

DIFFUSION COEFFICIENT MEASUREMENT BASED ON DIFFUSION-INDUCED FOCUSING IN OPTOFLUIDIC WAVEGUIDE

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ABSTRACT

This paper presents a new optofluidic method to measure the mutual diffusion coefficient in binary solutions by directly analyzing the diffusion-induced focusing pattern based on a single optical micrograph. The average diffusion coefficient of ethylene glycol (EG) and deionized water binary solution is measured under different mass fractions and flow rates to demonstrate the proposed method. Experimental results show good agreement with the reported data and indicate that the mutual diffusion coefficient is slightly decreased with the increase of the flow rate and is inversely proportional to the mass fraction of EG.

KEYWORDS: Diffusion coefficient, Single optical micrograph, Diffusion-induced focusing, Optofluidic waveguide

INTRODUCTION

Diffusion is an elementary phenomenon, which describes the mass transport from the zones of higher concentration to the ones of lower concentration. This phenomenon has drawn great interest since its first discovery in the nineteenth century, not only because it reveals the basic molecular behavior at the atomic level, but also due to its increasing influence in fields such as chemical, medicine and physiology [1].

Due to this interest, different experimental methods are developed to determine the molecular diffusion coefficient, which is an important parameter indicating the diffusion mobility. The conventional measurement methods include total internal reflection fluorescence (TIRF) microscopy [2,3], spectroscopy [4,5] and interferometry [6-8]. The TIRF microscopy approach utilizes TIRF microscope to monitor the position of fluorescence as a function of time and therefore determines the diffusion coefficient of fluorescent molecules. This approach is apparently simple but is limited to fluorescent substance. The other approaches generally measure the concentration change as a function of time by using spectroscope or interferometer and show good accuracy and universality. However, they require complicated experimental setup and procedures.

In this paper, an optofluidic system is designed to measure the average mutual diffusion coefficient in a binary solution directly based on a single optical micrograph by analyzing the diffusion-induced focusing pattern [9] in the microchannel. Ethylene glycol (EG) and deionized (DI) water are used to demonstrate the proposed method.

WORKING PRINCIPLE

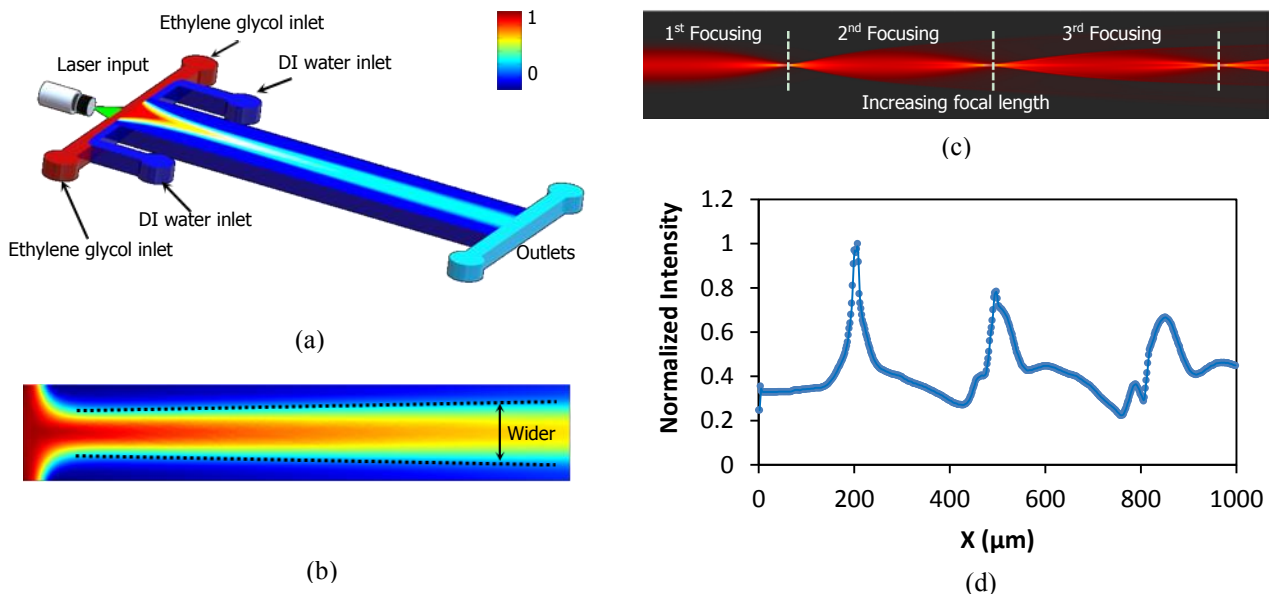


Figure 1: Illustration of the diffusion-induced focusing in optofluidic waveguide. (a) Schematic of the optofluidic microchip for diffusion coefficient measurement, (b) concentration distribution in the microchannel, showing the equivalent waveguide width is getting wider, (c) light propagation pattern, showing light focuses periodically with increasing focal length, and (d) normalized light intensity along the central line of the microchannel.

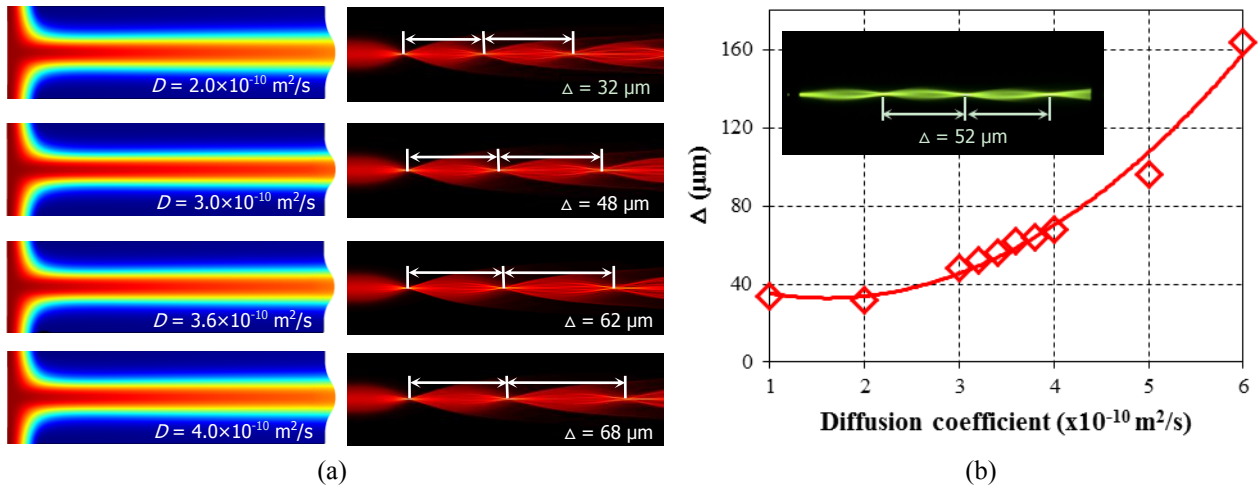


Figure 2: Illustration of the proposed method to measure diffusion coefficient in binary solutions. (a) Simulation results of concentration distribution and light propagation pattern with different diffusion coefficients, and (b) diffusion coefficient vs 2nd and 3rd focal length difference. Inset shows the captured microphoto with a focal length difference of 52 μm .

Figure 1(a) shows the designed optofluidic microchip for the measurement of the diffusion coefficient in a binary solution. The microchip is composed of two EG inlets, two DI water inlets, two outlets, a fiber slot and a microchannel. The dimensions of the microchannel are 1000 (Length) \times 150 (Width) \times 100 (Height) μm^3 . EG (with higher refractive index, RI) and DI water (with lower RI) are injected into the microchannel to form an optofluidic waveguide. With the effect of mutual diffusion between the flows, the RI contrast of the core and cladding streams is reduced along the microchannel, i.e. the equivalent waveguide width is wider as illustrated in Fig. 1(b). The increase of equivalent waveguide width eventually leads to the phenomenon that light focuses periodically with an increasing focal length. As shown in Fig. 1(c), the 3rd focal length is larger than the 2nd focal length and the focal length difference is correlated to the diffusion coefficient of the binary solution. Therefore, the diffusion coefficient can be easily determined by measuring the 2nd and 3rd focal length difference (Δ). Figure 1(d) plots the normalized light intensity of the focusing pattern along the central line of the microchannel. Three peaks, which could be used in the determination of the focal length, are clearly seen.

In order to quantify the relationship of diffusion coefficient and Δ , numerical models are set up in COMSOL 4.3a and Rsoft. Figure 2(a) shows the simulated concentration distributions and light propagation patterns under different diffusion coefficients. When the diffusion coefficient increases from 2 to $4 \times 10^{-10} \text{ m}^2/\text{s}$, Δ increases from 32 μm to 68 μm . The relationship is plotted in Fig. 2(b). In the experiment, an optical micrograph is captured and Δ is measured to obtain the diffusion coefficient based on Fig. 2(b). For example, the inset shows a focal length difference of 52 μm , which corresponds to a diffusion coefficient of $3.2 \times 10^{-10} \text{ m}^2/\text{s}$.

RESULTS AND DISCUSSION

The average diffusion coefficient of EG and DI water binary solution is measured under different mass fractions and flow rates. Figure 3(a) shows the measured average diffusion coefficient of the EG-DI water binary solution as a function of mass fraction with a fixed flow rate of 1 $\mu\text{L}/\text{min}$. The diffusion coefficients of the binary solution with 30%, 50%, and 100% EG are calculated to be $8.1 \times 10^{-10} \text{ m}^2/\text{s}$, $6.6 \times 10^{-10} \text{ m}^2/\text{s}$ and $3.2 \times 10^{-10} \text{ m}^2/\text{s}$, respectively. The results show that the diffusion coefficient is inversely proportional to the mass fraction, which agree with previously reported results [10]. The slight mismatch could possibly be explained by several reasons. Firstly, the reference data is not accurate considering diffusion coefficient is generally within about five or ten-percent accuracy [11]. Secondly, the flow velocity in the 2D numerical model is generally smaller than that in the real case. Therefore, a smaller diffusion coefficient is used in the 2D model to offset the velocity inaccuracy based on our current diffusion coefficient determination strategy. An optimized numerical model could improve the reliability of the results. Thirdly, the instability of flow streams brings some errors to the experiment results. This instability is especially significant with flow rates below 0.5 $\mu\text{L}/\text{min}$. A better pump or an optimized flow rate should be effective to reduce the influence of flow instability.

Figure 3(b) shows the measured diffusion coefficient of the binary solutions under different flow rates with initial ethylene glycol's mass fraction of 0.3 (Triangle), 0.5 (Square) and 1.0 (circle), respectively. Each data point in Fig. 3(b) is the average value of five experiment results. Under the same initial mass fraction of EG, the average diffusion coefficient decreases slightly with flow rates increasing from 1 $\mu\text{L}/\text{min}$ to 2 $\mu\text{L}/\text{min}$. This is caused by the fact that the diffusion time is limited by the higher flow rate, and consequently higher mass fraction of EG remains throughout the microchannel.

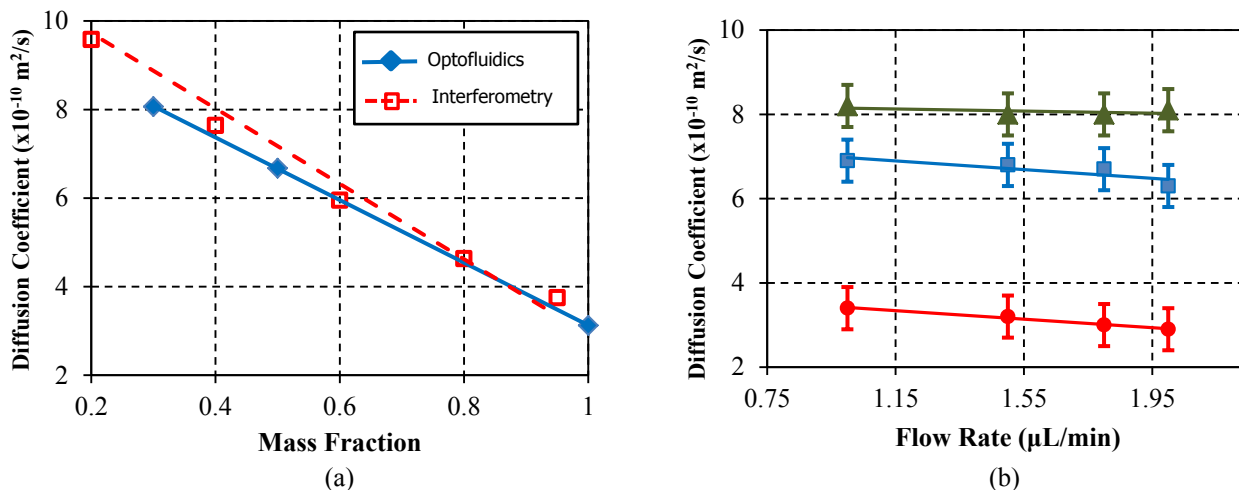


Figure 3: Measured diffusion coefficient of ethylene glycol-water binary solution. (a) Measured relationship between diffusion coefficient and ethylene glycol's mass fraction by using optofluidic platform and interferometry [2], respectively, and (b) measured diffusion coefficients under different flow rates with initial ethylene glycol's mass fraction of 0.3 (Triangle), 0.5 (Square) and 1.0 (circle), respectively.

CONCLUSION

This paper presents a new optofluidic method to measure the mutual diffusion coefficient in binary solutions in microchannel by directly analyzing the diffusion-induced focusing pattern based on a single optical micrograph. Ethylene glycol and deionized water are used to demonstrate the proposed method. Experimental results show good agreement with the reported data and indicate that the mutual diffusion coefficient is slightly decreased with the increase of the flow rate and is inversely proportional to the mass fraction of EG. This proposed method have potential applications in concentration mapping and chemical reaction monitoring.

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REFERENCES

- [1] M. Larranaga, M.M. Bou-Ali, D. Soler, M. Martinez-Agirre, A. Mialdun, V. Shevtsova, "Remarks on the analysis method for determining diffusion coefficient in ternary mixtures", *C.R. Mec.*, 341, 356–364, 2013.
- [2] K. Pappaert, J. Biesemans, D. Clicq, S. Vankrunkelsven and G. Desmet, "Measurements of diffusion coefficients in 1-D micro- and nanochannels using shear-driven flows", *Lab Chip*, 5, 1104, 2005.
- [3] A. E. Kamholz, B. H. Weigl, B. A. Finlayson, and P. Yager, "Quantitative Analysis of Molecular Interaction in a Microfluidic Channel: The T-Sensor", *Anal. Chem.*, 71, 23, 5340–5347, 1999.
- [4] Y. Lin, X. Yu, Z. Wang, S. Tu, and Z. Wang, "Measurement of temperature-dependent diffusion coefficients using a confocal Raman microscope with microfluidic chips considering laser-induced heating effect", *Anal. Chim. Acta.*, 667, 103–112, 2010.
- [5] R. Foord, E. Jakeman, C. J. Oliver, E. R. Pike, R. J. Blagrove, E. Wood and A. R. Peacocke, Determination of Diffusion Coefficients of Haemocyanin at Low Concentration by Intensity Fluctuation Spectroscopy of Scattered Laser Light, *Nature*, 227, 242 – 245, 1970.
- [6] K. Swinney, D. Markov and D. J. Bornhop, "Chip-Scale Universal Detection Based on Backscatter Interferometry", *Anal. Chem.*, 72, 13, 2690–2695, 2000.
- [7] J. F. Torres, A. Komiya, E. Shoji, J. Okajima, and S. Maruyama, "Development of phase-shifting interferometry for measurement of isothermal diffusion coefficients in binary solutions", *Opt. Laser Eng.*, 50 1287–1296, 2012.
- [8] L. K. Chin, A. Q. Liu, Y. C. Soh, C. S. Limb and C. L. Lin, "A reconfigurable optofluidic Michelson interferometer using tunable droplet grating", *Lab Chip*, 10, 1072-1078, 2010.
- [9] Y. Yang, A. Q. Liu, L. K. Chin, X. M. Zhang, D. P. Tsai, C. L. Lin, G. P. Wang, and N. I. Zheludev, "Optofluidic waveguide as a transformation optics device for lightwave bending and manipulation", *Opt. Express*, 3, 651, 2012.
- [10] G. Ternström, A. Sjostrand, G. Aly, and A. Jernqvist, "Mutual Diffusion Coefficients of Water + Ethylene Glycol and Water + Glycerol Mixtures", *J. Chem. Eng. Data*, 41, 876, 1996.
- [11] E. L. Cussler, *Diffusion: Mass Transfer in Fluid Systems*, Cambridge University Press, New York, 2009.

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