Liquid Diffusion Measurements by Digital Speckle Photography (DSP): Analysis of the PIV-Mode Approach

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Abstract - The aim of this work is to evaluate Digital Speckle photography (DSP) in liquid diffusion measurements. The experimental setup is based on a recorded speckle pattern. The light is passed through a transparent test section, where the diffusion process occurs. Two different exposures, recorded on a photosensor, are taken. Speckle images can be evaluated by decorrelation or by using cross-correlation algorithms borrowed from PIV (Particle Image Velocimetry). After a brief description of the method, we discuss the experimental results obtained during the diffusion of a 1.75 M (moles l^{-1}) solution of common salt (NaCl) in pure water at 26°C.

Keywords: Mass transfer, diffusion, liquid mixtures, correlation, speckle photography, flow visualization

1. Introduction

Mass transfer occurs in many different fields, from engineering (mechanical, chemical and aerospace engineering) to science (physics, chemistry, biology). It is also central to our daily lives (Cussler, 1997).

According to Crank (1980), diffusion is "the process by which matter is transported from one part of a system to another as a result of random molecular motions".

Since many years, optical methods are used to investigate fluid flow phenomena in transparent media; in particular, theyhave been used in diffusion measurements since the 19th century (Jost, 1960).

Major features of optical techniques are good accuracy and non-invasivity.

Classic interferometers (Jamin, Mach-Zender, etc.) (Jost, 1960) have been routinely employed: they offer the highest accuracy at a great cost in equipment and experimental effort (Cussler, 1997). The advent of holographic interferometric techniques, although not yielding the same precision, offers some advantages (Ruiz-Bevia et al., 1985).

Speckle photography techniques are based on the properties of speckles, typical granular images, which appear when objects are illuminated by laser light. Introduced also in the 1970's years, speckle techniques are well known in fluid mechanics and heat transfer problems (Fomin, 1988). In diffusivity measurements, they mostly concern ESPI (Electronic Speckle Pattern Interferometry) (Rastogi, 2001; Ambrosini et al., 2008; Axelsson and Marucci, 2008). Recent studies involve digital techniques and/or improved data processing (Zhang et al., 2012; Mialdun et al., 2013; Chhaniwal et al., 2014; He et al., 2015).

The aim of this work is to discuss speckle-based deflection techniques, proposed in (Ambrosini et al., 2002; Ambrosini et al., 2011), in the evaluation of liquid diffusion.

In the experimental setup, the light is passed through a transparent test section, where the diffusion process occurs. Two different exposures, recorded on a photosensor, are taken. Speckle images can be

evaluated by decorrelation (Ambrosini et al., 2002) or by using algorithms based on correlation algorithms of the type used in PIV (Particle Image Velocimetry) (Raffel et al., 1998; Ambrosini et al., 2011).

In this paper, attention is given to the PIV approach, with the aim of evaluating the performance of the correlation algorithms. After a very brief description of the method, we report a discussion and some experiments, considering the diffusion of a 1.75 M (moles l^{-1}) solution of common salt (NaCl) in pure water at 26°C.

2. Experimental Setup and Basic Theory



Fig. 1. Experimental setup for diffusivity measurement by DSP

The experimental arrangement is depicted schematically in Fig. 1. The spectrophotometric glass diffusion cell is equipped with a Teflon shutting device to avoid evaporation phenomena. The cell, filled through a capillary tube from the bottom to minimize turbulence and mixing, is illuminated by an expanded and collimated laser beam by means of a diffuser (ground glass). The light source is a laser diode (Lasiris by StockerYale, wavelength 638.5 nm, output power 5 mW).

The light rays diffracted by the ground glass, form a speckle pattern on the photosensor of a CCD video camera which images the diffuser. The video signal is finally processed by a frame grabber that allows arithmetic operations on images in real-time.

The TV camera was a Silicon Video® 9T001C with PIXCI® D2X imaging board by EPIX Inc., with a resolution of 2048×1536 pixels. The camera lens was a TEC-55 55mm F/2.8 Telecentric Computar Lens, which reduces viewing angle error and magnification error while providing good resolution and contrast with low distortion.

If refractive index n in the test section is non-uniform, the rays through the test object will be deflected. In other words, if the refractive index changes, the deflection angle will also change (Ambrosini et al., 2002). The change of the deflection angle can be regarded as a local translation of the speckle pattern and this displacement is proportional to the derivative of the refractive index.

The change in the index of refraction $\Delta n(x,t_1,t_2) = n(x,t_2) - n(x,t_1)$, for two given times t_1 and $t_2 > t_1$ has two characteristic extremes (say x_A and x_B), whose separation, say w, can be used to obtain the diffusion coefficient D. In fact, the following relation holds (Ambrosini et al., 2002):

$$D = \frac{w^2 [(1/t_1) - (1/t_2)]}{8 \ln(t_2/t_1)}$$
(1)

Eq. 1, first obtained by Bochner and Pipman (1976), was widely used in holographic interferometry and ESPI measurements (Ruiz-Bevia et al., 1985; Ambrosini et al., 2008).

In speckle-based deflection techniques, different exposures, recorded on a photosensor, are taken at different times from the beginning of the diffusion experiment. The distance *w* can then be obtained simply by subtracting the two images, as described in (Ambrosini et al., 2002) or by applying cross-correlation algorithms, as described in (Ambrosini et al., 2011), the latter method being more accurate.

3. Data Processing by the PIV-Mode Approach

In this type of data processing, sub-images are extracted from the first image (recorded at time t_1) and the second image (recorded at time t_2), then the correlation surface is obtained using suitable correlation filters. The peak location in the correlation surface gives the relative displacement between the two sub-images (Sjödahl, 2000). It can be shown (Ambrosini et al., 2002) that x_A and x_B are object points not exhibiting speckle displacement, therefore their separation w can be easily obtained also from the PIV mode displacement map, see Fig. 2.

PIV-mode is one of the simplest quantitative data processing, due to the possibility of borrowing existing PIV software with only minor modifications. In this work, pattern displacements are evaluated using correlation algorithms based on the toolbox MatPIV 1.6.1 by J.K. Sveen (Web-1).

The interrogated images are divided into smaller regions, also known as sub-windows, interrogationwindows or interrogation-regions. Each sub-window in the first image is compared with the corresponding sub-window in the second image.

The following options are available in MatPIV:

- single, which calculates the displacement with a single iteration through the images, using crosscorrelation;
- multi, that, using cross-correlation, performs three iterations through the images. This will start
 off using whatever window size specified, but the final iteration is performed using half this size.
- *multin*, which is an extension of *multi* and makes the calculations with n iterations through the images. In this case the size of the sub-window for each iteration should be inserted manually.
- mqd, which calculates the displacement in a single pass using a more general form (Web-1).
- *norm*, which is very similar to *single* but uses a normalized equation.



Fig. 2. A typical displacement map obtained by MatPIV. The arrows show the distance w to be used in Eq. (1).

4. Experiments

For gaining insight about the performance of DSP used with the PIV-mode approach, the measurement campaign was realized in the same conditions of (Ambrosini et al., 2011). In particular, experimental measurements were performed considering the diffusion of a 1.75 M (moles l^{-1}) solution of common salt (NaCl) in pure water at 26 °C.



Fig. 3. A typical speckle image, recorded 50 minutes after the beginning of diffusion phenomena.

As regards the MatPIV options, the following choices were made:

- the *single* method was considered because of its simplicity;
- the *norm* method was not considered, being very similar to a single iteration;
- in previous investigations (Ambrosini et al., 2012) no significant difference was found between the methods *multi* and *multin*, therefore only the performance of the *multi* method is considered here;
- in previous investigations (Ambrosini et al., 2012) the *mqd* method exhibits slightly better performance, with respect to the *multi* method, however it seems more sensitive to noise, therefore it is not considered here.

| $t_{\rm c}$ (min) | $t_{\rm e}$ (min) | single | single | single | multi | multi | multi | Riquelme et al., |
|----------------------------|----------------------------|--------|--------|--------|-------|-------|-------|------------------|
| <i>v</i> ₁ (mm) | <i>v</i> ₂ (mm) | 16 | 32 | 64 | 16 | 32 | 64 | 2007 |
| 35 | 40 | 1.78 | 1.85 | 1.99 | 1.88 | 1.85 | 1.85 | 1.611 |
| 45 | 50 | 1.86 | 1.92 | 1.8 | 1.86 | 1.8 | 1.92 | 1.584 |
| 45 | 55 | 1.78 | 1.72 | 1.72 | 1.72 | 1.66 | 1.72 | 1.634 |
| 50 | 55 | 1.52 | 1.52 | 1.63 | 1.63 | 1.57 | 1.52 | 1.627 |
| 55 | 65 | 1.83 | 1.94 | 2.06 | 1.86 | 1.89 | 1.94 | 1.561 |
| 65 | 70 | 1.82 | 1.62 | 1.82 | 1.85 | 1.67 | 1.82 | 1.598 |
| 65 | 75 | 1.76 | 1.66 | 1.57 | 1.76 | 1.76 | 1.66 | 1.605 |
| 70 | 75 | 1.74 | 1.60 | 1.51 | 1.65 | 1.74 | 1.6 | 1.527 |
| 70 | 80 | 1.69 | 1.55 | 1.64 | 1.67 | 1.69 | 1.55 | 1.53 |
| 75 | 80 | 1.68 | 1.59 | 1.59 | 1.7 | 1.81 | 1.7 | 1.499 |

Table. 1. Diffusion coefficient of NaCl in water at 26 °C in units of 10^{-9} m² s⁻¹.

There are few data available for comparison: Riquelme et al. (2007) recalculated some data from literature obtaining a diffusion coefficient of 1.538 u, being u defined to be 10^{-9} m² s⁻¹ for conciseness. More interestingly, they gave several experimental results for different diffusion times, which, with some caution, can be used for a more detailed comparison.

As shown in Table 1, for each correlation method, three windows size were considered: 16, 32 and 64 pixels. Although the data of (Riquelme et al., 2007) did not match perfectly with other values in literature and some systematic error may exist (Riquelme et al., 2007), several considerations can be made.

The multi method generally performs better with respect to the single method on the same window size (this is largely due to the fact that the final iteration is made on a window size which is half the original one).

The experimental results show a large dispersion, percent errors range from less than 1% to 30%. This dispersion depends on the difficulty to find, with good accuracy and repeatability, the values of the distance w to be used in Eq. (1).

4. Conclusion

This paper is devoted to evaluating the performance of digital speckle photography (DSP) in liquid diffusion measurements.

Speckle-based deflection techniques are less sensitive than holographic or ESPI methods, however they are very simple to construct and to align. Furthermore, they present a remarkable simplicity in experimental procedure.

A straightforward data processing can be based on correlation algorithms of the type used in PIV and then applying Eq. 1.

However, similarly to holographic interferometry and ESPI measurements (to whom Eq. 1 also applies), the evaluation of the distance w, to be inserted in Eq. 1, can introduce errors. In fact, let us consider Eq. 1. It involves only three experimental variables: the times t_1 and t_2 and the distance w.

The zero of time could be a possible source of error. However, the usual filling procedure, proposed by Gabelmann-Gray and Fenichel (1979), has proven capable to give an accuracy of the order of 1-2% (respectively, using holographic interferometry and ESPI). Therefore we may consider as the most probable source of errors the separation distance *w*.

When this distance is evaluated directly from the speckle displacements, as in this paper, a dispersion of results of the order of 10-15% is found, mostly induced by the separation distance *w*.

In order to maintain the interesting features of DSP, refined data processing should be introduced, such as a least squares fitting of the speckle displacement data.

References

- Ambrosini, D., Paoletti, D., Ponticiello, A., & Schirripa Spagnolo, G. (2002). Speckle Decorrelation Study Of Liquid Diffusion. Opt. Lasers. Eng., 37, 341–353.
- Ambrosini, D., Paoletti, D., & Rashidnia, N. (2008). Overview Of Diffusion Measurements By Optical Techniques. Opt. Las. Engin., 46, 852-864.
- Ambrosini, D., Paoletti, D., Di Biase, R., & Galli, G. (2011). Speckle-Based Deflection Techniques In Diffusivity Measurements. *Defects and Diffusion Forum*, 312-315, 912-917.
- Ambrosini, D., Paoletti, D., Di Biase, R., Rastogi, P.K., & Gorthi, S.S. (2012). Role Of Data Processing In Measuring Temperature Gradients With DOE Schardin's Schlieren #2. Opt. Las. Engin., 50, 1069-1074.
- Axelsson, A., & Marucci, M. (2008). The Use Of Holographic Interferometry And Electron Speckle Pattern Interferometry For Diffusion Measurement In Biochemical And Pharmaceutical Engineering Application. Opt. Las. Engin., 46, 865-876.
- Bochner, N., & Pipman, J. (1976). A Simple Method Of Determining Diffusion Constants Byholographic Interferometry. *J Phys D: Appl Phys*, 9, 1825–1830.
- Chhaniwal, V.K., Narayanamurthy, C.S., & Anand, A. (2014). Imaging Of Mass Transfer Using Artificial Fringe Deflection. *Opt. Eng.*, 53, 074106.
- Crank, J. (1980). The Mathematics Of Diffusion. Oxford Science Publication.
- Cussler, E.L. (1997). Diffusion Mass Transfer in Fluid Systems. 2nd ed. Cambridge University Press.
- Fomin, N.A. (1998). Speckle Photography For Fluid Mechanics Measurements. Springer-Verlag.
- Gabelmann-Gray, L., & Fenichel, H. (1979). Holographic Interferometric Study Of Liquid Diffusion. *Appl. Opt.*, 18, 343-345.

He, M.G., Zhang, S., Zhang, Y., & Peng, S.G. (2015). Development Of Measuring Diffusion Coefficient By Digital Holographic Interferometry In Transparent Liquid Mixtures. *Opt. Express*, 23, 10884-10899.

Jost, W. (1960). Diffusion In Solids, Liquids, Gases. Academic Press.

Mialdun, A., Sechenyh, V., Legros, J.C., Ortiz de Zarate, J.M., & Shevtsova, V. (2013). Investigation Of Fickian Diffusion In The Ternary Mixture Of 1,2,3,4-Tetrahydronaphthalene, Isobutylbenzene, And Dodecane. J. Chem. Phys., 139, 104903.

Raffel, M., Willert, C., & Kompenhans, J. (1998). Particle image velocimetry. Springer.

Rastogi, P.K. (Ed.) (2001). Digital Speckle Pattern Interferometry And Related Techniques. Wiley.

- Riquelme, R., Lira, I., Perez-Lopez, C., Rayas, J.A., & Rodriguez-Vera, R. (2007). Interferometric Measurement Of A Diffusion Coefficient: Comparison Of Two Methods And Uncertainty Analysis. *J. Phys. D: Appl. Phys.*, 40, 2769-2776.
- Ruiz-Bevia, F., Celdran-Mallol, A., Santos-Garcia, C., & Fernandez-Sempere, J. (1985). Liquid Diffusion Measurement By Holographic Interferometry. *Can. J. Chem. Eng.*, 63, 765–771.
- Sjödahl, M. (2000). Digital Speckle Photography. In P.K. Rastogi, D. Inaudi (Eds.), *Trends In Optical* Non-Destructive Testing And Inspection, Elsevier.
- Zhang, Y., Zhao, J., Di, J., Jiang, H., Wang, K., Wang, J., Guo, Y., & Yin, D. (2012). Real-Time Monitoring Of The Solution Concentration Variation During The Crystallization Process Of Protein-Lysozyme By Using Digital Holographic Interferometry. *Opt. Express*, 20, 18415-18421.

Web sites:

Web-1: https://www.mn.uio.no/math/english/people/aca/jks/matpiv/, consulted January 2015.