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Determination of the Effect of Suspended Nanosized Particles on Mass Transfer Rate in an Agitated Vessel

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Abstract- Mass transfer of ferricyanide ions through electrolyte nanofluids in an un-baffled, agitated vessel with a Rushton turbine was investigated over a range of Reynolds number from a laminer flow region to turbulent. Nanofluids consisted of 30-50 nm CuO nanosized particles suspended in ferri-ferrocyanide and aqueous potassium carbonate as the base fluid. A well-known electrochemical limiting diffusion current technique (ELDCT) was used to measure local mass transfer coefficients. It has been established that mass transfer rate strongly depends on the hydrodynamic parameters such as the rotational speed of the turbine, the blade angle of turbine (θ), and the dimensionless distance of the turbine from the bottom surface (H/d). The mass transfer coefficients were measured in the nanofluid and in the base fluid with no nanosized particles. Measurements showed that as the Re numbers increased, so enhancement ratio (k_1/k_2) which is defined as the ratio of the mass transfer coefficient of the nanofluid to the mass transfer coefficient of the base fluid, increased up to 8 times in axial direction and up to 3 times in radial direction. The highest values of (k_1/k_2) were measured for turbine with 30^o in axial direction and for the turbine as 45^o in radial direction. The effect of (H/d) was observed only in axial direction.

Keywords**:** Heat-mass transfer coeficient; Agitated vessel, Electrochemical limiting diffusion current technique, Nanofluid, Nanosized powder.

1. Introduction

Mechanically agitated vessels are widely used in chemical engineering systems because they play an important role in enhancing mass and heat transfer between a solid and liquid phase. Improved mixing is of great importance for liquid-to-liquid and liquid-to-solid systems, and it can be used for a variety of purposes, e.g., homogenisation of physical properties and composition, prevention of stratification or deposition of suspended particles, for improved rates of heat, mass transfer and chemical reaction (Geankoplis, C.J., 1993; Busciglio et al., 2012). Heat and mass transfer rates can be managed applying various techniques in several processes. Effective mixing is particularly important for vessels in which an exothermic reaction is forming or a vessel wall is reacting (Nguyen et al., 2007). However, the addition of non-reacting nanosized particles to the agitated liquid phase is one of the heat/mass transfer enhancement tehniques (Şara et al., 2011).

Many researchers have investigated heat transfer characteristics in non-Newtonian media agitated by using the electrochemical techniques. They reported that the processes proceeded most intensively in an agitated vessel equipped with the Rushton turbine and that mixing of liquid phases plays an important role in producing and increasing essential interfacial area to improve mass and heat transfer between liquid phases (Broniarz-Press et al., 2005; Ghotli et al., 2013). Mixing time in agitated vessels equipped with Rushton turbine have been measured by many researchers (Hiraoka et al., 2001; Busciglio et al., 2012).

Many researchers have studied about the addition of nanosized solid particles on the mass transfer rate in the liquid media for different geometries. In these studies, the effects of the addition of solid particles into the liquid phase on mass transfer have been investigated by using the electrochemical limiting diffusion current techniques (Şara et al., 2011; Sirajuddin and Talbot, 2011). In the experimental studies, such as CuO, SiC, Al_2O_3, B_4C, $CaCO_3$, glass, and some polymers have been widely used as particle materials.

The mass transfer rates between the vessel wall and the liquid can be controlled by means of changing the hydrodynamic parameters, such as, Re number depending on rotational speed, the angle of turbine blades and the distance between the turbine and the bottom surface of the vessel and by using nanosized powder. In order to elucidate the effect of nanoparticles on mass transfer, further researches with nanosized particles are still in demand, and this investigation was planned and carried out to make some contribution to this demand. The goal of this study is to determine the effects of the hydrodynamic parameters on mass transfer coefficient in ferri-ferrocyanide and potassium carbonate solution with CuO nanosized particles and in the base solution (without CuO nanoparticles). The investigation is performed for both laminar and turbulent flow regimes in a vertically-oriented vessel using ELDCT to measure mass transfer coefficient on the wall and bottom surface.

2. Experimental

The scheme of experimental system in the study is shown in detail in Figure 1. The jacketted vessel with a 12 cm inner diameter and 20 cm height is made of PVC material. Fourteen isolated nickel cathodes of 2 mm diameter for measuring the local mass transfer coefficients were mounted at 1cm intervals in the vertical direction on the vessel wall while a very thin nickel plate (10cm×10cm) located vertically on the vessel wall made up the anode. Additionally, twelve nickel cathodes with a diameter of 2mm for measuring local mass transfer coefficients were placed at 1cm intervals in the radial direction on the bottom surface of the vessel. The nickel electrodes are made of pure nickel wire (99.98%). Before each series of experimental measurements, the electroactive surface areas of nickel cathodes were determined using electrolytic methods according to Cottrell Equations (1) (Rieger, 1994), and all precautions reported by Berger and Ziai (Berger and Ziai, 1983) were taken.

$$I = nFAC_{\infty}\sqrt{\frac{D}{\pi * t}} \tag{1}$$

Where I is the current, n is the number of electrons entering the redox reaction, F is the Faraday constant, A is the electrode area, D is the diffusion coefficient of the ferricyanide ion in the solution, C_{∞} is the concentration of the ferricyanie ions in the bulk and t is time.

Distance between the center of cathodes and the bottom of the vessel was taken as the value of r (the radial coordinate in the cylindrical coordinate system), which was used to calculate dimensionless radial distance. Similarly, the axial coordinate (z) was used to calculate the dimensionless distance in z direction. The experimental studies were carried out for two types of solutions: the first type of solution was the mixture 5 $molm^{-3}$ potassium ferricyanide and 20 $molm^{-3}$ ferrocyanide with a supporting electrolyte of 500 $molm^{-3}$ potassium carbonate, and the second type of solution consisted of the first type of electrolyte with nanosized CuO particle, in the vessel equipped with Ruston turbine, and a paddle with four pitched blades with angles of 0^o, of 30^o, of 45^o and of 60^o in Figure 2.

A- Vertically located cathodes, **B-** Radially located cathodes, **C-** Rushton turbine, **D-** Mechanical Mixer and tachometer, **E-** Anode, **F-** Voltmeter, **G-** DC Power supply, **H-** Data Card, **I-** Computer.

The used turbines were made of 318 Cr-Ni stainless steel material with a diameter of 4cm. Nanosized CuO particles were provided by the NanoAmor Company, which are accepted to be spherical with a diameter of 30-50nm and density of $6300kgm^{-3}$. The quantity of nanosized particles in the electrolyte was expressed as volume fraction (ϕ). The volume fraction given with this equation is as following: ϕ=volume of fluid/volume of solution (Sara et al., 2011). In this study, the value of ϕ is taken as 0.94, since the optimum volume fraction value is in the range of 0.94-1.94% in the literature. Otherwise, the viscosity value of the electrolyte will have changed with the addition of nanoparticles. Nguyen et al. (2003) reported that the addition of CuO or Al_2O_3 nanosized particles up to a volume fraction of 13% into the water did not change its viscosity.

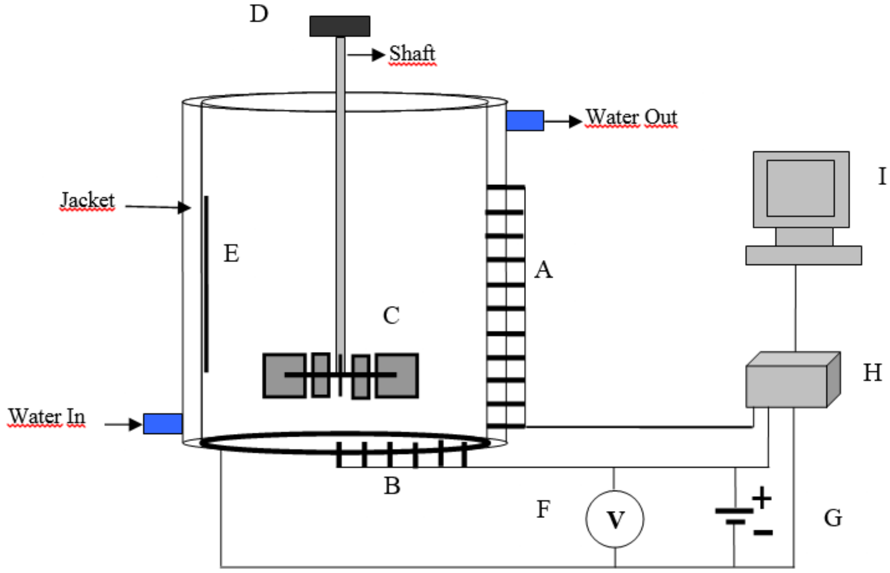


Fig. 1. The jacketted vessel agitated with Rushton turbine.

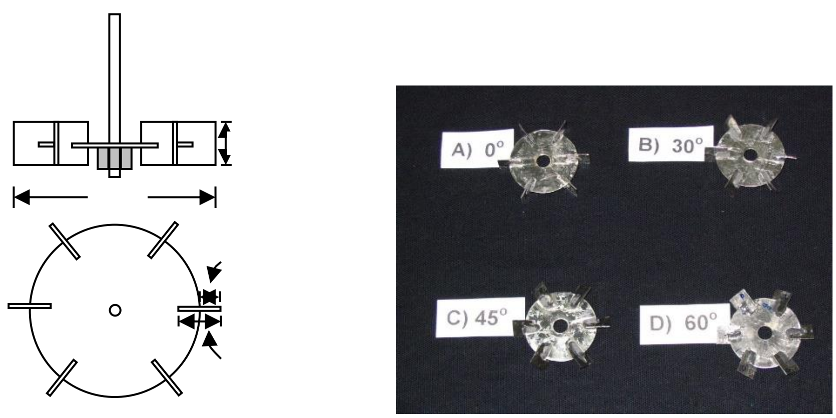


Fig. 2. Rushton turbine with four blades and the photographs used in the experiments.

Before the measurements of local current values, a working voltage value corresponding to the limiting current value for different impeller speeds was determined to be that of 0.60 V. At the operating conditions of 23±0.5°C, the physical properties of the electrolyte were determined from the study of Eroğlu et al. (Eroğlu et al., 2011). A controlled DC power supply was employed to apply a potential (0.6V) between two electrodes. A computer equipped with a data acquisition card was used for data recording.

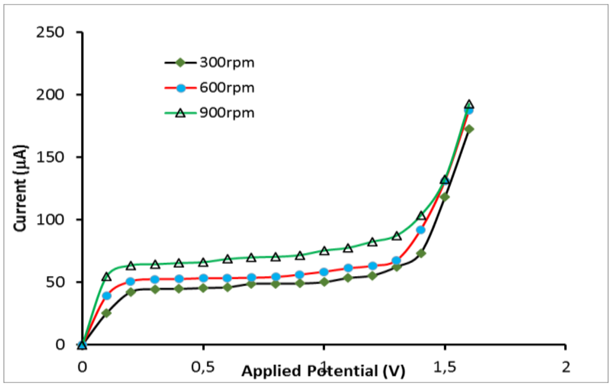


Fig. 3. Polarization Curves for different turbine rotational speeds.

The ELDCT is based on a diffusion controlled reaction of ferricyanide ions on the cathode surface. When the anode surface area and the concentration of ferrocyanide ions are bigger than cathode surface and the concentration of ferricyanide ions in the solutions, the chemical reaction is cathodically controlled. The local mass transfer coefficients (k) were calculated from the following equation derived for the limiting current diffusion controlled conditions (Selman, 1981 ; Mizushina, 1971).

$$k = \frac{I_{\lim}}{nFAC_{\infty}} \tag{2}$$

Where I_{lim} is the limiting current, n is the exchanged electron number, F is Faraday coefficient, A is electrode active surface area, C_{∞} is the concentration of the ferricyanide ions in the bulk. To calculate Sherwood numbers from the local current values, the following equation is used.

$$Sh = \frac{k * R}{D} \tag{3}$$

Where R is the electrode diameter, D is the diffusivity of ferricyanide ion. The velocity (w) in an agitated vessel is generally described with the following equation:

$$w = \pi * d * n$$
(4)

Where n is turbine rotational speed in s^{-1} and d is impeller diameter. The rotational speed is converted to Re numbers with the following equation:

$$\mathrm{Re} = \frac{w * d}{\nu} \tag{5}$$

The turbine rotational speeds converted to Re numbers to obtain the dimensionless form of the turbine speeds (Table 1).

Table 1. Reynolds numbers corresponding to the studied turbine speeds

Turbine speed (rpm)	300	600	900	1200
Re Numbers	7150	14,300	21,440	28,600

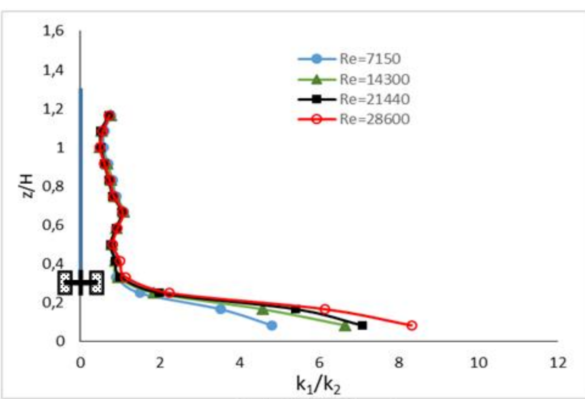
3. Results

Determination of mass transfer coefficient distribution on the inner surface of the vessel and the bottom surface of the vessel are of great importance for mass transfer applications for electrocoating applications in a vessel, for the situations of a catalytic reaction on the vessel wall, and particularly for the situation of forming heat transfer with high speed between a wall and a fluid in a vessel.

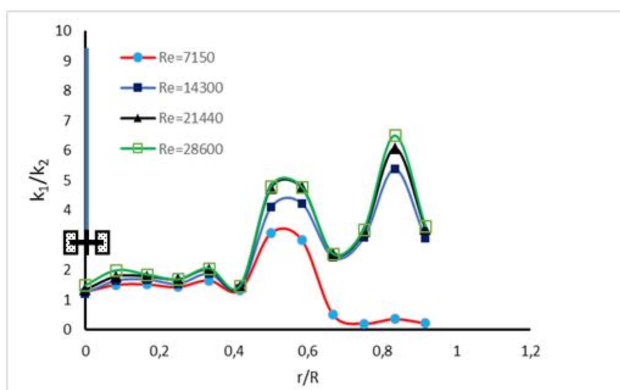
Generally, mixing refers to forcing a fluid by mechanical means to flow in a circulatory manner in a vessel. Mixing of a fluid in the vessel can cause a decrease in the thickness of the concentration boundary layer on the vessel wall, and thus the mass transfer rate interface of wall-fluid can be improved. Furthermore, the addition of non-reacting nanosized powder into fluid may help to increase the heat/mass transfer rate forming microconvective vortices near the electrode. The following graphs show the effects of working parameters and nanosized particles on distribution of the local mass transfer coefficients. These graphs show only the effects of the working parameters on the behavior of mass transfer coefficients in the vessel.

3. 1. Effect of Re Numbers on (k_1/k_2) Distribution

An increase in k_1/k_2 ratio was observed as seen in Figure 4a. k_1 was 5 times of k_2 in laminar flow at Re=7150 while k_1 was 8.5 times of k_2 in turbulent flow at Re=28600. The increase in the mass transfer was attributed to microconvective vortices formed by nanoparticles added into the solution, with the increase in Reynold`s number (Şara et al.,2011). The change of (k1/k2) ratio in terms of (r/R) as a function of increasing Reynold`s number at (H/d)=1 and $\theta=45^o$ is seen in Figure 4b. (k_1/k_2) ratio increased a bit as a function of Reynold`s number in the range of 0-0.4 (r/R) at the bottom of the tank since the disc part of the Rushton turbine could not generate a powerful axial flow at the center of the base region of the tank. There observed an increase as a function of the increasing Reynold`s number at the location where (r/R) is greater than 0.4. It was attributed to the better suspension obtained by the vigorous turbulent flow at the higher Reynold`s numbers and microconvective vortices formed by nanoparticles.



a) Axial distribution

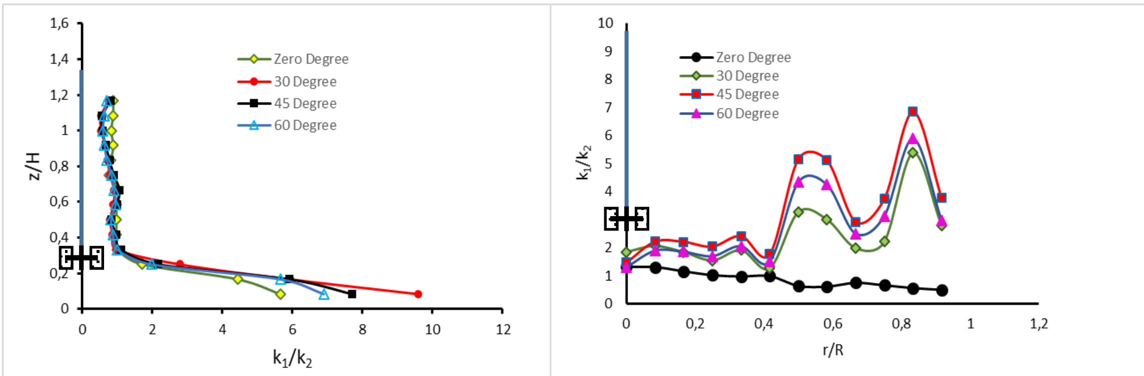


b) Radial distribution

Fig. 4. Distribution of mass transfer coefficient ratio with nanopowder and no powder for (H/d)=1 and $\theta=45^o$.

3. 2. Effect of angle of turbine blades on (k_1/k_2) distribution

The distribution of (k_1/k_2) in axial direction is shown in the agitated vessel equipped with the Rushton turbine with different blade angles in Figure 5a. It is clear that the use of nanopowder is not so effective in the range in which (z/H) is greater than 0.3, and that the best suspension can be obtained by the 30^o turbine angle while the worst can be obtained by zero degree turbine angle depending on swirl intensities when it is examined as a function of turbine angles at the (z/H) ranges 0 to 0.3.



a) Axial distribution **b)** Radial distribution

Fig. 5. Distribution of mass transfer coefficient ratio with nanopowder and no powder for Re=21440 and (H/d)=1.

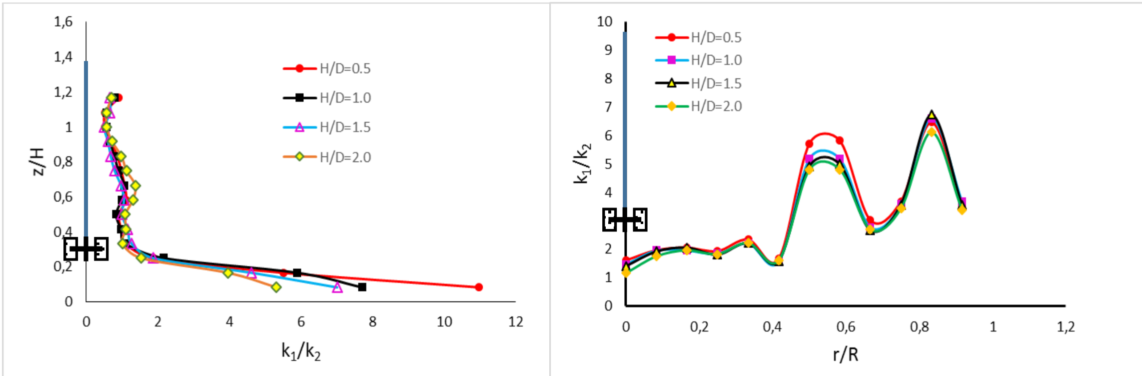
(k_1/k_2) change in radial direction for the Rushton turbines with different angles is shown in Figure 5b. Folded double increase in the (k_1/k_2) ratio is obtained in the distance from the center of the tank base to the (r/R)=0.4 in radial position. This is because that turbine cannot generate a strong flow owing to the fact that the disk region of the propeller (up to r/R=0.4) blocks the flow. The 30^o angle turbine has a lower effect on the (k_1/k_2) at the range in which (r/R) is greater than 0.4 because it cannot generate a powerful flow in axial direction. However, turbines with 30^o, 60^o and 45^o angles generate a powerful flow in axial as well as radial direction and form a better suspension. It has been observed that the ratio of (k_1/k_2) as a function of turbine angle in the region of (r/R)>0.4 increased from 0.5 to 7 by means of microconvective vortices formed by nanopowders at the bottom of the tank.

3. 3. Effect of (H/d) on (k_1/k_2) distribution

The axial change in (k_1/k_2) for the different (H/d) values when a Rushton turbine with 45^o angle is used and Re is 21440, is shown in Figure 6a. No nanopowder effect has been observed in vertical direction in the wall zone in which (z/H) is greater than 0.35. That the turbine in lower (H/d) locations gives higher (k_1/k_2) values may be attributed to the more powerful turbulent flow created by the turbine and therefore better suspension formation in the range of (z/H) 0 to 0.35. In Figure 6b, it is seen that (k_1/k_2) is not affected by (H/d) in the range in which (r/R) is between zero and 0.4 from the center at he bottom of the tank when 45^o Rushton tubine is used at Re=21140. On the other hand, in some locations where (r/R) is 0.58 and 0.83, (H/d) is slightly effective.

In the agitated vessels equipped with turbine, Sh (Sherwood) number is generally correlated as a function of Re number, (H/d), Sc, the shaft eccentricity (Broniarz-Pres, 2007). In the present system, the shaft is located concentrically in the vessel. For this reason, Sh numbers are correlated separately for each turbine blades angles by neglecting the shaft eccentricity as given in Eq. (6).

$$Sh = a\,\mathrm{Re}^b \left(\frac{H}{d}\right)^c Sc^d \qquad (6)$$



a) Axial distribution **b)** Radial distribution

Fig. 6. Distribution of mass transfer coefficient ratio with nanopowder and no powder for Re=21440 and θ=45^o.

In the equation (6), unknown constants for each turbine with different blade angles are given in Table 2 (in axial direction).

Table 2. The values of constants in Eq. (6)

Constants	**a**	**b**	**c**	**d**	**r^2**
0^o turbine with nanosized CuO	0.066	0.47	0.016	0.340	0.99
0^o turbine without nanosized CuO	0.040	0.240	-0.037	0.715	0.98
30^o turbine with nanosized CuO	0.227	0.313	-0.034	0.377	0.97
30^o turbine without nanosized CuO	0.05	0.422	-0.001	0.45	0.99
45^o turbine with nanosized CuO	0.003	0.43	0.018	0.83	0.99
45^o turbine without nanosized CuO	0.49	0.46	0.014	0.076	0.99
60^o turbine with nanosized CuO	0.211	0.490	0.023	0.153	0.99
60^o turbine without nanosized CuO	0.080	0.470	-0.037	-0.025	0.98

4. Conclusion

In this study, the effects of suspended nanosized particles on mass transfer are studied by the cooperated mixing system and electrochemical limiting diffusion current technique, and the following results are obtained,

- The addition of nanosized particles into the base electrolyte resulted in an increase of mass transfer coefficients,
- When the value of Re number increased up to 28600, the mass transfer coefficients with nanosized particles in comparison with those of no powder increased about eight times in axial direction. However in the radial direction, mass transfer coefficients with nanosized particles in comparison with those of no powder increased about three times.
- As the values of blade angles of Rushton turbines used in present investigation were compared, the maximum value of (k_1/k_2) increased about ten times in axial direction for the turbine of 30^o and about six times in radial direction for the turbine of 45^o.
- When (H/d) values were decreased from 2.0 to 0.5, the maximum values of (k_1/k_2) increased from 5.5 to 11 in axial direction.
- The measured mass transfer coefficients for different situations can be converted to heat transfer coefficient by using analogy. As a result, obtained mass transfer distribution shows only the heat transfer coefficient behavior in the axial and radial direction for the same geometry.

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