Concentration of N^{15} by Chemical Exchange Reaction Method

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Introduction

For the purpose of the utilization of concentrated N^{15} as tracer works, we have been studying the concentration of N^{15} during these three years, and we are now able to concentrate the heavy nitrogen isotope up to nearly 10 atomic percent. The experimental results will be reported here briefly.

The method of the concentration of N^{15} was quite the same as that of Urey and others,(1) namely the chemical exchange reaction between ammonia gas and ammonium ion in the packed concentrating column.

Experimental Apparatus

The experimental apparatus consisted of two stages and their configurations were as shown in Fig. 1. The principal parts of the apparatus were as follows:

(1) **Concentrating column.**—The concentrating columns used in this work were made of glass tubes and total heights and inner diameters of the columns were 8 m. and 2.2 cm. for the 1st stage respectively and 6 m. and 1.0 cm. for the 2nd stage.

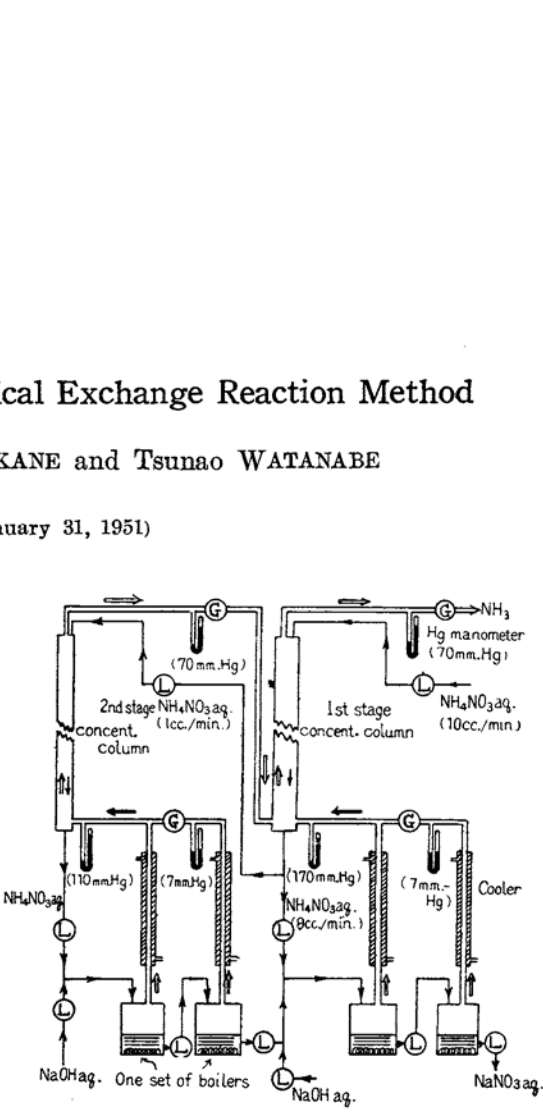

Fig. 1.
L: Liquid Pump.　G: Gas Pump.

Each column was packed with the following; Fenske type (one turn helix) made of thin aluminum wire (0.3 mm. in diameter). The diameters of the helixes were 2.4 mm for the case of the 1st stage and 2.0 mm. for the case of the 2nd stage.

They were cleaned and boiled in water solution of Na_2CO_3. By this treatment, the surfaces of them were changed to gamma crystals

(1) H. C. Urey, J. R. Huffman, H. G. Thode and M. Fox, *J. Chem. Phys.*, **5**, 856 (1937); H. G. Thode and H. C. Urey, *J. Chem. Phys.*, **7**, 34 (1939).

of Al_2O_3.[2] These packing materials proved to be very satisfactory for the concentrating work as they were easy to produce on a large scale and also quite safe for the corrosion by the water solution of NH_4NO_3 and ammonia gas.

The space factors of the packing materials in the columns were 16.7% for the 1st stage column and 19.2% for the 2nd one.

(2) **Boilers.**—For each stage two sets of boilers were used. Each set was composed of five boilers made of copper tubes 10 cm. in diameter, four of which were 50 cm. in length and were placed horizontally while one was 20 cm. in height and was placed vertically. The five boilers were connected in series, the vertical one being placed at the end. Each boiler was heated with a 400 watt alumica heater. Water vapour was removed from the flow of ammonia gas at cold traps placed before each gas pump. The outsides of traps were cooled by methylalcohol-water mixture of about −15° circulatiug from a tank of a methylchloride refrigerating machine by a circulating pump. By introducing an auxiliary gas pump between two sets of boilers of each stage, we could reduce the residual amount of ammonium ions in the solution of $NaNO_3$ which would be drained from the last boiler of the stage less than one hundred thousandth (1/100,000) of the initial amount fed at the top of the column.

To keep the liquid levels in the boilers constant, we attached to the last boiler of each set a liquid level regulator as shown in Fig. 2. The liquid-contact electrodes were made of thin platinum foils. The magnetic valves were constructed especially for this purpose and were operated by the compression of rubber tubes by the action of electromagnets.

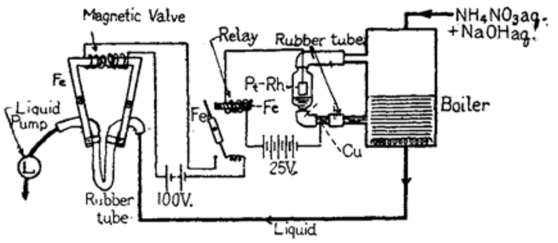

Fig. 2.

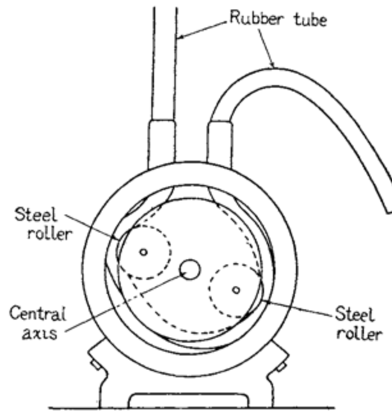

Fig. 3.

(3) **Liquid pump.**—For the transport of the chemical solutions (NH_4NO_3, NaCH, $NaNO_3$), we constructed a special type of liquid pump, the working mechanism of which can be easily seen in Fig. 3. By changing the velocity of rotation

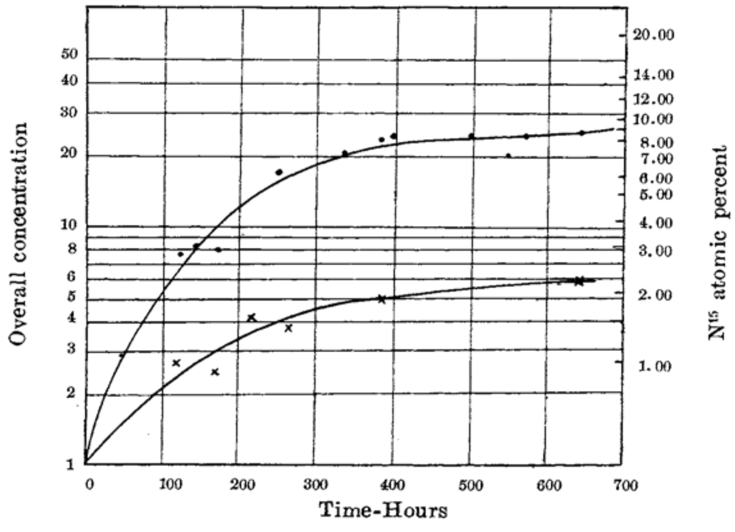

Fig. 4.—●, Concn. at the bottom of the second stage column; ×, Concn. at the bottom of the first stage column.

(2) We are greatly indebted to Miyata Laboratory of this institute for this surface treatment of the packing materials.

of the central axis, and inner diameter of the rubber tube, we could change the speed of the transport of liquid from 0.01 cc./min. to 30 cc./min. by a single pump. As for the lubricant between the steel rollers and the rubber tube, we used glycerine which proved to be very satisfactory.

(4) **Chemicals.**—As for the solution of the ammonium ion we used 60% solution of NH_4NO_3, to which 1% of methylalcohol was added. The feeding velocities of the ammonium nitrate solution were 10 cc./min. for the 1st column and 1 cc./min. for the 2nd column. As for the solution of NaOH, 30% solution of commercial caustic soda was used.

Experimental Results

We had operated the apparatus mentioned above from June 21 to July 19, nearly 670 hrs. The pressure at the top of the column of each stage was kept at 70 mm. Hg and the pressure drops in the columns were 10 mm. Hg per meter for 1st stage and 6 mm. Hg per meter for the 2nd one.

The changing rate of the concentration of N^{15} at the bottom of the column of each stage was shown in Fig. 4. From 420 hrs. we began to produce samples of concentrated N^{15} in the form of $(NH_4)_2SO_4$ by sending bubbles of ammonia gas from the bottom of the 2nd stage column through H_2SO_4. Thus we could get totally 20 g. of $(NH_4)_2SO_4$ during this work. The N^{15} contents of the samples varied from 8 to 9.3 atomic percent. The overall concentration factor of the two stages was 27 and that of the 1st stage only was 5.9.

In conclusion we express our thanks to the late Dr. Yoshio Nishina, the President of this Institute, for his kind encouragement throughout this work. We are also greatly indebted to the Ministry of Agriculture and Forestry for grants without which this work could not have been accomplished. We also wish to thank Messrs. Sadao Matsuo, Shyohei Isomura, Ryuzo Shibata, Shohichi Motonaga, Kozo Morishita for their assistance in connection with this experiment.

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