On the Chemical Differences Between Nitrogen Isotopes

We wish to report on some preliminary experiments concerning the possibility of a chemical separation of the nitrogen isotopes. Although our work in this field has not yet been completed, a publication by E. Ogawa[1] causes us to publish a short summary of our conclusions at this time.

The analyses were performed with a mass spectrometer in the laboratory of Professor Bleakney in Princeton University as described by Wahl, Huffman and Hipple.[2] As these authors found indications of a small difference in vapor pressure between $N^{14}H_3$ and $N^{15}H_3$ and because distillation, if effective, would be by far the easiest way to obtain a change in the isotope rates, we first experimented on this possibility. We used the method by which Wahl and Urey investigated the vapor pressure ratios in water.[3] Distillations were carried out without boiling at different temperatures, the final residue varying between 1/15 and 1/70 of the amount of starting material. No definite trend of the vapor pressure ratio with temperature could be distinguished. As an average value for the pressure ratio $P(N^{14}H_3) : P(N^{15}H_3)$ we obtained 1.0025 in six experiments, with an average error of 0.003. It may be mentioned that Wahl, Huffman and Hipple give 1.0052 as their best figure for this proportion. It is probable that $N^{14}H_3$ is slightly more volatile than is $N^{15}H_3$, but we feel convinced that the vapor pressure ratio differs from unity only a few tenths of a percent at most. Therefore the nitrogen separation by the distillation of ammonia does not seem promising. Next we investigated the reaction of $N^{15}H_3 + N^{14}H_4OH \rightleftarrows N^{14}H_3 + N^{15}H_4OH$ for which Wahl, Huffman and Hipple had reported a possible but uncertain concentration of the heavy isotope in the NH_4OH. Purified air was passed through a bottle containing concentrated ammonia till only a small fraction of the original amount of ammonia was left in the solution. From the isotopic composition of the original material and the final sample we calculated the equilibrium constant from the Rayleigh formula. We found that the N^{15} possibly concentrates slightly in the liquid but the effect is so small at any rate, that it could not be recognized with certainty.

Then we turned to the exchange reaction $N^{15}H_3 + N^{14}H_4^+ \rightleftarrows N^{14}H_3 + N^{15}H_4^+$, already considered by Urey and Greiff.[4] We started from a concentrated solution of ammonium chloride, to which at intervals we added small amounts of sodium hydroxide solution. A current of purified air removed the NH_3 formed after each addition. At the moment we have the analysis of only one run in which we started with 0.61 mole NH_4Cl and finished with 0.052 mole. During this process the ratio of $N^{14}N^{15}$ decreased from 250.2 to 233.8. (These numbers are not corrected for diffuse background and hence the analysis of the original material does not agree with the generally accepted value). This gives for the fractionation factor the value 1.027. We hope to check this value soon with other samples we have prepared, but we have been forced to abandon this matter temporarily because it will not be possible to obtain analyses by the mass spectrometer for some time.

The last experiment described seems to be the same as one performed by Ogawa, an extract of which publication is printed in *Nature*.[5] From the figures given fractionation factors ranging from 1.7 to 1.8 can be calculated which are very much higher than ours. We feel quite certain that as large an increase in concentration of N^{15} could not remain undetected by the mass spectrometer and that Ogawa's values, which are obtained from atomic weight determinations, are too high.

The importance of finding the most favorable reaction is clearly seen from the calculations by Urey and Greiff on the efficiency of a column as depending on the separating factor of the process used.

Finally, it may be mentioned that a preliminary experiment on the exchange $HCN^{15} + CN^{14-} \rightleftarrows HCN^{14} + CN^{15-}$ indicated that N^{15} was concentrated in the HCN gas to quite an appreciable extent.

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H. C. Urey
A. H. W. Aten, Jr.

Department of Chemistry,
Columbia University,
New York, N. Y.

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Effect of Hydrocarbons at Low Temperatures on Slow Neutrons

Investigations of the properties of slow neutrons passed through paraffin at various temperatures have established the fact that temperature equilibrium must be attained at least partially.[1–5] The somewhat remarkable character of this fact, considering the size of the energy losses and the probable mechanisms involved led the authors to investigate the phenomenon in more detail, first with paraffin and later with pure normal heptane.

The apparatus used is shown in Fig. 1. It consists of a metal Dewar (brass and monel metal walls 1 mm thick) surrounded by a large cylindrical block of paraffin in which the radon-beryllium neutron source (100–220 millicuries) was imbedded. The inside diameter of the Dewar was 14 cm and the height one meter. A soft glass Dewar (3 cm outside diameter) was mounted along the axis of the metal Dewar, to allow irradiation of the cylindrical silver targets without alteration of the temperature of the hydrocarbon. The cylindrical copper container shown was used for the heptane only, and was the same length as the silver targets (11 cm), and 4.5 cm thick, with a wall thickness of 0.3 mm. Copper-constantan thermocouples were

TABLE I.

Thickness of paraffin cylinder	Temperature (°K)	Activity (% 300°K activity)
1.5	300°–200°	100±2
	100°– 11°	113±2
2.5	70°	94±2
	21°	86±2
3.5	90°	103±2
	55°	71±2