

determinations and the present neutron determination. An examination of the curve (Fig. 5) shows that the number of neighbors under the peak at 3.9A falls rapidly from 12 at 84°K to about 7 at 90°K, beyond which the curve would appear to fall more slowly. This means that the structure of the liquid changes rapidly for temperatures just above the melting point. Since the number of atoms per unit volume is approximately constant, a decrease in the number of atoms at a given spacing requires that these atoms appear elsewhere. It is intended to find the effect of temperature on the posi-

tions and heights of the peaks and to check the present results when experimental work can be resumed.

ACKNOWLEDGMENT

The authors wish to thank Dr. H. Gellman of the Theoretical Physics Branch for calculations of the Fourier transforms on the FERUT Computer and Miss B. Sears for assistance in similar calculations. Thanks are also due to Mr. W. McAlpin for the mechanical design of the cryostat, and to Mr. W. Woytowich for technical assistance.

Concentration of Uranium and Lead and the Isotopic Composition of Lead in Meteoritic Material

C. PATTERSON AND H. BROWN, *California Institute of Technology, Pasadena, California*
G. TILTON, *Department of Terrestrial Magnetism, Carnegie Institution, Washington, D. C.*

AND

M. INGRAM, *University of Chicago, Chicago, Illinois and Argonne National Laboratory, Lemont, Illinois*

(Received July 27, 1953)

The cosmic abundance of lead and uranium have been determined by studying the lead and uranium contents of meteoritic materials. Lead is found to be present to 8×10^{-3} atom/10 000 atoms of silicon, and uranium to 1×10^{-4} atom/10 000 atom of silicon. The new value for lead removes the hump in the cosmic abundance curve in the 206–208 mass region. The relative primordial abundances of lead isotopes of mass 204, 206, 207, and 208 are found to be 1:9.4:10.3:29.2, respectively.

THE concentrations of uranium in some stone meteorites and troilite from an iron meteorite were determined by the isotope dilution method.^{1,2} The concentrations of lead in a stone and iron meteorite and in troilite from an iron meteorite were determined colorimetrically, using Pb²¹⁰ for yield corrections.³ The lead samples so isolated were analyzed in a mass spectrometer, using the surface ionization method.² The effect of contamination upon the composition of the lead in the sample from the stone meteorite is as yet uncertain and, therefore, only the composition of the

troilite and the iron meteorite lead is reported here. The results are given in Table I.

Of the uranium concentrations in stone meteorites that have been previously reported,^{4–10} only the radon determinations of G. Davis agree with the values reported here. His values are given in Table II. It is believed that the higher values obtained by the early investigators resulted from a failure to exercise proper precautions in excluding moisture from electroscopes during radon measurements. Evans, Kip, and Moberg¹¹ recognized the critical nature of this phenomenon when they obtained anomalously high values for the radon content of sea water until the most stringent precautions were taken to dry the gases in the electroscop chamber. Other possible sources of error might be contamination

TABLE I. Uranium and lead in meteoritic material.

Sample	U concentration (ppm)	Pb concentration (ppm)	Pb composition (atomic ratios)		
			206/204	207/204	208/204
Canyon diablo troilite	0.009 ± 0.003	18 ± 1	9.41	10.27	29.16
Canyon diablo metal phase	—	(0.37 ± 0.05) ^a	(9.7) ^a	(10.5) ^a	(29.3) ^a
Modoc (total)	0.011 ± 0.002	0.9 ± 0.1	—	—	—
Norton county (total)	0.010 ± 0.003	—	—	—	—

^a Corrected for terrestrial lead contamination.

¹ G. Tilton, Atomic Energy Commission Report AECD-3182, 1951 (unpublished).

² G. Tilton *et al.*, Bull. Geol. Soc. Amer. (to be published).

³ C. Patterson, Atomic Energy Commission Report AECD-3180, 1951 (unpublished).

⁴ R. J. Strutt, Proc. Roy. Soc. (London) A77, 481 (1906).

⁵ T. Quirke and L. Finkelstein, Am. J. Sci. 44, 237 (1917).

⁶ G. Halledauer, Wien. Ber. 134 (IIa), 39 (1925).

⁷ A. Holmes, Bull. Natl. Research Council 81, 418 (1931).

⁸ F. Paneth and W. Koeck, Z. physik. Chem., Bodenstein-Festband, 145 (1931).

⁹ J. R. Horan, Oak Ridge Institute of Nuclear Studies, Oak Ridge, Tennessee reports a uranium concentration greater than 3 ppm in a carbonaceous chondrite from Murray, Callaway County, Kentucky (to be published in a new journal, Meteoritics). Mr. W. Nichiporuk of the California Institute of Technology has checked this meteorite by alpha counting and finds less than about 0.4 ppm of uranium plus thorium (the limit of the method).

¹⁰ G. L. Davis, Am. J. Sci. 243, 107 (1950).

¹¹ Evans, Kip, and Moberg, Am. J. Sci. 36, 241 (1938).

from reagents or glass vessels. Some of the values of uranium concentrations in iron meteorites reported by Paneth^{12,13} and by Davis¹⁰ are given in Table II.

The lead concentrations in Table I agree with the spectroscopic value for troilite reported by Goldschmidt and Hormann,¹⁴ but are in disagreement with their values for stone meteorites and the values for stone and iron meteorites and troilite reported by the Noddacks.¹⁵⁻¹⁷ These values are given in Table III.

Studies of meteoritic uranium and lead are of interest because of their relation to problems concerning cosmology and geochemistry. The values given in Tables I and II indicate that the relative cosmic abundance of uranium is 1×10^{-4} atom/10 000 atoms of silicon and that of lead is 8×10^{-3} atom/10 000 atoms of silicon according to the conventions of Brown¹⁸ and Urey,¹⁹

TABLE II. Previously-reported uranium concentrations on meteoritic material.

Stone meteorite	U concentration (ppm)	Observer
Cumberland	0.010	G. Davis ^a
Shaw	0.048	G. Davis ^a
Iron meteorites		
Average of seven	0.0011	G. Davis ^b
Average of six	0.0025	Arrol, Jacobi, and Paneth ^c

^a See reference 10.
^b See reference 10.
^c See reference 13.

and that (1) a large distinct maximum in the mass 206, mass 208 region of the cosmic-abundance curve, as was evident from previously reported data, does not exist; (2) the previously reported significant decrease between mass 209 and mass 238 is confirmed by our data; (3) the relative primordial abundances of the isotopes of lead are 1:9.4:10.3:29.2 with increasing mass respectively.

¹² F. Paneth, *Naturwiss.* **19**, 164 (1931).

¹³ Arrol, Jacobi, and Paneth, *Nature* **149**, 235 (1942).

¹⁴ V. M. Goldschmidt, *Skrifter Norske Videnskaps-Akad. Oslo. I. Mat.-Natur. Kl.* **1937**, No. 4 (1938).

¹⁵ I. Noddack and W. Noddack, *Naturwiss.* **18**, 757 (1930).

¹⁶ I. Noddack and W. Noddack, *Z. physik. Chem.* **A154**, 207 (1931).

¹⁷ I. Noddack and W. Noddack, *Svenk. kemisk Tidskrift* **46**, 173 (1934).

¹⁸ H. Brown, *Revs. Modern Phys.* **21**, 625 (1949).

¹⁹ H. Urey, *Phys. Rev.* **83**, 248 (1952).

TABLE III. Previously-reported lead concentrations in meteoritic material.

Pb concentration (ppm)			Observer
Stone meteorites	Iron meteorites	Troilite from iron meteorites	
4	...	20	Goldschmidt and Hormann ^a
5	53	710	I. and W. Noddack ^b
2	60	470	I. and W. Noddack ^c
18	I. and W. Noddack ^d

^a See reference 14.

^b See reference 15.

^c See reference 16.

^d See reference 17.

Goldberg, Uchiyama, and Brown²⁰ have shown by a detailed study of some trace elements in a large number of iron meteorites that extensive, though perhaps systematic, variations in the concentrations of those trace elements occur. For this reason, the values given here might differ somewhat from those chosen as significant after a detailed study of these two elements in meteoritic material has been made. High concentrations in minor phases of heterogeneous occurrence, as indicated by the concentration of lead in troilite, may produce an uncertainty of a factor of about 2 in the concentration of these two elements. The values given here are probably significant to within a factor of about 5 of the meteoritic concentrations, assuming they do not represent extremes of possible variations. Brown has proposed that lead isolated from iron meteorites might be used as a measure of the isotopic composition of lead at the time of formation of the solar system.^{21,22} On the basis of these results the concentrations of uranium relative to lead are so small that the isotopic composition of the lead in Canon Diablo troilite could be that present at the time the meteorite was formed. It must be emphasized, however, that in addition to the obscure nature of the relation between meteoritic and other stellar material, the effects of chemical fractionation during the process of meteorite formation upon estimates of the cosmic abundances of uranium and lead are as yet unknown.

²⁰ E. Goldberg *et al.*, *Geochim. et Cosmochim. Acta* **2**, 1 (1951).

²¹ H. Bruhn, *Phys. Rev.* **72**, 348 (1947).

²² Initial attempts to study this lead were unsuccessful because of contamination by terrestrial lead. [H. Suess and A. Nier, 1948 (unpublished); Patterson, Brown, and Nier, 1948 (unpublished).]