A Preliminary Study of the Determination of Hydrogen Content in Heavy Water by Means of Neutron Transmission

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The aim of this work is to examine the applicability of a neutron transmission procedure to the determination of the hydrogen content in heavy water, although the mass spectrometric and infrared absorption spectrophotometric methods[1,2] are most commonly used for this purpose.

The principle suggested here is based on the difference in total cross sections of thermal neutrons between light and heavy water.[3]

1) I. Kirshenbaum, "Physical Properties and Analysis of Heavy Water," McGraw Hill, New York (1951), p. 201.
2) G. Gordon and H. Yamatera, *Anal. Chem.*, **36**, 1866 (1964).
3) ANL-5800, Reactor Physics Constants.

The following formula is derived to give the sensitivity as a function of the amount of light water in heavy water, while the relation is shown in Fig. 1.;

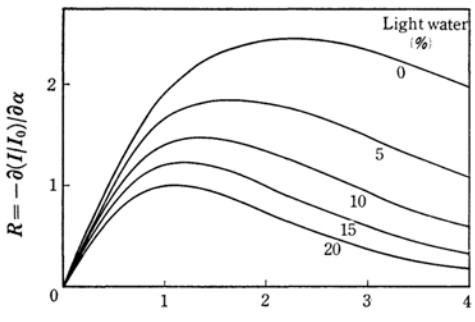

Fig. 1. Relation of R to thickness d of cell.

$$R = -\frac{\partial(I/I_0)}{\partial\alpha} = a.d.\exp\{-(a\alpha + b)d\} \qquad (1)$$

where I_0 is the number of well-collimated incident neutrons falling on a small area; I, the number of neutrons which succeed in passing through the thickness, d, of the target over the same area; a, the difference between the macroscopic total cross sections for light and heavy waters; b, the macroscopic total cross section of heavy water, and α, the fractional molar concentration of light water.

From each maximum of the curves in Fig. 1 one can predict the optimum thickness, d, which will give the highest sensitivity of measurement for any arbitrary concentration of light water.

A preliminary experiment was performed by the arrangement shown in Fig. 2.

A series of samples were prepared by diluting pure heavy water (99.94 mol.%). The samples were poured into a dried cell by nitrogen-gas pressure. A cell 2 cm. thick was used, since this thickness, according to the curves in Fig. 1, seemed to give the maximum sensitivity for a 95—100 percent concentration of heavy water.

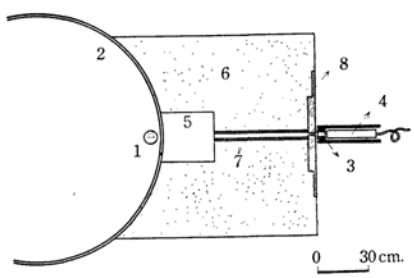

Fig. 2. Experimental arrangement.

(1): Neutron source (Am-Be, 6 curies)
(2): H_2O tank
(3): Cell (Al, 10×70 mm$^2\times20$ mm.)
(4): BF_3 counter (25 mm$\phi\times30$ cm.)
(5): Paraffin
(6): Boron paraffin
(7): Coarse collimeter
(8): Cadmium plate.

The counting rate of incident thermal neutrons through the cadmium slit (1.5×30 mm.) was 5.92 ± 0.01 c.p.m. during 12 hr. of measurement, and the background was 1.02 ± 0.025 c. p. m. during a 24 hr. period.

Figure 3 shows the curves obtained by the experiments and by calculations based on the neutron linear attenuation formula. This formula gives the highest sensitivity which can be expected by this method.

The fairly large disagreement between the two curves seems to be caused by more complicated phenomena than are taken into account by a simplified consideration like the linear attenuation formula. However, the results obtained experimentally show that this measurement

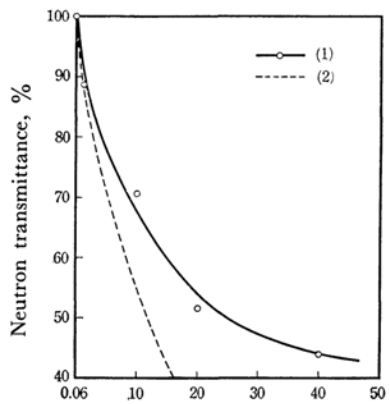

Fig. 3. Relation between neutron transmittance and concentration of light water (normalized to D_2O 99.94 mol. percent concentration).

(1): Experimental
(2): Calculated for linear attenuation formula $I/I_0 = \exp\{-(a\alpha+b)d\}$

procedure can be established as a method of chemical chemical analysis.

The transmittance of the arbitrary sample to the standard (99.94 mol.%) at high concentrations of heavy water (98—100%) decreased to 11.1 percent for the concentration of light water shown in Fig. 3.

If a higher neutron flux, such as that from a nuclear reactor were used, this procedure could be established as a method for hydrogen determination of an even higher accuracy and an even greater rapidity.