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# Measurement of mass diffusivity by light streak imaging

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## ABSTRACT

Mass diffusion relates to migration of one species in another and is a consequence of local heterogeneity of concentration distribution and energy. The present study describes an optical method based on the shadowgraph technique for measuring a binary mass diffusivity of a solute in a solvent using a collimated beam of light. The measurement technique exploits the principle that refraction of a light beam due to concentration gradient creates a sharp visible streak in an image. The streak is a bright, visible linear feature in a shadowgraph image and opens up a new method of diffusivity measurement. The movement of the streak with time is due to a finite mass diffusivity of a solute in a solvent. Experiments with glucose, salt and glycerin diffusion in water have been conducted within a time window obtained from the sensitivity analysis. The respective mass diffusivities are determined by the light streak as well as the linearized shadowgraph technique, and compared with the values reported in the literature. The proposed approach promises to yield a simple method of determining mass diffusivity in a variety of binary systems without explicit measurement of the species concentration.

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## 1. Introduction

Determination of mass diffusivity in binary systems is essential for widespread applications such as dispersion of contaminants, mass transfer phenomena with chemical reactions, dissolution process of a solid phase in a solvent, and in many pharmaceutical and biotechnological applications. Furthermore, modeling and analysis of engineering processes often require the knowledge of mass diffusivity. The conventional methods of measuring mass diffusivity in the binary diffusion systems are elaborate and time consuming. Most require the measurement of species concentration that by itself is a difficult task. In addition, measurement techniques often intrude the diffusion process thereby inducing errors in the measurements. Many obstacles in the estimation of mass diffusivity can be overcome by employing optical techniques owing to their inertia free and nonintrusive nature. Moreover, since refractive index is very sensitive to change in density

(or concentration of species), optical techniques allow very accurate determination of concentration field. In this work we demonstrate use of an optical technique based on shadowgraph to determine mass diffusivity of a binary system. While shadowgraph accurately determines mass diffusivity it requires sufficiently powerful laser source and high resolution imaging techniques to precisely measure the refractive index variation in the diffusion field, and is also time consuming. To address this issue we propose a novel method based on light streak imaging which allows rapid determination of mass diffusivity by using commonly available white light and low resolution image grabber.

In the literature many methods have been employed to estimate mass diffusivity. One of the most commonly employed technique is the Stokes type of diaphragm cell wherein quasi-steady state diffusion occurs through a porous diaphragm (Cussler, 2009; Stokes, 1950). Here, diffusion takes place through a porous barrier (diaphragm) connecting two

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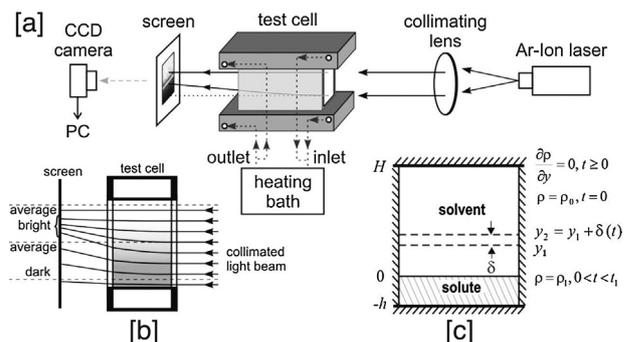
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cells, which are maintained initially at distinct but uniform liquid concentration by stirring. A sintered glass piece or filter paper may be used as diaphragm. After a known time interval, concentration is measured in both compartments, which leads to estimation of mass diffusivity. The decaying pulse technique takes advantage of the transient behavior of a semi-infinite system subjected to a concentration pulse of a limited duration at its boundary (Capobianchi et al., 1998). The mass diffusivity is determined by analyzing the pulse movement against the appropriate analytical solution.

Mass and thermal diffusivity, particularly in biological tissues and porous media have been measured using radiation techniques. These media are non-transparent and the measurements of species concentration and temperature are based on scattering (Blagoveshchenskii et al., 2012; Georgalis et al., 2012). Interferometry [Mach–Zehnder (Ahadi et al., 2014; Nimdeo et al., 2014; Riquelme et al., 2007), Rayleigh (Miller et al., 1988; Zhang and Annunziata, 2008), Gouy (Vitagliano et al., 2005), speckle pattern (Kaunisto et al., 2011), holographic (Axelsson and Marucci, 2008), and Michelson (Abramov et al., 2007)] are optical methods in which an unsteady refractive index profile in a transparent system is measured, thus making it particularly useful for aqueous solutions. Concentration can be easily measured because of a unique relationship between density and refractive index. In a Mach–Zehnder interferometer, a beam of light passes through a density or refractive index field formed by diffusion of solute. Consequently there is a phase lag in the light beam passing through the test cell as compared to the reference beam passing through the reference cell. The phase difference results in interferograms containing fringe patterns leading to concentration/refractive index distribution. Measured concentration distribution in space and time is correlated to mass diffusivity through a one dimensional analytical model. The accuracy of such experiments is high but the method is affected by complexity in experimentation, need for high quality optics, stringent alignment conditions, and high cost.

In a beam deflection technique, the light intensity distribution emerging from the test cell changes with time due to continuous diffusion causing change in the refractive index gradient. Such images can be used to determine the time dependent deflection angle of light at the exit plane of the test apparatus to quantify the mass diffusivity. Optical methods have an obvious advantage of not requiring direct species concentration measurement. However, accuracy of parameter estimation depends on resolution of the intensity measurement and hence the bit resolution of a CCD camera (Ambrosini et al., 2008; Königer et al., 2009; Mialdun and Shevtsova, 2008).

The approach proposed in this study for mass diffusivity measurement is related to the shadowgraph technique. In the proposed technique, a light streak formed on the screen is located and followed in a pre-defined mass transfer process. This information relates in a simple manner with the mass diffusivity. Neither phase nor intensity is required in the data analysis. Hence, unlike measurements based on coherent optics, it does not require a laser, laser-grade optics, or a high resolution detector for light intensity measurement. Streak formation is related to beam deflection and occurs even with a white light source. The entire approach is robust, cost-effective, and is ideal for creating a portable instrument. There are several implications of working with an image feature and not the intensity distribution. The light streak is bright and clearly identifiable. Its position can be determined by feature extraction techniques and light intensity itself need not be



**Fig. 1 – Schematic drawing of (a) Experimental setup for light streak imaging. (b) Passage of a light beam through a field of concentration gradient and formation of the streak of light, and (c) initial condition inside the diffusion cell.**

measured. Therefore, inexpensive detectors such as photodiodes or digital cameras can be used. Moreover, data analysis is simpler than in the original shadowgraph method.

## 2. Description of the diffusion field

The schematic diagram of the diffusion field is shown in Fig. 1c. Here,  $H$  is the height of the solvent placed over the solute having height  $h$ . If a mass diffusion coefficient  $D$  is assumed to be a constant, the time dependent concentration profile along the vertical coordinate ( $y$ ) of the test cell is obtained by applying Fick's second law of diffusion for the density distribution  $\rho(y, t)$  (Bird et al., 2009):

$$\frac{\partial^2 \rho}{\partial y^2} = \frac{1}{D} \frac{\partial \rho}{\partial t} \quad -h < y < H, \quad t > 0 \quad (1)$$

If  $\rho_1$  and  $\rho_0$  are the respective densities of the solute and the solvent, an initial condition is given by

$$\rho = \rho_0 \quad 0 < y < H \quad \text{at } t = 0, \quad (2)$$

and

$$\rho = \rho_1 \quad -h < y < 0 \quad \text{at } t = 0 \quad (3)$$

The boundary conditions at the impermeable top and bottom surfaces of the cavity are

$$\frac{\partial \rho}{\partial y} = 0 \quad \text{at } y = H \quad \text{and } y = -h \quad (4)$$

Eq. (1) can be solved analytically for initial conditions given by Eqs. (2) and (3), and the boundary conditions given by Eq. (4). The solution suggests that for  $t > 0$  the concentration at the interface drops from  $\rho_1$  to  $(\rho_1 + \rho_0)/2$  almost instantaneously and remains constant for a prolonged period of time (Nimdeo et al., 2014). Consequently, the problem can be solved by considering a constant concentration boundary condition at the initial solute–solvent interface ( $y = 0$ ) over  $0 \leq y \leq H$  with a boundary condition given by

$$\rho = \frac{\rho_1 + \rho_0}{2} \quad \text{at } y = 0 \quad \text{for } t \geq 0 \quad (5)$$

Substituting  $\tau = Dt/H^2$ ,  $\eta = y/H$  and  $\theta = (2\rho - \rho_1 - \rho_0)/(\rho_0 - \rho_1)$ , the non-dimensional form of Eq. (1) is obtained as

$$\frac{\partial^2 \theta}{\partial \eta^2} = \frac{\partial \theta}{\partial \tau} \quad \text{for } 0 < \eta < 1 \quad (6)$$

The dimensionless initial and boundary conditions are:

$$\theta = 1 \quad \text{over } 0 < \eta < 1 \quad \text{at } \tau = 0 \quad (7)$$

and

$$\frac{\partial \theta}{\partial \eta} = 0 \quad \text{at } \eta = 1 \quad \text{and} \quad \theta = 0 \quad \text{at } \eta = 0 \quad \text{for } \tau \geq 0 \quad (8)$$

Eq. (6) can be solved analytically by the method of separation of variables leading to the following closed form solution:

$$\theta = \frac{4}{\pi} \sum_{m=0}^{\infty} \left\{ \left( \frac{1}{(2m+1)} \right) \exp \left( -\frac{(2m+1)^2}{4} \pi^2 \tau \right) \sin \left[ \left( \frac{2m+1}{2} \right) \pi \eta \right] \right\} \quad (9)$$

As discussed in Section 3, shadowgraph yields concentration distribution, and its comparison with Eq. (9) gives binary mass diffusivity. We also employ a light streak technique, wherein an approximated but more convenient form of Eq. (9) is compared with the movement of the light streak leading to a rapid estimation of the mass diffusivity.

### 3. Imaging techniques and analysis

Shadowgraph and light streak imaging are discussed in the following sections.

#### 3.1. Shadowgraph

Shadowgraph employs a collimated beam of parallel light rays from an Ar-ion laser, which passes through the solution in a test section and falls on a screen. The image formed on the screen is recorded by a CCD camera. If there are no refractive index gradients, all rays are parallel and the image has a distribution of light intensity identical to that from a pinhole of a spatial filter. In presence of a refractive index gradient, light is deflected from its original path, which creates a dark or bright spread on the image as shown in Fig. 1b.

The passage of a ray of light in a variable refractive index field can be described using Fermat's variational principle (Königer et al., 2009). Under an assumption of small displacement of a light beam inside an inhomogeneous field and outside an apparatus up to a screen, a linearized governing equation of the shadowgraph process can be derived as (Schopf et al., 1996):

$$\frac{I_0(x, y) - I_s(x, y)}{I_s(x, y)} = (ld) \nabla^2 [\log \{n(x, y)\}] \quad (10)$$

where  $I_s$  is the intensity on the screen for an inhomogeneous refractive index field,  $I_0$  is the original undisturbed intensity,  $d$  is the length of the test chamber having fluid,  $l$  is the distance between the screen and the exit window of the apparatus, and  $n(x, y)$  is the refractive index at a point  $(x, y)$ . Shadowgraph relies on careful measurement of the light intensity distribution. The light intensity variation over the screen can be measured to recover the refractive index field from Eq. (10).

The refractive index–density–species concentration relation can be used to obtain the spatio-temporal distribution of concentration (Goldstein, 1996). Consequently, in conjunction with an expression for concentration distribution given by Eq. (9), mass diffusivity can be determined.

In principle, the mass diffusivity can be estimated from the entire duration of the experiment. However, over a certain time interval, the gradient of concentration (or refractive index) is more sensitive to variation in mass diffusivity compared to other times intervals. Consequently a fit of Eq. (9) to the experimentally obtained concentration distribution in the highly sensitive time window leads to the best value of the diffusivity. To fix the experimental time window for an estimation of mass diffusivity from the intensity distribution, we conduct sensitivity analysis by integrating the second derivative of density (Eq. (1)) over a time span as follows:

$$S_y(y, D; t) = \int_0^t \frac{d}{dD} \left( \frac{d^2 \rho / dy^2}{dD} \right) dt' \quad (11)$$

Combining Eqs. (9) and (11), the dimensionless form of the sensitivity parameter  $S(\eta, \xi; \tau')$  can be derived as

$$S(\eta, \xi; \tau') = \frac{4}{\pi \tau' \xi^2} \sum_{m=0}^{\infty} \frac{1}{(2m+1)} \sin \left[ \frac{(2m+1)\pi}{2} \eta \right] \times \left\{ -\exp(-M\xi\tau') [M\xi\tau' + 1] + 1 \right\} \quad (12)$$

where  $M = (2m+1)^2 \pi^2 / 4$ ,  $S(\eta, \xi; \tau') = H^2 D_R S_y(y, D; t) / [t(\rho_0 - \rho_1)]$ ,  $\eta = y/H$ ,  $\tau' = D_R t / H^2$ ,  $\xi = D/D_R$ . Here  $D$  is an estimate of mass diffusivity of a binary system being studied;  $D_R$  is a reference diffusion coefficient for any known binary system. To obtain mass diffusivity by the shadowgraph method, Eq. (9) is fitted to the experimental data of density variation in that time window, where the sensitivity function  $S(\eta, \xi; \tau')$  defined by Eq. (12) is high and disturbance initially created due to interface formation is negligible.

While the procedure to obtain mass diffusivity from the shadowgraph aided by sensitivity analysis is rigorous, it is cumbersome to obtain the refractive index by solving the partial differential Eq. (10). Furthermore, Eq. (10) presumes a high resolution image detection setup to record intensity variation in the light beam. Availability of a high resolution image is of great significance in shadowgraph as intensity needs to be integrated twice in order to obtain refractive index at the image pixels. In order to overcome these constraints, we present below a simple approach for determination of the mass diffusivity applicable in an unsteady, one dimensional experiment in liquid-liquid as well as solid-liquid diffusion systems without homogeneous chemical reactions.

#### 3.2. Light streak imaging

The proposed measurement technique relies on an appearance of a bright horizontal light streak on the imaging plane when the mass diffusion process is exposed to a parallel beam of light. The speed of a movement of the horizontal light streak is determined by the mass diffusivity. On recording its position as a function of time, mass diffusivity can be obtained. Since the streak is a zone of high intensity contrast and light intensity itself is not needed to be measured, inexpensive detectors such as digital cameras can be employed to record its

movement. The streak position can be determined by feature-extraction techniques available in the image processing tools. The technique requires a medium to be transparent. This condition is fulfilled in numerous engineering applications, including ionic and non-ionic liquids, radioisotope separation, chemical and colloidal gels, and industrial pastes.

The principle underlying the light streak imaging method is similar to the shadowgraph, as shown in Fig. 1a. Owing to the solute concentration variation in the vertical direction inside a test cell, the incoming light rays bend toward the regions of lower density by refraction. The extent of beam deflection depends on the concentration gradient, length of the apparatus in the viewing direction, and a position of the screen. Usually, small values of mass diffusivity cause large gradients in the species concentration. Consequently, light passing through the zone having a large density gradient bends into a small region, giving rise to the appearance of a light streak. With continuing diffusion, concentration field changes with time causing a movement in a position of the light streak. At long times, as concentration of a solute starts becoming uniform, the light streak broadens and finally vanishes from the screen. Interestingly, light streak is a characteristic feature of mass diffusion process due to the possibility of large gradients in refractive index. The light streak does not appear in the heat transfer experiments because the thermal diffusivity is orders of magnitude greater than the mass diffusivity (Bhandari et al., 2013).

The use of the light streak method to determine mass diffusivity is motivated by computational simplicity and speed of analysis. The formation of a light streak is predicted by the full nonlinear system of shadowgraph equations (Goldstein, 1996; Schopf et al., 1996). Although these equations can be numerically inverted along with the light streak position, it is mathematically cumbersome to obtain diffusivity. Instead, we use dimensional analysis which is simpler and equally effective in estimating diffusivity. For a given diffusion cell and wavelength of light, an instantaneous position of a light streak ( $\delta$ ) above the interface is a function of system variables such as mass diffusivity ( $D$ ), thickness of the diffusing media ( $H$ ) and time ( $t$ ) leading to:  $\delta = \delta(H, D, t)$ . Consequently the dimensional analysis yields two dimensionless groups:  $\delta/H$  and  $Dt/H^2$  leading to the functional form:

$$\frac{\delta}{H} = f\left(\frac{Dt}{H^2}\right) \quad (13)$$

As discussed below we experimentally measure  $\delta$  as a function of time for a solute whose diffusivity is a priori known from the literature. We fit a suitable functional form to the calibration experimental data and obtain parameters of the function. The Buckingham  $\pi$  theorem associated with dimensional analysis compels that the parameters depend on the features of the experimental setup but not on the choice of the solute and the solvent.

#### 4. Apparatus and instrumentation

The experimental setup developed for the shadowgraph as well as the light streak imaging technique is shown in Fig. 1. An optical setup is equipped with a 1 W continuous wave Ar-Ion laser (wavelength = 514.5 nm) as the light source. Independent experiments were also carried out with a white light source to confirm the formation and movement of the light streak. A spatial filter—concave mirror assembly is used to

develop a collimated light beam. The diffusion test cell, optical components and the laser have a common centerline. The collimated beam passes along the length of the isothermal test cell as shown in Fig. 1, wherein mass diffusion is in progress in the vertically upward direction. The refracted light rays fall on a screen that are recorded by a camera. In this work we use a high speed 8-bit Mikrotron MC-1302 CCD camera of spatial resolution of  $640 \times 576$ . The CCD camera is connected to a frame-grabber. For a light streak measurement, a 10 MP digital camera with 6-bit resolution can also serve the purpose.

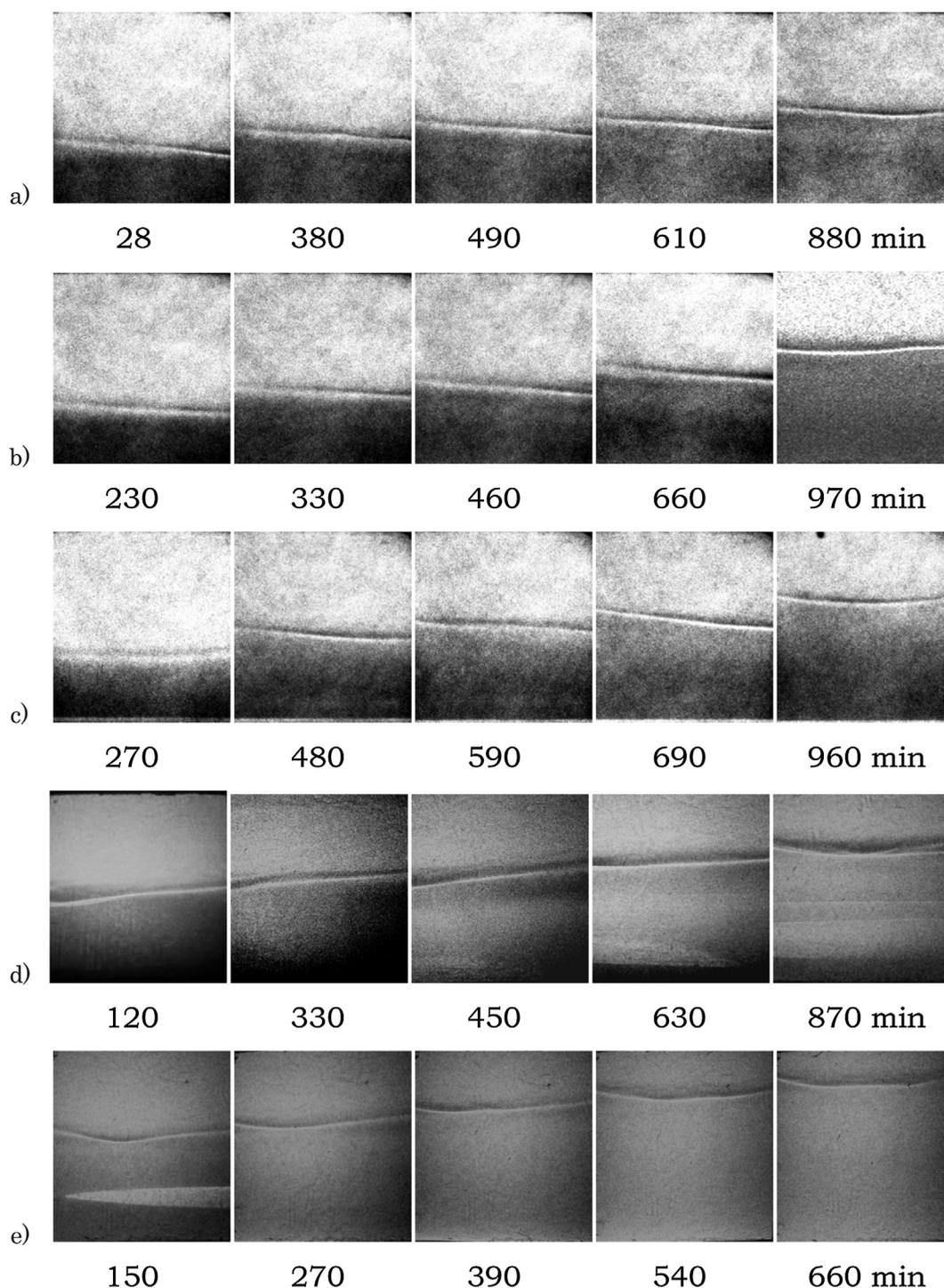
The experimental test cell shown in Fig. 1 consists of three sections, namely the top tank, the test section (cavity) and the lower tank. The test section is rectangular in plan, and is made of two square optical windows. It has a nominal length of 100 mm and a height of 50 mm. The size of the test section in the direction the light beam, namely its length, plays an important role in fixing the sensitivity of concentration measurement. The right side of Eq. (10) has cavity length as a multiplicative factor. Due to greater deflection of the light beam, longer test cells increase the thickness of the light streak in an experiment. The solution is confined by two copper plates of thickness 2 mm above and below the cavity. The test cavity through which a laser beam passes is made of  $50 \times 50 \text{ mm}^2$  optical windows made of glass (Bk-7 material,  $\lambda/4$  surface flatness with 30–40 scratch-dig surface quality). Parallelism of the optical glass windows is established by ensuring that the passage of the light beam remains unchanged with and without the test cell. The top and bottom surfaces of the cavity are kept at uniform temperatures by using constant temperature bath. The wall temperatures are monitored by thermocouples mounted on surface connected to a multi-channel temperature recorder (DBK temperature scanner). The room temperature is maintained constant within  $\pm 0.2 \text{ K}$  during the experiment. The sides of the test cell are covered by 4 mm thick Bakelite sheets which insulate the test section. This arrangement is particularly relevant at elevated temperatures. In addition, the diffusion apparatus is required to be free of external vibrations.

The experiment is initiated by adding a layer of concentrated solution of the solute being studied at the base of the test cell. For the data reported in the present work, the initial solution layer was less than 15% of the cavity height. A time series of the recorded images are shown in Fig. 2. These images reveal a temporal evolution of the horizontal light-streak; however, the same images are also used to carry out shadowgraph analysis. Each snap shot of the image sequence is stored as an intensity matrix of integers in the range 0–255 with the matrix size being  $640 \times 576$ . However, a few columns of intensity data per image is adequate for the analysis. Provided that the light streak has moved sufficiently away from the interface, the initial layer thickness does not influence an estimate of the diffusivity.

In this work we study solutes such as: 10–20 wt% glucose solution, pure glycerin, and 20 wt% NaCl solution. Experiments have been carried out at  $20^\circ \text{C}$  unless specified otherwise.

#### 5. Results and discussion

In Fig. 2 we see shadowgraph images at various times after starting the diffusion experiment in distilled water for the following solute compositions: (a) 10 wt% glucose solution,



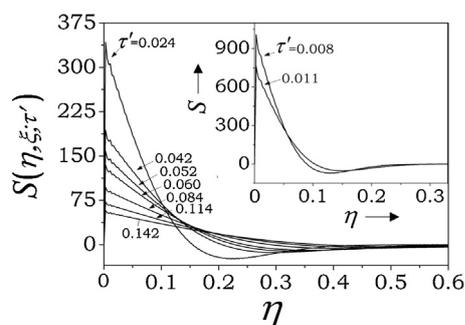
**Fig. 2 – Evolution of the shadowgraphs during mass diffusion of (a) 10 wt% glucose solution, (b) 15 wt% glucose solution, (c) 20 wt% glucose solution, (d) pure glycerin, and (e) 20 wt% NaCl solution in deionized water at various time instants. The light streak contained in the shadowgraph is not strictly horizontal due to the slight non-parallelism of the incoming light beam.**

(b) 15 wt% glucose solution, (c) 20 wt% glucose solution, (d) pure glycerin, and (e) 20 wt% NaCl solution. Images in Fig. 2 also clearly show a formation and movement of the light streak. Before starting the experiments, a time window required to study diffusion of a particular solute, where the sensitivity is the highest is estimated by calculating the sensitivity function using Eq. (12). Since the solutes have distinct diffusivities, sensitivity function allows estimation of that time window, over which images can be analyzed. However, in order to solve Eq. (12), an approximate (having same order of magnitude) estimate of diffusivity is necessary. Such estimate can be obtained by analyzing any shadowgraph image

using Eq. (10). In this work we, use a known value of diffusivity of infinitely diluted KCl solution in water from literature ( $D_R = 1.89 \times 10^{-9} \text{ m}^2/\text{s}$ ) (Weast, 1989) as a reference to estimate the sensitivity function for the shadowgraph measurement.

### 5.1. Shadowgraph analysis

In Fig. 3, we plot sensitivity of the light contrast as a function of position at different time intervals for diffusion of glucose in water. It can be seen that irrespective of time, the contrast sensitivity is high near the solvent–solute interface, and decreases away from it as a concentration gradient decreases. Fig. 3 also

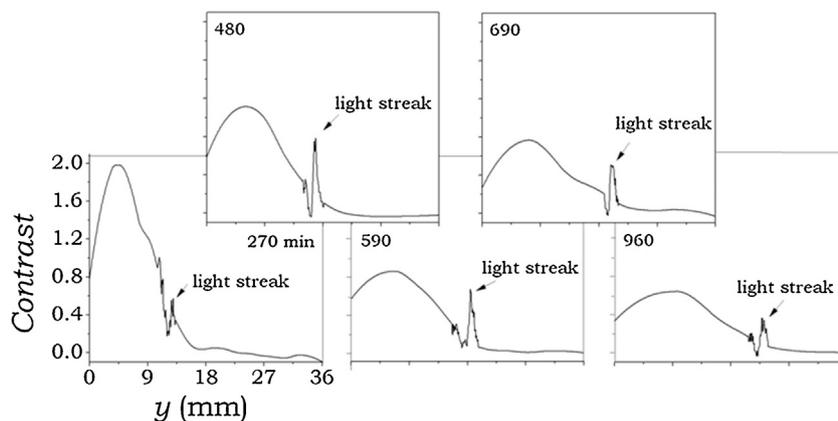


**Fig. 3 – Contrast sensitivity plotted as a function of dimensionless position for diffusion of glucose in deionized water. The inset shows the data of absolute sensitivity with position for short durations ( $\tau'$ ).**

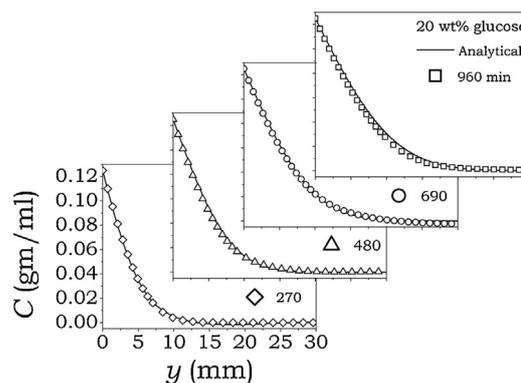
shows that the sensitivity is higher at small times, which can be attributed to the large light intensity contrast at early times. At such early stage of an experiment, slight non-parallelism of the light beam entering the test cell produces substantial error due to interface creation. Hence, the data can be recorded later when an error due to interface creation is eliminated. Therefore we use the data collected between 120 ( $\tau' = 0.011$ ) to 1300 ( $\tau' = 0.114$ ) min for calculating the mass diffusion coefficient.

Data analysis discussed below pertains to shadowgraph imaging carried out within a sensitivity window. Diffusion of a glucose solution with 20 wt% initial concentration into fresh water at 20 °C is first considered. We extract light intensity distribution from the CCD images in order to determine contrast at every pixel in the image along the height of the test cell. Contrast distributions at various time instants are shown in Fig. 4. It can be seen that the light intensity contrast increases from the initial solution–solvent interface, and after achieving a maximum decreases gradually. However the decreasing part of the intensity curve is intercepted by a sharp fluctuation which arises from the bright light streak. It can be seen that a position of the light streak moves upwards continuously as a function of time.

From the contrast data we evaluate the second derivative of concentration by using Eq. (10). The differential equation for density is discretized over the physical domain by a finite-difference method (FDM) with boundary conditions mentioned in Eq. (5). The system of algebraic equations can be solved for the concentration distribution along the height of the test cell and is shown in Fig. 5. The position–concentration data at various time instants shows good fit except at very



**Fig. 4 – Evolution of light intensity contrast as a function of position along the y coordinate of the test cell during diffusion of 20 wt% glucose solution in deionized water at 20 °C.**

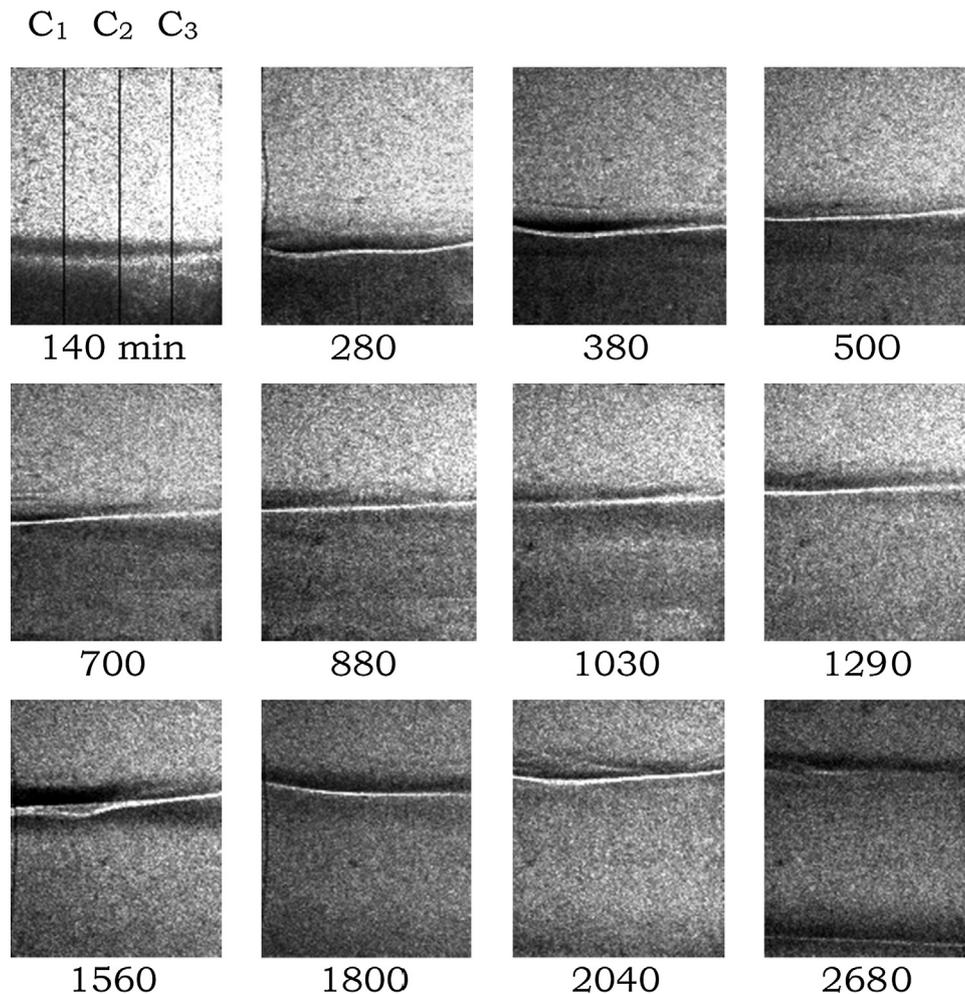


**Fig. 5 – Evolution of concentration  $C$  as a function of position along the vertical coordinate of the test cell during diffusion of 20 wt% glucose solution in deionized water at 20 °C.**

long times when the sensitivity function has diminished substantially, relative to the early time values. Mass diffusivity of glucose in water is then determined by regressing Eq. (9) with the experimental data shown in Fig. 5. The average mass diffusivity for diffusion of 20 wt% glucose solution in water was found to be  $D = 6.86 \times 10^{-10} \text{ m}^2/\text{s}$ , which matches with the value quoted in the literature  $D = 6.73 \times 10^{-10} \text{ m}^2/\text{s}$  (Haynes, 2012–2013).

## 5.2. Light streak method

In the light streak method, a calibration experiment is necessary to obtain functional form associated with expression (13), which facilitates estimation of mass diffusivity. We therefore perform a calibration experiment with 20 wt% sucrose solution in deionized water at 25 °C. The corresponding shadowgraph images collected at selected time instants during the mass diffusion experiment are shown in Fig. 6. It can be seen that the light streak is near horizontal and its movement is in the vertically upward direction. Non-flatness of the light streak arises from slight non-parallelism of the light beam entering the test cell. The light streak is broader at early times, becomes sharper and moves vertically upwards with a passage of time. Later on, the light streak becomes broader again and is immobilized in time. Vertical lines in the first image of Fig. 6 are columns considered for locating the y-coordinate of the light streak. Primary data in the form of position of horizontal light-streak as a function of time are shown in Fig. 7. The

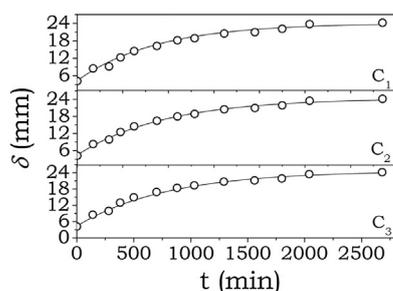


**Fig. 6** – Vertical movement of the light streak recorded as a function of time for a diffusion of 20 wt% sucrose solutions in deionized water at 25 °C. Vertical lines in the first shadowgraph are columns  $C_1$ ,  $C_2$ ,  $C_3$  at which the y-coordinate of the streak position is determined.

plot shows clear movement of the light streak at early time. However, as time passes the streak movement slows down. This behavior of the light streak can be interpreted in terms of sensitivity.

In order to fit the position data we propose a following functional form given by

$$\delta = \delta_0 + a \exp\left(\frac{-bDt}{H^2}\right) \quad (14)$$



**Fig. 7** – Positions of the light streak as a function of time at column  $C_1$ ,  $C_2$  and  $C_3$  during diffusion of 20 wt% sucrose solution in deionized water. Temperature of the apparatus is 25 °C. Open circles are the experimental data while solid lines are the best fit of Eq. (14) to the same.

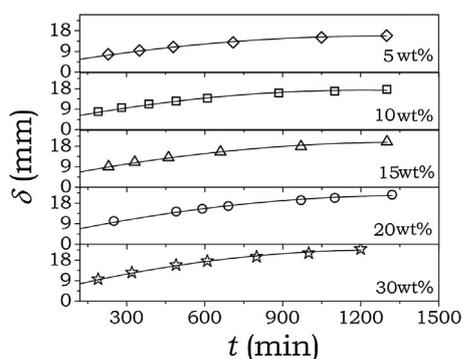
where  $\delta_0$ ,  $a$  and  $b$  are the parameters. We use the known value of diffusion coefficient for sucrose solution in deionized water at 25 °C:  $D = 5.22 \times 10^{-10} \text{ m}^2/\text{s}$  from the literature (Haynes, 2012–2013; Nimdeo et al., 2014). Regression of Eq. (14) against the image data yields the following model parameters for the apparatus used in the present study:  $\delta_0 = 2.485$ ,  $a = -2.0466$ ,  $b = 3.788H^2$ . Although the parameters are apparatus specific, these are independent of the choice of the diffusing species and the solution. In a given setup, these parameters can be used for data analysis with other temperatures, solute–solvent combinations with unknown mass diffusivity.

Subsequent to calibration and determination of constants, the light streak method was applied to several solute–solvent combinations. The extracted position–time data of the light streak over sensitivity time window for various glucose concentrations are shown in Fig. 8. The movement is seen to be greater for higher glucose concentrations. With parameters  $\delta_0$ ,  $a$  and  $b$  determined from the calibration experiment, mass diffusivity of glucose in water has been determined by curve-fitting the data of Fig. 9 with Eq. (14). Fig. 9 summarizes the least squares error for various glucose concentrations at 20 °C with the diffusion coefficient as a free parameter. A clear minimum attained enables unambiguous determination of mass diffusivity.

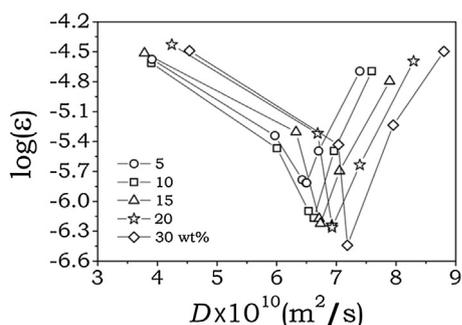
For 5, 10, 15, 20 and 30 wt% glucose solution in deionized water at various temperatures, the mass diffusivity values retrieved are plotted in Fig. 10 and tabulated in Table 1. It can

**Table 1 – Measured mass diffusivities ( $D$ ) for various solute–solvent combinations at temperature  $T$  and their comparison with the literature.**

Name of the technique	Binary system	Weight %	$T$ ( $^{\circ}\text{C}$ )	$D$ ( $\times 10^{-9}$ $\text{m}^2/\text{s}$ )		
Light streak imaging	Glucose + $\text{H}_2\text{O}$	5	20	0.65		
		5	25	0.661		
		5	30	0.676		
		10	20	0.662		
		15	20	0.675		
		15	25	0.705		
		15	30	0.732		
		20	20	0.691		
		30	20	0.718		
		30	25	0.75		
		30	30	0.779		
		Shadowgraphy	NaCl + $\text{H}_2\text{O}$	20	20	1.524
			Glycerin + $\text{H}_2\text{O}$	99.9	20	1.042
Glucose + $\text{H}_2\text{O}$	20		20	0.686		
Taylor dispersion Ribeiro et al. (2006)	Glucose + $\text{H}_2\text{O}$		1.77	25 to 39	0.65–0.92	
Taylor dispersion Haynes (2012–2013)	Glucose + $\text{H}_2\text{O}$		–	25	0.673	
Rayleigh interferometry Rard and Miller (1979)	NaCl + $\text{H}_2\text{O}$		1 to 23	25	1.48–1.57	
Rayleigh interferometry Haynes (2012–2013)	Glycerin + $\text{H}_2\text{O}$		99.9	25	1.064	
M–Z interferometry Nimdeo et al. (2014)	Sucrose + $\text{H}_2\text{O}$		5 to 50	25	0.421–0.521	
M–Z interferometry Haynes (2012–2013)	Sucrose + $\text{H}_2\text{O}$		–	25	0.522	

**Fig. 8 – Position of the horizontal light streak (symbols) along the  $y$  co-ordinate of the test cell (at column  $C_2$ ) as a function of time during diffusion of glucose in deionized water at  $20\text{ }^{\circ}\text{C}$ . The lines through the data are fits of Eq. (14).**

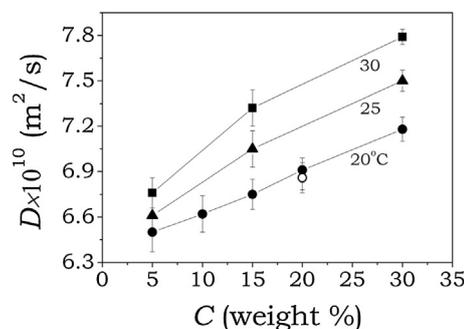
now be inferred that for glucose mass diffusivity increases with an increase in solute concentration as well as temperature. A value of  $D=6.73 \times 10^{-10}$   $\text{m}^2/\text{s}$  at  $25\text{ }^{\circ}\text{C}$  has been reported in the literature (Haynes, 2012–2013) and is consistent with values obtained from the present technique. For 20 wt% NaCl in water, the present approach yielded  $D=1.524 \times 10^{-9}$   $\text{m}^2/\text{s}$ . For glycerin in water, diffusivity was

**Fig. 9 – Variation of logarithmic error during estimation of mass diffusivity of glucose in water at  $20\text{ }^{\circ}\text{C}$  at different solute concentrations.**

estimated to be  $D=1.042 \times 10^{-9}$   $\text{m}^2/\text{s}$ . These are within  $\pm 2\%$  of reported values in the literature as shown in Table 1.

Experiments were also carried out with a halogen lamp as a white light source and a clear light streak was observed (not shown). In view of the wavelength dependence of refractive index, there is a possibility of a beam of white light at normal incidence splitting into colors due to refraction. Refractive index of an aqueous solution is a slightly decreasing function with increasing wavelength. However, white light falls in a narrow range of wavelengths (400–700 nm) and the corresponding extent of variation of refractive index is small, varying from 1.343 to 1.332, with a refractive index of 1.3330 for white light. The shadowgraph measurement relies primarily on the derivative  $dn/dC$ , where  $n$  is refractive index and  $C$  is solute concentration, which shows negligible dependence on wavelength. Hence, wavelength distribution is not expected to affect diffusivity measurement in when white light sources are used in the light streak imaging technique.

From the above discussion it is clear that the light streak method is very simple in instrument arrangement as well as in analysis as compared to shadowgraph. It requires only a

**Fig. 10 – Plot of mass diffusion coefficient of glucose in deionized water at various temperatures and concentrations of glucose solution obtained by light streak imaging technique (filled symbols). The mass diffusivity value obtained from shadowgraph technique for 20 wt% glucose solution at  $20\text{ }^{\circ}\text{C}$  is shown by open symbol.**

position of the light streak and is evaluated easily with image processing tools. On the other hand shadowgraph requires measurement of intensity followed by a solution of a second order differential equation involving measured intensity. Furthermore, the parameters retrieved are sensitive to light intensity fluctuations from the laser and camera noise. This drawback is serious over a long duration experiments. The light streak method overcomes these limitations.

## 6. Conclusions

A new method has been proposed to measure a diffusion coefficient of the transparent and miscible binary liquid pairs. It relies on formation of the light streak, when a beam of light at parallel incidence passes through a medium wherein mass diffusion is underway. In this method, a movement and speed of the light streak along a vertical coordinate of the diffusion cell is monitored. This technique is a non-invasive data generation approach that significantly simplifies the analysis. It can work with readily available components including lasers and white light sources and a digital camera. It is cost effective as compared to the conventional shadowgraph or interferometry techniques that require high power monochromatic laser, laser-grade optics, and a high resolution detector. Mass diffusivity values determined by light streak technique are found to be in good agreement with the literature.

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