Determination of the diffusion length of thermal neutrons in heavy water

By **R. u. K. Dopel** and **W. Heisenberg-**

1. Problem

For the question of the usability of D20 in the 238-machine, the absorption of neutrons in D20 is of decisive importance. This absorption is probably very small, and it was the task of the experiments described below to establish at least an upper limit for the absorption which determines the usability of D20. The absorption is most simply measured by the "diffusion length," which in Report I of one of us has been estimated at about 34 cm. If the diffusion length is greater than about 30 cm, then, according to previous knowledge of the constant of 238, the heavy water is usable for the machine.

Since for the experiments in Leipzig only about 91 D20 were available, a quantity for which the neutron absorption influences the neutron intensity at most in the order of a few percent, an experimental arrangement had to be chosen for which the theoretical intensity course is known completely exactly and for which the measurements may show only relative errors of a few thousandths.

The theory of the Mef3 arrangement is based on the following superposition: If in any region of space the production of thermal neutrons can be dispensed with, then in that region the distribution of thermal neutrons will be
<by the diffusion equation (*e* density, *I* diffusion length).

$$l^2 \Delta \varrho - \varrho = 0 \quad (1)$$

is determined. If, furthermore, the thermal neutrons are emitted from a spherical surface of radius **R** with uniform intensity, then, according to (1), in the ln nern of the sphere the density distribution <is determined by the equation

$$e = \frac{\sinh r //}{r/1} \quad (2)$$

so for *R*   *I* <lurch   2

$$e = ea \left(1 +- \frac{1}{6} \frac{r-2}{/2} \right) \quad (3)$$

must be given. A measurement of *e* in the center and at the edge of the sphere therefore immediately leads to a determination of the diffusion length.

The effect of an arbitrarily designed source of thermal neutrons can always be investigated experimentally by preparing a sink of thermal neutrons of the desired form from Cd instead of the source and placing it in a vessel in which neutrons of arbitrary energy distribution are present due to some other source. Then at any point the neutron intensity with and without Cd is compared. The difference corresponds to the neutron distribution produced by the (negative!) Cd neutron source.

The spherical-flat source of thermal neutrons, which is used to distribute (2) was thus produced in the following manner:

2. Experimental setup

a) Neutron source

An Al sphere ($r =$ 12 cm; wall thickness 2 mm) is suspended in a cylindrical vat (h = 80 cm; r = 40 cm) filled with Hz(), which can be filled with Dz() through a 7 cm wide and 35 cm long neck. The neck is closed by a paraffin-filled brass tube which is covered with Al at its lower end; the Dz() sphere is therefore bounded on all sides by Al.

The neutron source (approx. 480 mg Ra + Be in a nickel sphere of 9 mm radius and 0.2 mm wall thickness) is now located in the centre of this sphere. The neutrons pass through the Dz() preferably as fast neutrons, are decelerated in the surrounding Hz() and are partly scattered back into the Dz() as slow neutrons. The decrease of these neutrons from the periphery of the Dz() Ku gel towards the centre is measured.

3   Now, however, the (Ra+ Be) neutron source already emits a small

In addition, a part of the fast neutrons (in particular of {Ray -+ D2} $=$ photon neutrons in Dz()) is already decelerated to thermal energy. We must be able to subtract these neutrons from the total number of available neutrons in order to get only those neutrons which are at

of the Hz() boundary run inwards. This is achieved by measuring the neutron intensity once in the mentioned Al sphere and a second time in an identical sphere, but externally occupied by 1 mm Cd. In the Al sphere all thermal neutrons are measured, in the Cd sphere only those that do not come from the outside; in addition, in both cases (depending on the indicator) a certain amount of non-thermal neutrons is measured, which falls out when the difference is formed. The difference therefore consists of the thermal neutrons which come into the Dz() sphere from the outside.

b) Indicator

A very thin layer of Dyz()$_{3 \text{ was}}$ chosen as an indicator of thermal neutrons; its P-activity is a measure of the neutron density at the exposure site. It was known that Dy responds almost exclusively to thermal neutrons.

However, its activation by thermal and non-thermal neutrons in paraffin was again determined in particular (activation without and with Cd coverage in paraffin). The activation by non-thermal neutrons is less than 1OJo of the activation by thermal neutrons. The $Dy_2 O_3$ was
with a little shellac onto a dtinnen Al plate (0 = 5 cm) and covered with Al foil. By means of Al-wire it could be attached to the bottom of the spherical closing cylinder and exposed at different heights of the $D_2 O$ sphere.


c) **Ionization chamber**

The small absorption effect to be expected requires great accuracy, so for the main measurement no /J-counting tube was used, but an ionization chamber.   With this !ass it is easy to use large /J intensities, so that any statistically induced inaccuracy becomes small (Fig. 1).
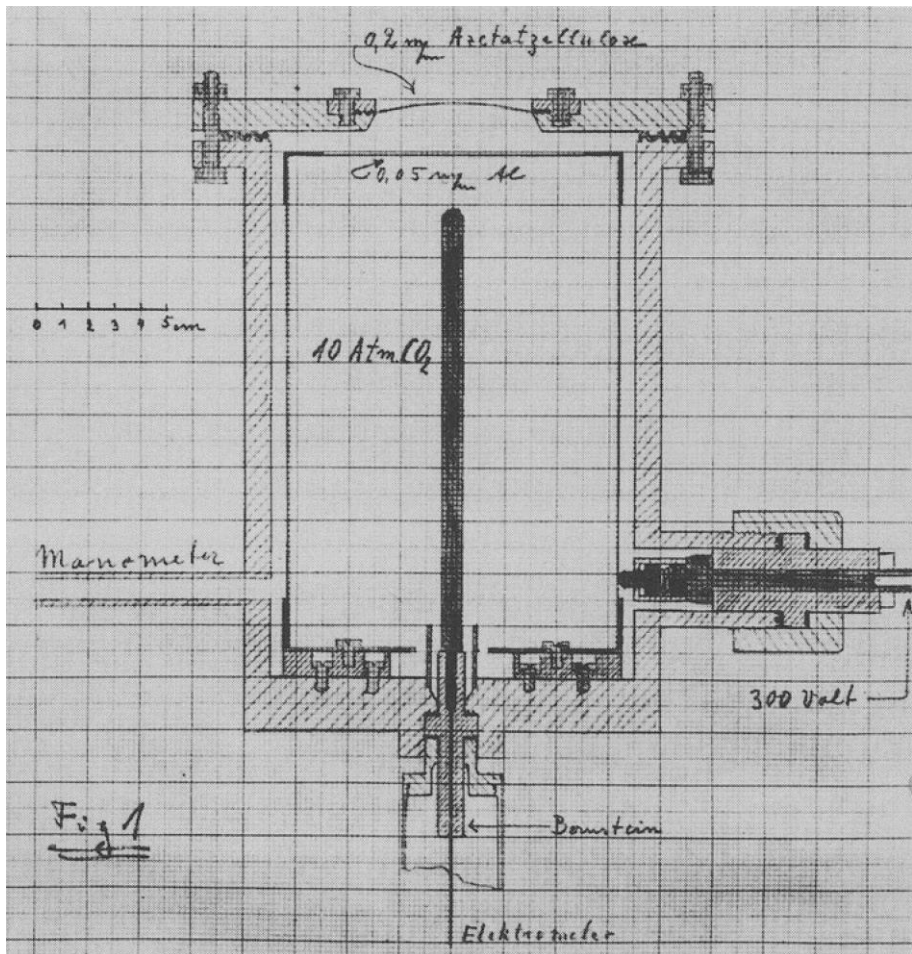
5 The ionization chamber consists of a pressure vessel filled with 10 Atii CO_2 - At the top it is <lled by a 0.2 mm thick acetate cellulose foil, diameter 6 cm, which is <lled by 6 steel wires about 1 mm thick, covering at most 80Jo of the total surface. The foil absorbs 750Jo of the striking Dy-P-rays; it was renewed every 8 days and is under these circumstances

never burst[2] . The actual ionisation chamber is located in the pressure chamber, covered at the top by aluminium foil (d = 0.05 mm). The chamber voltage was 300 volts (dry battery).

d) Charge measurement

The internal charge was determined with a Wulf's electrometer (1 scale division 2 - 10-[2] volts) by means of compensation (Fig. 2).

Fig. 2. Schaltschema
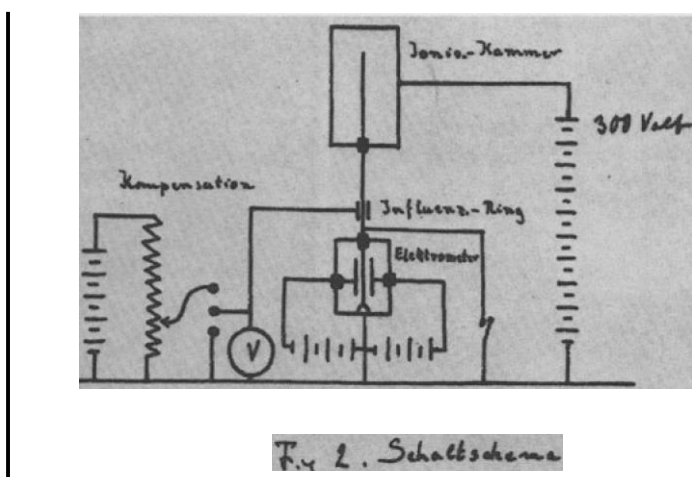
6 The electrometer filament was charged with continuous compensation until the compensation voltage to be applied always reached the same value. Thus the measurement was reduced to a pure time comparison measurement. In order to be independent of any changes in the sensitivity of the overall arrangement, the Dy activity was always divided by the activity of a calibrated Th-P calibration preparation.

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[2] However, difficulties were encountered with another consignment of acetate cellulose film, which we used in a subsequent control measurement, so that we had to take the film twice in this last measurement.

3. Course of a measurement and evaluation

The Dy-indicator was exposed overnight for 15 hours. Then first the residual current was measured, which the ionization chamber supplied even without additional irradiation, subsequently determined by the ion yield of the Th calibration preparation, during which time (approx. 45 **min.)** the also excited activity of the Dy carrier consisting of Al could completely decay. Only then was the Dy activity measurement started. Because of the temporal decrease of this activity, the compensation apparatus had to be controlled by hand while continuously observing the electrometer filament until a selected compensation voltage of 15 or 60 volts was reached. Depending on the time of the measurement, the place of exposure, etc., this was done in 2 to 12 minutes; then the dy-activity measurement was repeated several times and observed for about an hour. Then the above residual current and calibration preparation measurements were repeated until their errors were close to those of the Dy measurement. The evaluation was done in the following way: From the compensation voltage required for a given chamber irradiation time was subtracted that voltage which would have been necessary to compensate for the residual current during that time. The remaining voltage divided by that time is a measure of the Dy activity at the time of measurement; Dy activities thus determined at successive times were reduced to a certain time after the conclusion of the exposure by means of the Dy decay time resulting from all activities and averaged. This mean
was diverged by Th oak activity determined in the same way.   7

4. Results

a) With the dysprosium indicator of 0.08 g $Dy_2 O_3$ the following values were found for the ratio of the intensity to the intensity of the calibration preparation (*r* distance of the indicator center from the center of the sphere):

Table 1

	Al without Cd		Al with Cd		Difference	
r=	0.9 cm	10.5 cm	0.9 cm	10.5 cm	0.9 cm	10.5 cm
1. mef3 series	1,357 ±0,0057	1,221 ±0,0045	0,2271 ±0,0027	0,0801 ±0,0022	1,130 ±0,0064	1,141 ±0,0050
2. mef3 series	1,350 ±0,0083	1,213 ±0,0060	0,2241 ±0,0028	0,0731 ±0,0024	1,126 ±0,0088	1,1395 ±0,0065
3. mef3 series	0,1355 ±0,00060	0,1209 ±0,00045	0,02498 ±0,00035	0,00905 ±0,00031	0,11049 ±0,00069	0,11184 ±0,00054

The individual values of the three Me/3 series cannot be compared directly with each other, since slow changes in the indicator were observed.

which perhaps indicate the faint echo of an activity of very long lifetime, and since a different calibration device was used for the third series. The ratios of the differences are, however, comparable:

Table 2

	1. mef3 series	2. mef3 series	3. mef3 series	Mean value
Diff. ($r = 10.5$) Diff. ($r = 0.9$)	$1{,}00975 \pm 0{,}0072$	$1{,}0120 \pm 0{,}0097$	$1{,}0122 \pm 0{,}0080$	$1{,}0111 \pm 0{,}0048$

Thus, from the three Mel3 series:

$$\frac{\text{Difference } (r = 10.5=)}{\text{Difference } (r = 0.9)} \quad 1.0ll \pm 0.0048.$$

b) This result cannot yet be used directly to determine the diffu-

8 The neutron distribution (2) can be used as a basis for the neutron distribution, since certain corrections have to be taken into account beforehand. The neutron distribution (2) can still be disturbed <by

1) The absorption of thermal neutrons in the Ra-Be source.
2) Absorption in the indicator.
3) By deviations from the spherical symmetry, which are caused by the neck of the bottle, for example.

The experiment was set up from the outset in such a way that the three corrections remained as small as possible. To determine corrections (1) and (2), a control measurement was made using a dysprosium indi cator about 6 times stiirer; at the same time, the Ra-Be source in nickel, where the main absorption was probably due to the nickel groove, was surrounded with a stiirer nickel groove, so that the total amount of nickel was also increased about 6 times. With this approximately 6-fold amount of Dy and Ni, the following values were found:

Table 3

$r =$	0.9 cm	10.5 cm
Al without Cd	$7{,}6750 \pm 0{,}0212$	$7{,}3517 \pm 0{,}0233$
Al with Cd	$1{,}1159 \pm 0{,}0048$	$0{,}4298 \pm 0{,}006$
Difference	$6{,}5591 \pm 0{,}022$	$6{,}9219 \pm 0{,}023$

For the ratio of the difference now results:

$$\frac{\text{Difference } (r = 10.5=)}{\text{Difference}} \quad (r = 0.9)$$

$1,055 \pm 0,005$

Theoretical considerations show that the absorption in the Ra-Be source should lead to an increase, the absorption in the indicator to a decrease of the ratio of the differences[3]. The experimental event shows that the influence of the absorption in the source is predominant.

The deviations from the spherical symmetry (3) were kept as small as possible, and the intensity measurements were always made in the lower sphere.

halves were carried out. In a control measurement at $r = 10.5$ cm I was found in the sphere 9

from Al without Cd, the neck of the bottle was used once without Cd and once *with* Cd. Even such a gross deviation from the kygel symmetry resulted in a variation of only a few percent. Even if the influence is considerably greater at 0.9 cm, the errors caused by the small deviations from the spherical symmetry can therefore be completely neglected.

If one extrapolates from the two measurements Table 2 and Table 3 to the decreasing amount of $Dy_2 O_3$ and Ni, one obtains the final value for the ratio of the differences:

$$\frac{\text{Difference } (r = 10.5 =)}{\text{Difference } (r = 0.9)} \quad 1{\cdot}0023 \pm 0{,}005 \tag{4}$$

fiir vanishing Ni- and Dy_2O_3 quantities. Thus it follows from (2):

$$\frac{r^2}{l^2} = 0{,}0138 \pm 0{,}030 \tag{5}$$

The error entered here is to be understood as the mean error. If we calculate the diffusion length l from (5), we can conclude that l is probably about 90 cm. A value of l smaller than 39 cm (which corresponds to twice the mean error) can be regarded as very improbable, but even this value is still higher than the previously estimated value of 34 cm.

If one calculates from the diffusion length or from (5) the sum of the capture cross sections of the absorbing atoms (referred to an O atom, i.e. $a = a_0 + t_{a0} + $ aver impurity), one obtains, if one assumes the free path length of the thermal neutrons for elastic collisions to be 1.7 cm (cf. Report I, p. 10, Tables 2 and 3):

$a = 0.0767 (r/l)^2 \cdot 10^{-24} cm^2$ so

$a = (0{,}0011 \pm 0{,}0023) \cdot 10^{-24}$ cm^2 -

In report I the value $a = 0.0075 \cdot 10^{-24}$ cm^2 was assumed.

5. Summary

By the experiments described, it is found that the diffusion length in O_2O is probably considerably higher, and the absorption thus considerably lower, than was assumed in Report I of the one of us. From this it must be concluded that

It can be concluded that, if the data hitherto assumed about the prepa rat 238 are correct, *a machine for the production of energy can be built from D;{) and 238.* The results of [Section] 4 even leave open the possibility that D and O are practically not absorbed at all, but rather that all the absorption we measured was due to the contamination of D20 by similar water (about 1/2 OJo $_{H20}$). In fact, contamination by 1/20/o H20 and disappearing absorption in D20 gave *a* value of $0.0012 - 10-24$ [cm2].