Measurements of D$_2$O-concentration in D$_2$O-H$_2$O mixture using neutron radiography technique

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This study is based on the optical density results measured from neutron radiographs of different samples of light water (H$_2$O) and heavy water (D$_2$O), taken separately, and also in the D$_2$O-H$_2$O mixture. A calibration curve was drawn through the results, extracted from a series of standard mixtures of D$_2$O-H$_2$O, with the known D$_2$O concentration. The scale along the abscissa is to be explained as percentage of deuterium atoms present in the liquid compared with all atoms of hydrogen isotopes present. It may be concluded that, as D$_2$O concentration added to light water increases, the optical density also increases, i.e., optical density is a function of D$_2$O concentration. Neutron radiographs were then taken from two other samples of D$_2$O-H$_2$O mixture with different and unknown D$_2$O concentration. Concentration of D$_2$O was determined using the calibration curve via measurement of average optical density values.

[Keywords: Neutron radiography technique, Radiography technique, D$_2$O concentration]

1 Introduction

Neutron radiography (NR) is a technique for making a radiograph of the internal details of an object by the selective absorption of a neutron beam by it. It is a non-destructive testing (NDT) technique, which is complementary to other NDT techniques, such as, X-ray and gamma ray radiography. Neutron radiography has some extra advantages such as, it can detect hydrogenous materials very efficiently due to a large neutron cross-section of hydrogen. This advantage and a few other ones have made it useful for various purposes. Radiography is commonly used to detect voids and inclusions within a material and so, it is often necessary to know the minimum size of void that can be detected. Using this technique, Peter was able to produce several radiographs of different objects from the Forschungsanstalder Deutschen Reichspost. A few authors have investigated the diffusion of hydrogen in metals using this method.

A neutron radiographic method to study the motion of hydrogen in metals was developed, in addition to the existing methods, because it promised to be a technique for measuring the hydrogen distribution in the sample directly. By such a method, the motion of hydrogen can be observed directly, and this leads to a very direct determination of the diffusion coefficient. In 1968, the first neutron radiographic measurements of the mutual diffusion of light water and heavy water were performed by a few scientists. Also, several other workers have measured the self-diffusion coefficients of ordinary water, in heavy water, using NR method. In the present work, the authors have studied the measurements of D$_2$O-concentration in D$_2$O-H$_2$O mixture, using NR technique.

2 Experimental Procedure

In the present study, the tangential beam port of the 3 MW TRIGA MK-II research reactor of AEERE, Savar, Dhaka, has been used as a source of neutrons. The tangential beam port has been chosen for neutron radiography, because the neutron beam coming out of this port contains lower amount of gamma rays compared to other three ports.

Direct film neutron radiography technique has been adopted in the present study. In the NR camera, gadolinium foil of 25 $\mu$m thickness was
used as converter and Agfa structurix D4, DW industrial X-ray films were used as detector in the experiment. The films have emulsion on one side only. The camera is a light-tight device for holding film and converter foil in close contact during exposure. During loading of the film and converter foil in the NR camera, the emulsion surface of the film was kept in contact with the converter foil. The camera was then closed tightly. All these procedures have been done in the dark room. Then sample containers made of aluminium having the same diameter (1.5 cm) were placed at the surface of the NR camera by aluminium tape. The camera was then placed at the camera holder table across the neutron beam.

To find out the optimum irradiation time of the samples, a series of experiments were performed with different exposure time. To do these experiments, the reactor was operated at 250 kW and the neutron flux at the radiography plane was $1.13 \times 10^6 \text{ n.cm}^{-2}\text{s}^{-1}$. Measured values of $L/D$ ratio and the Cd-ratio of the beam at the neutron radiography plane are 60 and 10.51, respectively.

The irradiated film was separated from the cassette in the dark room after irradiating the sample. Post-irradiation procedures were then adopted, which include, developing, washing, fixing, final washing, drying, etc. These were carried out to make the image of the irradiated sample visible.

Details of the NR facility of the present work have been found elsewhere [2-3]. Heavy water and light water samples have been used in the present experiment. In the experiments, the first sample was a single component liquid, like heavy water ($D_2O$) and the second sample was also a single constituent, namely, light water ($H_2O$). The third to tenth samples were mixtures of (i) known (but same amount) concentration of $D_2O$ (1 cc) and $H_2O$ (1 cc), in which case, diffusion time was 16 min and (ii) unknown and known (but different) concentration of $D_2O$ (like: 18%, 23%, 28%, 33%, 38%) and $H_2O$, in which case, the diffusion time was kept at 16 min. In each case, for the preparation of the complex samples, the light water ($H_2O$) was first put into the container and then $D_2O$ was added, very carefully, on the top of the $H_2O$ with the help of a syringe. Normally, one may proceed using a suitable vessel containing the first liquid ($H_2O$) and then carefully adding $D_2O$ into the same vessel without stirring it. Before or after taking the sample in the container, the film was loaded in the radiography cassette and closed tightly. To expose the sample in every case, NR cassette was placed in the cassette holder table of the NR facility on the neutron beam. Here, exposure time depends mainly on the intensity of the neutron beam, density and thickness of the sample and neutron cross-section. So, to find out the optimum exposure time of each of the samples, a series of experiments have been performed with varying exposure time. Optimum exposure time so measured was 8 min.

<table>
<thead>
<tr>
<th>Sample No.</th>
<th>Average optical density of the film at the different regions</th>
<th>Average optical density of the film</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$U_{av}$</td>
<td>$M_{av}$</td>
</tr>
<tr>
<td>1</td>
<td>1.103</td>
<td>1.103</td>
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<tr>
<td>2</td>
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<tr>
<td>3</td>
<td>0.948</td>
<td>0.907</td>
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<tr>
<td>4</td>
<td>0.964</td>
<td>0.967</td>
</tr>
<tr>
<td>5</td>
<td>0.968</td>
<td>0.976</td>
</tr>
<tr>
<td>6</td>
<td>0.969</td>
<td>0.975</td>
</tr>
<tr>
<td>7</td>
<td>0.995</td>
<td>1.004</td>
</tr>
<tr>
<td>8</td>
<td>1.037</td>
<td>1.051</td>
</tr>
<tr>
<td>9</td>
<td>0.976</td>
<td>0.990</td>
</tr>
<tr>
<td>10</td>
<td>1.018</td>
<td>1.022</td>
</tr>
</tbody>
</table>

After exposure to each sample, the irradiated film was developed, washed, fixed and finally washed. Kodak D-19 developer and Kodak Unifix powder were used as developer and fixer in the experiment. The optical density values of radiographs of the samples were measured using a digital densitometer (Model 07-424, S-23285, Victoreen, Inc., USA). Variation of optical density ($D$) values has then been studied, using the following relation:

$$D = \log \left( \frac{A_0}{A} \right)$$

Here, $A_0$ = response of densitometer without the image

$A$ = response of densitometer with the image

Knowledge, so gathered, has been adopted for finding out unknown $D_2O$ concentration in the
mixture of heavy water in light water. The results are shown in Tables 1 and 2.

Table 2 — Background optical density of the film for samples (1-10)

<table>
<thead>
<tr>
<th>Sample No.</th>
<th>Average background optical density of the film at the different regions</th>
<th>Average background optical density of the film</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Up_{ film }</td>
<td>Mid_{ film }</td>
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<tr>
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<td>1.418</td>
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<td>1.402</td>
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<tr>
<td>10</td>
<td>1.554</td>
<td>1.550</td>
</tr>
</tbody>
</table>

3 Results and Discussion

An exchange of the $^1$H and $^2$H (i.e., D) in the molecules plays a significant role in the diffusion process. Many liquids contain hydrogen in a chemical bond that has been detected by NR technique. Deuterium atoms may replace the hydrogen atoms in one of the liquids; as a result, the mutual diffusion is observed between the two liquids. The experimental results of the average density of the upper, middle and lower areas for the samples and the background of the film are shown in Tables 1 and 2. Non-variation of the results in sample 1 indicates that, the present radiography results are reliable. Constant optical density values at all places in sample 2 prove that, in this sample, the same molecules are uniformly distributed. For sample 3, increase in optical density is noted in the lower area of the radiograph.

Accordingly, a decrease in optical density in the upper area is observed. This result indicates diffusion of D$_2$O into the lower area of the mixture. The observed density values at upper, middle and lower areas for sample 4 are different. This variation indicates occurrence of the diffusion processes. The results of such measurements show that some molecules of the two liquids mutually diffuse. To be even more specific, some deuterium molecules diffuse toward the lower region. The radiograph allows one to differentiate the two liquids from each other, but the results specify the presence of D$_2$O and H$_2$O. The obtained results of the three areas for the sample 5 show a variation, which indicates occurrence of diffusion among the areas. The average optical density of the total points is larger than that of sample 4. This increase occurs due to the increase in D$_2$O concentration. In case of samples 6, 7 and 8, one can observe an increase in optical density for each sample, compared to the previous one, and this has happened due to further increase in D$_2$O concentration. The average optical density values of the three areas for the samples 9 and 10, show that, some molecules between the two liquids mutually diffuse.

![Optical density versus D$_2$O concentration (calibration curve)](image_url)
It is important to note that in Fig. 1, the abscissa does not represent the actual concentration of D$_2$O present in the liquid. This concentration is lower, due to the dynamic equilibrium:

$$\text{D}_2\text{O} + \text{H}_2\text{O} \leftrightarrow 2\text{HDO}$$

Hence, the scale along the abscissa has to be explained as percentage of deuterium atoms present in the liquid compared with all atoms of hydrogen isotopes present. So, it may be concluded that, if D$_2$O concentration added to light water increases, the optical density also increases (calibration curve). That is, optical density is a function of D$_2$O concentration. So, the unknown concentration of D$_2$O in D$_2$O-H$_2$O mixture can be easily calculated from the calibration curve of known samples. Samples numbered 9 and 10 contained unknown D$_2$O concentration in the mixture of D$_2$O-H$_2$O. The optical density values of these two unknown samples as measured from the curve in Fig. 3 are 0.989 and 1.028 for the D$_2$O concentration 32.5% and 35.5%, respectively. Additionally, it may be mentioned that, if diffusion time is increased, the two liquids in the mixture could be separately visualized in the neutron radiographs.

References