Holographic Interferometry Measurement of Diffusion Coefficient

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Abstract
Holographic Interferometry has considerably broadened our traditional perception of Interferometry. It is concerned with the formation and interpretation of fringe patterns, which appear when a wave generated at some earlier time and stored in a hologram is later reconstructed by interfering with a comparison wave. Diffusion in transparent mixtures can conveniently be studied by means of Holographic Interferometry. I have studied the diffusion process of 0.5 N and 1 N methanol solutions. I have obtained distinct and well resolved interference fringes that are used for determining diffusion coefficients. It is observed that as the concentration of solution decreases the values of diffusion coefficients increases by very small amount. As diffusion progresses the diffusing boundary between the two solutions increases. The method gives reasonable agreement with the other methods.

Key words: Diffusion coefficient, holographic interferometry, interference fringes.

1. Introduction
Diffusion is the tendency of molecules to migrate from a region of high concentration to a region of lower concentration and is a direct result of Brownian motion. Diffusion is the movement of the molecules due to their thermal energy under the influence of concentration gradient, temperature and electrical potential. The coefficient of diffusion of some substances in a fluid can be predicted theoretically or found from available experimental correlations [1-3]. Diffusion in transparent mixtures can conveniently be studied by optical Interferometry [4-5]. A number of investigators have used real time Interferometry to calculate diffusion coefficients of various mixtures from data obtained in diffusion experiments [6-8]. The diffusion coefficient of the potassium dihydrogen phosphate in water was investigated by using Holographic Interferometry [9]. The Holographic Interferometry technique is an advanced optical tool used to investigate liquid media.

In this paper, I have reported on the use of Double Exposure Holographic Interferometry technique to obtain the diffusion coefficient of methanol in distilled water. This technique involves the measurement of the distance between two peaks in the concentration profile as shown in Fig. 1. The advantage of this approach is that it does not depend on the identification of the initial boundary between the solutions. By measuring only three parameters and substituting it in a mathematical formula allows immediate deduction of the diffusion coefficients of Methanol solution.

2. Basic Principles of Diffusion Process in Liquids
Consider a free diffusion process with a diffusion coefficient (D) independent of concentration (c). According to the Fick’s second law, which can be expressed for one-dimensional diffusion (along the x-axis), as in reference [10-11].
\[
\frac{\partial c(x,t)}{\partial t} = D \frac{\partial^2 c(x,t)}{\partial x^2} \tag{1}
\]

The solution of Eq. (1) for two binary liquid mixtures initially \((t = 0)\) separated at \(x = 0\) with concentration \(c_1\) and \(c_2\) is \([1]\).

\[
c(x,t) = \frac{c_1 + c_2}{2} + \frac{c_2 - c_1}{2} \text{erf}\left(\frac{x}{2\sqrt{Dt}}\right), \tag{2}
\]

where the error function \(\text{erf}\) is defined as \([12]\)

\[
erf(u) = \frac{2}{\sqrt{\pi}} \int_0^u \exp(-\eta^2) d\eta.
\]

In the diffusion cell, the refractive index \((\eta)\) can be treated as a linear function of the concentration, call \(c(x, t)\), especially when the concentration gradient is small, then, as a first approximation, we can write \([11]\).

\[
n(x,t) = \left(\frac{dn}{dc}\right)_0 c(x,t) = n_0,
\]

where \(\left(\frac{dn}{dc}\right)_0\) is the mean value of the derivative \(\frac{dn}{dc}\) in the applied concentration range and \(n_0\) is a constant.

The change in the index of reflection \(\Delta n[(x,t_1,t_2)]\) as a function of \(x\) for two given times \(t_1\) and \(t_2 > t_1\), is

\[
\Delta n(x,t_1,t_2) = n(x,t_2) - n(x,t_1) = \left(\frac{dn}{dc}\right)_0 \frac{c_2 - c_1}{2} \left[\text{erf}\left(\frac{x}{2\sqrt{Dt_2}}\right) - \text{erf}\left(\frac{x}{2\sqrt{Dt_1}}\right)\right]. \tag{4}
\]

Equation (4) is plotted in Fig. 1. The curve has two characteristic extremes. Their positions, call \(x_A\) and \(x_B\), may be found from the condition,
Using Eqs. (4) and (5), we can write

\[
\frac{\partial}{\partial x} \left[ \text{erf} \left( \frac{x}{2\sqrt{D t_2}} \right) \right] - \frac{\partial}{\partial x} \left[ \text{erf} \left( \frac{x}{2\sqrt{D t_1}} \right) \right] = 0
\]

Therefore

\[
\frac{\exp \left( -\left( \frac{x}{\sqrt{4Dt_2}} \right)^2 \right)}{2\sqrt{Dt_2}} = \frac{\exp \left( -\left( \frac{x}{\sqrt{4Dt_1}} \right)^2 \right)}{2\sqrt{Dt_1}}
\]

Taking the logarithms of the left and right hand sides of Eq. (7) we obtain

\[
-\left( \frac{x}{2\sqrt{Dt_2}} \right)^2 - \ln(2\sqrt{Dt_2}) = \left( \frac{x}{2\sqrt{Dt_1}} \right)^2 - \ln(2\sqrt{Dt_1}).
\]

from which

\[
x^2 = \frac{2D\ln(t_2/t_1)}{(t_1/t_q) - (t_2/t_q)}.
\]

Hence we have

\[
x_A = \left[ \frac{2D\ln(t_2/t_1)}{(t_1/t_q) - (t_2/t_q)} \right]^{1/2}
\]

and

\[
x_B = -\left[ \frac{2D\ln(t_2/t_1)}{(t_1/t_q) - (t_2/t_q)} \right]^{1/2}
\]

In this way we obtain the separation of the two extremes on the x axis:

\[
\omega = x_A - x_B = 2\left[ \frac{2D\ln(t_2/t_1)}{(t_1/t_q) - (t_2/t_q)} \right]^{1/2}
\]

And the diffusion coefficient is given by,

\[
D = \frac{\omega^2}{8\ln(t_2/t_q)}
\]

This formula has several important features. To calculate the diffusion coefficient it is sufficient to measure the distance between the two extrema i.e. \(\omega\) and the times \(t_1\) and \(t_2\). There is no need to establish the wavelength of light, thickness of the cell, concentration of solutions, values of \(dn/dc\) and the rigorous location of the initial boundary between the solutions. Since the refractive index at different planes in the cell is different, the light passing through the cell will have different optical paths at different planes given by [13],

\[
\Delta_1(x_1) = n_1(x_1)l, \Delta_2(x_2) = n_2(x_2)l, \ldots
\]

where \(\Delta_1, \Delta_2\) are the optical paths of light rays through different planes situated at \(x_1, x_2, \ldots\) and \(n_1, n_2, \ldots\) are the corresponding refractive indices of the planes and \(L\) is thickness of the cell.

The local correlation of laser speckle images can be used to determine \(\omega\) without employing interferometry techniques. This leads to a simplification of the experimental procedure.

3. Experimental Procedure

A schematic diagram of the optical system is shown in Fig. 2. The system under investigation was placed in the glass cell of the dimensions 1 x 1 x 4.2 cm³. The longer dimension gave the direction of diffusion. Light from a 5 mW He-Ne laser (632.8 nm and 2 mm diameter) was used for double exposure experiments. The beam from the source was incident on beam splitter BS. 50% of light was transmitted through BS and incident on highly reflecting mirror M₁ and 50% was reflected and incident on another highly reflecting mirror M₂. The beam reflected from M₁ was expanded using a spatial filtering assembly SF₁ and was collimated using lens L₁. The collimated light was then allowed to pass through the cell containing diffusing solution. The reflected beam from mirror M₂, was expanded and filtered using spatial filtering assembly SF₂ and
allowed to fall directly on the hologram plane through lens \(L_2\). The path lengths of object and reference beam are made exactly equal.

The cell is filled through a capillary tube from the bottom to minimize turbulence and mixing. The experimental cell is connected via a thin flexible tube to a glass cylinder with a valve at the centre of tube. First the lighter fluid is put into the cell. Next the denser liquid is introduced slowly from below, lifting the lighter liquid without convection movements. This filling procedure was developed by Gabelmann-Gray and Fenichel [14]. I have studied the solution of methanol in pure distilled water to observe the diffusion process at 25°C.

I have studied the diffusion process of 0.5 N and 1 N methanol solutions. After forming the interface level between methanol solution and distilled water, the diffusion process started. The first exposure was taken at 9000 seconds and second exposure was taken at 9900 seconds.

All the holograms were recorded on the 8E 75 HD Agfa holographic plates having dimensions 1 cm \(\times\) 1 cm \(\times\) 0.2 cm and the exposure time taken for each exposure was 30 sec. The reconstructed images of the interferograms were captured with high resolution Q-imaging Retiga-1300 CCD camera (Pixel size 6.7 \(\mu\)m \(\times\) 6.7 \(\mu\)m).

4. Results

The fringes are rather broad because the moment of first exposure for each hologram is made when the concentration distribution is already flatter. This is also the main reason for the increase in the distance between fringe extrema. I have recorded
the double exposure holograms of transparent mixtures and studied the process of diffusion in case of methanol solutions.

4.1 Diffusion Coefficients of Methanol

The system was tested with the methanol solution for the two different concentrations viz. 0.5 N and 1 N. Initially the first exposure was taken at relatively greater time i.e. at 9000 sec. The second exposure was taken after 900 sec. i.e. at 9900 seconds. After that the successive double exposure holograms were recorded for every 900 seconds. The 15 minutes time interval was allotted for processing the plate. Figures 3.1 and 3.2 represent the horizontal fringes recorded on 8E75HD holographic plates.

Figure 3.1 (a) gives the interferogram after 9000 sec. of diffusion. The separation between the two extreme points in this is 0.289 cm and its corresponding diffusion coefficient is $1.11 \times 10^{-5}$ cm$^2$/sec.

Fig. 3.1 Photographs of Interferograms for diffusion studies of methanol solution for 0.5 N solution: (a) after 9000 s; (b) after 10800 s; and (c) after 12600 s
Fig. 3.2 Photographs of interferograms for diffusion studies of methanol solution for 1 N solution: (a) after 9000 s; and (b) after 10800 s

<table>
<thead>
<tr>
<th>Interferogram</th>
<th>t₁ (seconds)</th>
<th>t₂ (seconds)</th>
<th>w (cm)</th>
<th>D (10⁻⁵ cm²/s)</th>
<th>Ave. D (10⁻⁵ cm²/s)</th>
</tr>
</thead>
<tbody>
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<td>13500</td>
<td>0.356</td>
<td>1.218</td>
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</table>

N Methanol solution

Table 2

<table>
<thead>
<tr>
<th>Interferogram</th>
<th>t₁ (seconds)</th>
<th>t₂ (seconds)</th>
<th>w (cm)</th>
<th>D (10⁻⁵ cm²/s)</th>
<th>Ave. D (10⁻⁵ cm²/s)</th>
</tr>
</thead>
<tbody>
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<tr>
<td>2</td>
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<td>11700</td>
<td>0.344</td>
<td>1.318</td>
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</tbody>
</table>
The corresponding results are displayed in Tables 1 and 2. In each interferograms all the measurable parameters are measured as described above. For 0.5 N methanol solution the average D values is $1.150 \times 10^{-5}$ cm$^2$/sec while for 1 N solution it is $1.313 \times 10^{-5}$ cm$^2$/sec. These values are in good agreement with the standard value $1.25 \times 10^{-5}$. The result shows that diffusion coefficient is independent of concentration. In each case it shows its value nearly equal to standard value. As diffusion progresses the diffusing boundary between the two solutions increases.

5. Conclusion

In this paper I have presented Holographic Interferometry technique to measure the diffusion coefficient of a transparent methanol solution. This method is relatively easy to implement and this can be performed even with small power lasers. The diffusion coefficient of methanol is independent of the concentration of the solution. The diffusion coefficients can be used to study the purity of the solutions.

References