23 March 1962, Volume 135, Number 3508

SCIENCE

Nuclear Clues to the Early History of the Solar System

Light nuclei were synthesized before planetary material had entirely separated from hydrogen.

William A. Fowler

The space age cometh, and with it come the problems associated with responsible scientific endeavor. There are those who wish to "climb" to the outer reaches of the solar system just because it is "there." This is the motivation of the mountaineer vis-à-vis mountains. It will probably satisfy the space explorer and discoverer in the new adventure, but it will not satisfy the scientist. The scientist must still ask the additional question, "What are we going to learn in space research?" This article will not suggest a general answer to this question. It will attempt to lay the background for two questions to which we might hope to obtain partial answers. The first question is this: What were the nuclear processes which synthesized the chemical elements in the matter which makes up the sun, the earth, the other planets, and the meteorites-in short, the matter of the solar system? This leads to a second question: Are there clues in nucleosynthesis to the events

which transpired during the early history of the solar system?

A knowledge of the nuclear composition of solar-system material is basic to all other problems concerning the system. For example, before the complicated chemical and geological structure of the planets can be completely understood, starting with atoms and molecules and progressing to the many complications of macroscopic matter, we will need detailed knowledge of the abundance of the elements and their isotopes. We need to know the absolute and relative abundance of each of the various nuclear species throughout the solar system.

Up until the present time we have been severely limited in our efforts to make direct observations on nuclear abundances. The material directly available to us has been that in a thin layer of the earth's crust and that in the meteorites and the tektites. The meteorites may come from the asteroidal belt and may represent fragments from the breakup of a planet or from the collision of two planetoids in that region of the solar system. If this is the case, the meteorites probably tell us more about the average nuclear composition of large solid bodies in the solar system than does the greatly differentiated material of the earth's crust. The tektites may be from the moon. Alternatively, they may represent material splashed from the surface of the earth in a collision with another object and returned to the earth by its gravitational pull. In any case, we have been able to make direct observations only on very special samples of solar-system material.

These direct observations have been supplemented by astronomical observations in which spectroscopic techniques for differentiating one element from another have been employed. Unfortunately, only in rare instances-for example, differentiation between C13 and C¹²—have isotopes been differentiated. Nevertheless, by the analysis of emitted, absorbed, and reflected light, much has been learned concerning the composition of the surface or atmosphere of the sun, the moon, and the planets. The composition of the interstellar and interplanetary medium has also been studied. Much has been learned, but not enough. In fact, it can be fairly said that what we have learned has raised more questions than it has solved. We have some scattered clues to the origin and history of the solar system, but that is all. Exploration of space may well break this field of investigation wide open. Instruments will soon be landed on the moon and the nearer planets. There are techniques by which information on the nuclear composition of the landing surfaces can be ascertained and sent back to the earth. Eventually we will be able to obtain samples of lunar and planetary matter. That will be the day!

In the meantime, let us consider one promising technique for obtaining information by landing instruments on the surface of the moon or planets. For years in our nuclear laboratory at California Institute of Technology we have analyzed the composition, by atomic mass, of the surface layers of target materials by scattering energetic protons, alpha particles, and other light nuclei from these targets (1). In elastic scattering the recoil energy of the scattered particle is a certain fraction of its incident energy which depends on

The author is professor of physics at the California Institute of Technology, Pasadena. At present he is on leave at St. John's College and the Department of Applied Mathematics and Theoretical Physics, Cambridge University, Cambridge, England. This article is the text of a lecture presented at the Space-Nuclear Conference sponsored by the American Rocket Society and Oak Ridge National Laboratory and held in Gatlinburg, Tennessee, from 3 to 5 May 1961. It is published with the permission of the American Rocket Society.

the ratio of the mass of the scattering nucleus to that of the scattered particle. For example, in the case of backward scattering, through an angle of 180 degrees, the recoil energy E_{rec} is related to the incident energy E_{inc} according to the equation

$E_{\rm rec}/E_{\rm inc} = (M_0 - M_1)^2/(M_0 + M_1)^2$,

where $M_0 \ge M_1$, M_0 is the mass of the scattering nucleus, and M_1 is the mass of the scattered nucleus. In the case $M_1 \ge M_0$, the foregoing expression can be used but the incident nucleus continues forward instead of reversing its direction during the collision.

There is nothing mysterious about this equation; it follows directly from the conservation of energy and momentum. It is well known that a billiard ball stops dead in a direct hit with no English on a similar billiard ball: $E_{rec} =$ 0 for $M_1 = M_0$. In an elastic recoil from a very heavy object, the billiard ball recoils with approximately its full energy: $E_{\rm rec}/E_{\rm inc} \approx 1$ for $M_1 \ll M_0$. Thus, the spectrum in energy (or velocity, or momentum) of the scattered nuclei of known mass reveals the otherwise unknown mass or masses of the target nuclei. This yields a unique determination for light nuclei, and even for heavier nuclei where isobars occur (those with atomic mass A = 36 and higher), the resulting ambiguity is the least of the difficult practical problems encountered even in laboratory application of the technique. I shall not elaborate upon these difficulties here. The important point is that a technique, simple in principle and involving no chemical or mechanical processing of the material, is available for the study of the nuclear constitution of planetary and satellite surfaces. Material at greater depths can also be studied, in situ or on being transferred to the surface. A group under A. Turkevich (2) at the University of Chicago is reportedly designing and developing apparatus for applying this technique in space research.

Origin of Nuclear Species

The remainder of this article is devoted to an account of a recent development in nuclear astrophysics which tells us something about the early history of the solar system. To begin at the beginning, it is necessary to recount current ideas concerning the origin of the nuclear species in the solar system, and this in turn requires some account of the origin and history of the Galaxy in which the solar system resides.

When we look out beyond the confines of the planetary system which surrounds the sun, we see our Galaxy as the majestic assemblage of stars which we call the Milky Way. We look at the Milky Way edge on, from a position approximately half-way out from the center of the Galaxy. The stars in the Galaxy populate a flat, disk-shaped structure with a spherical nucleus from which radiate several spiral arms trailing the direction of rotation. Twenty billion years ago our Galaxy was vastly different. It was then a rotating extended cloud of gas hanging nebulous and diffuse in space. The gas consisted of hydrogen, which is the lightest of all the elements. That a small amount of helium or heavier elements was also present cannot be absolutely ruled out, but for the moment the simplest hypothesis is that the primordial gas of the Galaxy was pure hydrogen. We do not know today whence this hydrogen came. It may have been created by processes we do not understand at the start of the expanding universe, as some believe, or it may be that hydrogen has been and is still being constantly created in a steady-state universe, as others believe.

It may also be that, during its lifetime, the Galaxy has continuously exchanged material with the intergalactic medium. We do not know whether our Galaxy is an "open" or "closed" system. These problems need not concern us here. The basic nuclear processes in the stars of the galaxy do not depend critically on small contaminations in material principally composed of hydrogen in any case.

In time, the hydrogen gas in some regions in the Galaxy collected to form the first stars of the system, and this process of star formation has continued until the present. The sun was formed 4.5 billion years ago, relatively late in the lifetime of our 20-billionyear-old Galaxy.

Upon condensation, the material of a star heats up as gravitational energy is converted into internal thermal energies, and the condensation continues until the temperature becomes high enough $(10^{7} \,^{\circ}\text{K})$ for nuclear burning to occur. In the first-generation stars this nuclear burning converts the pure hydrogen into helium by a series of nuclear reactions which have been extensively studied experimentally and theoretically. The overall process can be symbolized as follows:

4 $H^1 \rightarrow He^4$.

Hydrogen is the fuel, and helium is the ash. The onset of the hydrogen burning (nuclear, not chemical) releases energy, the internal temperature and pressure rise, and the gravitational collapse is stopped. The star is stabilized with an internal fusion reactor as its source of energy, and it shines steadily forever after-or almost forever! Eventually, even the prodigious nuclear energy source is exhausted; the central hydrogen is all converted into helium, which does not burn at the temperature at which hydrogen is consumed. Hydrogen nuclei or protons have a single positive charge equal in magnitude to the charge of the electron. Helium nuclei are doubly charged, and greater relative velocities, and thus greater temperatures, are required to bring them close enough together for fusion against their mutual electrostatic repulsions. At this point, gravity takes over once again and contracts and heats the inner helium core of the star until the temperature at which helium burning begins (10° °K) is reached. The helium burning results in the production of carbon. The process takes place through the transient formation of short-lived Be⁸ in the collision of two He⁴ nuclei. In rare instances, before it breaks apart, the Be^s captures a third He⁴ nucleus to form stable C^{12} . The over-all process can be symbolized by

3 He⁴ \rightarrow C¹².

The production of helium and carbon in successive stages from pure hydrogen will serve as an example of the nuclear processes by which, it is now known, all of the isotopes of the heavier elements-those beyond carbon in the periodic table, including unstable transuranic elements-can be synthesized, with hydrogen as the basic building block. As successive nuclear processes take place, the composition of a star changes and the star is said to evolve as its internal structure and external appearance vary in response to the composition changes which develop in various layers of its interior. The hydrogen burning occurs in the stellar core during the main-sequence stage of stellar evolution; the helium burning and the burning of heavier nuclei occur during the red-giant stage. During this

last stage, hydrogen burning continues outside the helium core.

It is essential, from the point of view of stellar nucleosynthesis, that instabilities should arise during the evolution and aging of a star. Stellar instabilities are indeed observed, the most spectacular being supernova events in which a star almost completely explodes and distributes its transmuted material to interstellar space. This material is then mixed with the uncondensed hydrogen gas in the Galaxy and thus is available for condensation into later-generation stars. In this way the heavy elements in the solar system were produced-not by the sun itself but by nuclear processes in countless stars which formed and died long before the sun came into being. The sun itself has been steadily converting hydrogen into helium since its formation, but this helium is still at the sun's center. What we see in the solar atmosphere is unburned primordial hydrogen (~ 90 percent, by number) plus the nuclear debris from a long period of previous stellar activity. A detailed account of this general point of view concerning the origin of the elements has been given by Burbidge, Burbidge, Fowler, and Hoyle (3), and I have given a nontechnical account (4). The announcement of a comprehensive scheme for the origin of the nuclear species by means of nuclear reactions in stars was first made in Science in 1956 (4a). The planets presumably formed from material similar to that in the primitive solar atmosphere. Condensation separated the solids from the hydrogen and other gases that were preponderant in the primitive material.

The outstanding exceptions in this chain of nuclear synthesis are the light nuclei other than He⁴. It is fairly well established that He³ is of secondary. radiogenic origin in terrestrial processes. No stable nuclear forms exist at mass 5 or mass 8. However, it has long been known that severe difficulties stand in the way of any theory (5) of the origin of deuterons (6) and of the isotopes of lithium, beryllium, and boron in terms of thermonuclear reactions in the interiors of stars. The problem of finding a mechanism for the synthesis of these light nuclei is a very difficult one, since D², Li⁶, Li⁷, Be⁹, B¹⁰, and \mathbf{B}^{11} would be rapidly destroyed in hydrogen burning in stellar interiors even if it is granted that they are produced at all. In the over-all process $4H^1 \rightarrow$

23 MARCH 1962

He⁴, in main-sequence stars, D² is produced in equilibrium abundance given by the ratio D²/H¹ $\approx 10^{-17}$, a very small value compared to the terrestrial ratio D²/H¹ = 1.5×10^{-4} . In helium burning (symbolized by 3He⁴ \rightarrow C¹²) in giant stars, the stable isotopes of lithium, beryllium, and boron are bypassed. Thus, the production of these nuclei does not lie in the main line of element synthesis. The low abundance of these light nuclei attests to this.

It has been realized, as a consequence, that the synthesis must almost certainly occur through spallation by high-energy, nonthermal particles interacting with relatively cool material of moderate density, irrespective of the detailed astrophysical environment in which it takes place. (I use the term spallation throughout this article to cover all mechanisms of production of deuterons, lithium, beryllium, boron, and other nuclei at high energy. Thus, the products of spallation include those ejected directly from nuclei, those which result from evaporation processes, and those remaining as residues after the direct-ejection and evaporation processes have ended.) The production of deuterons, lithium, beryllium, and boron nuclei by spallation of heavier nuclei in the cosmic radiation is an example. We take it that terrestrial and meteoritic deuterium, lithium, beryllium, and boron were produced in a similar way. The deuterium resulted, for example, when the spallation produced deuterons combined with electrons to form neutral atoms. When I use the term deuterium below, I imply the atomic processes subsequent to the primary nuclear processes.

Astrophysical Setting

The angular momentum of the solar system resides principally in the planets, not in the sun. Hoyle (7) has suggested that the primitive condensing sun transferred its angular momentum of rotation to the planets by means of a magnetic torque coupling between the solar condensation and the disk of primitive planetary material. This transfer swept away the hydrogen, helium, and other gases which constituted the bulk of solar material and left behind those solid bodies or planetesimals which formed by condensation at the low ambient temperatures (130° to 200°K). These planetesimals ultimately

aggregated to form the planets. In the angular-momentum transfer, magnetic energy of the order of 5 \times 10⁴⁵ ergs was dissipated in a sequence of powerful solar flares similar to those which occur on the surface of the present sun. A characteristic of the solar flare is that the stored energy of the magnetic field is converted into the energy of high-speed particles with surprising efficiency, as high as 10 to 20 percent. Because the lines of the magnetic field passed directly from the solar condensation to the partially ionized planetary gases, accelerated particles traveled readily outward, spiraling about the lines of force, from the sun to the planetary material. The high-energy particles were not dissipated isotropically in space.

Thus, the solid condensations, immersed in the out-flowing gas, were bombarded by highly energetic particles, largely protons. The total energy of this bombardment was of the order of 10⁴⁵ ergs. The angular-momentum transfer and the acceleration and bombardment processes occurred in a period of the order of the Kelvin-Helmholtz estimate for contraction of the sun, an interval of approximately 10⁷ years. The dissipation rate was thus ~ 10^{33} ergs per year. At the present time, the sun emits high-energy particles with total energy equal to ~ 10^{31} ergs per year. Thus the early, active sun dissipated energy in the form of fast charged particles at a rate approximately 10⁷ times as high as that observed at the present time. These fast particles produced spallation products and neutrons in the surface layers of the small planetesimals orbiting about the sun. These planetesimals were destined eventually to form the planets and the parent bodies of the meteorites. The high-energy particles also produced spallation products and neutrons in the surface layers of the sun, but this interesting aspect of the over-all process will not be discussed in detail here.

The discovery of Bonsack and Greenstein (8) that the surface layers of the T Tauri stars contain more than ten times as much lithium as the nebular material from which they formed supports this theory of stellar nucleosynthesis. The T Tauri stars are stars still in the process of formation, and it is suggested that the lithium has been produced in or near their surfaces by irradiation of the condensing material by high-energy particles. It is presumed

that the T Tauri stars have not yet developed outer convective layers which mix to high enough temperatures to destroy the lithium which has been produced. Our sun probably passed through a stage similar to that through which the T Tauri stars are now passing. The difficulties in the synthesis of deuterium, lithium, beryllium, and boron nuclei that I have mentioned suggest that the primitive material from which the solar system formed did not necessarily contain these nuclei. They were produced by processes in the planetary material itself during the early history of the solar system.

Synthesis of Lithium, Beryllium, Boron

It is well here to examine the abundances and cross sections listed in Table 1. In column 2 the meteoritic or terrestrial abundances of interest are listed. The abundances are given relative to the usual standard, silicon =



Fig. 1. Typical spallation and neutron processes induced in the planetesimals by high-energy protons from the sun. The top proton reaches the end of its range without interaction. The second proton interacts with an O¹⁶ nucleus to produce several protons (p) and neutrons (n), an alpha particle (α) , and a Li⁶ nucleus. The path of one of the neutrons is shown in detail. After scattering and thermalization it is captured by a proton in a hydrogen atom to form a deuteron with the emission of gamma radiation. The third high-energy proton produces a B¹⁰ nucleus among other spallation products. One of the spallation-produced neutrons is captured by the Li⁶ nucleus to yield an alpha particle and a triton (t). The triton eventually decays to He⁸ through beta radiation. The fourth proton interacts with Mg²⁴ to produce Be⁷, Li⁸, and so on. The Be⁷ particle captures an electron to form Li⁷ while the Li⁸ particle decays to Be⁸, which breaks up into two alpha particles. Aneutron interacts with B¹⁰ to produce Li⁷ and an alpha particle. The neutron interactions illustrate the depletion of spallation-produced Li⁶ and B¹⁰ and the increase in abundances of He⁸ and Li⁷.

10⁸. Cross sections of products of spallation are given in column 3. Cross sections of products of thermal neutron destruction are given in column 5, after the appropriate reaction (col. 4). Inspection of the abundances of lithium, beryllium, and boron nuclei and of the cross sections of spallation products leads to several interesting conclusions. It does not seem reasonable at all, on the basis of spallation production, that the even-A isotopes Li⁶ and B¹⁰ should be so low in abundance relative to the odd-A isotopes Li⁷, Be⁹, and B¹¹. The finding that abundances of Be⁹ and B¹¹ nuclei are approximately equal is not surprising, but then why should the abundance of Li⁷ nuclei be approximately five times as great? An immediate explanation is given on qualitative grounds for these otherwise puzzling abundance ratios if it is assumed that nuclei were subjected to slow neutron reactions simultaneously with their production. Then the reactions

$$\operatorname{Li}^{6}(n,\alpha) \operatorname{T}^{3}(\beta^{-}v^{*}) \operatorname{He}^{3}$$

and

$$B^{10}(n,\alpha)Li,$$

which have large low-energy cross sections, will decrease the abundances of Li° and B^{10} and increase those of Li^{7} and He³. The thermal cross sections are 945 and 3813 barns, respectively. As noted previously, terrestrial He³ is thought to be due to secondary radiogenic processes in the earth's crust. (It can be shown that the He³ produced by spallation escapes by diffusion from the planetesimals in a relatively short time.) The (n,α) reactions on Li⁷, Be⁹, and \mathbf{B}^{11} are endoergic and do not occur at low energy. The (n,γ) reactions on these nuclei have very low cross sections. Some of the spallation and neutron reactions are illustrated in Fig. 1.

Thus there would seem to be unmistakable qualitative evidence that the isotopes of lithium, beryllium, and boron were subject to neutron irradiation simultaneously with their synthesis by spallation. The existence of neutrons occasions no surprise in itself, for the spallation reactions themselves produce neutrons.

Quantitative calculations in which the lithium, beryllium, and boron cross sections and abundances are used lead to a value for the neutron flux which irradiated the solid planetesimals. This irradiation occurred over the period $T = 10^7$ years (= 3×10^{14} seconds) ²³ MARCH 1962 Table 1. Abundances and cross sections. Cross sections are given in barns (1 barn = 10^{-24} cm²) and millibarns (mbarn).

Isotope	Meteoritic or terrestrial abundance	Spallation prod- ucts, cross section (mbarn)	Neutron-destruction products	
			Reaction	Cross section
H ¹	4×10^{3}	······································	$H^1(n \propto) D^2$	0.33 barn
D^2	0.6 (1.5 \times 10 ⁻⁴ H ¹)	100	$D^2(n,\gamma)T^3$	0.55 barn
Li ⁶	7.4	15	$Li^{6}(\mathbf{n},\alpha)\mathbf{T}^{3}$	945 harn
Li ⁷	92.6	15	$Li^7(n \propto) Li^8$	33 mbarn
Be ⁹	20.0	11	$Be^{9}(n \propto) Be^{10}$	10 mbarn
B10	4.5	38	$B^{10}(n_{\alpha})Li^{7}$	3813 harn
B11	19.5	11	$\mathbf{B}^{11}(\mathbf{n},\boldsymbol{\gamma})\mathbf{B}^{12}$	< 50 mbarn

and millibarns (mbarn).

during which the contracting sun was active in producing high-energy particles which in turn produced the neutrons. Since the spallation-product cross sections are not too well established experimentally (some of those in Table 1 are estimates), it is necessary to lean heavily on all of the abundance data and the well-established neutron cross sections. However, the calculation can be illustrated by using only a few of the numbers in Table 1. Consider the production of B^{11} per unit time. If we let the nuclear symbol B^{11} represent the abundance of B^{11} nuclei, then

$$\frac{\mathrm{d}\mathbf{B}^{11}}{\mathrm{d}t} = s_{11}\phi_{\mathrm{p}}N$$

where s_{11} is the spallation-product cross section for B¹¹ (= 11 millibarns = 11 × 10^{-27} cm²) (Table 1); ϕ_P is the flux of high-energy particles which induce the spallation reactions; and N is the number of spallation targets. The abundances B¹¹ and N must refer to the same sample of matter. For our purposes it is sufficient to take $\phi_P N$ as constant in time, so that

$$B^{11} = s_{11}\phi_n NT$$

For the case of B^{10} the destruction by the (n,α) reaction must also be considered. Thus,

$$\frac{\mathrm{dB}^{10}}{\mathrm{d}t} = s_{10}\phi_{\mathrm{p}}N - \sigma_{10}\phi_{\mathrm{n}}\mathrm{B}^{10}$$

where s_{10} is the spallation-product cross section for B^{10} (= 38 millibarns = 38 $\times 10^{-27}$ cm²); σ_{10} is the thermal neutron cross section for B^{10} (= 3813 barns = 3813 $\times 10^{-24}$ cm²); and ϕ_n is the neutron flux.

Upon integration, with ϕ_n as well as $\phi_n N$ taken as constant in time, it is found that

$$B^{10} = \frac{s_{10}\phi_{\rm p}}{\sigma_{10}\phi_{\rm n}} N \left[1 - \exp\left(-\sigma_{10}\phi_{\rm n}T\right)\right]$$
$$\approx \frac{s_{10}\phi_{\rm p}}{\sigma_{10}\phi_{\rm n}} N \text{ for } \sigma_{10}\phi_{\rm n}T >> 1$$

Using the last approximation for B^{10} , which we find to be justifiable, one finds

$$\frac{\mathbf{B}^{11}}{\mathbf{B}^{10}} = \frac{s_{11}}{s_{10}} \, \sigma_{10} \phi_{n} T = 1100 \, \phi_{n} T \times 10^{-24}$$

With $B^{11}/B^{10} = 19.5/4.5 = 4.3$, the final result for the time-integrated neutron flux is $\phi_n T = 4 \times 10^{21}$ neutrons per square centimeter. Hence, $\sigma_{10}\phi_n T \approx 15$, justifying the use of the approximation valid for $\sigma_{10}\phi_n T >> 1$. The same value for $\phi_n T$ is found on analysis of the depletion of Li⁶ and the increase in Li⁷. If T is taken to be 3×10^{14} seconds, the thermal neutron flux is then $\phi_n = 1.3 \times 10^7$ neutrons per square centimeter per second. For comparison it may be noted that the neutron flux in pile thermal columns has reached $\sim 10^{10} \text{ n/cm}^3$ sec.

Deuteronomy: The Synthesis of Deuterons

With the determination of $\phi_n T$ it becomes possible to make quantitative calculations on neutron interactions with other nuclei. One of the most important of these is the capture of neutrons by protons to form deuterons. The cross section for this capture is given in Table 1 as $\sigma_1 = 0.33$ barn. Thus, through neutron capture alone,

$$D^2/H^1 = \sigma_1 \phi_n T = 1.3 \times 10^{-3}$$

More detailed analysis indicates that this ratio is increased by the direct production of deuterons in spallation processes to

$$D^2/H^1 = \sigma_1 \phi_n T = 1.3 \times 10^{-3}$$

It is immediately apparent that this value is just an order of magnitude greater than the terrestrial-abundance ratio given in Table 1:

$$D^2/H^1 = 1/6500 = 1.5 \times 10^{-4}$$
.

This value is also known to hold ap-

1041

proximately for the carbonaceous chondrites, a type of meteorite relatively rich in hydrogen. This is strong evidence that material which formed the parent bodies of the meteorites suffered the same irradiation as material which formed the earth. It is very gratifying to find that the neutron irradiation which was necessary to give the observed modification of the abundances of Li⁶, Li⁷, and B¹⁰ was great enough to produce a D^2/H^1 ratio greater than the terrestrial ratio in the material irradiated. In fact, this must be an essential feature of any synthesis for deuterium, lithium, beryllium, and boron nuclei, because not all of the terrestrial material can have been irradiated by thermal neutrons totaling such a number that $\phi_n T = 4 \times 10^{21} \ n/\text{cm}^2$. Otherwise, nuclear species with cross sections greater than 1000 barns would have been greatly reduced in number from their primordial abundance in the material of the solar nebula.

At this point, one example will suffice, that of Gd¹⁵⁷, for which $\sigma(n,\gamma) = 2.4 \times 10^5$ barns. The cross section for Gd¹⁵⁶ has not been measured but must be comparable to that for Gd¹⁵⁸, which is 4 barns. On the other hand, the isotopic ratios relative to total gadolinium are as follows:

$Gd^{156}: Gd^{157}: Gd^{158} = 0.20: 0.16: 0.25.$

The odd-A/even-A ratio of somewhat less than unity seems to be quite normal. If all the terrestrial material had been irradiated by neutrons, the result would be

$Gd^{157} = Gd_0^{157}exp(-960),$

where Gd_0^{157} is the abundance before irradiation. This and other examples (including U²³⁵, of unhappy destiny) indicate definitely that only a small fraction of the material was irradiated with thermal neutrons and then was subsequently mixed with the unaltered material. This situation can, of course, come about in several ways; the most probable explanation is that material formed bodies large in radius compared to the radii of the high-energy charged particles which produced the neutrons and that the neutrons were thermalized and captured close to their point of origin. Only the outer layers of the planetesimals were irradiated. An estimate could be made for a dilution factor F_d on the basis of examples like Gd¹⁵⁷. However, it seems preferable to determine F_d on the basis of the terrestrial ratio for D^2/H^1 . If we define F_a in general as the ratio of the total amount of material to the amount irradiated, then we have

 $F_{a} = 1.5 \times 10^{-3} / 1.5 \times 10^{-4} = 10,$

for terrestrial and meteoritic matter. We return to this point later.

An Intermediate Stage in the Primitive Solar System

That some hydrogen was present during the synthesis of terrestrial deuterium, lithium, beryllium, and boron is qualitatively clear for several reasons. The hydrogen is effective in thermalizing the neutrons before they decay. Some hydrogen is necessary to yield deuterium upon neutron capture, since the production from direct spallation is small (~15 percent). However, it is also clear from qualitative data that the amount of hydrogen could not have equalled the overwhelming amount in primordial solar matter, since then all neutrons would have been absorbed in hydrogen and none would have been available for the reactions $Li^6(n,\alpha)T^3$ or $B^{10}(n,\alpha)Li^7$. A quantitative calculation of the hydrogen content yields $H^1 = 8$ \times 10⁶ on the silicon = 10⁶ scale. Now, in the sun, on the same scale $H^1 = 3.2$ \times 10¹⁰, while terrestrially, as Table 1 indicates, $H^1 = 4 \times 10^3$. Thus, in the planetesimals hydrogen was relatively less abundant than it is in the sun and more abundant than it is in the earth, where it occurs principally in ocean water. The abundances of deuterium, lithium, beryllium, and boron demand a stage for primitive terrestrial material intermediate between the original material of the sun and the present material of the earth. In other words, the material of the terrestrial planets was irradiated by high-energy particles from the sun at a stage of development of the planetary system when the material had become very largely, but not completely, separated from hydrogen.

Many others have reached a somewhat similar conclusion from chemical and geophysical evidence which has been summarized by Urey (9). The reduction in the abundance of hydrogen from that in the primitive solar material could have been brought about, as far as we can see, only through condensation of solid bodies or planetesimals from the primitive gas.

It is noteworthy that $H = 8 \times 10^{6}$ is of the order of magnitude (in fact, close to twice the value) of the present value for terrestrial oxygen, $O = 3.5 \times$

10⁶. This would be expected if the metals were not oxidized and if the hydrogen and oxygen were trapped in the well-known chemical combination H₂O, presumably as ice, in the primitive planetesimals of preterrestrial material. (We assume that the same hydrogen-to-oxygen ratio held throughout the shielded inner parts of the planetesimals and through the irradiated outer parts.) According to this point of view, in the subsequent formation of the earth, most of the hydrogen but very little or none of the oxygen was lost, the oxygen serving to oxidize the metals.

Size of the Planetesimals

It is thus indicated that the intermediate-stage planetesimals consisted of an icy matrix in which were embedded the metals, probably in the form of small grains. The ice constituted about 35 percent, by mass, of the planetesimals, and the density of these bodies was about 2 grams per cubic centimeter.

It is possible to make an estimate of the average size of the planetesimals from the conclusion, given earlier, that only 10 percent of the material suffered irradiation. This would indicate that the high-energy particles, mostly protons, penetrated through a surface layer which constituted 10 percent, by mass, of the planetesimals, as indicated in Fig. 2. The mean free path of high-energy protons can be calculated on the basis of geometrical cross sections for the nuclei in the planetesimals; it is found to be about 40 centimeters. The condition that the mass of the shielded interior of a body of radius a be 10 times the mass of the exposed surface skin of thickness $\lambda \sim 40$ centimeters gives

 $4\pi a^3 3 \approx 10(4\pi a^2 \lambda),$

$$a \approx 30 \lambda \sim 12$$
 meters.

or

Thus, the planetesimals were of "metric" dimensions during irradiation. Hoyle (7) has already shown that these bodies had to be at least 1 meter in radius, otherwise they would have been swept along with the outflowing gas. It can also be shown that the planetesimals could not have been very large, since for a given total mass, that of the inner planets, the target area presented to the high-energy particles would then have been too small. A detailed calculation indicates that planetesimals of approximately 12-meter radius would

SCIENCE, VOL. 135

have been struck by about 10 percent of the solar particles. The same calculation indicates that the high-energy particle acceleration required only 5×10^{44} ergs, or 10 percent of the total magnetic energy dissipated by the flares. For this calculation it is assumed that the primitive matter of only the inner or "terrestrial" planets (Mercury, Venus, Earth, and Mars) was irradiated. A uniform distribution in planetesimal radii from 1 to 50 meters also meets all the requirements discussed above.

Time Required for

Large Solid Bodies To Form

It has recently become clear from the observations of Reynolds (10), Murthy (11), and Fish, Goles, and Anders (12) that the time interval between the last event of nucleosynthesis and various stages in the formation of large solid bodies in the solar system could not have exceeded 10⁶ to 10⁸ years. These observations essentially indicate that I¹²⁰ ($\bar{\tau}$ [the mean lifetime of the radioactivity] = 2.5 × 10⁷ years), Pd¹⁰⁷ ($\bar{\tau} = 10^7$ years), and Al²⁸ ($\bar{\tau} = 10^6$ years) were incorporated in the parent bodies of the meteorites before these radioactive nuclei completely decayed.

It is difficult to understand how time between a last event of galactic nucleosynthesis in stars or supernovae and the formation of the parent bodies of the meteorites could have been so short. The "mixing" time in the Galaxy is alone thought to have been greater than $2 \times 10^{\rm s}$ years, the period of galactic rotation. It can be argued that a supernova event triggered the condensation of the solar system (see 3, p. 639), or that the sun originated in an "association" of stars such as the young stars which seem to be moving radially outward from a common point of explosive origin. If we discount these interesting possibilities, it would seem reasonable to ascribe the relatively late production of the I129, Pd107, and Al26 to the synthesis in the solar nebula described in this article

Quantitative calculations indicate that sufficient quantities of these radioactive nuclei were indeed produced. The case of I^{120} will suffice as an example of the calculations. In studying abundances of isotopic xenon in the Richardton stone meteorite, Reynolds (10) found, among other things, that Xe^{120} was 42 percent more abundant in the meteorite than in atmospheric xenon. Reynolds attributed this anomalous or radiogenic Xe^{120} to the radioactive decay reaction

$$I^{129}(\beta^{-}\nu^{*})Xe^{129},$$

which has a mean lifetime $\overline{\tau}_{120} = 2.5 \times 10^{\circ}$ years, as indicated previously. This explanation assumes that when the iodine-xenon ratio was finally fixed in the meteoritic material, some I¹²⁰ remained from a nucleosynthesis which had occurred not too long before. The subsequent decay of the I¹²⁰ and trapping of the resultant Xe¹²⁰ yielded the anomaly. For the time Δt between the termination of nucleosynthesis and the final establishment of the iodine-xenon ratio in the meteorite, Reynolds wrote

$$\frac{\mathrm{X}\mathrm{e}^{120^{*}}}{\mathrm{I}^{127}} = \left(\frac{\mathrm{I}^{129}}{\mathrm{I}^{127}}\right)_{\mathrm{o}} \exp\left(-\Delta t/\overline{\tau}^{129}\right)$$

where I^{127} is taken as a convenient standard, as determined in the meteorite in question; Xe^{120*} is the amount of Xe^{120} attributed to the decay of I^{120} in the



Fig. 2. The bombardment of an icy planetesimal by protons from the sun having an average energy near 500 Mev and a mean free path λ of approximately 40 centimeters. The protons spiral with large radii of curvature about magnetic lines of force and enter the planetesimal uniformly over all angles facing the sun. The rotation then results in the uniform irradiation of the outer layers of the planetesimals. Nuclear evidence indicates that only about 10 percent of the material was irradiated; hence, through a simple calculation (see text) a mean radius of approximately 12 meters is derived. A spread in radius of from 1 to 50 meters is a satisfactory solution of the problem. Metallic grains probably predominated over the silicates and oxides indicated in the figure.

meteorite; and $(I^{129}/I^{127})_{\circ}$ is the ratio of I^{129} to I^{127} at the end of nucleosynthesis. Solving for Δt yields

$$\Delta t = 5.7 \times 10^7 \left[\log \frac{I^{127}}{X e^{120^*}} + \log \left(\frac{I^{129}}{I^{127}} \right) \right]$$

By an ingenious combination of activation and mass spectrometric techniques Reynolds has been able to determine the ratio I^{127}/Xe^{120^*} in meteorites. For the Richardton meteorite he finds (I^{127}/Xe^{120^*}) = 1.02 ± 0.11 × 10⁵, and thus log (I^{127}/Xe^{120^*}) = 5.0.

The radioactive nucleus I^{120} is produced primarily by neutron capture in Te¹²⁸, which has a thermal-neutron-capture cross section $\sigma_{128} = 0.15$ barn and an abundance in solar-system material three times that of I^{127} . Thus

$$\left(rac{\Pi^{120}}{\Pi^{127}}
ight)_{0} pprox rac{\sigma_{128}\phi_{n}T}{F_{d}}\left(rac{\mathrm{Te}^{128}}{\Pi^{127}}
ight)_{0}$$

 $pprox 0.15 imes 10^{-24} imes 4 imes 10^{21} imes 3 \div 10$
 $pprox 2 imes 10^{-4}$

We neglect the small loss of I^{120} during the production period. Thus, log (I^{120}/I^{127}) ≈ -3.7 . When this value and log (I^{127}/Xe^{120^*}) = 5.0 are substituted into the equation for Δt in the preceding paragraph, one finds

$$\Delta t \approx 7.5 \times 10^7$$
 years.

Thus, the parent body of the Richardton meteorite had formed and cooled to the point where degassing of xenon had ended within 7.5×10^7 years of the formation of I^{120} in the planetesimals of metric dimensions which formed this body. Thus, anomalous Xe^{120} is a nuclear clue to the duration of an important epoch in the history of the solar system. The isotopes Pb^{107} and Ag^{106} yield clues to significant events in this early history on an even shorter time scale (10^6 to 10^7 years).

Speculations

It is apparent from the remarks on the energy budget made in the last section but one that the major planets, with mass 200 times that of the terrestrial planets, were not appreciably affected by the irradiation. Thus, they should contain deuterium, lithium, beryllium, and boron only in abundances comparable to those in the material of the sun when it first formed and in the material which then constituted and still constitutes the interstellar medium. It is the thesis of this article that these abundances are considerably smaller than those observed in the earth and the meteorites and considerably smaller than those expected in the other minor planets. Predicted upper limits for the abundances in the interstellar medium and the major planets relative to abundances in the terrestrial planets and the meteorites are ≤ 50 percent for deuterium and ≤ 10 percent for lithium, beryllium, and boron. Space explorers should not find much deuterium, lithium, beryllium, or boron on Jupiter! If they do, then perhaps we have underestimated the youthful vigor of the sun.

We have seen that there is a very strong requirement that the material of the terrestrial planets became very largely separated from hydrogen at a stage when the material was condensed into bodies of metric dimensions. This requirement serves to cast considerable doubt on the validity of the theory that the terrestrial planets evolved from very much larger proto-planets in which hydrogen was present in the proportion in which it is normally present in the sun, and on the validity of the idea that the earth itself once had a strongly reducing atmosphere predominantly composed of hydrogen.

The present argument has certain biological implications as follows. Conditions during the formation of the planetesimals were highly favorable to the building of biologically interesting molecules. At that stage, H2O, NH3, and CH4 were present in high concentrations in the surrounding medium. Free hydrogen was overwhelmingly abundant, and fine solid particles presented a large surface area on which absorption processes could take place. Electromagnetic activity on the surface of the sun supplied energy of the order of 1045 ergs in the form of ultraviolet light over a period of 10⁷ years. In contrast to this biologically favorable situation, conditions seem to have become much less favorable after separation of the solid planetesimals from the gaseous component. Thereafter, oxidizing conditions appear to have replaced the former reducing conditions. These remarks apply to the subsequent formation of the earth from the planetesimals of metric dimensions. It is suggested that prebiotic or even biotic materials could have been formed in the planetesimals prior to the formation of the earth. I will not elaborate on this speculation at this time.

Summary

Space research seeks to ascertain, among other things, the history of the origin of the solar system. There already exist certain nuclear clues to this history, and additional evidence will be forthcoming from exploration of the moon and the planets.

In this article (see 13) I have discussed the present implication of the observed abundances of D^2 , Li^6 , Li^7 , Be^0 , B^{10} , and B^{11} in terrestrial and meteoritic matter. The abundances indicate that the synthesis of these light nuclei occurred during an intermediate stage in the early history of the solar system.

In this intermediate stage the planetary material had become largely, but not completely, separated from the hydrogen which was the main constituent of primitive solar material. Appropriate physical conditions were satisfied by solid planetesimals of 1- to 50-meter radius, consisting of metallic grains embedded in an icy matrix. The synthesis occurred through spallation and neutron reactions simultaneously induced in the outer layers of the planetesimals by the bombardment of highenergy charged particles, mostly protons, accelerated in magnetic flares at the surface of the condensing sun. The importance of the (n,α) reactions for Li⁶ and B¹⁰ is indicated by the relatively low abundances of these two nuclei.

Anomalous abundances of Xe^{120} and Ag^{107} observed in meteorites can be attributed to the decay of radioactive I^{120} and Pd^{107} produced in the planetesimals. The interval between the irradiation of the small planetesimals and various stages of the formation of large bodies in the solar system is found to be between 10^{6} and 10^{8} years.

Conclusion

We may conclude, then, that the isotopic compositions of hydrogen, lithium, and boron and the existence of abundance anomalies in meteorites serve as clues to a remarkable and interesting stage in the development of the solar system. It was well said: "And the Lord shewed signs and wonders \dots " (14).

To learn more, our motto must be a paraphrase of an earlier exhortation: "Spaceward ho!"

References and Notes

- 1. C. W. Snyder, S. Rubin, W. A. Fowler, C. C.
- L. G. W. Shyder, S. Rubin, W. A. Fowler, C. C. Lauritsen, *Rev. Sci. Instr.* 21, 852 (1950).
 A. Turkevich, "Chemical analysis of surfaces using large angle scattering of heavy charged particles," in preparation.
- E. M. Burbidge, G. R. Burbidge, W. A. Fowler, F. Hoyle, *Revs. Modern Phys.* 29, 547 (1957).
- 547 (1957).
 W. A. Fowler, in Modern Physics for the Engineer, L. N. Ridenour and W. A. Nierenberg, Eds. (McGraw-Hill, New York, 1961), ser. 2, chap. 9.
 F. Hoyle, W. A. Fowler, G. R. Burbidge, E. M. Burbidge, Science 124, 611 (1956).
 W. A. Fowler, G. R. Burbidge, E. M. Burbidge, Astrophys. J. 122, suppl. 2, 167 (1955).
 The reader will note that reference is made

- 6. The reader will note that reference is made

to nuclei, not to nuclides or atoms; nucleo-synthesis primarily involves nuclear, not atomic, processes. The word *isotope* is taken to refer to a nucleus, not an atom. The sym-bols used for nuclei are the same as those used commonly for the chemical elements, as follower: As always at a charge a proused commonly for the chemical elements, as follows: Ag, silver; Al, aluminum; B, boron; Be, beryllium; C, carbon; D, deuterium; Gd, gadolinium; H, hydrogen; He, helium; I, iodine; Li, lithium; Pb, lead; Pd, palladium; T, tantalum; Te, tellurium; and Xe, xenon.
7. F. Hoyle, *Quart. J. Roy. Astron. Soc.* 1, 28 (1960)

- (1960).
- (1960).
 8. W. K. Bonsack and J. L. Greenstein, Astrophys. J. 131, 83 (1960).
 9. H. C. Urey, The Planets (Yale Univ. Press, New Haven, Conn., 1952). This book gives an excellent account of the chemistry and geophysics of the origin of the solar system. For a more recent account and a more recent. For a more recent account and a more recent

bibliography, see H. C. Urey, in *Physics and* Chemistry of the Earth, L. H. Ahrens et al., Eds. (Pergamon, New York, 1957), vol. 2, Eds. p. 46.

- 10. J. H. Reynolds, Phys. Rev. Letters 4, 8 (1960). 11. V. R. Murthy, ibid. 5, 539 (1960).
- 12. R. A. Fish, G. G. Goles, E. Anders, Astrophys. J. 132, 243 (1960).
- phys. J. 132, 243 (1960).
 13. This article contains material abstracted from two articles by W. A. Fowler, J. L. Greenstein, and F. Hoyle [Am. J. Phys. 29, 393 (1961); Geophys. J., in press]. A detailed list of references may be found in these two articles. The research which served as the basis of the present article is supported in part by the joint program of the Office of Naval Research and the U.S. Atomic Energy Commission. Commission.
- 14. Deuteronomy 6:22.

INSTRUMENTS AND TECHNIQUES

Glass Electrodes Sensitive to Divalent Cations

Ion exchange models for new electrode glasses simplify analyses for calcium and other ions.

R. M. Garrels, M. Sato, M. E. Thompson, A. H. Truesdell

Glass electrodes, whose electrode response is analogous to that of pH glass electrodes, have been prepared and used to measure the concentrations of divalent cations, such as Ca++, Mg++, Ba++, Sr++, Mn++, and Zn++. Certain electrodes or combinations of electrodes can be used to determine the activities of individual divalent cations or the sum of the activities of two or more divalent cations in solutions that may contain moderate concentrations of monovalent cations. The electrode response apparently is not affected by the nature of the anions.

These electrodes may be used for measuring the hardness of drinking water, the calcium and magnesium in sea water, and the calcium in urine. It is anticipated that with some further

23 MARCH 1962

development it will be possible to measure the activities of calcium and magnesium in whole blood or serum. Measurement of one divalent cation in the presence of others may also soon become practical.

Glass electrodes for measuring pHhave been used during much of the last half century. The change in potential of the best of these electrodes in solutions of varying pH is found to be nearly identical to the change in potential measured by the standard hydrogen electrode in the same solutions. For this reason, with appropriate safeguards, it is customary formally to equate pH, as measured by the glass electrode, with $-\log a_{H^+}(1)$.

The extent of the "alkali error" of pH glass electrodes is known to be a function of the composition of the glass. Advantage was taken of this fact by Eisenman and his co-workers (2), who developed practical sodium- and potassium-sensitive glass electrodes. The response of these electrodes to monovalent cations is sufficiently close to that predicted by the Nernst equation to permit postulation of a formal relation between measured potential and $-\log a_{ion}$, as defined by the relation

$E = E^{o} + (2.303 RT/F) \log(a_{ion})$

where R is the gas constant, T is the absolute temperature, and F is the faraday.

Sensitivity of some glass electrodes to divalent cations has been noted before (3), but these glasses have strong pH functions and unreproducible responses and therefore never came into general use. In other attempts to measure divalent cation activities, particularly of calcium, use has been made of the electrical properties of other types of membrane surfaces-for example, clay membranes (4), ion-exchange resin membranes (5), and multilayer stearate membranes (6).

We chose the glasses that we tested for several reasons. Because the ion selectivity of glass electrodes is related to the differential strength of bonding of cations to the glass structure, we tried glasses close in composition to insoluble crystalline silicates-that is, natural glasses, such as obsidians or tektites.

We tested Pyrex, because it is a borosilicate glass and alkaline earth borates are much less soluble than alkali borates. We made and tested the potassium barium aluminum silicate because it was thought that a glass containing large cations might more easily accommodate divalent cations.

The experimental electrodes are glass tubes; they contain an inner solution of potassium chloride or hydrogen chloride and a chloridized silver wire that is connected by a shielded lead to an amplifier. The sensitive glass, in the form of a membrane, is at the lower end

Dr. Garrels is professor of geology at Har-vard; Dr. Sato is assistant professor of geo-chemistry at Okayama University (Misasa, Ja-pan); Miss Thompson is a research assistant in geology at Harvard; Mr. Truesdell is a geologist with the U.S. Geological Survey, Washington, D.C. At the time this article was written Dr Sato D.C. At the time this article was written, Dr. Sato was a research associate at Harvard and Mr. Truesdell was a graduate student at Harvard.