Radiation Age of a Meteorite from Cosmic-Ray-Produced He^3 and H^3†

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The tritium and He^3 contents of the Norton County stone meteorite were measured. At the time of fall (1948) the tritium activity of two different specimens were (0.28 ± 0.02) and (0.25 ± 0.02) disintegrations min^{-1} g^{-1}. The measured He^3 contents per gram were (2.27 ± 0.11) and $(2.35\pm0.11)\times10^{-6}$ cc STP respectively. Experiments were made which indicate that there has been no appreciable loss of He^3. The amount of He^3 accumulated and the tritium production rate combined give apparent He^3-H^3 ages for irradiation of 420 and 480 million years respectively. If an assumption is made as to the direct production rate of He^3 by spallation, these ages reduce to 240 and 280 million years. The $A^{40}-K^{40}$ age of this meteorite is 4400^{+640}_{-740} million years. The possible significance of this difference in age is discussed.

INTRODUCTION

IN recent years much interest has been given to the spallation products produced in meteorites by cosmic radiation.[1–12] On the basis of the observed contents of He^3 and neon attempts have been made to estimate the length of time the meteorites were exposed to cosmic radiation. These calculations are hampered by the facts that (a) the cross sections for the production of He^3 and Ne are not well known, and (b) the actual irradiation rate at the site in question is difficult to evaluate. Both uncertainties are partly due to the fact that the meteorites change their size and shape considerably during their passage through the atmosphere.

These difficulties can be overcome, however, in such cases where it is possible to measure the production rate of a certain isotope as well as the amount accumulated. Probably the spallation product where such measurements are most promising is He^3 because (a) the amounts accumulated can be measured with high accuracy and do not have to be corrected for any original He^3 content of the meteorite, and (b) the production rate of tritium is high enough to be determined by standard low level counting techniques. The tritium production rate allows the total He^3 production rate to be calculated if corrections are applied for the amount of He^3 produced directly.[13]

EXPERIMENTAL TECHNIQUES

Aliquots of the powdered meteorite samples were placed in a Ni crucible and heated by induction in order to extract the gases. In such cases where a He^4 tracer and/or H_2 carrier were used they were added before the heating was started. Where necessary purified CO_2 was added to maintain a minimum pressure of a few mm Hg in order to prevent a gas discharge.

The evolved gases were passed through CuO at 600°C and the condensable part frozen out with liquid nitrogen. This process was repeated until no further decrease of the pressure was detectable. The remaining amount of gas, always less than 1% of the initial, was exposed to hot Ca and to a tungsten wire at about 2000°C. After that it was transferred by means of a Toepler pump into a sample tube and sealed off.

The CO_2 and H_2O from the condensed fraction were carefully separated by repeated fractional distillation at dry ice temperature. The water was converted to hydrogen with a mixture of CaO and Zn dust at 550°C,[14] the evolved gas passed through a liquid nitrogen trap and transferred into a counter (850 cm^3) with a Toepler pump. 4 cm of argon and 2 cm of ethylene were added and the mixture was counted inside a ring of anticoincidence counters surrounded by an 8-inch iron shield. The background of this counter with hydrogen prepared from essentially tritium-free water by the same process as mentioned before was (6.6 ± 0.1) counts/min. The given error includes the statistical error and shows the reproducibility of the background measurements with hydrogen prepared in different runs.

The He samples were analyzed with a 12-inch radius,

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TABLE I. Results of the tests for completeness of extraction.

Sample	Run	Sample weight g	Temp deg C	Time of heating hours	Carrier cc STP H_2	Flux $Na_2B_4O_7$ g	H^3, counts per min	H^3 at time of fall dis/min g	He^3, 10^{-6} cc STP/g
Norton Co.	18	49.60	1050	7	3	...	8.0 ±0.8	0.26 ±0.03	...
Norton Co.	34	29.46	1300	7	3	...	5.38±0.3	0.296±0.015	2.24±0.11
Norton Co.	43	20.09	1150	7	...	25	1.73±0.16	0.139±0.015	...
Norton Co.	48	18.836	1200	7	30	25	2.83±0.24	0.245±0.02	2.29±0.11
Furnas Co.	55	21.307	1200	7	3	...	3.03±0.25	0.236±0.02	2.35±0.11
Furnas Co.	57	31.50	1400	7	20	...	4.91±0.20	0.255±0.015	...

60° single-focusing mass spectrometer. Ions were produced by electron bombardment and the resolved ion beam measured by an electron multiplier. The amount of He^3 was determined by isotopic dilution. Corrections for discrimination in the mass spectrometer were made by comparison with mixtures of He^3 and He^4 of known composition.

EXPERIMENTAL RESULTS

The results of the various tests for completeness of extraction are given in Table I. These experiments were performed on aliquots of samples of the Norton County achondrite.

The fact that within the limits of error the H^3 yield is the same when heating the sample without flux to 1050°C and 1400°C (with and without carrier) indicates this yield to be 100%.

As a further check, an attempt was made to extract the tritium by fusing the sample with $Na_2B_4O_7$. The low yield in case of run No. 43 is believed to be due to absorption of tritium in the white deposit on the cooled jacket above the crucible, formed during the reaction of the sample with the flux material. Although this effect can be eliminated almost completely by adding H_2 as carrier (run No. 48), heating the sample without flux to a temperature below the melting point still seems to be the most favorable method for the extraction of tritium from the stone meteorite for two reasons.

(a) Keeping the temperature below the melting point avoids the formation of sublimates which also occur when melting the stone without flux.

(b) The difference in the diffusion constant at temperatures just above and just below the melting point should be more than compensated by the increase of the diffusion length due to melting.

The He^3 yield appears to be 100%, independent of the method of extraction. Table II gives the final results for the different meteoritic samples.

The He^3-H^3 ages given in column 5 of Table II are obtained by dividing the total number of He^3 atoms by the number of tritium atoms decaying per time unit. Taking into account that some of the He^3 is produced directly, these ages have to be corrected. In column 6, the corrected ages are given, assuming a value of 0.7 for the ratio of the spallation production rates, He^3 to H^3.[13]

In the sample of the Marion chondrite (21.0 g) which fell in 1847 and thus should contain no tritium unless small amounts are still produced after the fall of the meteorites, no tritium could be detected. This is somewhat in contradiction to the finding of Fireman and Schwarzer[10] who found about equal amounts of tritium in terrestrial material (iron) and their most recent meteorite (Para de Minas, fell 1934).‡

DISCUSSION

The difference in the He^3-H^3 ages of the two specimens from the Norton County meteorite are not regarded to be significant as the experimental values are almost within the limits of error. Moreover, the ratio of the production rates of He^3 and H^3 will be dependent on the energy of the particles causing spallation, and thus be different for specimens from different locations in the same meteorite, depending on the shielding. This uncertainty, however, does not influence the results seriously because this ratio certainly has to be between zero and one, thus leading to a possible range of the He^3-H^3 age of $(200\text{–}500)\times10^6$ years. This would even be true if all the He^3 were produced by the process $Li^6(n,\alpha)H^3$. Taking for the lithium content of the Norton County meteorite the highest value given by Fireman and Schwarzer for achondrites[10] and making a reasonable assumption for the slow-neutron flux in the meteorite, the possibility of a considerable contribution of He^3 from this process can be excluded. Therefore, the only available experimental value for the He^3-H^3 production ratio by spallation, obtained with 340-Mev protons on iron,[13] was used to calculate the most probable He^3-H^3 ages (Table II, column 6).

The most striking result is that for the same specimen of this meteorite the He^3-H^3 age is more than a factor of ten lower than the $A^{40}-K^{40}$ age. A trivial explanation for this difference would be a preferential loss of He^3 compared with A^{40}. In order to investigate this possibility an aliquot of the Norton County sample was heated for two hours at 700°C and the evolved gases

‡ *Note added in proof.*—Dr. Fireman pointed out to us that, in agreement with our result, no tritium was detected in their oldest meteorite (Charcas, 1804). He considers the use of terrestrial material for background determinations—as was implied here—to be misleading.

TABLE II. Final results for the various meteoritic samples.

	Tritium counts/min g measured	Tritium dis/min g at time of fall	He^3, 10^{-6} cc STP/g	He^3-H^3 age [10^6 years] Uncorrected	Corrected for direct prod	A-K age 10^6 years[a]
Norton County[b]	0.175±0.012	0.28±0.02	2.27±0.11	420	240	4400^{+640}_{-740}
Furnas County[b]						
stone fraction	0.156±0.012	0.25±0.02	2.35±0.11	480	280	...
metal fraction	...	...	1.0 ±0.1	...	...	...
Marion[c]	<0.01	...	...	...	...	...

[a] See reference 11.
[b] Norton County and Furnas County are two specimens of the same fall. Date of fall: February 18, 1948.
[c] Stone meteorite (Chondrite). Date of fall: February 25, 1847.

were taken out. Then the sample was completely reacted with $Na_2B_4O_7$ flux at 1200°C and a second gas sample removed. The first fraction contained about 75% of both the A^{40} and He^3. This result shows that a heating of the meteorite to a temperature of 700°C or more would influence both gases equally. However, the possibility of a preferential loss of He^3 at lower temperatures over a long period of time cannot be excluded on the basis of this experiment.

With this possibility in mind, the He^3 content of a sample of several small metal inclusions from the Furnas County specimen was measured. Unfortunately, the small amount of this sample (1 g) precluded a measurement of the tritium content. As can be seen from Table II, the amount of He^3 found in the metal phase is only one-half that found in the stone phase. Diffusion losses, however, certainly would cause a greater loss in the stone phase than in the metal phase.

The metal and stone phases were intermingled. Thus the irradiation of both phases must have been the same. As the dimensions of the metal inclusions were large compared to the range of the H^3 and He^3 atoms produced by irradiation, the amount of He^3 found in the metal phase cannot have been influenced seriously by its smaller stopping power compared with that of the stone phase. The He^3 which was found must have been produced locally so that the amounts obtained from the stone and iron phases should be proportional to the cross sections per gram. Using the measured cross sections for tritium production in various elements[12,15,16] and the chemical composition of the stone phase of the Norton County meteorite as given by Beck and LaPaz,[17] a calculation shows the cross sections per gram for the stone phase to be twice that of iron. If one assumes that the ratio of the rates of direct production of He^3 and H^3 does not change appreciably between $Z=28$ and $Z=8$, this is in agreement with the results given in Table II.

CONCLUSION

The difference between the He^3-H^3 and the $A^{40}-K^{40}$ age of the Norton County achondrite appears to be real and not due to preferential loss of He^3. This low He^3-H^3 age can be explained in several ways:

1. The cosmic radiation has not been constant for the past 4400×10^6 years; it has not had its present intensity for more than about 500×10^6 years.
2. The cosmic radiation has not varied greatly with time (C^{14} dating experiments have indicated that it has been roughly constant for the past 10^4 years[18,19]). Thus the He^3-H^3 age defines the time at which the breakup of a much larger body produced this meteorite.

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