The Concentration of the Oxygen Isotopes

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number of analyses in connection with these experiments. The apparatus used by him was built from funds furnished by the American Philosophical Society. We should also say that we are indebted to Dr. Walker Bleakney of Princeton University for many courtesies in connection with the analysis of our samples.

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[1] Urey and Greiff, J. Am. Chem. Soc. **57**, 321 (1935).
[2] Meldrum and Roughton, J. Physiol. **80**, 113 (1933).

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As Lewis and Cornish[1] showed, there is a difference in vapor pressures of the O^{16} and O^{18} varieties of water. Experiments by Wahl and Urey show that this ratio of vapor pressures amounts to approximately 1.003 at the boiling point of water, and approximately 1.008 at 45° centigrade. This small difference in vapor pressure may be used for the concentration of the O^{16} isotope if sufficiently efficient fractionation columns can be constructed. Simple calculations show that such a column must have approximately 500 theoretical plates in order to produce increased concentrations of the isotopes sufficient to make possible an eventually complete separation of O^{18}. At the time this work was begun no such columns had been devised. One of us (G. B. P.)[2] suggested the use of alternate rotating and stationary cones in order to secure a very long path without making the column prohibitively long. The apparatus as at present constructed is 35 feet long and contains 619 rotating cones and 619 stationary cones. The rotating cones are attached to a shaft placed in the center of a 6″ tube. The stationary cones are attached to the tube. The distance between two stationary cones is $\frac{5}{8}$ of an inch and the rotating cones are placed as nearly as possible midway between stationary cones. In this way water drops from a fixed cone to a rotating cone, is carried by centrifugal force up and off to the walls of the tube and then drains under gravity to the center and drops to the next rotating cone. A complete description of this apparatus will be published later.

First experiments were made on a column containing 14 stationary and 14 rotating cones and later with a 5-foot section containing 90 cones of each variety and finally experiments have been made with a 35-foot column as described above. The initial experiment indicated that a theoretical plate is secured for approximately each pair of cones. In the case of the 5-foot section the results were not quite so good, approximately 75 theoretical plates being secured as shown by the separation of the oxygen isotopes of water using the ratio of vapor pressures as determined by Wahl and Urey. Using the 35-foot column a change in the ratios of the oxygen isotopes by a factor of 2 was secured. In these experiments a large container of water was used at the bottom and total reflux was used at the top. In this way water containing less O^{18} than the natural abundance was produced at the top. It required approximately four days for a stationary state to be established, after which no change in the concentration of the isotopes occurred. This is very much less than was expected on the basis of our experiments with shorter columns. The water in the boiler at the bottom was heated by steam and no careful attempts were made to control the rate of boiling precisely. Approximately 100 cc of water were boiled through the column per unit time and considerable fluctuations were noted in the back pressure on the column.

In the most recent experiments with the 35-foot column a flash boiler has been placed at the bottom of the column capable of boiling approximately 50 cc of water per minute. Water has been pumped into the top of the column with a proportioning pump and the water vapor issuing from the top of the column has been condensed and run to the sewer. The run was made under 20 centimeters pressure at the top of the column and water was introduced at the rate of approximately 45 cc per minute. The apparatus operated steadily, day and night, for eight days. At the end of this time the oxygen isotope concentration at the bottom of the column had been increased by the factor of 3 from the natural abundance and the deuterium content by a factor of approximately 40. These figures would indicate that the column operated at approximately 230 theoretical plates, a considerably smaller number than expected on the basis of preliminary experiments. Considerable difficulty was encountered in keeping the proportioning pump operating steadily because of rust collecting in the valves of the pump. It stopped operation completely on several occasions for periods of time that were unknown since the column was not attended except occasionally during this time. Also, an accident occurred in adjusting the pump which caused it to introduce approximately 150 cc of water per minute for a period of perhaps one hour. Such irregularities are certain to cause a decreased efficiency of the column. Considering these difficulties the increased concentration appears to be as good as could be expected. A steady state was not reached as shown by analyses on the last two days.

Analyses were made by passing water samples mixed with hydrogen over platinized asbestos at a temperature of approximately 350° centigrade to promote the exchange between water and hydrogen. In this way it was possible to bring the concentration of deuterium to very near normal concentration. The water samples were then diluted with distilled water and density difference determined by the pressure float method of Gilfillan.[3]

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[3] Gilfillan, J. Am. Chem. Soc. **56**, 406 (1934).