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Experimental validation and critical analysis of inverse method in mass transfer using electrochemical sensor

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ABSTRACT

Rehimi et al. [1] numerically studied the frequency response of an electrochemical probe. They applied an inverse sequential algorithm of the convection diffusion equation to simulated periodic mass transfer signals in order to determine the wall shear rate. This paper presents an experimental validation of the inverse method in mass transfer and a critical analysis of its advantages and limits of application. An electrochemical sensor (probe) was used to determine the mass transfer from the current delivered by the probe. The mass transfer is related to the wall shear rate via the convection diffusion equation. The database, obtained by electrochemical method also known as "Electro-Diffusion Method", was exploited to validate the inverse method experimentally. The inverse method was checked over a specific range of Péclet numbers varied from 4.58×10^3 to 1.06×10^5 with a well-controlled shear flow with known wall shear rate. An optimized computational algorithm using Matlab® was developed for the post-processing, the filtering of the electrochemical results and for programming available models, mostly used in polarography for the determination of the wall shear rate (Lévêque [2]; Sobolik et al. [3]; Deslouis et al. [4]). The comparison of these methods with the inverse method and the experimental wall shear rate allows a critical analysis of the restrictions of the different approaches. We demonstrated experimentally, that the difference between the real wall shear rate and the quasi-steady one of Lévêque [2], can reach 9% for dimensionless frequencies $f^* = \frac{fl^2}{D} \leqslant 205$ and oscillation amplitudes $\beta \geqslant 0.3$. For low frequencies ($f^* \leqslant 205$), Sobolik et al. [3] solution and Deslouis et al. [4] transfer function are in agreement with the experimental wall shear rate. The inverse method is validated for high frequencies of oscillations, for which the linear approaches lead to amplitude attenuation and phase shift of wall shear stress fluctuations with respect to the experimental one.

1. Introduction

The electrochemical method is one of the few non-intrusive techniques used for the measurement of the local wall shear rate, which is a very important parameter for flow characteristics in many experimental devices. The method is based on the determination of the limiting diffusion current delivered by a small probe placed on an inert wall in contact with the liquid flow. By solving the convection–diffusion equation in steady regime and without the axial diffusion term, a solution, the "Lévêque solution" [2], relating the limiting diffusion current and the wall shear rate was proposed for high Péclet numbers and by neglecting the axial diffusion. Sobolik et al. [3] have introduced another technique based on the correction of the wall shear rate obtained by the Lévêque [2] solution by adding a term deduced from the transitory response:

$$S_{Sob}(t) = S_q(t) + \frac{2}{3}\theta(t)\left(\frac{\partial S_q(t)}{\partial t}\right) \quad (1)$$

where

$$S_q = S_{Lev} = \frac{D}{l^2}\left(\frac{\mathrm{Sh}(t)}{0.807}\right)^3 \quad (2)$$

and

$$\theta(t) = 0.486 l^{\frac{2}{3}} D^{-\frac{1}{3}} S_q(t)^{-\frac{2}{3}} \quad (3)$$

This method correctly predicts the wall shear rate at high average Péclet numbers when the sampling rate is sufficient.

The most common approaches related to the wall shear stress probes are focused on the research of transfer functions between

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Nomenclature

A active area of the probe (m^2)
c molar concentration of the active ion (mol.m^{-3})
c_0 bulk concentration (mol/m^3)
C dimensionless concentration of the active ion $\left(C = \frac{c}{c_0}\right)$
D coefficient of diffusion ($m^2\,s^{-1}$)
f frequency of the superposed harmonic oscillations (Hz)
f^* dimensionless frequency, $f^* = \frac{fl^2}{D}$
f^+ dimensionless frequency, $f^+ = f^* \mathrm{Pe}^{-\frac{2}{3}} = f\left(\frac{l^4}{S^2 D}\right)^{\frac{1}{3}}$
F Faraday number (C mol^{-1})
h thickness of the gap between the rotating disk and the upper fixed one (m)
H transfer function
I limiting diffusion current delivered by the ED probe (A)
$K = I/(n\mathrm{Fc_0A})$ mass transfer coefficient (m s^{-1})
l length of the probe (m)
n stoechiometric number
$\mathrm{Pe} = Sl^2/D$ Péclet number (dimensionless)
r radial distance (mm)
$R = 2\pi fh^2/\upsilon$ oscillatory Reynolds number (dimensionless)
$\mathrm{Re}^* = \Omega h^2/\upsilon$ Reynolds number (dimensionless)
S wall shear rate (s^{-1})
$\bar{S}$ mean wall shear rate (s^{-1})
S_0 base shear rate (s^{-1})
$\mathrm{Sh} = lK/D$ Sherwood number (dimensionless)
t time (s)
$t^* = t(\omega/2\Pi)$ dimensionless time
U velocity (m s^{-1})
U_p polarization voltage (V)
x, y coordinates (m)

Greek symbols
β relative amplitude of superposed harmonic oscillations
ε constant
η dynamic viscosity of the fluid (Pa s)
υ kinematic viscosity ($m^2\,s^{-1}$)
ω normalized complex amplitude
Ω rotation speed of the disk (rad s^{-1})
σ dimensionless frequency defined in Deslouis et al.[4]
Δ laplace operator (dimensionless)

Superscripts and subscripts
– time average
* dimensionless form
↔ complex function or parameter
circ *circular*
exp experimental
grad gradient
Lev Lévêque
num numerical
q quasi-steady state
rect rectangular
Sob Sobolik et al. method

Shorthands
ED electro-diffusion
PC personal computer
PPDs parallel plate disks

wall shear stress and limiting diffusion current (Deslouis et al. [4]). The axial diffusion mass transfer was neglected and the approach was based on a linearization of the problem by assuming that the wall shear rate fluctuations remained small in comparison with the average value. The transfer function of Deslouis et al. [4] is proposed for rectangular and circular probes. For a rectangular probe, it can be written as:

– If the dimensionless frequency $\sigma_{rect} = 2\pi f\left(\frac{l^2}{D\bar{S}^2}\right)^{\frac{1}{3}} \leqslant 6$:

$$\begin{cases} \left|\frac{H(\sigma_{rect})}{H(0)}\right| = (1+0{,}056\sigma_{rect}^2+0{,}00126\sigma_{rect}^4)^{-\frac{1}{2}} \\ \arg(H(\sigma_{rect})) = -\arctan(0{,}276\sigma_{rect}(1+0{,}02\sigma_{rect}^2-0{,}00026\sigma_{rect}^4)) \end{cases} \tag{4}$$

– If $\sigma_{rect} = 2\pi f\left(\frac{l^2}{D\bar{S}^2}\right)^{\frac{1}{3}} \geqslant 6$:

$$\begin{cases} \left|\frac{H(\sigma_{rect})}{H(0)}\right| = \frac{\sqrt{55{,}2049+\left(5{,}64-7{,}49\sigma_{rect}^{\frac{1}{2}}\right)^2}}{2\sigma_{rect}^{\frac{3}{2}}} \\ \arg(H(\sigma_{rect})) = -\arctan\left(\frac{7{,}43\sigma^{\frac{1}{2}}-3{,}99\sqrt{2}}{3{,}99\sqrt{2}}\right) \end{cases} \tag{5}$$

For a circular probe of diameter d_s, the transfer function of Deslouis et al. [4] can be written:

– If $\sigma_{circ} = 2\pi f\left(\frac{d_s^2}{D\bar{S}^2}\right)^{\frac{1}{3}} \leqslant 6$:

$$\begin{cases} \left|\frac{H(\sigma_{circ})}{H(0)}\right| = (1+0{,}049\sigma_{circ}^2+0{,}0006\sigma_{circ}^4)^{-\frac{1}{2}} \\ \arg(H(\sigma_{circ})) = -\arctan(0{,}246\sigma_{circ}(1+0{,}0124\sigma_{circ}^2-0{,}00015\sigma_{circ}^4)) \end{cases} \tag{6}$$

– If $\sigma_{circ} = 2\pi f\left(\frac{d_s^2}{D\bar{S}^2}\right)^{\frac{1}{3}} \geqslant 6$:

$$\begin{cases} \left|\frac{H(\sigma_{circ})}{H(0)}\right| = \frac{\sqrt{56{,}18+\left(7{,}495-8{,}832\sigma_{circ}^{\frac{1}{2}}\right)^2}}{2\sigma_{circ}^{\frac{3}{2}}} \\ \arg(H(\sigma_{circ})) = -\arctan\left(\frac{7{,}495-8{,}832\sqrt{\sigma_{circ}}}{7{,}495}\right) \end{cases} \tag{7}$$

Deslouis et al. [4] used a rotating disk for which a periodic flow was imposed. They placed simple circular probe of a 0.03 cm diameter at 1.1 cm from the disk axis to validate the transfer function experimentally. The approach is no longer valid for high frequencies.

When the fluctuations are important, the linear approach cannot be applied. For low Péclet numbers and for high flow fluctuations, the wall shear stress has to be determined by solving the inverse problem by using the inverse "technique". The inverse method was the subject of few studies to solve mass transfer problems. Rehimi et al. [1] applied the inverse method using simulated data to determine wall shear rate. They tested the sequential estimation for a known wall shear rate of the form:

$$S^*(t) = 1 + \beta\cos(2\pi ft^* + \varphi) \tag{8}$$

The time evolution of the Sherwood number was determined for different values of β by solving the direct problem. They simulated the mass transfer for a fixed wall shear rate given by Eq. (8) for high Péclet number ($\mathrm{Pe} = 10^4$) which allows to consider negligible axial diffusion. No previous work was done to validate the inverse method experimentally for different ranges of frequencies and amplitudes of flow oscillations and different Péclet numbers. For highly fluctuating oscillatory flows, the quasi-steady solution of Lévêque [2] is no longer applicable [1]. We validated the inverse method experimentally to determine the wall shear rate in controlled fluctuating flows using a circular probe for different ranges of frequencies and amplitudes of flow oscillations and different