Production of Helium in Iron Meteorites by the Action of Cosmic Rays*

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The helium distribution in a slice from the iron meteorite, Grant, has been measured and plotted in the form of contour maps. The contours of constant helium show a minimum helium content and isotopic ratio, He^3/He^4, near the center of the slice, the isotopic ratio varying from 0.26 near the center to 0.30 at the surface. A cosmogenic helium production rate equation was fitted to the data giving a He^3/He^4 production ratio by primary cosmic rays of 0.50 and by secondary particles of 0.14. Primary and secondary particle interaction cross sections were found to be 540 mb and 720 mb, respectively. The ratio of the average post-atmospheric radius to the pre-atmospheric radius of Grant was calculated to be 0.65.

BAUER[1] and Huntley[2] independently proposed that cosmic rays interacting with meteorites should produce measurable amounts of helium. Bauer[1] also suggested that the He^3/He^4 abundance ratio might be quite different from that found in terrestrial helium. Evidence to support this hypothesis was obtained by Paneth *et al.*[3] who showed that, in accordance with predictions from evaporation theory,[4] the He^3/He^4 abundance ratio in iron meteorites lay in the range 0.2 to 0.3.

Inasmuch as cosmic rays are attenuated as they pass through matter, one should observe a "depth effect" if the helium at different distances from the surface of a meteorite were investigated. Paneth *et al.*[5] studied the isotopic composition as well as the absolute amount of helium found in drillings taken from two holes bored in the iron meteorite Carbo, and indeed found a depth effect. Martin[6] made a theoretical analysis of the problem. More recently Ebert and Wänke,[7] working in Paneth's laboratory, have reported on a redetermination of the helium (and neon) contents of a number of meteorites. They discussed in some detail the depth effect, and the ablation of meteorites as they pass through the atmosphere.

The purpose of the present investigation was to make a systematic study of the distribution of He^3 and He^4 in the one-half ton iron meteorite, Grant. The meteorite had a cross-sectional slice cut from it which in turn was cut into a number of bars. Sets of these bars were distributed to several laboratories for rare-gas analyses.[8] Preliminary reports on the present investigation appear elsewhere.[9,10]

APPARATUS AND METHODS

A double-focusing mass spectrometer calibrated with standard helium samples was used to determine both the absolute amount and the isotopic ratio He^3/He^4 of the helium in the gas extracted from the meteorite samples. The instrument was similar to one earlier described[11,12] for determining precise atomic masses. In order to improve the sensitivity, the slits were made wider. However, the resolution was ample to separate the He^3 and HD ion peaks.

One feature of the present instrument was a double collection system for He^3 and He^4, permitting simultaneous reading of the two ion beams. Separate vacuum systems were employed for evacuating the ion source region and the analyzer. Because the conductance between the two regions was extremely low (determined by the source slit area), samples could be analyzed by a recirculation technique. The gas was admitted to a manifold of metal vacuum valves and copper tubing and pumped continuously (by the mass-spectrometer ion-source diffusion pump) through the source region, past a titanium sponge getter, and back to the ion

TABLE I. Helium concentration in Bar B of meteorite Grant. L and R in position column refer to distances left or right from reference line scribed on slice (see Fig. 2).

Position cm L or R	He^3 (10^{-6} std. cc/g)[a]	He^4 (10^{-6} std. cc/g)	Total He (10^{-6} std. cc/g)	Ratio He^3/He^4
37.0 L	6.17	21.88	28.05	0.282
33.0 L	5.75	21.09	26.84	0.273
23.0 L	5.17	19.47	24.64	0.265
13.2 L	4.92	18.57	23.49	0.265
8.0 L	4.88	18.51	23.39	0.264
4.0 L	4.99	18.84	23.83	0.265
3.0 L	4.97	18.70	23.67	0.266
1.0 R	5.18	19.69	24.87	0.262
7.0 R	5.61	20.34	25.95	0.275
12.8 R	5.94	20.93	26.87	0.283

[a] cc of gas at 0°C and 760-mm Hg pressure per gram of meteorite.

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[1] C. A. Bauer, Phys. Rev. **72**, 354 (1947); **74**, 225 (1948); **74**, 501 (1948).
[2] H. E. Huntley, Nature **161**, 356 (1948).
[3] Paneth, Reasbeck, and Mayne, Geochim. et Cosmochim. Acta **2**, 300 (1952).
[4] K. J. LeCouteur, Proc. Phys. Soc. (London) **A63**, 259 (1950).
[5] Paneth, Reasbeck, and Mayne, Nature **172**, 200 (1953).
[6] G. R. Martin, Geochim. et Cosmochim. Acta **3**, 288 (1953).
[7] K. H. Ebert and H. Wanke, Z. Naturforsch. **12a**, 766 (1957).
[8] The authors are indebted to Dr. E. P. Henderson of the U. S. National Museum, Smithsonian Institution of Washington, D. C., who made the Grant Meteorite available and arranged for its cutting by the Battelle Memorial Institute, Columbus, Ohio.
[9] J. H. Hoffman and A. O. Nier, Bull. Am. Phys. Soc. Ser. II, **2**, 349 (1957); Ser. II, **3**, 221 (1958).
[10] J. H. Hoffman and A. O. Nier, Program of the Thirty Ninth Annual Meeting of the American Geophysical Union, 1958 (unpublished).
[11] A. O. Nier and T. R. Roberts, Phys. Rev. **81**, 507 (1951).
[12] A.O. Nier, National Bureau of Standards Circular No. 522 (U. S. Government Printing Office, Washington, D. C., 1953), p. 29.

source. In this manner He^3 samples as small as 10^{-10} cc (NTP) could be detected. The useful sensitivity for He^4 was not as favorable due to an in-leakage of approximately 2×10^{-9} cc (NTP)/min of atmospheric helium as a result of the permeability to helium of the Pyrex parts of the vacuum system. Approximately 3% of the sample was lost to the analyzer region per minute and pumped away.

To extract the gas from an iron meteorite, a sample of about 0.150 g was loaded in an alumina crucible inside a tantalum cylinder and heated in vacuum by induction heating. The gas thus released was pumped continuously by a metal oil diffusion pump into the titanium getter in the spectrometer recirculation line where it was purified and stored until the heating cycle was completed. The helium was then recirculated through the mass-spectrometer ion source and the He^3 and He^4 peak heights were measured on a two-channel recording potentiometer.

In practice, 85 to 98% of the helium was extracted in a single heating cycle. A second or third heating cycle was performed until the entire sample was evaporated. It is believed that at least 99% of the helium was extracted from the sample.

To calibrate the mass spectrometer, a standard containing known amounts of He^3 and He^4 was run immediately after and in the same manner as the unknown. The standard was prepared as follows: known amounts of pure He^3 and He^4 were mixed to give a stock supply of helium having an isotopic composition approximately equal to that found in meteorites. A standard volume (1.05 cc) was filled to a pressure of several cm Hg with the standard helium mixture. By a series of four expansions into known volumes the original quantity of helium was reduced to a known amount near 3×10^{-6} cc (NTP) which was then admitted to the recirculation line of the mass spectrometer and measured.

To determine whether all the helium released by a sample was collected and measured, the following experiment was performed. A small copper tube of about 0.3 cc volume was filled with a quantity (known to approximately 1%) of helium and sealed. This was placed in the alumina crucible and subjected to the same procedure as a meteorite sample. The measured amounts of He^3 and He^4 both agreed to within 2% of the amounts sealed into the tube.

In order to determine the atmospheric helium in-leakage under actual operating conditions, blank runs were made periodically using empty alumina crucibles, and a correction made. In a typical meteorite sample, the correction amounted to from 1 to 5% of the He^4.

From these tests and those of the standards, it is believed that all the meteorite analyses were made to an accuracy of 3% in the relative quantities of He^3 and of He^4 per gram of meteorite. The absolute accuracy of the helium content measurements is approximately 5%.

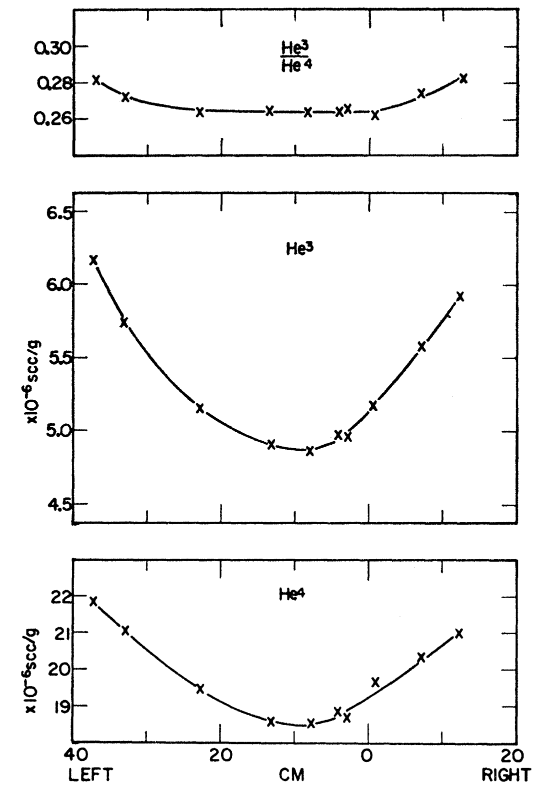

FIG. 1. Helium concentration in Bar B of Grant meteorite. Distances are with respect to reference line scribed on slice (see Fig. 2).

RESULTS

The Grant meteorite is a 1060-pound fine octahedrite iron meteorite found in 1929 in the Zuni Mountains, forty-five miles south of Grant, New Mexico. It was cut in half and a slice 1 cm thick was removed (parallel to the cut surface) from one of the halves. The slice was cut into a number of parallel bars. Those marked B, F, J, N, and R were made available to us for the present research. Helium analyses were made on samples from various points along each of these bars, and the results from a typical bar, Bar B, are shown in Table I and plotted in Fig. 1. Distances are referred to a line scribed on the slice perpendicular to all the bars. (See Fig. 2 for reference line and bar positions.)

Contours of constant He^3 and constant He^4 were drawn from the helium-content data, the results being the contour maps shown in Fig. 2 and Fig. 3. These contour maps indicate that the minimum helium content and minimum isotopic ratio in the meteorite slice both lie near the center and increase monotonically towards each surface. It should be pointed out that these contours show only a two-dimensional picture of the helium distribution. The slice was cut so as to pass through the center of mass of the post-atmospheric meteorite. While this may not coincide with the pre-atmospheric center of mass or the "radiation center" of the meteorite, it is probably a fairly close approxima-

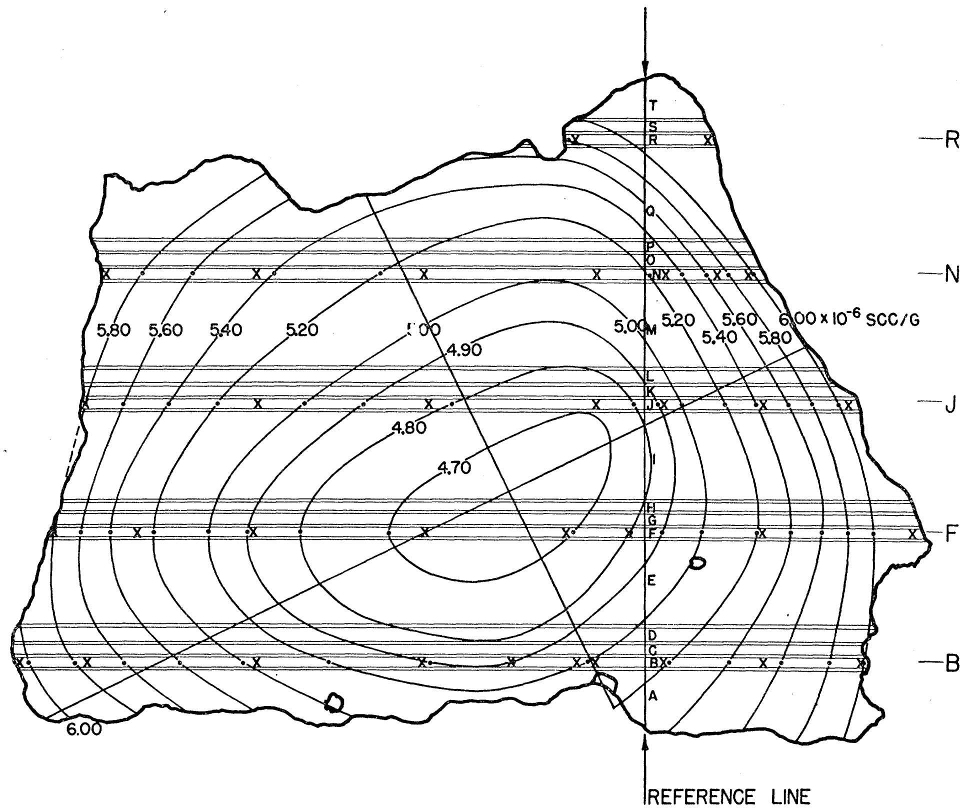

FIG. 2. Contours of constant He^3 in Grant meteorite slice. Sets of horizontal lines indicate bars into which slice was cut. Crosses (×) are points at which helium analyses were actually made. Dots (•) are interpolated helium concentrations taken from graphs such as shown in Fig. 1. Diagonal lines indicate directions along which radial helium concentration averages were made.

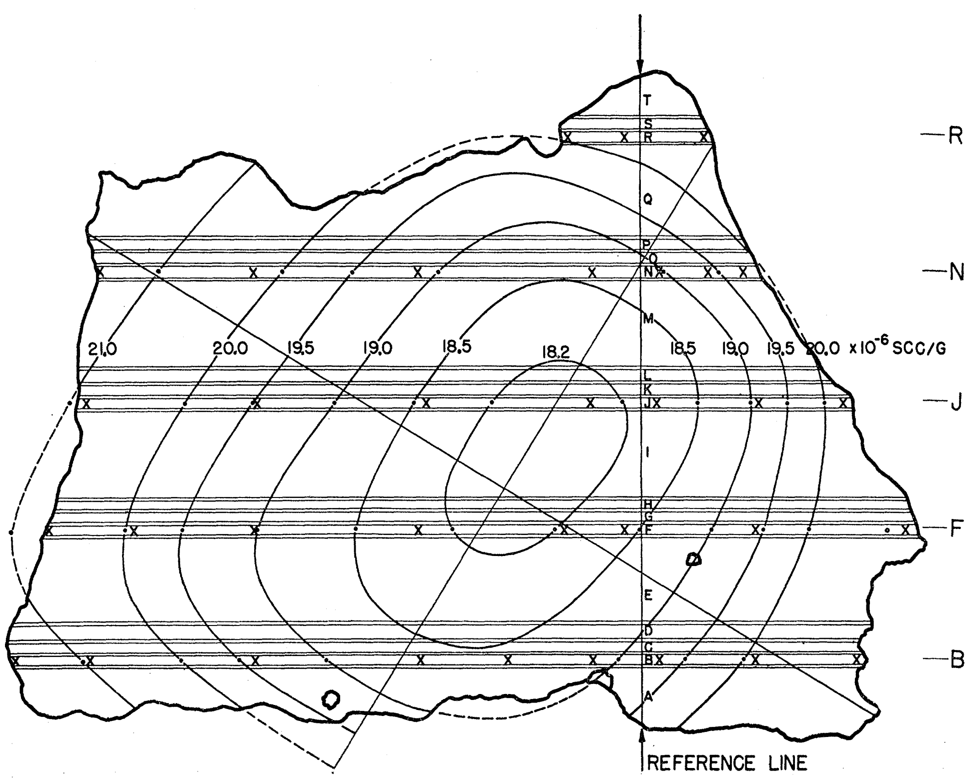

FIG. 3. Contours of constant He^4 in Grant meteorite slice.

tion since in the slice itself the point of minimum helium content lies near the geometric center. If ever samples become available from another plane of the meteorite, this question could be settled.

In order to interpret the experimental data, a comparison was made with an equation representing the production of helium by cosmic rays. Parameters appearing in the equation were evaluated by fitting the equation to the contour data, and the results were compared with data obtained from other types of experiments. To facilitate the evaluation of the parameters in the equation, the average radial He^3 and He^4 contents and the He^3/He^4 ratio were plotted (in effect the contours were thus replaced by "equivalent" spheres). The results of this averaging appear as the solid portions of the curves in Fig. 4. The dashed portions will be explained later.

Helium is produced in meteorites by the interaction of cosmic-ray particles with the nuclei of the meteorite material. When a nucleus is struck by a very high-energy particle (cosmic-ray proton), a large quantity of energy can be transferred to the nucleus by the creation and reabsorption of π mesons in the nucleus.[13] A few very high-energy nucleons or π mesons will escape from the struck nucleus. These are knock-on particles and are seen as minimum-ionizing tracks in nuclear emulsions. Their multiplicity is about 0.2 times the primary flux.[14] Since their energy is in the range of primary cosmic-ray energy, they will be considered to interact in a meteorite in the same manner as primary cosmic-ray particles. Let σ_p be the total interaction cross section of primary cosmic rays with matter. Then the effective absorption cross section, σ_a, of the primary plus the high-energy knock-on particles will be smaller than the primary interaction cross section. Thus, $\sigma_a=\sigma_p/1.2$.

Lower energy nucleons and π mesons will also be emitted (grey tracks in emulsions) with a multiplicity of about 3 per primary incident particle.[14] These are termed secondaries and have an interaction cross section σ_s. It is assumed here that all secondary particles are emitted in the forward direction with respect to the primary particles. While Camerini *et al.*[14] have shown that there is a finite backscatter, the gray tracks in emulsions are strongly oriented in the forward direction. Since no data are available for this phenomenon in iron and the backscatter is small in emulsions, we will neglect it here. In this analysis, both σ_a and σ_s are considered constant over the energy ranges of primaries and secondaries, respectively, since actually they do vary only slowly with energy.

After a nucleus has been highly excited and the high-energy secondary particles have escaped and a near equipartition of energy has been achieved, the nucleus

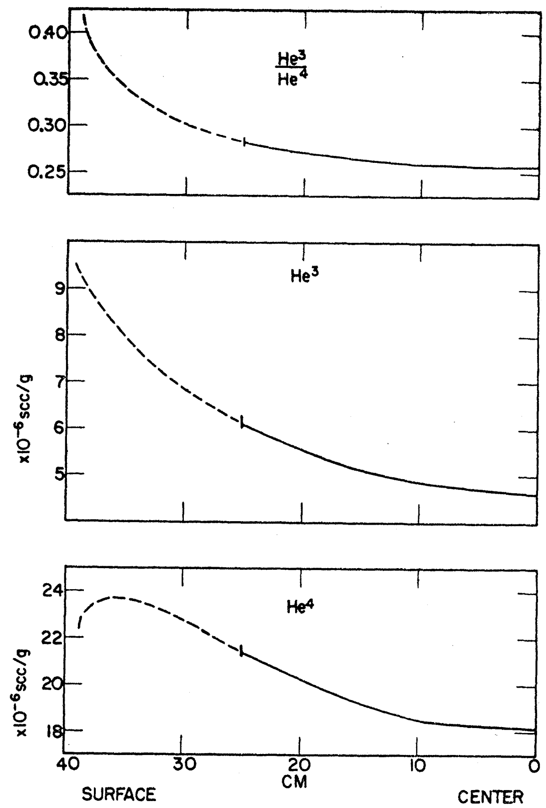

FIG. 4. Average radial helium distribution in Grant meteorite slice. Solid portions of curves obtained from contour maps shown in Fig. 2 and Fig. 3. Entire curves (solid plus dashed) are plots of helium production equation fitted to experimental results.

may dissipate its energy by either spallation or fragmentation. The former process consists of evaporation of light-mass particles such as n, p, d, t, He^3 and He^4. This process accounts for most of the cosmogenic He^4 and some of the He^3 produced in a meteorite. He^3 may also be produced by knock-on nucleons picking up two additional nucleons and subsequently being emitted as He^3 or H^3. The residual nuclei after the evaporation process is completed are called the spallation products. These may be any isotope of any element lighter than iron and include neon and argon which have also been measured in meteorites.[15–17]

Under the assumption that a unidirectional beam of primary cosmic rays incident normally on a semi-infinite meteorite will be attenuated exponentially and that its energy spectrum is invariant with penetration depth, an equation which represents the He^3 production rate at a distance l below the surface may be written

[13] Wolfgang, Baker, Caretto, Cumming, Friedlander, and Hudis, Phys. Rev. **103**, 394 (1956).

[14] Camerini, Davies, Fowler, Franzinetti, Murhead, Lock, Perkins, and Yekutieli, Phil. Mag. **42**, 1241 (1951).

[15] P. Reasbeck and K. I. Mayne, Nature **176**, 733 (1955).

[16] W. Genter and J. Zähringer, Geochim. et Cosmochim. Acta **11**, 60 (1957).

[17] R. Bieri, Bull. Am. Phys. Soc. Ser. II, **3**, 221 (1958); also private communication.

TABLE II. Summary of numerical quantities involved in helium-production analysis of Grant meteorite.

$r/R=0.65^a$	
$r=25$ cm[b]	
$R=39$ cm	
$\mu_a=0.039^a$	$\sigma_a=450$ mb
$\mu_p=0.046$	$\sigma_p=540$ mb
$\mu_s=0.062^a$	$\sigma_s=720$ mb
$M_{He^3}/M_{He^4}=0.50^a$	$m_{He^3}/m_{He^4}=0.14^a$
$M_{He^3}=1.1$	$\sigma_{pp3}=600$ mb[c]
$M_{He^4}=2.2$	$\sigma_{pp4}=1200$ mb
$m_{He^3}=0.14$	$\sigma_{ps3}=120$ mb
$m_{He^4}=1.0$	$\sigma_{ps4}=850$ mb

[a] Determinations from curve fitting process (Fig. 4).
[b] Average radius taken along diagonal lines in Fig. 2.
[c] Extrapolated from data in reference 18. See text. The other numbers were calculated from the items designated by a, b, and c.

as follows[7]:

$$P_{He^3}=M_{He^3}\mu_p I_0 \exp(-\mu_a l) + m_{He^3}\frac{\mu_s}{\mu_s-\mu_a}S\mu_p I_0[\exp(-\mu_a l)-\exp(-\mu_s l)], \quad (1)$$

where M_{He^3}= multiplicity of He^3, i.e., the number of He^3 particles (including H^3) produced per primary interaction; m_{He^3}= the number of He^3 particles (including H^3) produced per secondary interaction; I_0= primary cosmic-ray intensity in space, and S= number of secondary particles emitted per primary interaction. Also $\mu_j=(N\rho\sigma_j)/A$, where N= Avogadro's number, A= average atomic weight of meteorite material, ρ= density of meteorite material, and $j=a, p, s$. The first and second terms of Eq. (1) represent the He^3 production rate by primary and secondary particles, respectively. A similar equation exists for He^4, the only difference being that the multiplicities for He^4 are written in place of those for He^3.

Equation (1) was derived assuming a unidirectional flux. Since the cosmic-ray flux is actually omnidirectional and the Grant meteorite radius is of the same order of magnitude as the cosmic-ray interaction length in iron, the helium production rate at a given point inside the meteorite is found by integrating Eq. (1), recognizing that cosmic rays reaching this point come from all possible directions. In performing the integration, the assumption is made that the pre-atmospheric meteorite could be approximated by a sphere. This assumption appears reasonable because the contours are approximately circular.

By adjusting the values of the parameters in the integrated form of Eq. (1), the equation was made to fit the He^3 data curve; likewise the He^4 form of the equation was made to fit the He^4 data curve of Fig. 4. From this curve-fitting process, values for μ_a, μ_s, and the ratios of the helium multiplicities were obtained. These are shown in Table II. Also obtained was the ratio of the post- to pre-atmospheric radius of the meteorite, which ratio, r/R, turned out to be 0.65. The average post-atmospheric radius is 25 cm, thus the pre-atmospheric radius, R, is 39 cm. The dashed portions of the curves of Fig. 4 show the radial helium distribution in the region ablated during passage through the atmosphere. Thus, the complete (dashed plus solid) curves give the helium distribution in the pre-atmospheric meteorite.

In Table II, $M_{He^3}/M_{He^4}=0.50$ is the ratio of He^3/He^4 (including decayed tritium) produced by primary cosmic rays in the meteorite. It must be pointed out that in deriving Eq. (1), it was assumed that this ratio is a constant over the energy range of primary cosmic rays (several Bev), and thus this number represents the average value of the He^3/He^4 ratio over this energy range. Likewise $m_{He^3}/m_{He^4}=0.14$ is the average value of the He^3/He^4 ratio (including decayed tritium) produced by secondary particles, again assumed a constant over the secondary energy range (several hundred Mev). It is reasonable to expect from a consideration of the processes by which helium is produced that the He^3/He^4 ratio should be higher at higher energy, since a fraction of the He^3 may be produced by a knock-on process which is thought to be quite energy-dependent, the amount of He^3 produced increasing with energy. Also since the α particle is a more tightly bound structure than He^3 or H^3 it would be relatively more abundantly evaporated at lower energy. Both these processes act to give a He^3/He^4 ratio higher for primaries than for secondaries.

The value 0.5 for the ratio He^3/He^4 produced by primaries will now be considered in view of other experimental data. Since the Grant meteorite size is of the same order of magnitude as the cosmic-ray interaction length in iron, at every point in the meteorite (including the pre-atmospheric surface) some of the helium will have been produced by primaries and some by secondaries. Since it is known from the work of Schaeffer and Zähringer[18] that lower energy protons (few hundred Mev) hitting an iron target produce a He^3/He^4 ratio (including decayed tritium) of 0.1 to 0.2, the primary cosmic-ray He^3/He^4 production ratio must be greater than the largest measured helium isotope ratio in Grant. This latter is 0.30. Also, Fig. 4 shows this ratio reaches 0.42 at the pre-atmospheric surface. Thus, the primary ratio of 0.50 seems reasonable from these considerations.

In comparing the results obtained for targets exposed to proton beams with those found for meteorites, allowance must be made for the fact that in a meteorite the He^3 found includes the decayed H^3 as well as the directly produced He^3; in target experiments, on the other hand, measurements are usually made in a time short compared with the half-life of H^3 (12 years) so that the He^3 found does not include any appreciable amount of decayed H^3.

[18] O. A. Schaeffer and J. Zähringer, Z. Naturforsch. **13a**, 346 (1958).

The literature gives two different values for the He^3/H^3 ratio produced by protons in iron. Martin *et al.*[19] give 0.6 for 340 Mev protons. More recently Schaeffer and Zähringer[18] report 1.4 at 430 Mev and 2.4 at 3 Bev. The latter also measured the He^3/He^4 ratio for protons of several different energies and found this ratio to increase fairly rapidly with energy, having a value of 0.18 at 3 Bev, the highest measured energy.

If one accepts the target He^3/He^4 ratio of 0.18 and attempts to calculate a He^3/He^4 ratio, which includes decayed tritium, one finds a value of 0.25 if a He^3/H^3 ratio of 2.4 is used and a value of 0.39 if Martin's value extrapolated to higher energies,[7] 0.85, is employed. The presently calculated value of 0.5 is not inconsistent with either of these results if the He^3/He^4 production ratio increases rapidly with energy beyond 3 Bev.

Bieri,[17] on the other hand, measured the He^3/He^4 ratio in a copper target exposed to 6 Bev protons and found a value of 0.17. The He^3/He^4 ratio which includes decayed H^3 will then be 0.24 for a He^3/H^3 ratio of 2.4 and 0.37 for a He^3/H^3 ratio of 0.85.

Since results on various meteorites as well as the analyses in the present paper point to a He^3/He^4 ratio (including decayed H^3) of at least 0.35 for protons of cosmic-ray energy on iron, it is obvious that more determinations of the He^3/H^3 and He^3/He^4 ratios are required for targets exposed to protons in the cosmic-ray energy range in order to clarify the picture.

From the curve-fitting process discussed earlier, only ratios of the multiplicities were obtained. In order to find absolute magnitudes of these quantities, one of them was evaluated from other data. Unfortunately, lack of knowledge of the cosmic-ray energy spectrum in space makes it somewhat difficult to obtain such a number. Schaeffer and Zähringer[18] have reported the (He^3+H^3) production cross section, σ_{pp3}, in iron by protons at several energies. They found that this cross section rose fairly rapidly with energy, reaching a value of 340 mb at 3 Bev. If one accepts these data, it appears reasonable to expect that σ_{pp3} will continue to rise with increasing energy and reach a value of perhaps 600 mb at the average energy of primary cosmic rays.

From the value of μ_p given in Table II, σ_p was calculated to be 540 mb. Hence $M_{He^3}=\sigma_{pp3}/\sigma_p=600$ mb/540 mb$=1.1$. The other three multiplicities are $M_{He^4}=\sigma_{pp4}/\sigma_p$, $m_{He^3}=\sigma_{ps3}/\sigma_s$, and $m_{He^4}=\sigma_{ps4}/\sigma_s$, where σ_{pp4} is the He^4 production cross section by primaries, σ_{ps3} is the He^3 production cross section by secondaries, and σ_{ps4} is the He^4 production cross section by secondaries. These quantities could then be calculated, the results appearing in Table II.

It is interesting to compare the helium multiplicities obtained for Ag and Br from cosmic-ray emulsion experiments with those calculated here for Fe. In emulsions the distinction is made only between charge 1 and charge 2 particles and not the isotopes of hydrogen and helium. Therefore we must calculate the He^3 (not including H^3) plus He^4 multiplicity. If one assumes that the He^3/H^3 ratio of 2.4 reported by Schaeffer and Zähringer reaches a value of about 3 at primary cosmic-ray energies, the He^3 production cross section (not including H^3) will be 450 mb and the multiplicity (without H^3) will be 0.8. From Table II $M_{He^4}=2.2$. Hence the total helium (charge 2) multiplicity is about 3. This is in good agreement with the value of the order of 3 to 4 from emulsion data.[20]

The secondary interaction cross section resulting from the curve fitting process is 720 mb. If it is assumed that this is the geometric cross section for an iron nucleus, $\pi(r_0A^{\frac{1}{3}})^2$, then $r_0=1.0\times10^{-13}$ cm. This lies near values of this constant as measured by other types of experiments.

Since the amount of helium found in a meteorite which is free of uranium and thorium is determined by the cosmic-ray intensity to which the body was exposed, integrated over time, one can in principle calculate an age for the Grant meteorite.[21]

ACKNOWLEDGMENTS

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[19] Martin, Thompson, Wardle, and Mayne, Phil. Mag. **45**, 410 (1954).

[20] E. P. Ney (private communication).

[21] This age, which is an exposure time to cosmic rays, depends on some factors which are not well established. One assumes that the space and time average of the cosmic rays to which the meteorite was exposed was the same as that measured near the top of our atmosphere at the present time, about 0.25 particle/cm^2-steradian. [J. A. Van Allen and S. F. Singer, Phys. Rev. **78**, 819 (1950); Meredith, VanAllen, and Gottlieb, Phys. Rev. **99**, 198 (1955)]. Further, values for the primary helium production cross sections which were used in the age calculations could be considerably in error since they were obtained by extrapolation from measurements that have been made at energies below those of average cosmic rays; moreover, there is considerable uncertainty in the actual energy distribution in cosmic rays. Finally, as previous sections indicate, the production cross sections by secondary particles are not known to any high degree of accuracy. On the other hand, loss of helium by diffusion apparently does not introduce an appreciable error in view of the fact that the highest helium concentration is found at the surfaces rather than the interior of the meteorite body. The "age" calculated from the present data is then 0.6×10^9 years. The value should be accepted with some reservations until the uncertainties mentioned have been clarified.