**Gas-Liquid Flows in Thin-Gap Electrochemical Microchannels – PIV Study**

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# Gas-liquid flows in thin-gap electrochemical microchannels – PIV study

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**Abstract**

We focused on the study of the local hydrodynamics of liquid in gas-liquid flow in thin-gap microchannels (width 10 mm, depth 2 mm - 0.1 mm). Two-phase flow was obtained by injecting bubbles into the channel to mimic gas generation in electrochemical reactions. The liquid flow velocity vectors were obtained by the PIV technique. Single phase measurements were used to verify our method by comparison with the theoretical velocity profiles. Local velocity fields around single bubble were evaluated.

**Keywords**

Thin-gap microchannel, gas-liquid hydrodynamics, PIV

## 1 Introduction

Microstructuring represents a novel approach for the design of chemical process equipment. The internal characteristic dimensions of microstructured equipment are typically below 1 mm leading to a very high surface to volume ratio, which allows intensified heat and mass transfer, thus providing solutions to the constantly increasing demands of chemical technologies [1].

Microstructured approach has been used intuitively in the design of the cells for electroorganic synthesis already since 1960s. The main reason represents extremely low conductivity of the organic solvents resulting in unacceptably high energy costs when using classical electrochemical cell arrangement. The typical representative of the construction optimized for such a type of process represents the socalled thin capillary gap cell. A typical example of a process utilizing this type of cell represents electrochemical methoxylation of 4-methylanisole developed and utilized by BASF [2]. Due to the extremely low interelectrode distance, reaching typically 0.5–1.0 mm, this cell may be nearly recognized as a microstructured one.

In electrochemical applications, microstructured reactors have the advantage of providing a smaller ohmic drop, which allows a lower concentration of supporting electrolytes to be used [3]. A properly designed electrochemical microreactor can provide high conversion and selectivity of a reaction in only a single pass of reaction mixture through the microreactor. This is thanks to the interelectrode distance being of the same order of magnitude as the thickness of the Nernst diffusion layer. Amongst the different designs, thin-gap microreactors are very promising for electrochemical synthesis [4]. In certain processes, microstructured reactors with a thin electrode gap can significantly decrease operational and investment costs arising from the separation and purification of products [5].

For the successful design of electrochemical microreactors it is necessary to understand the hydrodynamic behaviour of a given system. This is particularly important in reaction

systems where gas bubbles are generated on the electrode create two-phase dispersion with the liquid reaction mixture. Bubbles can significantly affect the conversion and selectivity of the reaction by hindering the active electrode surface and corrupting the current density distribution in the liquid around the bubble [4].

Objectives of this work were to develop an experimental methodology based on PIV for the measurement of velocity fields around bubbles in thin-gap microreactors and to study the effect of bubbles on non-reactive liquid flow fields in the thin-gap device using the methodology developed.

## 2 Experimental methods

### *2.1 Experimental apparatus*

Our experimental apparatus (Figure 1) was "once through" system, where liquid flowed from the syringe pump, through the microchannel and then it was discharged to the waste tank. Whole experimental setup can be divided into several parts: hydrodynamic cell with microchannel; visualization and PIV part with camera and backlight; supporting equipment with data acquisition system.

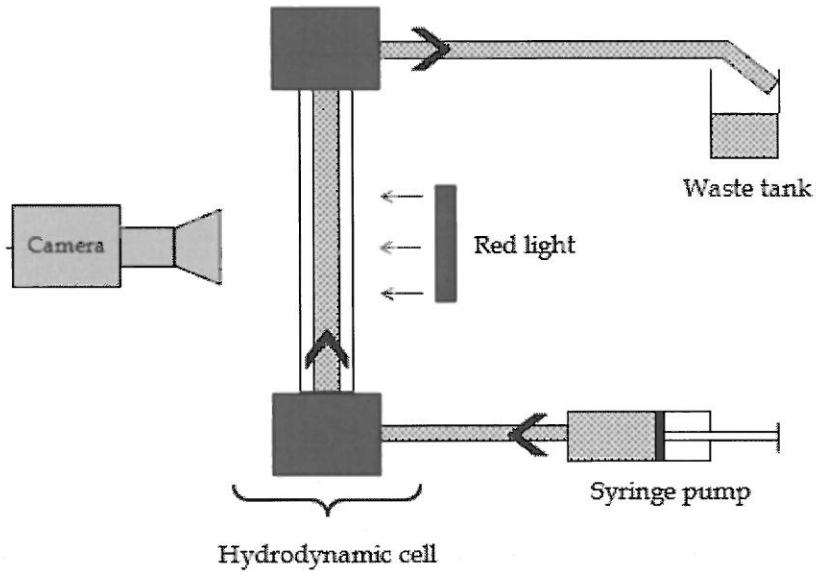


Figure 1: Experimental apparatus

The hydrodynamic cell (Figure 2a) is a main part of the experimental apparatus. This cell, designed in our laboratory and manufactured by Optical Development workshop Turnov, consists of two main parts. First part is the microchannel itself, second are inlet and outlet connections, which are provided by two plastic connector heads, in which the microchannel was fitted. One inlet section head have also another inlet position with septum in the microchannel axis, thus needle can be placed directly into the microchannel as a gas inlet for artificial bubble generation.

Used microchannels had rectangular cross-section and were oriented vertically (Figure 2b). Length of all microchannels was 150 mm, width 10 mm and depth (represents also possible interelectrode distance) varied between 0.1 mm to 2 mm. Glass used in construction of hydrodynamical cells had a superior quality, was optically clear and chemically stable. In one wall, a channel with desired depth was grinded, covered with the second wall and fixed together with capillary glue. The distance accuracy of resulting microchannel was about 1 %.

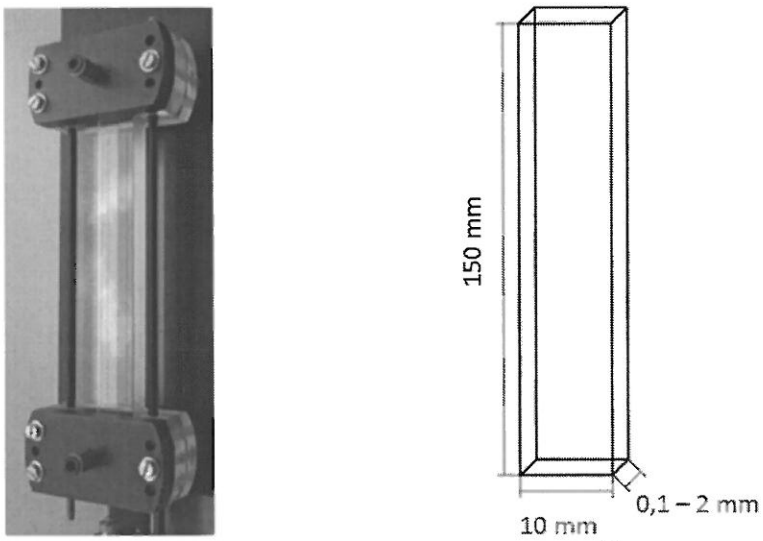


Figure 2: Hydrodynamic cell; (a) photo of the hydrodynamic cell; (b) scheme of the microchannel;

Photron SA1.1 high speed camera was used for PIV measurements. It is capable of over 5,000 full frames per second (fps) at mega pixel (1K x 1K) resolution. Nikkor F-mount was used to attach optics. Optics consisted of Sigma Macro 105 mm f/2.8 EX DG Nikon objective and 56 mm (20 mm and 36 mm) extension rings. Depth of focus of was in order of $10^{-5}$ m. LED circular light was used to illuminate seeding particles. Light source was installed as an indirect backlight. Light scattered on particles was recorded by the camera. For proper camera positioning, 3-D positioning table was used. Focus point can be determined (0.01 mm precision) by camera movement (fixed optics adjustment). Combination of an aperture of F2.8, exposure time of 1/5000 s and precise focusing provided sharp and contrast photographs suitable for subsequent PIV and image processing.

Ethanol was used as model liquid and air bubbles were formed with a needle directly inside the microchannel to mimic the gas generation by the electrochemical reaction. Polyamide seeding particles with mean diameter 20 μm (PSP-20, Dantec) or silver-coated hollow glass spheres with mean diameter 10 μm (S-HGS-10, Dantec) were used. A controllable syringe pump was used as a liquid supply. Bronkhorst Hi-Tec mass flow controller was used as a gas supply.

### *2.2 Experiments and data evaluation*

Experiments were divided into two main sections. First section dealt with one-phase flow experiments for verification of whole experimental and data evaluation process. Second experimental section dealt with two-phase gas-liquid flow in which gas was artificially injected to the microchannel by the needle.

Liquid with seeding particles was mixed on an electromagnetic stirrer to obtain homogeneously distributed suspension. This suspension was put into the syringe and placed to the pump. Apparatus was flooded by higher flow rate (up to 50 ml/min, depends on the microchannel used) and tubing was bled to eliminate any trapped gas. In case of two-phase measurements gas desired gas flow rate was set on the controller.

After reaching steady conditions flow video recording was done capturing at least 1000 images (figure 3a), which covers few seconds, depending on the microchannel and flow rates. Size of our region of interest was approximately $1.2 \times 1.2$ cm$^2$, but images were cropped to the channel walls immediately.

Whole PIV procedure was processed using MATLAB with Image Processing Toolbox (MIPT) and mpiv toolbox [6]. Images were preprocessed by median filtering to get rid of unfocused particles and other disturbances. PIV procedure was recursive correlation with decreasing interrogation area size from 64x64 pix to 32x32 pix. Window overlap for all steps was 0.5. Peak filtering value was 1.3. Example of evaluated liquid velocity vectors from one-phase measurements is shown on Figure 3b

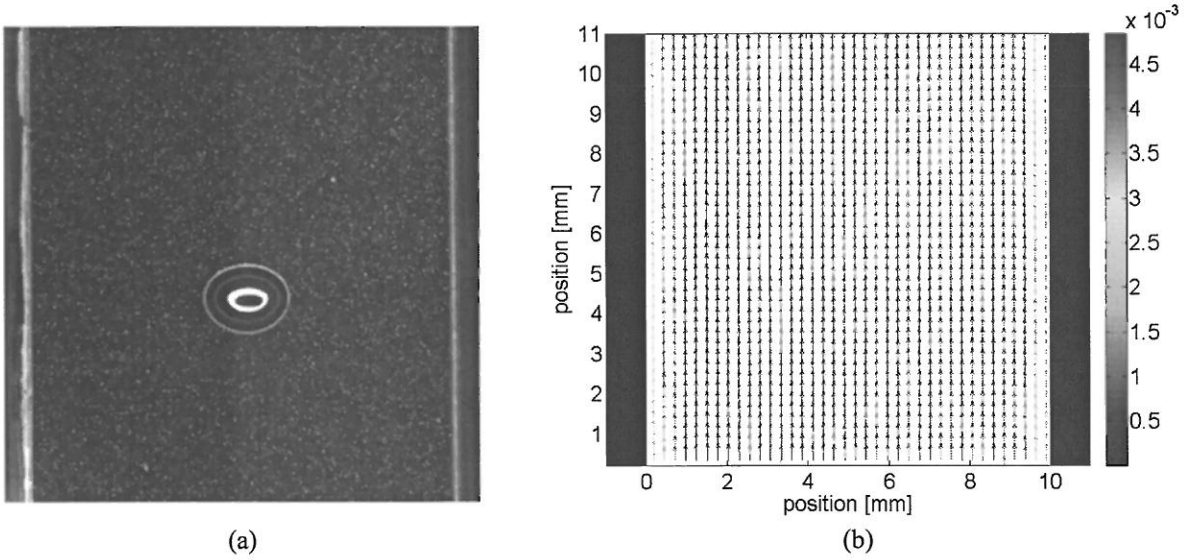


Figure 3: (a) PIV image of rising bubble in the microchannel; (b) one-phase velocity vectors in the microchannel

## 3 Results and discussion

Results of single-phase experiments show good agreement with theoretical predictions (figure 4).

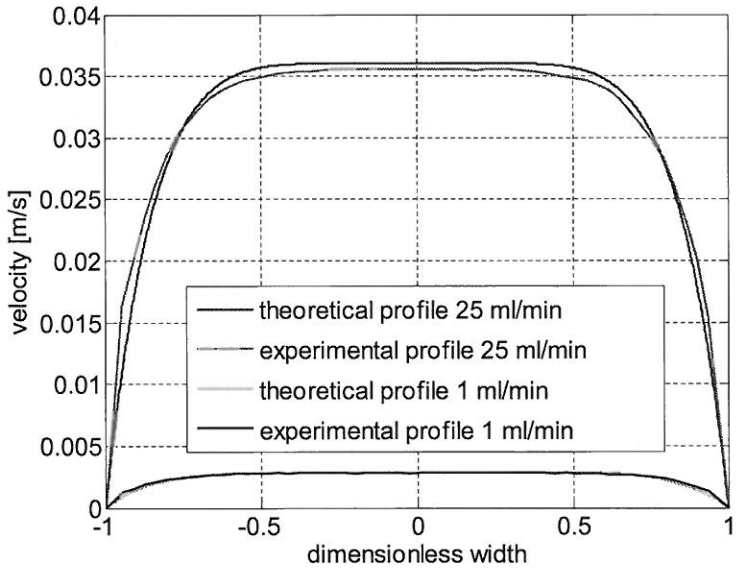


Figure 4: Validation of method – comparison of theoretical and experimental velocity profiles in the middle of the 2 mm microchannel

The bubbles create a dragging region behind them. The fluid acceleration in the center of the channel is compensated by a decrease of velocity near the walls (Figure 5a). At low liquid

flow rates (Figure 5b) recirculation of the fluid between bubble and the channel walls was observed, creating two distinct flow directions.

The steady flow without recirculation is optimal for the electrochemical reaction, the recirculation can lower the reaction selectivity. Also the bubbles affect the electrochemical reaction by blocking the electrode surface and distorting the current density distribution.

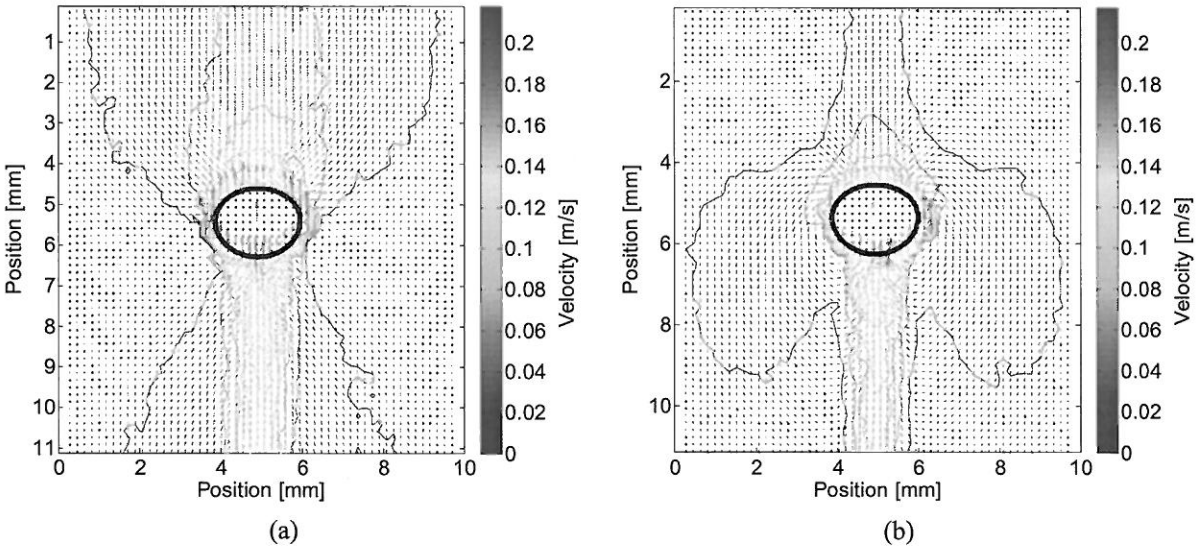


Figure 5: Liquid velocity vectors around single rising bubble (a) $Q_L$ = 25 ml/min, $Q_G$ = 1 ml/min (b) $Q_L$ = 1 ml/min, $Q_G$ = 1 ml/min

## 4 Conclusions

A PIV methodology for the measurement of liquid velocity fields in a thin-gap electrochemical microreactor has been developed. Single-phase measurements are in the good agreement with theoretical velocity profiles, thereby validating the experimental method.

Measurements in a non-reactive gas-liquid system show that the presence of bubbles cause the generation of large heterogeneities in the liquid flow field, which is quite different from the single-phase system.

Further analysis of the data set will aim at correlating the effect of bubble velocity, size and position on the liquid flow field in the thin-gap microchannel. This will be complemented by similar experiments in a reactive electrochemical system. The ensemble of this information will be used for the development of design rules for electrochemical microreactors.

## 5 Acknowledgments

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