pothesis ($S-A-P$) and also the Tolhoek-DeGroot[27] symmetry principle. The latter leads one to expect either an SAP form or a VT form for the β-interaction. This may mean that the neutron and proton are not quite as equivalent during Fermi interaction as is presumed in the symmetry principle.

We wish to acknowledge the help of Professor A. C. G. Mitchell in making this work possible. This work was assisted by the joint program of the ONR and AEC.

---

[27] H. Tolhoek and W. DeGroot, Phys. Rev. 84, 150 (1951).

PHYSICAL REVIEW VOLUME 88, NUMBER 6 DECEMBER 15, 1952

Variations in the Relative Abundances of the Isotopes of Common Lead

C. B. COLLINS, R. M. FARQUHAR, AND R. D. RUSSELL
Department of Physics, University of Toronto, Toronto, Canada

(Received July 18, 1952)

Mass spectrometer measurements of the relative isotopic abundances of samples of lead ores from Archean-type rocks showed larger variations than reported by Nier. The isotopic constitution of one lead was within the limits calculated by Holmes and by Bullard and Stanley. From the new measurements combined with existing data estimates of the time of formation of the earth's crust of 3.5 billion years and of a maximum time of formation of the elements of 5.5 billion years have been calculated. These values are in reasonable agreement with previous estimates.

NIER[1] reported the first, and still the only, thorough investigation of the variations of the isotopic constitution of ordinary and radiogenic leads. Several authors have since used Nier's data to calculate the time of solidification of the earth's crust (Holmes,[2] 3.35 billion years, Bullard and Stanley,[3] 3.29 billion years) and a limit on the time of formation of the elements (Alpher and Herman,[4] 5.3 billion years). All these estimates have been based upon extrapolations from the abundances of ordinary leads ranging in age from 25 to 1400 million years. More analyses of ordinary leads dated at 2000 million years or older have been needed to check these estimates. We have therefore analyzed a number of common lead samples, dating as many as possible from Nier's ages and from analyses of radiogenic leads in our own laboratory. Calculations of the age of the earth's crust and of a maximum age of the elements have been made, using the new measurements listed in Table I combined with Nier's data.[1]

The experimental work was carried out with a 180° direction-focusing Nier-type mass spectrometer with a resolution of about 1/300 of an atomic mass unit. Analyses were made using lead tetramethyl as previously reported by the authors[5,6] and by Dibeler and Mohler.[7] The isotopic lead abundances listed in Table I are believed to be accurate to within 1 percent on an absolute basis, although comparative measurements are good to 0.1 percent.

Since Pb^{204} is not believed to be of radiogenic origin, the total amount of this isotope is assumed to have remained constant from the time of formation of the elements to the present. The abundances of Pb^{206}, Pb^{207} and Pb^{208} are therefore given with respect to Pb^{204}.

A value of 5.5±0.2 billion years for the maximum limit on the time of formation of the elements has been calculated from the data given in Table I combined with Nier's measurements of the isotopic constitution of common leads of known age. The method of calculation and the assumptions made were essentially the same as those of Alpher and Herman,[4] and all values of constants were the same except the half-life of U^{235}, which was taken as 7.07×10^8 years.[8]

Using Nier's data, Alpher and Herman reported a figure of 5.3 billion years. The average abundances of the lead isotopes at different times have been listed in Table II.

The calculated relative number of atoms of U, Th, and Pb in the earth's crust at the present time are U—1.00, Th—3.83, and Pb—7.35.

The isotopic abundances of those lead minerals (Table I) which could be assigned an age were combined with Nier's measurements[1] on other dated lead minerals and used to calculate a value for the age of the earth's crust after the method of Bullard and Stanley.[3] This method assumes that the isotopic constitution of lead

---

[1] A. O. Nier, J. Am. Chem. Soc. 60, 1571 (1938); Phys. Rev. 55, 150 (1939); Nier, Thompson, and Murphey, Phys. Rev. 60, 112 (1941).

[2] A. Holmes, Nature 157, 680 (1946); 159, 127 (1947); 163, 453 (1949).

[3] E. C. Bullard and J. P. Stanley, Suomen Geeodeetisen Laitoksen Julkaisuja, Veroffentlichungen des Finnischen Geodatischen Institutes, No. 36, 33 (1949).

[4] R. A. Alpher and R. C. Herman, Phys. Rev. 84, 1111 (1951).

[5] Collins, Freeman, and Wilson, Phys. Rev. 82, 966 (1951); C. B. Collins and J. R. Freeman, Trans. Roy. Soc. Canada 45, Sec. IV, 23 (1951).

[6] Collins, Lang, Robinson, and Farquhar, Geol. Assoc. Canada (to be published).

[7] V. H. Dibeler and F. L. Mohler, J. Research Natl. Bur. Standards 47, 337 (1951).

[8] Fleming, Ghiorso, and Cunningham, Phys. Rev. 82, 967 (1951).

Table I. Isotopic abundances of "common" lead.

No.[a]	Source of lead locality	Isotopic abundances referred to $Pb^{204} = 1.00$ 206	207	208	Age in 10^6 years
1	Bolivia galena	18.94	15.86	39.70	25[b]
2	Kengere, Belgian Congo	17.58	15.94	38.34	630[c]
3	Frontenac Co., Ontario, Canada	16.79	15.40	36.49	1000[c]
4	Matachewan, Ontario, Canada	15.83	15.41	34.88	
5	Negus Mine, Yellowknife, N.W.T., Canada	14.63	15.27	34.46	1850[d]
6	Sioux Lookout galena, Ontario, Canada	14.03	14.89	33.78	2480[e]
7	Rex property, Great Slave Lake, N.W.T., Canada	16.37	15.65	36.34	1850[d]
8	Rex property, Great Slave Lake, N.W.T., Canada	16.42	15.75	36.54	1850[d]
9	Horseshoe Island, Yellowknife, N.W.T., Canada	14.16	15.08	34.28	
10	Delhi Township, Ontario, Canada	16.07	15.47	35.17	
11	Garson Mine, Sudbury, Ontario, Canada	23.35	16.98	45.57	
12	Worthington Mine, Sudbury, Ontario, Canada	26.00	16.94	52.21	
13	Upper Canada Mine, Ontario, Canada	19.6	15.2	33.6	
14	Rosetta Mine, Barberton, Transvaal, South Africa	12.65	14.27	32.78	
15	Great Bear Lake, N.W.T., Canada	15.93	15.30	35.3	1400[c]
16	Ivigtut, Greenland	14.65	14.65	34.48	
17	Galena, Joplin, Missouri	22.35	16.15	41.8	

[a] Samples Nos. 1 to 14 were analyzed by the authors. Samples Nos. 15, 16, and 17, were analyzed by Nier *et al.* (see reference 1).

[b] Geological dating by J. T. Wilson.

[c] See references 6 and 1. Both authors agree on these ages.

[d] See reference 6.

[e] Nier's two isotopic ages (see reference 1) for the Huron claim have been changed slightly using the new half-life value for U^{235} (see reference 8) of 7.07×10^8 yr, to 2465 and 2580×10^6 yr. These values were averaged with Ahren's age [L. H. Ahrens, Geochim. Cosmochim. Acto. 1, 312 (1951)] obtained by the Rb–Sr method of 2400×10^6 yr. The average of 2480 has been applied to the Sioux Lookout galena which occurs in the same geological area.

Table II. Calculated average isotopic abundances of lead at different times.

Isotope	$t = 5.5 \times 10^9$ yr	$t = 3.5 \times 10^9$ yr	$t = 0$ (present)
204	1.00	1.00	1.00
206	5.12	11.36	18.45
207	0.00	13.48	15.61
208	26.31	31.10	38.40

was identical everywhere when the crust solidified, but has differed from place to place at any subsequent time. A least squares analysis is used to determine the time at which all lead had the same isotopic composition and to calculate its composition at that time. The values obtained were

$$t_0 = (3.5 \pm 0.2) \times 10^9 \text{ years, at which time}$$

$$Pb^{206}/Pb^{204} = 11.33$$

and

$$Pb^{207}/Pb^{204} = 13.55.$$

The value of 3.5±0.2 billion years for the age of the earth's crust is higher than that of Holmes (3.35±0.2 billion years) and of Bullard (3.29±0.2 billion years), whose calculations were based entirely on Nier's data. The abundances of Pb^{206} and Pb^{207} relative to Pb^{204} listed above agree well with those in Table II from the mean abundance-time curves used for setting a limit on the age of the elements. This agreement indicates that the two independent methods of calculation are consistent with each other.

DISCUSSION

The calculations carried out by the authors have been based on a set of dated leads of known isotopic constitution. While the two calculations are different in principle, they both depend on the total effect of all the determinations, rather than on any individual one. Single samples such as Rosetta Mine, No. 14, cannot be dated from abundance-time curves such as those of Alpher and Herman,[4] who also pointed out this difficulty. These findings do not support the abundance-time scale for common lead reported by McCrady.[9] The same argument applies to samples Nos. 11 and 12. These discrepancies show that individual samples which have a high or low abundance of Pb^{204} are the result of differences in the relative amounts of Pb, U, and Th from place to place and from time to time, in certain cases. For most samples it is possible to apply a criterion based on the ratio of Pb^{207}/Pb^{206} to find whether the sample is anomalous.

The investigations whose results have been given briefly here will be reported more fully in another publication.

---

[9] E. McCrady, Trans. Am. Geophys. Union **33**, No. 2 (1952).