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International Journal of Heat and Mass Transfer 51 (2008) 910–919

International Journal of HEAT and MASS TRANSFER

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Determination of the flow and heat transfer characteristics in non-Newtonian media agitated using the electrochemical technique

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Received 29 January 2007; received in revised form 16 July 2007

Abstract

In the study the results of the friction factor in boundary layer and the distribution of heat transfer coefficient in non-Newtonian liquid agitated by different impellers, have been presented. It has been established that for studies in Na-CMC and guar gum aqueous solutions by the electrochemical method the following solution of 0.005 (kmol m^{-3}) $K_3[Fe(CN)_6]$, 0.005 (kmol m^{-3}) $K_4[Fe(CN)_6]$ and 0.3 (kmol m^{-3}) K_2SO_4 can be recommended. The common relationship (for a given type of an impeller) between local values of friction coefficient and heat transfer coefficient and Reynolds number proposed by Metzner and Otto [A.B. Metzner, R.E. Otto, Agitation of non-Newtonian fluids, AIChe J. 3 (1957) 3–10] for all power-law fluids, have been obtained.

Keywords: Agitation; Non-Newtonian fluids; Electrochemical method; Heat transfer coefficient distribution; Shear rate distribution; Friction factor distribution

1. Introduction

The electrochemical method is based on a diffusion-controlled reaction at the electrode surface and mass transfer between electrode surface and electrolyte solution. When an electric potential is applied between two electrodes in an aqueous solution of an electrolyte [1] an ionic reduction occurs at the cathode and an oxidation at the anode. As a result, a current which is proportional to the number of ions reacting at the electrodes per unit time, flows through the circuit. When the potential on the electrode is gradually increased the current first increases until a stable value is reached. This value is called the limiting current and it corresponds to the condition when the concentration of the reacting species of ions on the surface of the electrode equals zero. At steady state, ions that are converted at the electrode have to be supplied from the bulk of the liquid. This can occur by a diffusion process under the effect of the concentration gradient and by migration of the ions in the electric field. To suppress the last effect or make it negligible compared to diffusion and convection, a high concentration of inert electrolyte is used. In the electrochemical method during the transport of substance and charge in the electrolyte stream, ions from the main mass solution are transferred to the surface of the electrode. If the process is controlled only by the diffusion of the ions to the surface electrode, mass flux at the wall surface ($y = 0$) obeys the first Fick law. From the measurements of limiting current density the mass transfer coefficient k_m can be determined:

$$k_m = \frac{I_d}{C_\infty z_e F} \quad (1)$$

The red-ox couple most often used in various studies is potassium ferricyanide–ferrocyanide. The indifferent electrolytes used were potassium sulphate and sodium hydroxide [2].

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doi:10.1016/j.ijheatmasstransfer.2007.11.003

Nomenclature

A_E	surface area (m^2)	Re_{MO}	Reynolds number for mixing of power-law liquid
B^*	the proportionality coefficient	Sc_{MO}	Schmidt number for mixing of power-law liquid
c_F	friction factor	T	temperature (K)
c_p	specific heat ($J\ kg^{-1}\ K^{-1}$)	w	velocity ($m\ s^{-1}$)
$C_{p,p}$	mass concentration of polymer (%)	z	axial coordinate
d	impeller diameter (m)	z_e	number of electrons taking part in a reaction
d_E	electrode diameter (m)		
D	tank diameter (m)	*Greek symbols*	
D_A	diffusivity ($m^2\ s^{-1}$)	α	heat transfer coefficient ($W\ m^{-2}\ K^{-1}$)
F	Faraday's constant ($C\ mol^{-1}$)	η	viscosity (Pa s)
H	liquid height in a tank (m)	η_e	equivalent viscosity (Pa s)
I_d	limiting diffusion current (A)	ρ	density ($kg\ m^{-3}$)
k_m	mass transfer coefficient ($m\ s^{-1}$)	τ	shear stress (Pa)
K	consistency index ($Pa\ s^m$)	λ	thermal conductivity ($W\ m^{-1}\ K^{-1}$)
m	flow behaviour index	$\dot{\gamma}_E$	shear rate (s^{-1})
n	impeller rotational speed (s^{-1})	$\dot{\gamma}_m$	average shear rate (s^{-1})
Pr_{MO}	Prandtl number for mixing power-law liquid	φ	angular coordinate
R_E	radius of cathode (m)		

The electrochemical method has previously been applied in a variety of convective heat transfer and wall shear stress studies making use of well known analogies between the various transport phenomena. This measuring technique has been especially used for prior studies of the processes occurring in the boundary layer at the wall of the apparatus [2]. Also in chemical engineering this method has been used, for example: in the studies of pulsate and oscillate flows between parallel plate (plate heat exchanger) [3–6], in evaluation of the thickness of boundary layer in natural convection mass transfer [7–9], in the measurement of liquid film thickness in Newtonian [10] and non-Newtonian fluids [11], in pipes flow [12–15], in packed column [16,17], in fluidisation processes [18,19] and membrane reactor [20].

The aim of the present work was to determine experimentally the local distributions of shear rate, friction factor and heat transfer coefficient in non-Newtonian liquid agitated by different impellers. Previous literature [21–27] has not paid sufficient attention to introduce the electrochemical method to determination of the flow and heat transfer characteristics of the stirring when the non-Newtonian liquids are the media agitated.

2. Experimental set-up

Detailed construction of agitated vessel of diameter of $D = 0.19$ (m) used in the study is shown in Fig. 1a. Twelve nickel electrodes ($d_E = 5 \times 10^{-3}$ m) were placed at various heights along the tank wall. It should be pointed out that if the previous electrochemical experiments were carried out in an agitated solution, the electrode used had the diameters of $d_E \in (0.0002; 0.012)$ (m) [2,23,25–27]. Distance of the centre of electrodes from the tank bottom was taken as the value of z (axial coordinate). Application of twelve electrodes enabled the authors to carry out the measurements within the range of dimensionless coordinates $z/H \in (0.105, 0.974)$. In each study before the starting of measurements, the electrode surfaces were cleaned using the powdered aluminium oxide (Al_2O_3). Additionally, the nitrogen was used to remove the oxygen from the electrolyte solution.

On the tank bottom the opposite electrode (anode) made of the nickel plate about 0.10 (m) diameter, was mounted. The tank was provided with a heating-cooling jacket. The effect of an angular coordinate φ on the diffusion limiting current for non-Newtonian fluids has also been studied. The experimental studies were carried out for two model polymer solutions, carboxymethylcellulose sodium salt and guar gum, in the tank equipped with four various impellers: Rushton turbine, paddle with six pitched blade with angles of 45° and 135°, and the paddle with six blades with angle of 90°, for standard geometric invariants of the agitated vessel (Fig. 1b–e). The exemplary data of the limiting diffusion current is presented in Fig. 2. During the measurements for different solutions tested the exemplary limiting current values have been obtained as follows:

– for electrolyte solutions and Rushton turbine $I_d \in (1.7 \times 10^{-4}, 5.8 \times 10^{-4})$ (A),
– for Na-CMC aqueous solutions $I_d \in (1.2 \times 10^{-4}, 6.0 \times 10^{-4})$ (A),
– for guar gum aqueous solutions $I_d \in (1.1 \times 10^{-4}, 4.4 \times 10^{-4})$ (A).

3. Heat–mass transfer analogy

Chilton and Colburn [28] demonstrated that in turbulent tube flow the analogy between heat and mass transfer

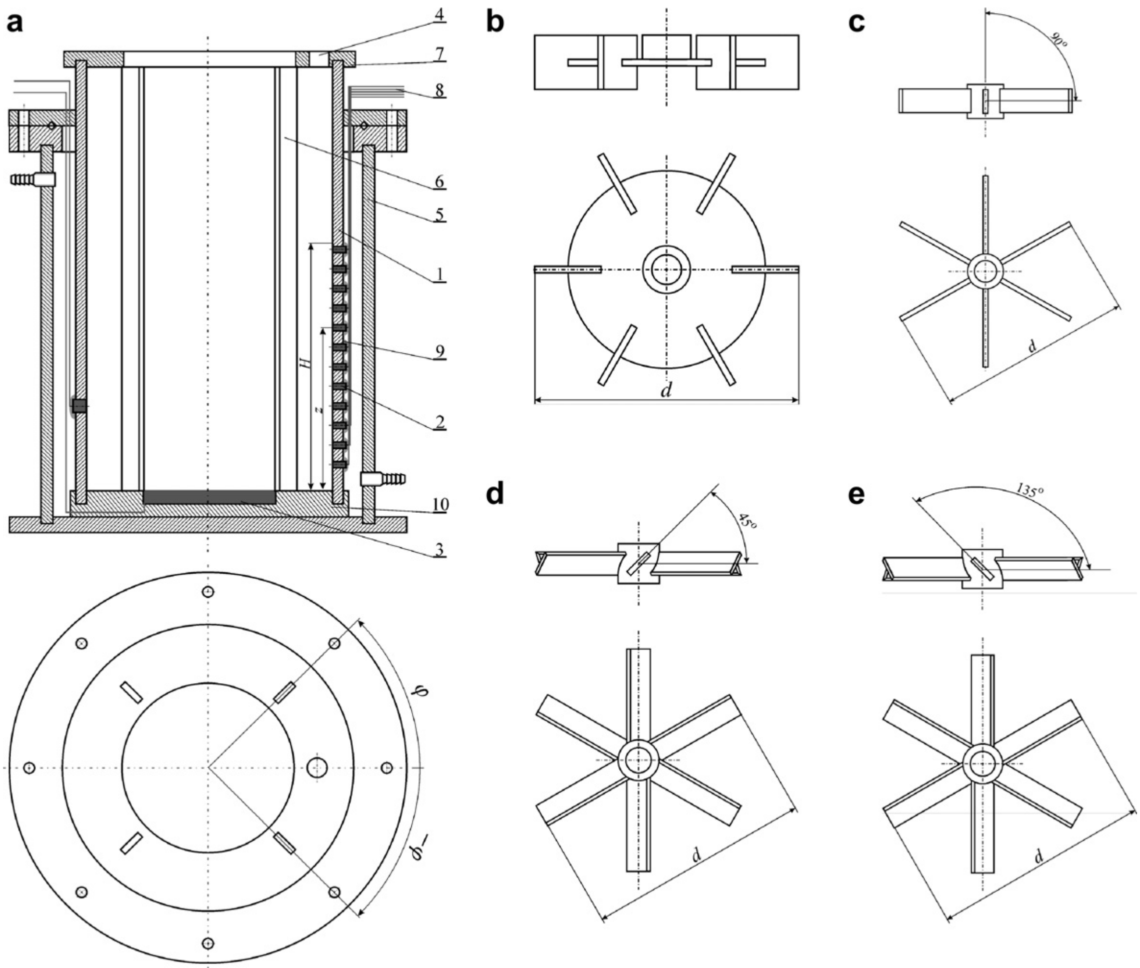

Fig. 1. The scheme of the measurement system (a) and impellers used in the study (b)–(e): 1 – tank wall, 2 – working electrodes (cathodes), 3 – opposite electrode (anode), 4 – hole on the reference electrode, 5 – heating jacket (cooling), 6 – baffles, 7 – flange, 8 – insulated nickel conduit, 9 – electric insulation, 10 – bottom of the tank, (b) Rushton turbine, (c) paddle with 6 straight blades with angle $\beta = 90°$, (d) paddle with 6 pitched blades with angle of $\beta = 45°$, (e) paddle with 6 pitched blades with angle of $\beta = 135°$.

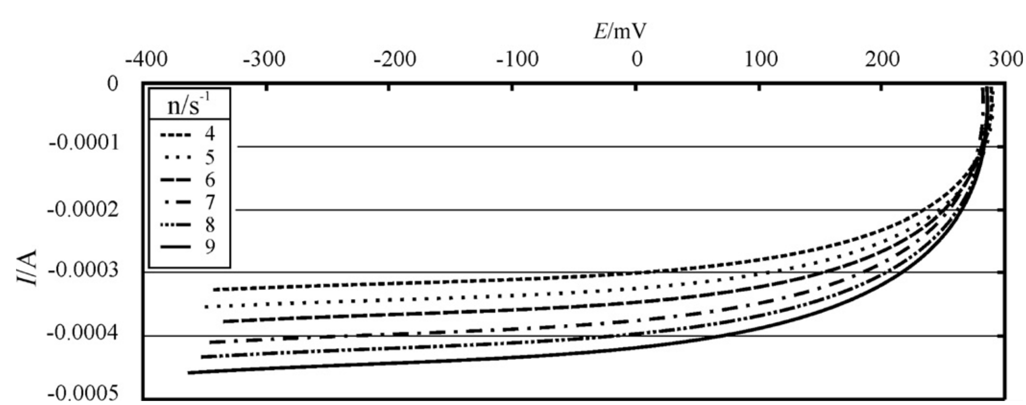

Fig. 2. The exemplary relation of current intensity on voltage for electrolyte solution with Na-CMC addition of the concentration of $C_{p,\text{p}} = 0.05\%$.

exists. As in a mixing the fluid flow is usually the turbulent one, this analogy permits to evaluate the relationship between the heat and mass transfer coefficients in a stirred tank too [29]. The resulting basic relation from Chilton and Colburn [28] analogy is as follows:

– for Newtonian fluids

$$j = \frac{\alpha}{gc_p} Pr^{2/3} = \frac{k_\text{m}\rho}{g} Sc^{2/3} \quad (2)$$

– for non-Newtonian fluids

$$j = \frac{\alpha}{g c_p} Pr_{MO}^{2/3} = \frac{k_m \rho}{g} Sc_{MO}^{2/3} \tag{3}$$

therefore

$$\frac{\alpha}{k_m} = c_p \rho \left(\frac{Sc}{Pr}\right)^{2/3} = c_p \rho \left(\frac{Sc_{MO}}{Pr_{MO}}\right)^{2/3} \tag{4}$$

4. Distribution of hydrodynamic and heat transfer coefficients in stirred tank

The distribution of the velocity in agitated vessel has extremely complicated character and is especially dependent on the geometry of vessel [30]. The knowledge of the distribution of velocity in the tank volume is very important from the point of view of pumping efficiency. The complexity of velocity distributions in agitated vessel brings up the difficulties in the correct definition of process dimensionless number where velocity is present. The velocity in an agitated vessel is usually described as follows:

$$w = \pi d n \tag{5}$$

The situation is more complicated in the case of the stirring of the non-Newtonian fluids because the viscosity is dependent on shear rate. The method of determination of the Reynolds number for shear-thinning fluid based on the average shear rate values has been proposed by Metzner and Otto [31]. For that purpose the equivalent viscosity η_e was defined as identical to Newtonian fluid viscosity, in the same geometrical and kinetical conditions within the laminar range of flow, when the same power for the liquid intermix is required. It is assumed that the average shear rate in agitated vessel is proportional to rotational speed of impeller:

$$\dot{\gamma}_m \propto n = B^* n \tag{6}$$

Constant B^* is characteristic for a given type of the agitated geometric system and for a given fluid group ($B^* = 4\pi$) [32]. For the power-law fluids:

$$\tau = K \dot{\gamma}^m \tag{7}$$

equivalent viscosity can be described by the following relation:

$$\eta_e = K(\dot{\gamma}_m)^{m-1} \tag{8}$$

the formula of the Reynolds number Re_{MO} proposed by Metzner and Otto [31]:

$$Re_{MO} = \frac{n^{2-m} d^2 \rho}{K} (4\pi)^{1-m} \tag{9}$$

has been used. The modified Prandtl number Pr_{MO} was resulted from the relation [32]

$$Re_{MO} Pr_{MO} = \frac{n d^2 c_p \rho}{\lambda} \tag{10}$$

hereby

$$Pr_{MO} = \frac{c_p K}{n^{1-m} \lambda} (4\pi)^{m-1} \tag{11}$$

and Schmidt number Sc_{MO} for power-law fluids:

$$Sc_{MO} = \frac{K}{\rho D_A} (4\pi)^{m-1} \tag{12}$$

The majority of previous studies were performed on local values of shear rate or heat transfer coefficient distribution in Newtonian fluids. The relationship between diffusion limiting current I_d, and shear rate $\dot{\gamma}_E$ in the surface electrode area [23] has been adopted:

$$\dot{\gamma}_E = \left(\frac{I_d}{2.156 z_e F C_{A0} D_A^{2/3} R_E^{5/3}}\right)^3 \tag{13}$$

The relationship used for values of $\dot{\gamma}_E$ and local shear stress is as follows:

$$\tau = \eta \dot{\gamma}_E \tag{14}$$

and for friction factor coefficient [24], accordingly:

$$c_F = \frac{D^2 \tau}{n^2 d^4 \rho} = \frac{D^2 \eta \dot{\gamma}_E}{n^2 d^4 \rho} \tag{15}$$

Firstly, in measurements in present studies the electrolytic solution used was as follows: 0.010 (kmol m^{-3}) $K_3[Fe(CN)_6]$, 0.050 (kmol m^{-3}) $K_4[Fe(CN)_6]$ and 0.5 (kmol m^{-3}) NaOH. It has been found that the presence of NaOH in a solution causes the change in non-Newtonian properties of Na-CMC solution [33]. This effect was

Table 1
The characteristics of the solutions tested

Solution tested	$C_{p,p}$ (%)	$K_3[Fe(CN)_6]$ (mol m^{-3})	$K_4[Fe(CN)_6]$ (mol m^{-3})	K_2SO_4 (mol m^{-3})	ρ (kg m^{-3})	K (Pa s^m)	m	$D_A \times 10^{10}$ (m^2 s^{-1})	pH
Electrolyte	0	5	5	300	1041	0.0031	1	6.68	9.64
Na-CMC	0.05					0.004	0.91		6.35
	0.1					0.011	0.83		6.83
	0.15					0.026	0.77		8.20
	0.2					0.052	0.72		6.57
	0.1					0.0108	0.83		6.35
Guar gum	0.21					0.0413	0.33	6.15	6.83
	0.25					0.0934	0.66		8.20

explained by Barai et al. [34]. The addition of NaOH to Na-CMC aqueous solution causes the reaction:

$$\text{Cell–OCH}_2\text{COONa} + \text{NaOH} \rightarrow \text{HOCH}_2\text{COONa} + \text{NaCl} \quad (16)$$

with the formation of sodium glycollate. Similar findings have been reported by Khalil et al. [35].

Taking into account this fact the basic measurements were performed in the following solution: 0.005 (kmol m^{-3}) $K_3[Fe(CN)_6]$, 0.005 (kmol m^{-3}) $K_4[Fe(CN)_6]$ and 0.3 (kmol m^{-3}) K_2SO_4. The characteristics and properties of the model Newtonian and aqueous solutions of the polymer used are presented in Table 1. The values of diffusivity coefficients were taken from [36–38]. It has been shown that the value of limiting diffusion current is constant in the whole range of angular coordinates $\varphi \in \langle -43°; 43° \rangle$ tested, confirmed by measurements for Newtonian fluids. The deviations from the average values did not exceed the measurement error boundaries.

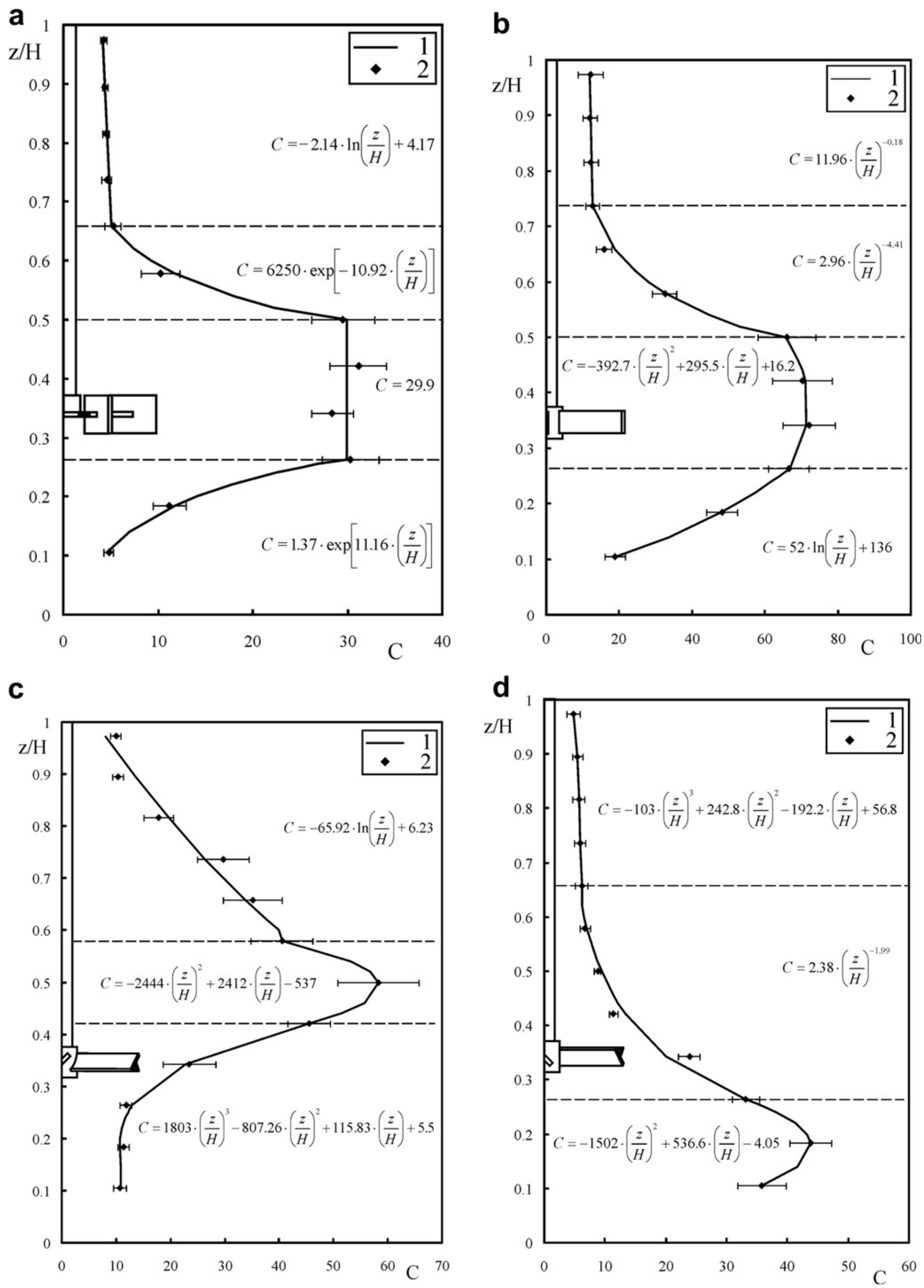

Fig. 3. Distributions of the constant C in Eq. (17): 1 – correlations equation, 2 – experimental points, (a) Rushton turbine, (b) the paddle with 6 blades ($\beta = 90°$), (c) the paddle with 6 pitched blades with angles of 45°, (d) the paddle with 6 pitched blades with angles of 135°.

The local values of heat transfer coefficients and friction factor coefficient were correlated in the form of dimensionless equations

$$Nu = CRe_{\mathrm{MO}}^{A}Pr_{\mathrm{MO}}^{B}V_{K}^{E} \tag{17}$$

$$c_{\mathrm{F}} = C_{1}Re_{\mathrm{MO}}^{A_1} \tag{18}$$

where coefficient C is a function of dimensionless coordinate z/H:

$$C = f\left(\frac{z}{H}\right) \tag{19}$$

and

$$V_{K} = \left(\frac{K}{K_{w}}\right)^{0.14} \tag{20}$$

As a result of the analysis of experimental data it was found that in the ranges of $Re_{\mathrm{MO}} \in (800, 22.300)$ and

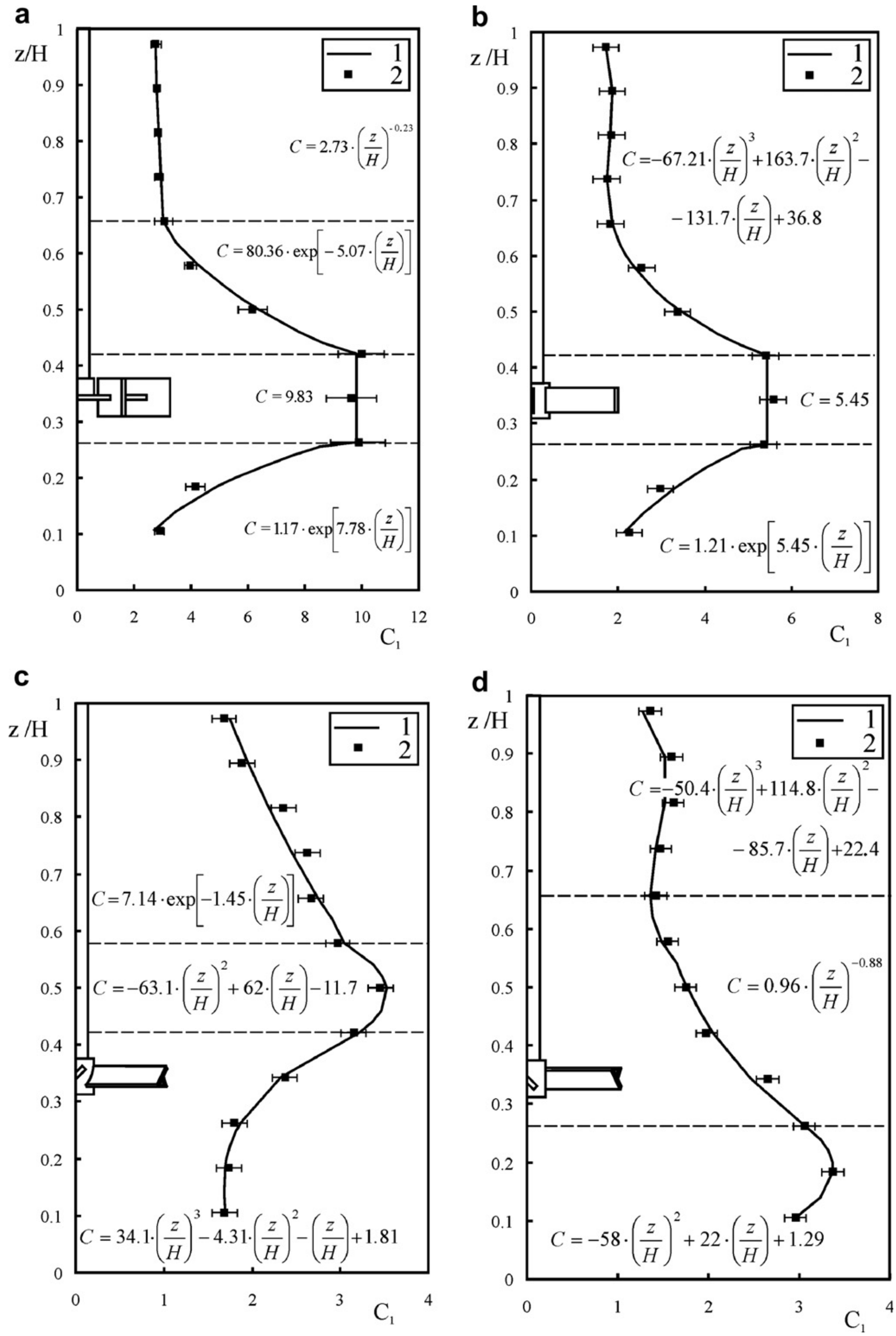

Fig. 4. Distributions of the coefficient C_1 in Eq. (18) – designation as in Fig. 3.

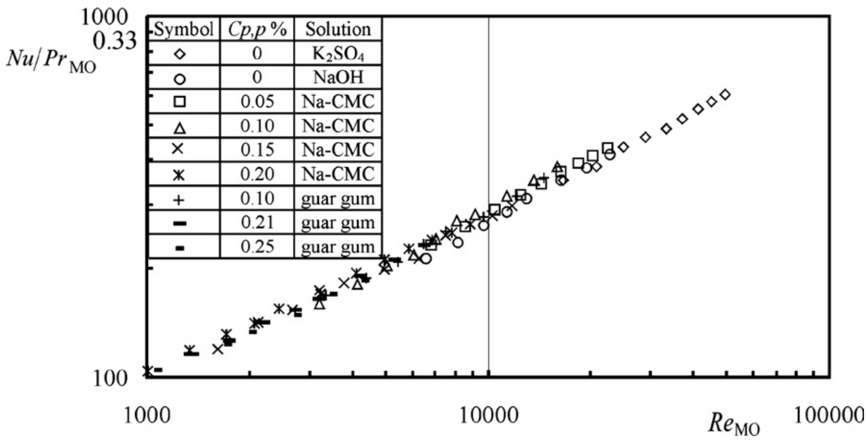

Fig. 5. The relation $Nu/Pr_{MO}^{0.33} = f(Re_{MO})$ for paddle with 6 pitched blades with angles of 45° and coordinate $z/H = 0.5$.

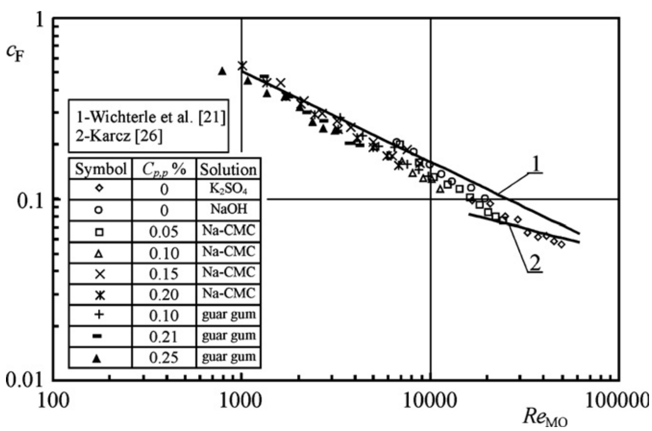

Fig. 6. The relationship of friction factor c_F on Reynolds number Re_{MO}, comparison of present experimental points with literature data for Newtonian fluids: 1 – Wichterle et al. [21], 2 – Karcz [23].

$Pr_{MO} \in (16, 157)$ tested the values of exponent A in Eq. (17) are independent of dimensionless coordinates z/H and the type of impeller used (for all impellers tested $A = 0.48$). The correlations of values of coefficient C on dimensionless coordinates z/H were characteristic for a given agitator construction (Fig. 3). The distributions of the coefficients C_1 in Eq. (18) for impellers tested are presented in Fig. 4.

The common relationships (for a given type of an impeller) between local values of heat transfer coefficient and Reynolds number Re_{MO} proposed by Metzner and Otto [31] for Newtonian and non-Newtonian fluids and given relationship z/H have been obtained. The exemplary relations of $Nu/Pr_{MO}^{0.33} = f(Re_{MO})$ are shown in Fig. 5. The common relationships for Newtonian and non-Newtonian fluids at values of $Re_{MO} \leqslant 22{,}300$ have been obtained. At greater values of Reynolds number only the single experi-

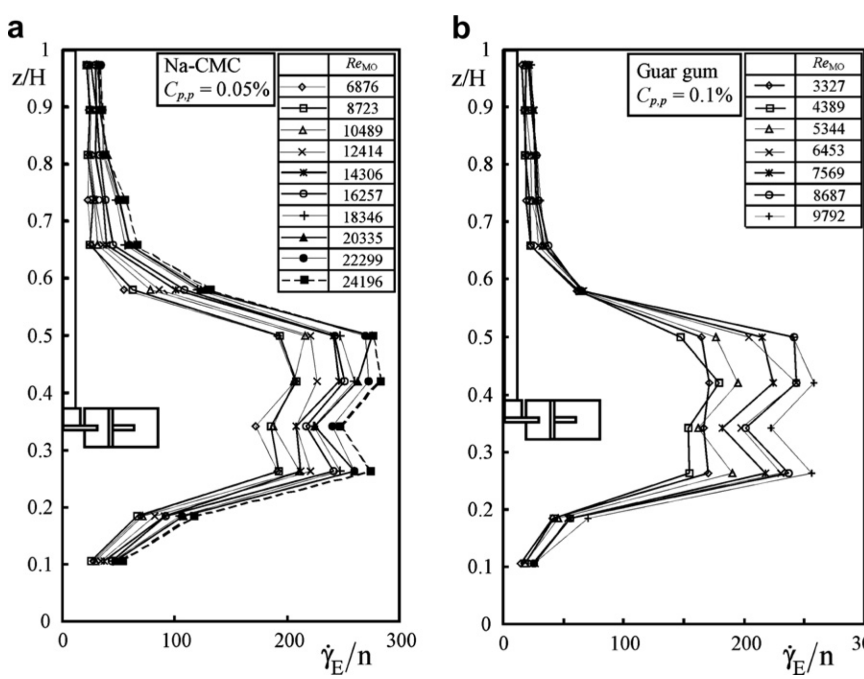

Fig. 7. Distributions of the dimensionless shear rate along agitated vessel wall for Rushton turbine: (a) Na-CMC aqueous solution, $C_{p,p} = 0.05\%$, (b) guar gum aqueous solution, $C_{p,p} = 0.1\%$.

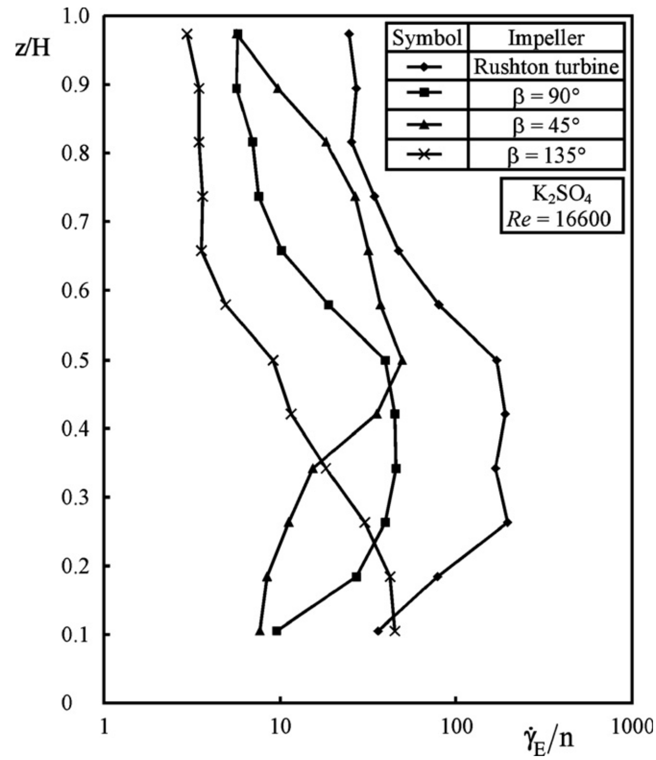

Fig. 8. Distribution of the dimensionless shear rate for impeller studied in Newtonian solutions tested.

mental points for Newtonian fluids have been obtained. The comparison of the distribution of local values of heat transfer coefficient for particular type of an impeller showed that the process proceeds most intensively in the

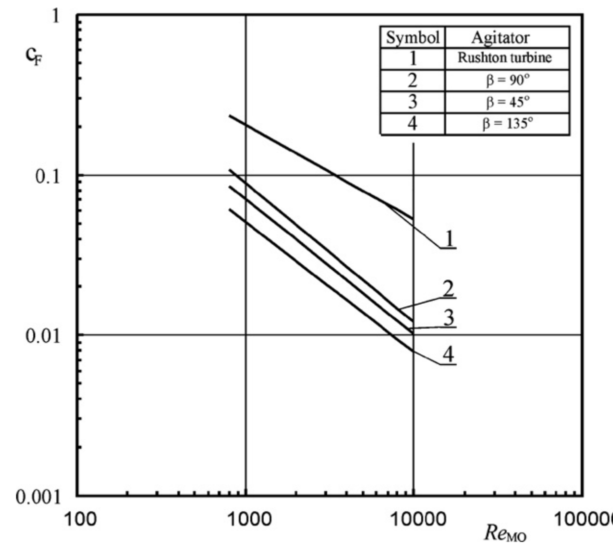

Fig. 10. Comparison of friction factor values averaged for impellers studied.

agitated vessel equipped with the Rushton turbine. For impellers with pitched blades the values of heat transfer coefficients are considerably lower possibly because the distribution of the adequate values along vertical wall is more uniform. As the result of the integration of equations for local values of friction and heat transfer coefficients the relationships for average values of these coefficients on total wall region of an apparatus were obtained.

Fig. 6 presents the comparison of experimental points obtained in our investigation with relationships from literature proposed for Newtonian fluids in the agitated vessel

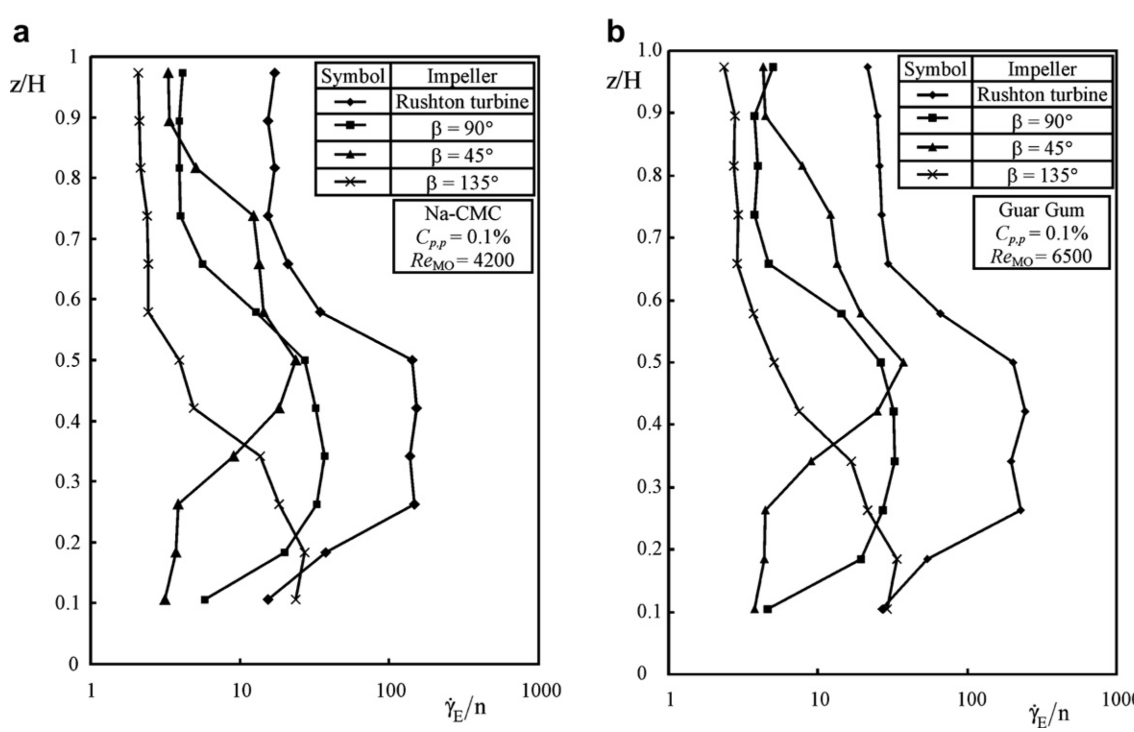

Fig. 9. Distributions of the dimensionless shear rate for impeller studied in Na-CMC (a) and guar gum (b) solutions of concentration of $C_{p,p} = 0.1\%$.

equipped with Rushton turbine. It is evident that experimental points at low values of Reynolds number (below about 8000) begin to overlap with the line described by Wichterle et al. [21] whereas at the highest Reynolds number values (above about 20,000) they begin to approach to the line plotted by Karcz [23]. Fig. 7 presents the distributions of the dimensionless shear rate on the agitated vessel wall equipped with the Rushton turbine. In the region of impeller stream impact (near $z/H = 0.34$) a decrease of dimensionless shear rate values has been observed. Possibly, it is connected with an appearance in this place of stagnation point, what is confirmed by observations of Wichterle et al. [21]. The distributions of the dimensionless shear rate along dimensionless coordinates z/H for impellers studied are shown in Figs. 8 and 9. A comparison of the distribution of local shear rate values showed that the process proceeds most intensively in the agitated vessel equipped with the Rushton turbine. The minimal values of shear rate are comparable with maximal ones for another impellers. This study has demonstrated that for a disc turbine and paddles with straight blades the maximum values occur in the region of impeller hanging. For impellers with pitched blades of angle 45° (pumping up a liquid) the maximum values occur above the impeller hanging region (at value $z/H = 0.5$), whereas for impellers with pitched blades of angle 135° (pumping down a liquid) maximum values are observed at the value of $z/H = 0.184$.

As a result of the integration of characteristic relations for local values of friction factor and heat transfer coefficient the common (for Newtonian and non-Newtonian fluids) relationships for average values of these coefficients on total wall region of an apparatus were obtained (Figs. 10 and 11). The adequate correlation relationships are listed in Table 2.

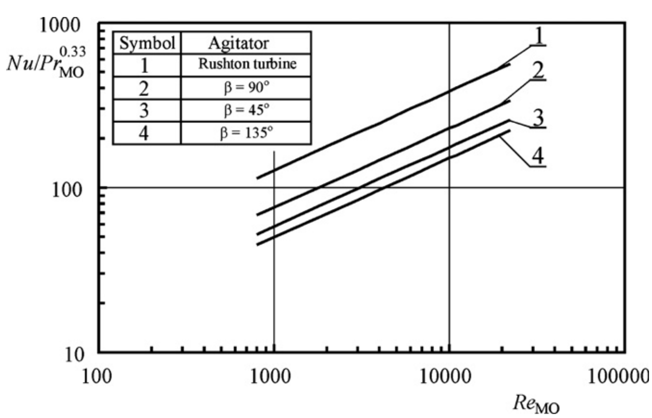

Fig. 11. Comparison of heat transfer coefficient values averaged for impellers studied.

Table 2
The correlation relationships for values of heat transfer coefficients and friction factors averaged for impellers tested

Impellers tested	Correlation equations	
	Average heat transfer coefficient α	Average friction factor c_F
Rushton turbine	$Nu = 4.6Re_{MO}^{0.48}Pr_{MO}^{0.33}V_K^{0.14}$	$c_F = 12.6Re_{MO}^{-0.59}$
paddle with 6 blades, $\beta = 90°$	$Nu = 2.74Re_{MO}^{0.48}Pr_{MO}^{0.33}V_K^{0.14}$	$c_F = 33.6Re_{MO}^{-0.86}$
paddle with 6 pitched blades, $\beta = 45°$	$Nu = 2.11Re_{MO}^{0.48}Pr_{MO}^{0.33}V_K^{0.14}$	$c_F = 23.4Re_{MO}^{-0.84}$
paddle with 6 pitched blades, $\beta = 135°$	$Nu = 1.81Re_{MO}^{0.48}Pr_{MO}^{0.33}V_K^{0.14}$	$c_F = 13.7Re_{MO}^{-0.81}$

5. Conclusions

The value of limiting diffusion current is constant in the whole range of angular coordinates $\varphi \in \langle -43°; 43° \rangle$ tested, confirmed by measurements for Newtonian fluids. The deviations from the average values did not exceed the measurement error boundaries. The common relationships (for a given type of an impeller) between local values of friction coefficient and heat transfer coefficient and Reynolds number (proposed by Metzner and Otto [31]) for Newtonian and non-Newtonian fluids have been obtained. The comparison of the distribution of local hydrodynamic and thermal parameters showed that the process proceeds most intensively in the agitated vessel equipped with the Rushton turbine. As a result of the integration of the relations determined by the local values of the both, the friction factors values and heat transfer coefficients, the correlation relationships for averaged values have been obtained. The Rushton turbine was characterized by the greatest values of mean heat transfer coefficients and friction factors. For all paddles tested the comparable efficiency has been obtained. This study has demonstrated that for determination of the flow and heat transfer characteristics in non-Newtonian media agitated the electrochemical method can be very useful. It has been found that the presence of NaOH in a solution causes the change of non-Newtonian properties of Na-CMC solution. It has been established that for studies on hydrodynamics in boundary layer and evaluation of local values of friction factor and heat transfer coefficients in Na-CMC and guar gum aqueous solutions by the electrochemical method the following solution of 0.005 (kmol m^{-3}) $K_3[Fe(CN)_6]$, 0.005 (kmol m^{-3}) $K_4[Fe(CN)_6]$ and 0.3 (kmol m^{-3}) K_2SO_4 can be recommended. Polymer aqueous solutions chosen in the study were very dilute that permitted to assume that the effect of a dissolved high-molecular substance on the diffusion kinetics of the electrode process is either negligible, or very small. The assumption needs the special confirmation in future.

Acknowledgement

This research was supported by Ministry of Science and High Education of Poland through Grant PUT DS ICt 32/113.

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