

THE CONCENTRATION OF DEUTERIUM IN SOME INDUSTRIAL WATERS.

By Masao HARADA and Toshizo TITANI.

Received August 28th, 1934. Published October 28th, 1934.

As the water from commercial electrolytic cells containing a little more deuterium than ordinary water is regarded as a convenient material for the preparation of heavy water, it seems worth while to examine the waters of other industrial sources on their deuterium contents. Thus, Newell and Ficklen⁽¹⁾ found that the water from some commercial chromium plating baths which had been operated for less than three years was heavier than ordinary water by 20 to 640 parts per million (p.p.m.); Washburn and Smith⁽²⁾ also examined the same sort of water but found that the excess in density was only 2 to 5 p.p.m.; Uhlmann⁽³⁾ called attention to the fact that the water from some laboratory storage batteries after six to ten years use was heavier than normal water by 33 p.p.m., suggesting that this water can be used as a starting material for the preparation of heavy water.

The present authors have also investigated some of the industrial waters on their deuterium contents by comparing their densities with that of normal water. The results are shown in the Table.

(1) L. L. Newell and J. B. Ficklen, *J. Am. Chem. Soc.*, **55** (1933), 2167.

(2) E. W. Washburn and E. R. Smith, *Bur. Standards J. Research*, **12** (1934), 305.

(3) W. Uhlmann, *Naturwissenschaften*, **22** (1934), 119.

Source of sample.	Period of operation	Readings in bouyancy balance.	Excess in density in p.p.m.	H ² : H ¹
(1) Commercial electrolytic cell	1 year	$0.39^{\circ} \pm 0.00$	28	1 : 2100
(2) Chromium plating bath	4 years	$0.12^{\circ} \pm 0.00$	7	1 : 3800
(3) Lead accumulator A.	9 years	$0.01^{\circ} \pm 0.01$	3	1 : 4400
" " B.	" "	$0.00^{\circ} \pm 0.01$	0	1 : 5000
" " C.	" "	$0.00^{\circ} \pm 0.02$	0	1 : 5000
(4) Edison battery A.	9 months	$0.41^{\circ} \pm 0.02$	28	1 : 2100
" " B.	6 months	$0.38^{\circ} \pm 0.02$	27	1 : 2100

The first sample to be investigated was the water from a commercial electrolytic cell, which, having been in use for all the year round, had been supplied with water sixty times its volume mainly owing to evaporation. The second sample was obtained from a commercial chromium plating bath running for nearly four years. During the process of plating, the evolution of hydrogen was hardly avoidable. The third samples were obtained from three different lead accumulators, all of which, having run for nearly nine years, had a supply of considerable amounts of water for the loss of the contents. The fourth samples were obtained from two Edison batteries, one being in use for about nine months in a part of coal mines, and the other for six months in another part of coal mines.

Various samples of water thus obtained were purified by the following processes: The water was filtered and distilled, after being neutralized if necessary, to free it from any dissolved substances; it was again distilled after being boiled with potassium permanganate in alkaline solution for nearly eight hours; then the distillate was redistilled twice, once with a few drops of sulphuric acid and once with some solid baryta. The water thus obtained was further subjected to three more successive distillations in the stream of air free from carbon dioxide, the first distillation being carried out under the addition of potassium permanganate and a few drops of sulphuric acid, the second under the addition of some solid baryta, and the final with a quartz condenser. The purity of the final distillate was determined by the conductivity method.

The density of the purified sample was determined referring to that of the ordinary water purified in the same way as above by the method of bouyancy balance using a quartz sinker. In order to increase the sensitivity of the balance, temperature a little lower than that of the room was employ-

ed, namely between 8° and 9°C., except the case (3) A, where the bath temperature was 23.80°C. The third column of the table shows the readings in the bouyancy balance in terms of degree, each being the mean of two independent determinations, except the case (3) A, where the mean of three independent determinations was taken.

If it is assumed that the increase in density is solely due to the enrichment of deuterium, the water from the commercial electrolytic cell contains about twice as much deuterium as ordinary water, which is almost the same for the cells which have been used for a longer period.⁽⁴⁾ Therefore, the water in the electrolytic cell seems to be capable of being concentrated up to this value, after which the rate of concentration becomes much slower. The present result for the chromium plating bath agrees fairly well with that of Washburn and Smith, but not with that of Newell and Ficklen. The result for the lead accumulators is found to be not in agreement with that of Uhlmann who found as much increase in density as 32 p.p.m. The degree of concentration for this case may be largely affected by the process of charging, as the evolution of hydrogen gas during this process can be regarded as the main cause of concentration. It seems rather plausible that our result for the lead accumulators is related in some way to the relatively poor separation efficiency obtained when dilute sulphuric acid solution is electrolysed between lead electrodes.⁽⁵⁾

The result for the Edison batteries is quite unexpected. The samples were taken from quite different sources and they were purified in the same way as the other samples. We are not yet sure whether the increase in density is really due to the concentration of deuterium or to some unseparable impurities included originally. We are still in the course of re-investigation of this case.

*Shiomi Institute for
Physical and Chemical Research,
and
Chemical Laboratory of the Osaka
Imperial University,*

(4) D. H. Rank, *J. Chem. Phys.*, **1** (1933), 750.

(5) T. Titani, K. Kurano, and M. Harada, *this Bulletin*, **9** (1934), 269.