and possibly photosynthetic life back to more than 2.7 billion years, an age in keeping with that suggested by the stromatolites of the Bulawayan limestones in South Africa (3, 12) and more than 0.7 billion years greater than that of the Gunflint Iron Formation (13).

> PRESTON E. CLOUD, JR., JOHN W. GRUNER, HANNELORE HAGEN

Department of Geology and

Geophysics and School of Mineral and Metallurgical Engineering, University of Minnesota, Minneapolis

References and Notes

- S. S. Goldich and others, *Minn. Geol. Surv. Bull.* 41, 5, 42-43, 51-52 (1961).
 D. Anderson, Ph.D. thesis, University of
- D. Anderson, Ph.D. thesis, University of Minnesota, March 1965; G. N. Hanson, Minnesota Geological Survey, unpublished. We are indebted to these gentlemen for permission to quote their data.
- permission to quote their data.
 3. L. O. Nicolaysen, in *Petrographic Studies:* A Volume in Honor of A. F. Buddington,
 A. E. J. Engel et al., Eds, (Geological Society of America, New York, 1962), pp. 569–598; see pp. 574–575.
 4. All samples here reported on were obtained from a carbonaceous lenge in the lower national sectors.
- from a carbonaceous lens in the lower part of the Soudan Iron Formation, 21st level of the Soudan Mine, Soudan, Minn. The exact location is 545 m below ground, directly beneath the center of NW¹/4, NW¹/4, SW¹/4, sec. 27, T62N, R15W. Locality X of 9 December 1963. For guidance and assistance underground in the Soudan Mine, Cloud is indebted to George Nemanich of Soudan, Minn. He is also grateful to U. W. Hella, director, Divi-sion of State Parks, Minnesota Department of Conservation, for permission to go under-ground and for authorizing the cooperation

of Nemanich in entering a closed-off area of

- the mine. B. M. French, *Science*, **146**, 917 (1964). Our treatment consisted of boiling for an hour in concentrated HF, followed by boiling 5. B. for another hour in a 50-percent solution of HCI.
- 6. For the analytical data, and for permission to quote, we are indebted not only to the analysts but also to F. S. Grimaldi and analysts Irving May who arranged for performance of these analyses in the laboratories of the
- of these analyses in the laboratories of the U.S. Geological Survey. W. G. Meinschein, J. Oró, M. Calvin, tele-phoned reports to Cloud. The occurrence of pristane and phytane has subsequently been reported in a paper from Calvin's laboratory [T. Belsky, R. B. Johns, E. D. McCarthy, A. L. Burlingame, W. Richter, M. Calvin, Nature 206 446 (1965)] 7. Berlingame, W. Bichter, M. Calvin, Nature 206, 446 (1965)].
 H. G. Thode and J. Monster, letters of 8 and 11 February 1965 to Cloud.
 J. F. Machamer, "Geology and origin of
- the iron ore deposits of the Zenith Mine, Ely, Minnesota," Minn. Geol. Surv. Spec.
- Ely, Minnesota," Minn. Geol. Surv. Spec. Publ., in press. E. G. Ehlers, J. M. Schopf, D. V. Stiles, J. D. Birle, "Fossil iron bacteria preserved in pyrite," in preparation. Information and authorization to use and publish by courtesy
- authorization to use and publish by courtesy of J. M. Schopf.
 11. 15 g KClO₃ + 150 ml H₂O + 300 ml concentrated HNO₃.
 12. A. M. Macgregor, Geol. Soc. So. Africa Trans. 43, 9 (1941); 54, xxvii (1952).
 13. E. S. Barghoorn and S. A. Tyler, Science 147, 563 (1965). P. E. Cloud Jr. Science 148. 563 (1965); P. E. Cloud, Jr., Science, 148,
- 27 (1965); P. E. Cloud, J., Science, 146, 27 (1965). Research leading to this publication was done under NSF grant GP-1807. The miner-alogy and microscopic organization of the 14. material has been investigated at the University of Minnesota by us. It was originally hoped that the results of all related studies might be coordinated in a single publication or group of simultaneously published papers to which this paper would be introductory. That not being feasible, would be infroducion. Servations, together with some previously unpublished data from others, in order that this information may be available for sub-sequent reference as appropriate.

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Cosmogenic Radionuclides in the Bondoc Meteorite

Abstract. Long-lived cosmogenic radionuclides were measured in the stone phase and in a mechanically separated metallic nodule from a fragment of the Bondoc mesosiderite. Activity levels of the various radionuclides in both phases, along with results of mass-spectrometric measurements of rare-gas isotopes in the stone phase, indicate that heavy shielding was the chief cause of the low specific activities observed.

The discovery and some of the properties of the Bondoc meteorite, a mesosiderite of unusual structure, have been discussed by Nininger (1). Measurement of its Al²⁶ content gave 5.0 ± 1.7 disintegrations per minute per kilogram (dpm kg⁻¹), about a factor of 10 lower than that in the average chondrite. The low Al²⁶ activity can be explained by one or more of three causes-namely, short period of exposure to cosmic rays, great terrestrial age, or shielding.

In order to better resolve these possibilities we have measured a number of cosmogenic radionuclides both in a metallic nodule and in the remaining silicate phase of the Bondoc meteorite, which was purchased from the American Meteorite Laboratory. The results are given in Table 1.

The fact that both the metallic nodule and the remaining phase contain Mn⁵³ activity ($T_1 = 2 \times 10^6$ years) in reasonable amounts precludes a short cosmic-ray exposure period of 4 to 10 \times 10⁴ years. Manganese-53 activity per unit mass of iron (the most probable target for production) is 0.13 ± 0.01 dpm g^{-1} in both samples; this fact strongly suggests that both nodule and stone phase were together in space at the same depth in the pre-atmospheric body for at least the last million years. The Be¹⁰ activity ($T_{\pm} = 2.7 \times 10^6 \text{ yr}$) of 1.8 ± 0.3 dpm kg⁻¹ is lower, by a factor of about 11, than that found in most chondrites. If this were due to a short cosmic-ray exposure (370,000 years), the expected Al²⁶ activity $(T_{\frac{1}{2}})$ = 7.4×10^5 yr) would be 29 percent of its normal saturated value-that is, $54 \times 0.29 = 16 \text{ dpm kg}^{-1}$ -which is not in agreement with the measured values.

Table 2 lists concentrations of stable, rare-gas isotopes in the silicate phase of Bondoc; Cobb's data (2) were obtained on a portion of the specimen we studied. The sample studied by Hintenberger et al. (3) may have come from the same 10-kg fragment from which ours was taken. Exposure ages can be calculated from the data in Table 2 by use of nuclide production rates of 2.0 and 0.249 \times 10⁻⁸ cm³ (STP) g⁻¹ per million years (4) for He³ and Ne²¹, respectively. The resulting ages range from 4 to 12 imes 10⁶ years. Even 4 imes10⁶ years is too long to account for the low activities that have been observed. In view of the observed deficiencies in radionuclide contents, the actual exposure age is probably greater than 20 million years.

The appreciable levels of Ni⁵⁹ ($T_{\frac{1}{2}}$ = 7.5 \times 10⁴ yr), Cl³⁶ (T = 3 \times 10⁵ yr), and Mn⁵³ activities in the nodule preclude the possibility that great terrestrial age is the cause of the general reduction in specific activities. The observed Ni⁵⁹ activity is equivalent to about 0.7 dpm g^{-1} nickel, to be compared with a maximum of 2.3 dpm g^{-1} nickel in irons (5). This lower Ni⁵⁹ activity corresponds to a terrestrial age of up to about 130,000 years, which is sufficient to reduce the saturated Cl³⁶ activity by about 25 percent and too low to have any effect on Mn53 activities. Yet the observed Cl36 activity is lower by a factor of 3 to 6 than average values in irons, and the Mn53 activity is lower by a factor of 2 to 3. Even allowing for the probability that production of Ni⁵⁹ from nickel may be enhanced by a factor of 2 to 4 in a stony matrix (6, 7) only increases the possible terrestrial age to a maximum of 300,000 years, still too low to account for the Cl³⁶ result, to say nothing of the Mn⁵³ result.

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Table 1. Cosmogenic radioactivities in the Bondoc meteorite. The nodule, of mass 120 g, contained 84 percent iron and 7.7 percent nickel; the silicate phase, of mass 360 g, contained 33.9 percent iron and 3.19 percent nickel.

Isotope	Activity (dpm kg ⁻¹)			
asotope	Nodule	Silicate		
Be10		1.8 ± 0.3		
A126		6.0 ± 0.9		
Mn^{53}	106 ± 7	43 ± 4		
Ni ⁵⁹	52 ± 6	≤ 40		
Cl ³⁶	3.0 ± 0.9			

A comparison of Be¹⁰ and Al²⁶ in the stone phase yields a similar conclusion. If the Be10 activity has been reduced to 10 percent of its value at saturation, because of great terrestrial age, then an age of 9×10^6 years is indicated, sufficient to eliminate measurable Al²⁶.

This leaves only shielding as the major cause of the low activities. From a radiochemical analysis of stone meteorites (7) we would expect, in a shielded sample containing 1.8 dpm Be10 per kilogram of Bondoc silicate, about 6 dpm Al²⁶, and 35 dpm Mn⁵³; these values are in good agreement with the activities observed in the stone phase of Bondoc. The expected Ni⁵⁹ activity is quite uncertain but should probably be in the neighborhood of 1 dpm g^{-1} nickel, or 32 dpm kg^{-1} silicate. The observed Cl36, Mn53, and Ni⁵⁹ activities in the nodule are compatible with a depth of 20 to 25 cm in a very large iron (5) or approximately 50 cm in a smaller stony-iron.

Assuming that the recovered Bondoc meteorite represents the center of the original body, we can estimate a lower limit for the pre-atmospheric mass. Our measurements were made on a fragment

Table	2.	Stable	rare-gas	isotopes	in	the	sili-
cate p	has	e.					

Isotope	Concn. $(10^{-8} \text{ cm}^3 \text{ g}^{-1})$ *				
isotope	Cobb (2)	Hintenberger (3)			
He ³	9.12	8.28			
He⁴	169	206			
Ne ²⁰	2.8	1.75			
Ne^{21}	2.9	1.75			
Ne^{22}	3.0	1.68			
Ar ³⁶	1.12				
Ar ³⁸	0.68				
Ar ⁴⁰	330				

* Standard temperature and pressure.

from the outer portion of the 890-kg mass recovered (equivalent to a stonyiron sphere of about 35-cm radius). Approximately 30 cm of stony-iron material would be required to equal the shielding effect of 20 to 25 cm of nickel-iron. The resulting 65-cm-radius stony-iron would have a mass of about 7000 kg.

In an independent calculation, we assumed that the primary cosmic radiation is attenuated in a meteorite with a mean absorption of 200 g cm⁻² and found that a pre-atmospheric mass of greater than 6000 kg is necessary to account for the factor-of-ten attenuation in Be10 and Al26 activities, assuming logarithmic dependence of specific activity on depth. Since both the Be¹⁰ and Al²⁶ are produced in significant amounts by secondary flux in stone meteorites (which increases initially with depth before dropping off), this last assumption is not completely valid; that is, the result is probably low. If the Bondoc specimen did not come from

the center of the original body, then, by either calculation, the pre-atmospheric mass was greater than 6000 to 7000 kg.

PHILLIP J. CRESSY, JR.* JULIAN P. SHEDLOVSKY[†] Department of Chemistry, Carnegie Institute of Technology, Pittsburgh, Pennsylvania

References and Notes

- 1. H. H. Nininger, Science 139, 345 (1963).
- H. H. Ninnger, science 139, 345 (1963).
 J. C. Cobb, unpublished data (1964).
 Von H. Hintenberger, H. Konig, L. Schultz, H. Wanke, Z. Naturforsch. 19a, 327 (1964).
 E. Anders, Space Sci. Rev. 3, 583 (1964).
- . H. Kaye, dissertation, Carnegie Institute of echnology; U.S. At. Energy Comm. Rept. Technology; U.S. NYO-8923 (1964)
- 6. M. Honda, S. Umenoto, J. R. Arnold, J.
- Geophys. Res. 66, 3541 (1961). 7. P. J. Cressy, Jr., dissertation, Carnegie Insti-
- P. J. Cressy, Jr., dissertation, Carnegie Institute of Technology; U.S. At. Energy Comm. Rept. NYO-8924 (1964).
 We thank J. H. Kaye for measuring the Ni⁵⁰ activity in the stone phase, and J. C. Cobb for use of unpublished data. Research supported by AEC contract At (30-1)-844.
 * Present address: Goddard Space Flight Center, Greenbelt Md
- Greenbelt, Md.
- † Present address: National Center for Atmo-spheric Research, Boulder, Colo. 16 April 1965

Crystal Multiplication without Nucleation

Abstract. Disk-shaped ice crystals grow out from the surface of polycrystalline ice in undercooled water. The rupture of the neck of the attached disk is a means of multiplying the number of viable crystals in the surrounding undercooled water. This is a source of frazil ice in streams and a source of new crystals in metal castings which are grain-refined by stirring.

It has frequently been observed in metals (1) and water (2) that the number of crystals that are formed during freezing can be greatly increased by causing the liquid to move, in relation to the solid, while freezing is taking place. A process of multiplication of the crystals has been identified and studied in water which is undercooled (that is, water below its equilibrium temperature, 0°C). This process of crystal multiplication accounts for the phenomenon of frazil ice, and for the multiplication of growing crystals (that is, grain refinement) caused by the agitation of metal castings during freezing.

Disk-shaped ice crystals were observed to grow on the surface of bulk ice when the bulk ice was placed in slightly undercooled water. These disks only touch the surface of the bulk ice at one place on their periphery (Fig. 1). The disks remain circular

during their growth and have been grown to a diameter of 1 cm (with a thickness of 0.7 mm) (3). A disk will sometimes be broken off by the force of buoyancy, and the point of attachment becomes the site for a new disk to grow. In this manner a single site can create two or three crystals per second, and these crystals trail away from the site much as bubbles trail away from a site in a beaker of boiling water where a crevice in the glass permits a vapor bubble to remain while new bubbles are generated (4). The "attached" disks have been described (5), but the important features for crystal multiplication are that (i) the disks grow with a fine bridge which is easily ruptured to set the disk free and (ii) a new disk grows at the site of detachment.

Frazil ice has been described (6) as being thin, free-floating, rounded ice disks which occur in streams and riv-