the coupling of the magnetic lines of force from the sun to the disk will focus the particles on the inner edge of the disk. If the particles travelled linearly from the edge of the disk, the flux would drop as 1/r since the divergence within the disk would now be in two dimensions rather than three. The complication is that the particles will follow the twisted field lines in the disk rather than diverge radially; nevertheless the flux would be expected to be significantly less, deep within the disk, than at the edge.

(b) All terrestrial and meteoritic material was thoroughly mixed either during or after the irradiation system. In the absence of large amounts of gas there would seem to be no good mechanism for the large-scale radial motion required by this hypothesis. Even with gas present there are difficulties
because only small planetesimals would be effectively moved by the gas and
the gas would tend to damp out any eccentricity in the orbits which would tend to promote mixing.

(c) The meteorites were formed from planetesimals which were irradiated at the same distance from the sun as those which eventually formed the earth. This would probably have to be somewhere near the present-day location of the earth. Meteorites would then be formed from planetesimals which were scattered out to the asteroid belt and formed "meteorite parent bodies" which then broke up yielding meteorites which eventually return to the earth. This is the most plausible of the three alternatives; however, one would expect that smaller bodies would be preferentially scattered out leading to a larger fraction of irradiated material in meteorites contrary to what is observed.

<u>Urey</u> [1965] has given arguments for a lunar origin for stone meteorites, in particular those having short cosmic-ray exposure ages. These are presumably knocked from the lunar surface by comet heads, iron meteorites from the asteroid belt, and possibly by those few types of stone meteorites which have high exposure ages. <u>Arnold</u> [1964, 1965] has shown how the statistical distributions of meteorite orbital elements and cosmic-ray exposure ages can be used to decide whether meteorites came from the moon or from the asteroid belt. If it turns out that those meteorites analyzed for Li, Gd, and K (all stones) came from the moon, then the requirements for the FGH model are most nearly met. Both types of material would have been in the same region of space and exposed to the same flux. It is not obvious that the moon would form from the same sampling of planetesimals as the earth, especially if the differences in density of the moon and earth reflect a difference in composition. One possibility is that the moon has a high water content, as discussed in Section IVA. Another possibility is that the moon formed in a different part of the

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solar system but has a surface coated with material of the type that formed the earth during or shortly after its capture by the earth. In this picture the meteorites would then be samples of this "terrestrial-like" lunar surface.

In summary, a lunar origin for stone meteorites could very well provide the required astrophysical situation for the FGH model to hold; whereas an asteroidal origin presents many more difficulties. We wish to again emphasize that we are not claiming — as a result of the calculations in this paper — that stone meteorites have a lunar origin. This question should be decided from independent considerations. The purpose of the present discussion is to point out its significance to the problem of the origin of DLiBeB.

It can be argued that the FGH model is ruled out because of the absence of any positive evidence that DLiBeB were produced in the solar system other than the plausibility and self-consistency of the model itself. This lack of independent confirming evidence in itself is serious and disturbing. However, if it were definitely established that the C^{13}/C^{12} ratio on the sun is much less than the terrestrial value, or if the variations in Li^7/Li^6 within a single meteorite reported by <u>Poschenrieder et al</u>. [1965] can be verified and extended to other meteorites and elements, then there would be experimental evidence which would be difficult to explain without the FGH model.

On the other hand the nuclear physics requirements of the model appear to present no great difficulties at the present time although more information on spallation yields and, perhaps, more study of the requirement that He^3 escape from the planetesimals would allow a more definite statement to be made on this point. This can be contrasted with the difficulties encountered in attempting to formulate models for the formation of DLiBeB in their terrestrial abundance and isotopic ratios in stellar atmospheres or in supernovae. The fact that the Li^7/Li^6 ratio in spallation (both of C and O) is much lower than

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the terrestrial abundance ratio definitely shows that something besides pure spallation must have been involved in the nucleosynthesis of terrestrial LiBeB. Neutrons are ineffective at low densities and in large H concentrations. Thus it is not clear what the second process would be in a star or especially in a supernova. One might appeal to convection on a stellar surface to destroy Li⁶; however, why then is Li about 5 times as abundant as B when the cloud chamber results show that, as expected, the B/Li spallation yield ratio is certainly greater than one? It is possible that the terrestrial B^{11}/B^{10} ratio can be obtained directly by spallation under reasonable astrophysical conditions? Further, D would be completely destroyed if convection destroyed most of the Li⁶. Where then does D originate? On a different type of star, perhaps? Stars have been observed which seem to be making Li, but no one has ever seen D in a star, let alone a D-rich star. (See, for example, Peimbert and Wallerstein [1965].) If a star or supernova can produce enough D to enrich the interstellar medium to the terrestrial D/H ratio, one would expect that copious amounts of LiBe and particularly B would have also been added to the interstellar medium making their terrestrial abundances several orders of magnitude higher than is observed. Also D [Weinrab, 1962], Li [Spitzer, 1949], Be [Spitzer and Field. 1955] cannot be detected in the interstellar medium. However, the upper limits which have been set for Li are high. For Be one can argue that it is preferentially locked in dust grains because Ca which would be expected to behave similarly in such an environment is also depleted. The D result ($\leq \frac{1}{2}$ the terrestrial D/H) is based on the 91.6 cm D line in the radio source Cas A, so one cannot say with certainty that the result is applicable to the interstellar medium as a whole. One can also argue that the earth is highly enriched in D due to chemical fractionation processes during the formation of the earth. The required fractionations are quite high however, and the above nuclear problems would remain even if D/H were an order of magnitude lower.

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In conclusion the present paper has brought the nuclear calculations of the FGH model up to date, generalized them in an approximate manner to include [•] the irradiation time dependence caused by the long life of Be¹⁰, and estimated the remaining uncertainties. The consequences of the revised calculation have been followed, particularly the effects of the nuclear irradiation on nuclei other than DLiBeB. The analysis of experimental data on the isotopic abundances of these elements leads to specific astrophysical conditions which must be met in order to retain the model. These conditions could provide the basis by which future space and meteorite research can decide what role nuclear physics has played in the formation of the solar system.

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APPENDIX A

ESTIMATION OF PARAMETERS USED IN CALCULATING THE NEUTRON FLUX

1. Abundance Data

As mentioned in the Introduction, there have been large changes in the LiBeB abundances since the work of FGH. The abundances used in the present calculations are given in Table Al and compared with the Suess and Urey [1956] (SU) values used by FGH. The recent abundance determinations are roughly a factor of 3 lower for Li and B and a factor of 30 lower for Be than those given by SU. Such changes had been predicted by H. C. Urey (see FGH, p. 181). The abundances in Table Al are all based on analyses of ordinary chondrites. The B/Li ratio assumed in the present work is based on the total Li and B abundances given by Shima and Honda [1963], Krankowsky and Miller (1964] and Shima [1962] along with terrestrial isotopic ratios. The meteoritic abundances are used to calculate the B/Li ratio appropriate to solar system material on the assumption that the B and Li in terrestrial rocks have been subjected to chemical fractionation. The meteoritic Li abundances vary only by about a factor of 2 among various classes of stone meteorites. Assuming this to be true for B also implies an uncertainty of about a factor of 3 in B/Li for unfractionated solar system material.

2. Determination of α_7/α_6

By mass spectrometry, <u>Gradsztajn</u>, <u>Epherre and Bernas</u> [1963] have measured cross-sections for the production of Li⁶ (9.8 ± 3 mb) and Li⁷ (13.7 ± 4 mb) from 0¹⁶ + 156-MeV protons. These cross-sections are relative to that for Be⁷ which <u>Gradsztajn</u> [1960] has measured as 5 ± 1.5 mb under the same conditions. The observed Li⁶ cross-section also includes He⁶. This gives $\alpha_7/\alpha_6 = 1.9 \pm 0.2$. Since it is this ratio that is measured directly, the uncertainty in it is

Al

considerably smaller than that obtained by combining the errors in the . cross-sections. The cross-sections will increase somewhat at higher energies; however, it should be a fairly good approximation to assume that the ratio is independent of proton energy. (Recent work by <u>Bernas et al</u>. [1965] suggest that the ratio is slightly lower at 600 MeV.)

Almost all of the L nuclei will be produced by the spallation of 0^{16} ; thus assuming that the same α_7/α_6 ratio holds for the other constituents of the planetesimals should introduce little error.

Table A2 shows the composite effect of changes in B/Li and α_7/α_6 on ψ_n for the uncertainties in these parameters given above for $T/\tau = \infty$. The sensitivity is expected to be similar for shorter times. Table A2 shows that deviations up to a factor of 2.5 in ψ_n are possible in unfavorable cases. This is not serious for our present purposes.

3. Estimation of $f_{10} = Be_s^{10}/B_s^{10}$

Evaluation of the ratio of Be_s^{10} to B_s^{10} produced in spallation was primarily based on the work of <u>Fuller</u> [1954] who analyzed the stars produced by 300 MeV neutrons in an oxygen-filled cloud chamber in the presence of an applied magnetic field. He was able to identify the charge and mass of all charged particles of $A \leq 4$; thus it was possible to obtain the charge spectrum for $3 \leq Z \leq 7$. Table A3 summarized the results of this experiment. The cross-section estimates are due to us. The cross-section for star formation of 225 mb was obtained by estimating 250 mb for the total inelastic cross-section for 0^{16} and subtracting 25 mb for (n, xn) reactions. To obtain f_{10} we must break down the observed charge distribution into individual isotopic cross-sections. For Be⁷ we will use the higher energy measurements of <u>Honda and Lal</u> [1960] from which we have adopted a cross-section of 9 ± 3 mb, although the work of

A2

Rayudu [1963] and Bernas et al. [1965] would suggest that this value is somewhat high. Honda and Lal [1964] report experiments from which we estimate that Be¹⁰/Be⁷ \approx 0.5 for protons on oxygen, <u>i.e.</u>, a spallation cross-section for Be¹⁰ of 4.5 \pm 2 mb. The total cross section for the Be isotopes in Table A3 is 16 mb; thus, the cross-section for Be⁹ is 2.5 ± 5 mb. Thus, the cross-section data can be interpreted as indicating a low yield for Be^9 in spallation corresponding to its low abundance in nature relative to the Li and B isotopes. The Be $^{9}/B^{11} \approx 1/10$ abundance ratio corresponds to the $Be^{9}/(Be^{11} + B^{11} + C^{11})$ yield ratio in spallation, <u>i.e.</u>, $\sigma(Be^{11}) + \sigma(B^{11}) + \sigma(B^{11})$ $\sigma(C^{11}) \approx 25 \text{ mb.}$ Be¹¹ would be expected to have a very small spallation yield because of its low (\approx 5 MeV) binding energy and large neutron to proton ratio and will be neglected here. The cross-section for C^{11} production from O^{16} is about 10 mb [Symonds et al., 1957]. This gives $\sigma(B^{11}) = 15$ mb and $\sigma(B^{10}) = 13$ mb from Table A3. From Symonds et al. [1957] $\sigma(C^{10}) \cong 6$ mb; thus $f_{10} = \sigma(Be)^{10}/$ $[\sigma(C^{10}) + \sigma(B^{10})] = 4.5/19 \approx 0.25$. The above analysis involves considerable error; however the answer obtained for f_{10} is reasonable. As with α_7/α_6 , this f_{10} value is assumed to hold for all energies and for all constituents of the planetesimals.

Table A4 indicates the sensitivity of ψ_n on f_{10} for $T/\tau = 0.5$ and $T/\tau = 3$ and shows that the large uncertainty in f_{10} does not seriously affect the calculated flux especially for larger values of T/τ .

All in all, the effect of uncertainties in T/τ , B/Li, α_7/α_6 and f_{10} are such that the adopted value of ψ_n is probably reliable to about a factor of 3.

A3

APPENDIX B

ESTIMATE OF α_{L} AND α_{2}

It is difficult to estimate α_{L} because the light elements are, for the most part, produced by the high energy primary flux; whereas, the majority of the neutrons are of secondary origin. Thus, the primary cross-sections given in Table B1 merely set an upper limit on α_{L} . The primary cross-sections have been estimated for a mean proton energy at time of interaction of roughly 300 MeV. For the purposes of averaging over chemical composition, the planetesimal constituents are divided into 0, Si-group, and Fe-group elements. As discussed in Appendix D. there are two choices for the chemical composition: (1) based on Type I carbonaceous chondrites with 0 = 55 per cent; Si = 35 per cent and Fe = 10 per cent by number, and (2) based on solar abundances with 0 = 70 per cent and Si = 30 per cent. The estimates for neutron cross-sections were guided by the work of Crandall and Millburn [1958], Skyrme [1951, 1962], Goldan'skii et al. [1958], Bercovitch et al. [1960] and Lavrukhina et al. [1963]. The O¹⁶ light element cross-section is based on the work of <u>Fuller</u> [1954]. Those for Si and Fe were obtained by taking 10 times the Be 7 cross-section for Al and Cu respectively, as given in the review article of Perfilov [1960]. For the carbonaceous chondrite composition Table Bl gives α_L = 0.075 and for the solar composition α_L = 0.12. FGH used α_L = 0.1 based on similar estimates; and, considering all the uncertainties, the FGH value can be taken as a reasonable upper limit.

A lower limit to $\alpha_{\rm L}$ can be obtained from estimates of neutron production by cosmic rays because the cosmic-ray spectrum is much "harder" (<u>i.e.</u>, drops off less rapidly with increasing energy) than any present-day solar flare

B1

spectrum. We assume this to be true at the time of formation of the solar system. The thick target neutron production rate will increase with the primary particle energy; whereas the light element production rate will increase less rapidly leading to a smaller α_L . The light element cross-section for 0^{16} will stay about constant for an increase in the average incident particle energy; however, the cross-section for Si and Fe will increase until it is comparable to that for 0^{16} . Therefore, for cosmic rays, $\sigma_L \approx 85$ mb. The mean interaction cross-sections given in Table Bl lead to roughly one L nucleus per four proton reactions for both compositions. Lingenfelter, <u>Canfield, and Hess</u> [1961] estimated 10 ± 3 neutrons per proton for material of chondritic composition based on measurements of the cosmic ray neutron flux in the earth's atmosphere. This gives $\alpha_L \sim 1/40 = 0.025$. Considering the uncertainties in the above estimates, $\alpha_L \gtrsim 0.02$ seems a reasonable lower limit. Table 4 in the main text indicates that the range $0.02 \leq \alpha_L \leq 0.1$ gives a wide spread in F_a and H/Si.

Another estimate can be made by considering how the energy of the average 500 MeV proton is dissipated, <u>i.e</u>., by estimating how many neutrons are produced in the development of the average nuclear cascade produced by the average incident particle. This was done for the carbonaceous chondrite composition by estimating the number and average energy of cascade ("knock-on") neutrons from the Monte-Carlo calculations of <u>Metropolis et al</u>. [1958]. Cascade protons were assumed to lose their energy by ionization. Then the reactions of the cascade neutrons were followed until the energy of the resulting neutrons was too low for any further multiplication. This gave an estimate for the length of the average cascade. The total neutron yield is thus the total number of evaporation neutrons at each step in the cascade were made in two ways: (a)

B2

from the experimental neutron production cross-sections referred to earlier. because these measurements are primarily for low energy neutrons or (b) from energy balance considerations based on the calculated average excitation energies remaining in the target nucleus following the cascade process as given by <u>Metropolis et al.</u> [1958]. Combining the neutron yields so obtained with the L yields based on primary cross-sections gave values of $\alpha_{\rm L}$ in the range 0.03 to 0.04; however, it should be emphasized that these estimates are very uncertain. Since the semi-empirical cosmic-ray estimate gave 0.025, values of 0.03-0.04 seem too low considering the large difference in the average primary particle energy in the two cases; thus, a higher value 0.05 has been adopted.

Deuterons will be produced by both the primary and secondary nuclear particles in the planetesimals. It is difficult to estimate the number of secondary deuterons; thus α_2 will be calculated for the extreme cases: (1) when the secondary deuterons are negligible and (2) when the ratio of primary to secondary deuterons is equal to that for the neutrons, in which case α_2 is just the ratio of the primary cross-sections. We consider case (1) to be the better approximation and shall adopt the value obtained in that case.

The deuteron production cross-section from 0^{16} given in Table Bl is from <u>Fuller</u> [1954]. Those for SiFe were obtained by assuming that the percentage of reactions producing deuterons is constant. Thus, very roughly, the cross sections will scale like the interaction cross-sections or as $A^{2/3}$.

For case (2) the primary cross-sections give $\alpha_2 = 0.18$ for the carbonaceous chondrite composition and $\alpha_2 = 0.22$ for the solar composition. These can be taken as giving a rough upper limit for α_2 of 0.2.

For case (1) the above value of $\alpha_L = 0.05$ for the carbonaceous chondrite composition leads to a total (primary + secondary) neutron cross-section of

BЗ

1100 mb which gives $\alpha_2 = 130/1100 \approx 0.1$ which is the adopted value for this . paper and is identical with the value used by FGH.

Table B2 shows that in any case the values of $F_d,$ and to a lesser extent, H/Si are not seriously affected by a factor of 2 uncertainty in α_2

ESTIMATION OF f

Even when the neutrons are at energies below 10 MeV where they are incapable of any further multiplication, there still remain three processes which can prevent the neutrons from becoming thermalized and reacting in the planetesimal: (a) beta-decay: this has negligible probability in a solid body, so we will neglect it in the following discussion. (b) Non-thermal capture. For all cases in these calculations the hydrogen concentration will be large (H/Si \sim 1); thus thermalization should be rapid, and non-thermal capture should be unimportant. Further, the neutrons need only be slowed into the 1/v region (below 0.1-1 keV) before they may be considered "thermalized" for the purposes of our calculations (see discussion in Section IIIA). Rough estimates indicate that about 10 per cent of the neutrons produced may be captured above the 1/v region. (c) Surface leakage. This is a finite effect, even for very large bodies. Hess, Canfield, and Lingenfelter [1961] calculate that 17 per cent of the neutrons produced in the earth's atmosphere escape. For the moon, assuming chondritic composition, Lingenfelter, Canfield, and Hess [1961] calculate 36 per cent leakage if H/Si = 0.04 (the chondritic value) or 17 per cent if H/Si = 1. The latter authors point out that the leakage rate is a sensitive function of the lunar H/Si ratio. In our calculations f_r is considered constant. We will now attempt to justify this. Although f_n would depend on H/Si for H/Si << 1, we are here concerned with bodies of $H/Si \sim l$. In these cases the mean distance travelled during thermalization will always be small compared to the mean distance which must be travelled in order to escape. Changes in H/Si will only affect the former distance, whereas the leakage rate is sensitive to the latter distance. An extension of this argument indicates that f will not be sensitive to the size of the

Cl

planetesimals as long as they are large (\geq 20-50 cm). As long as this "mean escape distance" is small compared to the radius of curvature or some other eppropriate measure of the dimensions of the planetesimals, the neutrons cannot distinguish their environment from that of a semi-infinite slab. From these calculations surface leakage may result in a 10-20 per cent loss.

Combining this with the above estimate of 10 per cent loss due to non-thermal capture, gives values of f_r of 0.7-0.8. The FGH calculations were based on $f_r = 1.0$; however, reference to Table 4 in the main text indicates that variations of this magnitude in f_r will not introduce an appreciable error in the calculated F_d and H/Si values.

C2

APPENDIX D

CALCULATION OF Σ' ; ADDITIONAL CHEMICAL CONSIDERATIONS

A value of Σ' follows directly from a choice of chemical composition for the planetesimals. There are two logical choices for this based on either meteoritic or solar abundances. For many elements these are in good agreement; however iron is much more abundant in the meteorites. Fe⁵⁶ has a fairly large neutron capture cross-section (2.53 b) and a large relative abundance, thus it will make an important contribution to Σ^{1} making the value calculated from solar abundances smaller than that from meteoritic abundances. The breakdown of the calculation for these two cases is shown in Table Dl. We have used meteoritic abundances based on the Type I carbonaceous chondrites as given by <u>Ringwood</u> [1962] based in turn on the chemical analyses of <u>Wiik</u> [1956]. There is considerable controvery as to what extent the composition of this class of meteorites approximates the primordial solar system abundances [Urey, 1964; Anders, 1963]; however, they are a logical choice for the present calculation since they are (with the exception of the enstatite chondrites) richest in total iron of all stony meteorite classes. The calculated value of $\Sigma' = 3.4$ is thus an approximate upper limit, whereas the solar value of $\Sigma' = 1.2$ constitutes a lower limit. This latter value differs only slightly from the value of 1.35 used by FGH. The solar abundances were taken from Aller [1961]. The source of the abundances not obtained from the above references (particularly P, S, Cl) are given by footnotes in Table Dl.

 N^{14} and A have been omitted from Table Dl following the basic assumption that gaseous substances were not incorporated into the planetesimals. As is discussed in the text, He³ has such a high neutron capture cross-section (5400 b) that we are not only forced to assume that it was highly depleted

Dl

in the formation of the planetesimals but also that the spallation produced He^3 (both as He^3 directly and as H^3) is able to diffuse out of the planetesimals in a time short compared to its lifetime for neutron capture.

The very large neutron capture cross-sections of Gd and Sm will cause the abundances of these elements to vary during the course of the irradiation. However, Table Dl indicates that taking Σ' to be a constant introduces negligible errors because the abundance of these elements is initially so small. Similar arguments can be made for other elements with large neutron capture cross-sections, including Li⁶ and B¹⁰.

Although we have used Si and Fe group abundances from carbonaceous chondrites, we have assumed that C - which is about 5 per cent by weight in the carbonaceous chondrites - was not present in any appreciable concentration in the planetesimals, i.e., we assume that these as well as all other meteorites are second generation bodies. This assumption does not significantly affect the calculation of Σ^{*} since C^{12} has a very low neutron capture cross section. We again emphasize that the significance of the carbonaceous chondrite abundances for the present calculation is that they provide an upper limit for Σ^{*} . More discussion of this assumption is given in the text in Sections IIC and IIIB.

From the abundances given in Table D1, the amount of 0^{16} in the planetesimals may be estimated assuming it to be present in proportions corresponding to the simple oxides of the metallic elements (e.g., the amount of 0 associated with Si = 1.0 is 2.0 corresponding to SiO₂). All of the iron group elements were lumped together and taken as (FeO + FeS). By this method one obtains for the carbonaceous chondrite composition $0 = 3.8 + \frac{1}{2}$ H and $3.0 + \frac{1}{2}$ H for the solar composition. For H/Si ~1 this corresponds to a gross chemical

D2

composition of 0 = 54 per cent, Si group = 34 per cent, Fe group = 12 per cent by number for the carbonaceous chondrite composition. As discussed in Appendix B, the gross chemical composition must be known in order to estimate α_L and α_2 which in turn are needed to calculate H/Si. In principle then one should - by iteration or some other means - make sure that the gross chemical composition, α_L , α_2 , and H/Si values adopted were all self-consistent; however, considering the other uncertainties involved in the calculation, it seemed pointless to include this complication in the calculation. We have just taken a "standard" gross chemical composition of 55 per cent 0, 35 per cent Si group and 10 per cent Fe group for the carbonaceous chondrite abundances and 70 per cent 0, 30 per cent Si group for the solar abundances.

Τ/τ	Ψ_n (neutrons/cm ²)	
0	5.9 × 10 ²¹	
0.5	5.7	
1.0	5.4	
3.0	4.6	
8	3,3	4.0 (FGH)

Values of Neutron Flux, ψ_n , as Function of Bombardment Time, T

 τ = mean life of Be¹⁰ \approx 4 \times 10⁶ yr

		Total L	B/Li	B _{ll}	B ¹⁰	Be ⁹	Li ⁷	Li ⁶
on (calculateo)	spallation	51.8	0.73	6.2	15.4	0.64	19.4	10.2
ic es (observed)	meteoritic abundances	44.3	0.21	6.2	1.5	0.64	33.3	2.7

Calculated Spallation Yields for $T/\tau\approx$ 7.5 (Si = $10^6)$

TABLE 2

Li _s /L	Be _s /L _s	B _s /L _s	(B/Li) _s		
0.77	0.03	0.20	0.26	calculated,	$T/\tau = 0$
0.64	0.06	0.30	0.47	**	$T/\tau = 3$
0.57	0.07	0.36	0.63	**	$T/\tau \approx 7.5$
0.49	0.08	0.43	0.88	••	$T/\tau = \infty$
0.34	0.23	0.43	1.3	11	$T/\tau = \infty$ (FGH)
0.39	0.08	0.53	1.4	0 ¹⁶ + 300 Me	Vn (Fuller)
0.24	0.12	0.64	3.7	C ¹² + 90 Me ³	Vn (Kellogg)

TABLE 3

Comparison of Calculated Spallation Yields with Cloud Chamber Data (Be¹⁰ included with Be)

TABLE 4

(F_d, H/Si) Solutions

	$\Sigma' = 3.4$	(Carbonaceous	Chondrites)	
fr		0.75	999 - 20 - 99 - 99 - 99 - 99 - 99 - 99 -	1.0
$\alpha_{\rm L}$	Fd	H/Si	Fd	H/Si
0.1	37	0.48	28	0.54
0.0 7 5	28	0.65	21	0.83
0.05	20	1.2	15	1.6
0.02	12	6.9	10.6	10
		Σ' = 1.2 (Sol	ar)	
fr		0 .7 5		1.0
a	Fd	H/Si	Fd	H/Si
0.1	15	0.76	12.3	1.2
0.075	13	1.5	11.1	2.2
0.05	11.5	3.1	10.4	4.5
0.02	10.3	12	9.9	16

TABLE	Al
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Abundances	of	LiBeB	(Si	=	10 ⁰	Scale)
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	SU	This Work	Ref.
Li ³	7.4	2.7	SH,KM
Li ⁷	92.6	33.3	SH,KM
Be ⁹	20	0.64	SW
B10	4.5	1.5	S
B ¹¹	19.5	6.2	S
	144.0	44.3	

 $B/Li = \frac{7.7}{36.0} = 0.214$

- SU = Suess and Urey, Rev. Mod. Phys. 28, 53 [1956]
- SH = <u>Shima and Honda</u>, J. Geophys. Res. <u>68</u> 2849 [1963]
- S = Shima, J. Geophys. Res. <u>67</u>, 4521 [1962]
- SW = Sill and Willis, Geochim. Cosmochim. Acta, 26, 1209 [1962]

KI4 = Krankowsky and Müller, Geochim. Cosmochim. Acta, 28, 1625 [1964]

TABLE A2

Effect of Variation of B/Li and α_7/α_6 on ψ_n

$$T/\tau = \infty$$

(Multiply entries by 10^{+21} to get ψ_n in n/cm²)

<u></u>				B/Li		
		0.07	0.1	0.21	0.3	0.6
<u> </u>	1.0	7.7	6.6	4.2	3.4	2.0
$\frac{\alpha_7}{\alpha_6}$	1.9	5.1	4.6	3.3	2.8	1.8
	3.0	3.6	3.3	2.6	2.2	1.6

TABLE A3

Spallation Yields of 300 MeV $n + 0^{16}$

from Fuller [1964]

Type of Event	Number Observed	Cross-Section (mb)
Total Number of Stars	602	225 ± 15
protons	619	231 ± 20
deuterons	284	106 ± 10
u ³	31	12 ± 3
He ³	52	19 ± 3
He	339	127 ± 12
Li	54	20 ± 3
Be	42	16 ± 3
В	74	28 ± 3

TABLE A4

	f ₁₀	0.1	0.25	0.5
T/τ				
0.5		4.7	5.7	6.2
3		3.9	4.6	4.8

Sensitivity of Calculated Neutron Flux to Changes in f_{10}

(Multiply by 10^{+21} to obtain ψ_n in n/cm²)

TABLE B1

	0 ¹⁶	Si	Fe	Average l	Average 2
neutrons	450	880	1800	740	5 70
deuterons	110	145	230	130	120
LiBeB	85	20	5	55	66
interaction cross-section	250	400	550	330	300

Estimated Primary Cross-Sections in mb

l = carbonaceous chondrite composition (Ringwood)

2 = solar composition (Aller)

TABLE B2

Sensitivity of Results to Variations in α_2

(Carbonaceous Chondrite Composition)

$\alpha_{\rm L} = 0.05$	$f_{r} = 0.75$		
α ₂	Fd	H/Si	
0.1	20	1.2	
0.2	22	2.1	

Calculation of Σ^*

Si Group	σ _A (barns)	N _A /Si	^N A σ _A	Fe G ro up	^σ A (barns)	n _a /si	σ _A ^N A		
Na	0.52	0.064	0.033	Sc	24.0	3.4 × 10 ⁻⁵	0.001		
Mg	0.063	1.05	0.066	Ti	5.8	0.0029	0.017		
Al	0.23	0.085	0.020	v	5.0	1.9×10^{-4}	0.001		
Si	0.16	1.000	0.160	Cr	3.1	0.0124	0.038		
P	0.20	0.013	0.003	Mn	13.2	0.0078	0.103		
S	0.52	0.51 ^a	0.266	Fe	2.53	0.889	2 .2 55		
Cl	33.6	0.0015 ^b	0 .0 52	Co	37.0	0.0023	0.085		
К	2.07	0.0039	0.008	Ni	4.8	0.0455	0.218		
Ca	0.44	0.075	0.033	Cu	3.8	2.38×10^{-4}	0.001		
		2.80	0.641	Gđ	4.6×10^{4}	$5.5 \times 10^{-7^{c}}$	0.025		
				Sm	5.8×10^{3}	$2.3 \times 10^{-7^{c}}$	0.001		
						0.96	2.745		
				Total $\Sigma^* = 3.39$					

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Case I: Carbonaceous Chondrite Abundances

(continued)

Si Group	σ _A (barns)	N _A /Si	σ _A N _A	Fe Group	σ _A (barns)	N _A /Si	σ _A ^N A
Na	0.52	0.063	0.033	Sc	24.0	2.1 × 10 ⁻⁵	0.001
Mg	0.063	0 .7 94	0.050	Ti	5.8	0.0015	0.009
Al	0.23	0.050	0.012	V	5.0	1.6 × 10 ⁻⁴	0.001
Si	0.16	1.000	0.160	Cr	3.1	0.0072	0.022
Ρ	0.20	0.007	0.001	Mn	13.2	0.0025	0.033
S	0.52	0.63	0.318	Fe	2.53	0.118	0.298
Cl	33.6	0.0015 ^d	0.052	Co	37.0	0.0014	0.052
K	2.07	0.0016	0.003	Ni	4.8	0.026	0.125
Ca	0.44	0.045	0.020	Cu	3.8	0.0035	0.013
		2.59	0.649	Gđ	4.6×10^{4}	$5.5 \times 10^{-7^{d}}$	0.025
				Sm	5.8×10^{3}	$2.3 \times 10^{-7^{d}}$	0.001
						0.160	0.580

Case II: Solar Abundances

Total $\Sigma' = 1.23$

^a Based on data given for Type I carbonaceous chondrites given by <u>Mason</u> [1962], p. 96.

^b Average carbonaceous chondrite value given by <u>Urey</u> [1964].

^c Value for Type I carbonaceous chondrites given by <u>Urey</u> [1964].

^d Assumed same as carbonaceous chondrites.

Fig. 1. The primeval solar system according to Hoyle.



Fig. 1