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The Relative Abundance of Hydrogen Isotopes in Natural Hydrogen

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The relative abundances of isotopes have been investigated extensively in recent years, but few researches of such ratios when one isotope is very rare have been reported. We have investigated a number of samples of hydrogen from various sources, water from the crater of Mt. Kilauea, an obsidian, Devonian water, and hydrogen from helium bearing natural gases. The spectrographic method used for detection is not free from objections, but the results show that there is no appreciable variation of abundance of H^2 in the samples investigated.

THE question as to whether the relative abundances of isotopes vary in different samples of elements has interested many investigators in recent years, because any variation would give evidence that transmutation reactions are progressing on the earth or that the elements as found on earth have not been thoroughly mixed.[1] These researches have shown no such variation with the possible exceptions of boron and lithium. It seems probable that the variations reported in the case of lithium are due to the different methods used and also that the observed differences in the atomic weights of boron may possibly be due to systematic errors due, for example, to variations in impurities. At least there exists no certain evidence for any variation in relative abundances of isotopes.

In spite of these negative results, it seemed worth while to investigate natural hydrogens to see whether any source of hydrogen containing appreciably larger amounts of H^2 than that contained in meteoric water could be found. The probable disintegration of nuclei by cosmic rays might produce small amounts of certain nuclei and if these are rare relative to other isotopes,

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[1] O. Hönigschmid and Birckenbach, Ber. **56**, (B) 1467 (1923); H. V. A. Briscoe and P. L. Robinson, J. Chem. Soc. **127**, 696 (1925); and Stephenson, ibid., 70 (1927); ibid., 150 (1925); and Smith, ibid., 282 (1927); Cousens and Turner, ibid., 2654 (1928); G. P. Baxter and T. Thorvaldson, J. Am. Chem. Soc. **33**, 337 (1911); Baxter and C. R. Hoover, ibid. **34**, 1657 (1912); and L. W. Parsons, ibid. **43**, 507 (1921); and Hilton, ibid. **45**, 694 (1923); and M. J. Dorcas, ibid. **46**, 357 (1924); Mlle. I. Curie, Compt. Rend. **172**, 1025 (1921); Mlle. E. Gleditsch and B. Samdahl, ibid. **174**, 746 (1922); W. D. Harkins and S. B. Stone, J. Am. Chem. Soc. **37**, 1367 (1926); J. N. Brönsted and Hevesy, Nature **109**, 813 (1922); Jaeger and Dijkstra, Proc. K. Akad. Wetensch. Amsterdam **27**, 393 (1924); A. D. Munro, J. Chem. Soc. **121**, 986 (1922); Muzaffar, J. Am. Chem. Soc. **45**, 2009 (1923); Willard and McAlpine, ibid. **43**, 797 (1924); Weatherill, ibid. **46**, 2437 (1924); A. Elliott, Zeits. f. Physik **67**, 75 (1931); Nature **126**, 203, 845 (1930); F. W. Aston, Nature **128**, 149 (1931); van Wijk and v. Koeveringe, Proc. Roy. Acad. **132**, 98 (1931); Morand, Thesis, Paris, (1929); H. Schüler, Die Naturwiss. **19**, 772 (1931); J. L. Hundley, Phys. Rev. **30**, 864 (1927).

a small amount of such a transmutation reaction might give a sufficient change in the isotopic ratios to be detected. Such a case of a rare isotope is furnished by the H^2 isotope reported by Urey, Brickwedde and Murphy.[2] Further Menzel[3] reports that the H^2 isotope must be very rare in the sun and thus it seemed to us well to be certain whether any large variation in abundance of this nuclear species could be detected in terrestrial sources of hydrogen.

SAMPLES AND EXPERIMENTAL METHOD

The samples of hydrogen investigated consisted of water collected by Dr. E. S. Shepherd from the crater of Mt. Kilauea in 1912, an obsidian containing about 0.75 percent of water from Cerro Noagua, New Mexico, a sample of water containing dissolved chlorides of sodium and magnesium trapped in the Devonian rocks of New York State secured by Professor C. P. Berkey in 1920, and hydrogen of helium bearing natural gases from the Cliffside and Panhandle gas fields in Texas. The water from the Kilauea samples was separated from other gases by collecting the water in a CO_2 trap and pumping the other gases out with a Toepler pump. The obsidian water was secured by heating the obsidian under vacuum in a quartz tube at $\sim 1000°$ C. The natural gases were passed over copper oxide to oxidize the hydrocarbons to water and carbon dioxide and the water was collected in a CO_2 trap. The connate water was used without purification.

These samples of water were introduced into the discharge tube used in a previous research[2] which was about 40 cm long, 8 mm inside diameter and carried about one ampere current. The $H\beta$ lines were photographed in the second order of a grating with a dispersion of 1.3A per millimeter. D. C. Ortho Eastman plates were used which gave lines free from excessive irradiation and hallation. The second order symmetrical ghosts in the second order of the grating appear to have about the same intensity as the $H^2\beta$ line from meteoric water. The exposure times used for all samples were ten and twenty minutes.

Visual observation as well as microphotometer curves show that there is no detectable difference between the special samples of water investigated and ordinary laboratory distilled water. The method of investigation is open to objection because of the partial reabsorption of the H^1 lines in the discharge tube.[4] However the conditions of the discharge tube were very nearly constant for all samples investigated and any marked differences in abundances would have been detected although the relative abundance cannot be satisfactorily determined.

We are indebted to Dr. E. S. Shepherd of the Carnegie Institution of Washington for supplying us with the Kilauea water and the sample of obsidian used; to Professor C. P. Berkey for the connate water and to Mr. R. A. Cattell of the United States Bureau of Mines for the helium bearing natural gases.

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[2] H. C. Urey, F. G. Brickwedde, and G. M. Murphy, Phys. Rev. **40**, 1 (1932).

[3] D. H. Menzel, Publ. Ast. Soc. Pacific **44**, 41 (1932).

[4] H. C. Urey, F. G. Brickwedde, and G. M. Murphy, Phys. Rev. **40**, 464 (1932).