Regular article

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A microscopic experimental study of nanoparticle motion for the enhancement of oxygen absorption in nanofluids

<https://doi.org/10.1515/ntrev-2018-0072>

Received June 28, 2018; accepted October 25, 2018; previously published online November 15, 2018

Abstract: The behavior of nanoparticle motion has a great influence on gas-liquid mass transfer. However, it has been very difficult to characterize the motion of nanoparticles from a micro view in mass transfer experiments. In this study, a novel method was proposed to investigate nanoparticle Brownian motion through the application of the total internal reflection fluorescence microscope in a self-designed sample (a quasi-static liquid micro-groove) and the mass transfer enhancement of nanoparticles. Nanoparticle movement behavior was photographed using an electron-multiplying charge coupled device, and 100 consecutive images were recorded using Micro-Manager software at a rate of 20 fps. The images were processed through the particle tracking velocimetry algorithm to calculate two-dimensional motion rates of nanoparticles caused by Brownian movement. It showed that nanoparticle loadings influenced the motion rates significantly, and the motion rates were larger with smaller particle sizes under the same operating condition. The mass transfer coefficients in the quasi-static gas-liquid mass transfer system were calculated and analyzed through microscopic measurement. Based on the above thought, three important non-dimensional numbers [Sherwood (Sh_p), Reynolds (Re_p), and Schmidt (Sc_p) numbers] for mass transfer theory were studied.

Keywords: image analysis; mass transfer; nanoparticle motion; TIRF microscope.

1 Introduction

In order to enhance heat and mass transfer, nanofluids produced by adding nanoparticles into liquids were used as efficient working fluids instead of conventional fluids. However, the structures and transfer characteristics of nanofluids were different from those of pure liquids or conventional liquid-solid mixtures due to the ultrafine size effect of nanoparticles [1]. Therefore, the traditional conclusions were not suitable for nanofluids. Numerous experimental and theoretical studies were carried out to investigate the mechanisms of heat and mass transfer enhanced by nanoparticles. Krishnamurthy et al. [2] observed the microscale mass transfer of a dye droplet in water-based nanofluid and pure water using a stereomicroscope, and found that the rate of dye diffusion is faster in nanofluids. The rate peaked when the volume fraction of nanoparticles was 0.5%. They thought that the time required for a dye to bypass a nanoparticle was shorter than that for a dye to bypass the particle under Brownian motion, but longer than that for convection current while traveling an equal distance. It indicated that not only did the nanoparticle Brownian motion cause the mass transport enhancement directly, but also the increased nanoscale stirring of the liquid induced by nanoparticle Brownian motion could be responsible for such enhancement.

The effects of nanoparticles on heat and mass transfer are multifaceted and complex. Koo and Kleinstreuer [3] compared the effects of Brownian motion, thermophoresis, and osmophoresis on thermal conductivity through relevant models. They found that the Brownian motion effect was more significant than the other two effects. The thermophoretic and osmophoretic motion effects were independent of particle size. However, the Brownian motion effect was influenced by particle size. Buongiorno [4] studied the convective transport in nanofluids and concluded seven slip mechanisms (inertia, Brownian diffusion, thermophoresis, diffusiophoresis, Magnus effect, fluid drainage, and gravity). It was considered that only Brownian diffusion and thermophoresis

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were important slip mechanisms in nanofluids [5, 6]. The nanofluid properties vary significantly within the boundary layer due to the effect of the temperature gradient and thermophoresis. Therefore, the boundary layer effect is one of the important factors influencing transfer characteristics, which should be studied further. Prasher et al. [7] found that convection caused by nanoparticle Brownian motion was the main factor for the enhancement of thermal conductivity through an order-of-magnitude analysis. They also obtained the correlations for the effective thermal conductivity of nanofluids based on the Brownian-motion-based convective-conductive model. Similarly, the transport mechanism in nanofluids has been widely studied [8–12], and it was universally recognized that micro-convection caused by nanoparticle Brownian motion was the main reason for heat and mass transfer enhancement.

Heat transfer and mass transfer are similar processes, and there is resemblance between them [13]. Nagy et al. [14] used the equation for the effective thermal conductivities of nanofluids to predict increased diffusion coefficient directly, and found that the theoretical results could agree well with experimental results. The effects of various nanoparticles on gas-liquid mass transfer were widely investigated through macroscopic experiments [15–18]. Komati and Suresh [19] studied the effects of magnetic iron oxide nanoparticles on nanofluid properties, which influenced mass transfer significantly. However, the Brownian motion effect could not be studied from the microscopic point of view by experimental methods due to limitations of technical equipment. Nie and Lin [20] studied nanoparticle Brownian motion through direct numerical simulation using a fluctuating lattice-Boltzmann model, and relevant simulation studies were reported by Veilleux and Coulombe [21]. Yoon et al. [22] concluded that particle hydrodynamics caused enhanced mass flux through experimental and computational studies. Although the Brownian motion theory had contributed substantially to our understanding of various microscopic phenomena [23], the effects of Brownian motion on mass transfer enhancement remains unclear and needs to be further investigated through advanced technology [24].

In this paper, nanoparticle behaviors caused by Brownian movement in quiescent nanofluids were observed using total internal reflection fluorescence (TIRF) microscopy, and the nanoparticle motion velocities and mass transfer coefficients in nanofluids were calculated through an image processing method. The local convection induced by nanoparticle Brownian motion resulted in additional mass diffusion, called the micro-convective mass transfer. Finally, the correlation of additional diffusion coefficient

caused by nanoparticle Brownian motion indirectly was obtained by dimensionless analysis.

2 Experiments

2.1 Experimental setup

The main devices of the experimental setup are a TIRF microscope (Olympus IX71, Olympus Corporation, Shinjuku City, Tokyo, Japan), electron-multiplying charge coupled device (EMCCD; iXon3-897, Andor, Oxford Instruments, Oxford, UK), image acquisition software (Micro-Manager, Oxford Instruments, Oxford, UK), reagents (Aladdin, Pudong New Area, Shanghai, China) and a sample cell, and the experimental system layout is shown in Figure 1. The laser is a continuous diode-pumped solid-state laser emitting at $\lambda_0 = 488$ nm (Figure 1 in dark) and at a power $P_0 = 10$ mW. The microscope objective has a magnification of 60 times and a large numerical aperture of 1.42. When the incident beam occurs at the interface between the cover glass and deionized water, an evanescent wave with exponentially decaying intensity is generated to excite the fluorescent microsphere near the interface. A fluorescent signal centered at $\lambda_e = 515$ nm (Figure 1 in green) is emitted by the fluorescent microsphere and collected by a 16-bit intensified EMCCD camera. The resolution of EMCCD is 512×512 pixels and each pixel measures $16 \times 16 \mu\text{m}^2$, which leads to a pixel-wise lateral resolution of $267 \times 267 \text{ nm}^2$ with a 60 times magnification. The region field of view is $136.5 \times 136.5 \mu\text{m}^2$. Image acquisition is performed at a rate of 20 fps, and the exposure time is set to 50 ms. The evanescent wave generated by the total internal reflection on the observed samples would excite fluorescent particles in the 400-nm optical thin layer, and these particles would be observed in TIRF. TIRF visualizes flow field with high accuracy because it measures fluid flow in a non-contact method, which could avoid the interference of the flow field. Moreover, the signal-to-noise ratio (SNR) of the experimental image is very high.

Figure 1: TIRF microscopy setup.

Figure 2: Schematic of the sample cell.

The structure of the sample cell is shown in Figure 2, and it mainly consists of an air chamber (oxygen) and a liquid micro-groove (nanofluids). The inner diameter and length of the liquid micro-groove and air chamber are 4 mm and 20 mm, 29 mm and 23 mm, respectively. Vaseline with a uniform thin layer is used for sealing the cover glass (Fisher Scientific) onto the bottom wall of the sample cell. The cover glass with a thickness of 170 μm is translucent for the wavelengths of interests (excitation at 488 nm and fluorescence at 515 nm).

2.2 Experimental procedure

Firstly, the sodium sulfite solution with the concentration of 0.5 mol/l was made up using deionized water and sodium sulfite solid particles. Nanofluids with fluorescence effect were prepared: nanoparticles (10/20 nm TiO_2 , 20/60 nm SiO_2) were added into the sodium sulfite solution, and the mixtures were dispersed by ultrasonic vibration to prepare nanofluids with good stability. Fluorescent particles prepared by corresponding nanoparticles using 6-carboxy-fluorescein were selected and dispersed into nanofluids as the tracer particles, and the mass ratio of fluorescent particles to nanoparticles remained at 1:50. Then, pure oxygen was introduced into the gas chamber through the vent. After that, nanofluids were injected into the liquid micro-groove through a syringe and then the vent was plugged.

The TIRF microscope was transferred to the best working condition, and propagating light beam was aligned with the gas-liquid interface of sample cell placed on the stage. The gas-liquid interface was moved into the central field of view through the x-axis and z-axis adjustment knob, and the sample was focused through the

coarse/fine focusing knob. Then, the sample was photographed at a rate of 20 fps with 100 consecutive images recorded using Micro-Manager software. Taking 20 nm TiO_2 nanoparticles with a solid content of 0.2 g/l as an example, the first five pictures photographed continuously at 0 min are shown in Figure 3.

3 Image processing

A self-made procedure based on the particle tracking velocimetry algorithm was used to process the images with high SNR collected by EMCCD. The 100 images were divided into 99 adjoining pairs, as two adjacent images were treated as one pair. Image processing was divided into the following four steps sequentially.

3.1 Noise reduction

Faculae on the images could be divided into two types: one was formed by fluorescent microspheres deposited on the cover glass and remained stationary; the other was formed by free-moving fluorescent microspheres in the focal plane. The gray matrices of 100 images were superimposed and averaged as background noise. Before detecting particles, each image gray matrix should subtract background noise to eliminate the interference of stationary particles.

3.2 Particle identification

In order to separate the lighter spot signal and darker background noise, the original images were converted to

Figure 3: Images recorded by EMCCD.

Figure 4: Detection ranges and matching windows (black box sections). (A) Particle identification. (B) Particle position at t . (C) Particle position at $t + \Delta t$.

binary images with a proper gray threshold. The number of pixels occupied by spots, all of which should be particles identified using connectivity, was recorded. The location with the maximum gray value was treated as the particle center position.

The spots with 1–25 pixels were regarded as effective particles because others with >25 pixels would contain many “false particles,” such as particles caused by agglomeration or halo. The gray threshold is increased to

further identify particles, and the above steps are repeated until all eligible particles are found.

In fact, gray distributions of spots generated by real particles should be consistent with Gaussian distributions, so the spots closest to the Gaussian distribution were screened to be real particles. As shown in Figure 4A, a 5×5 detection range was established with the gray peak point as center, a 3×3 matching window was established with each pixel in the range as center, and the correlation

coefficients between the gray distribution of each window and the Gaussian distribution were calculated. The pixel location with the maximum correlation coefficient was the true position of the particle. In addition, the center position of these particles was developed to sub-pixel level to improve speed accuracy.

3.3 Particle match

As shown in Figure 4B and C, 21×21 detection windows were established with particle position as the center for the two adjacent images at time t and $t + \Delta t$, respectively. Because of more than one particle within Figure 4B, the particle position in Figure 4B needed to be found to match that in Figure 4A best. A 3×3 detection window was established with the particle position in Figure 4B as the center, and correlation coefficients of gray distributions

between the detection figure and Figure 4A were calculated. The particle with the maximum correlation coefficient was the best match. The particle match results for the images in Figure 3 are shown in Figure 5, where a red dot (t) corresponds to a nearby green dot ($t + \Delta t$), showing the locations of a particle in two adjacent exposure times, namely adjacent images between 1–2, 2–3, 3–4, and 4–5 in Figure 3.

Compared with the experimental results (images in Figure 3), it could be calculated through Eqs. (1) and (2) that the average detection efficiency of effective particles was 87% and the average particle matching efficiency reached 95%, both of which showed that the accuracy of data processing was very high.

$$E_d = \frac{1}{5} \sum_{n=1}^5 \frac{\text{The number of particles in detection}}{\text{The number of particles photographed}}, \quad (1)$$

Figure 5: Particle match of the images.

$$E_t = \frac{1}{4} \sum_{n=1}^4 \frac{\text{The number of matched pairs in detection}}{\text{The number of matched pairs for effective particles photographed}}, \quad (2)$$

where E_d and E_t represent the average detection efficiency of effective particles and the average particle matching efficiency, respectively.

3.4 Rate solving

Two-dimensional Brownian motion velocities on the focal plane could be calculated without the determination of distances in the z direction. The distances between a pair of particles in the x -direction and y -direction were assumed to be Δx and Δy , respectively. The interval was Δt , and the particle Brownian motion velocities could be calculated using the following equation:

$$\bar{V} = \sqrt{(\Delta x / \Delta t)^2 + (\Delta y / \Delta t)^2}. \quad (3)$$

4 Results and discussion

In order to improve the reliability and repeatability of the experiment, every experiment under the same condition was carried out five times, and the following results were the average of three parallel data values from multiple experiments.

4.1 Brownian motion velocity measurements

The experimental images of various nanoparticle conditions (10/20 nm TiO₂, 20/60 nm SiO₂) in the Na₂SO₃ solution with the concentration of 0.5 mol/l were processed using the above method, and Brownian motion velocities are shown in Figure 6, which could verify the correctness of the results through comparison with the studies by Li et al. [25]. As can be seen from Figure 6, the Brownian motion velocity of 10 nm TiO₂ nanoparticles at the lowest solid loading was 0.174 mm/s, which was the biggest among four kinds of nanoparticles. It was mainly because the mass of nanoparticles with smaller diameters was slight and the viscous resistance of nanoparticles was relatively small according to the Stokes theorem [26]; therefore, the effect of random Brown force on the particle motion would be more obvious. In addition, Brownian velocity decreased generally with the increase of solid loading for four kinds of nanoparticles; for example, the

Figure 6: Brownian motion velocities for various nanoparticle loadings.

velocity of 60 nm SiO₂ was decreased gradually from 0.150 to 0.141 mm/s by the solid loading increase from 0.2 to 1.0 g/l. The distance between nanoparticles was decreased by the increase of solid loading and the mutual effect between nanoparticles was strengthened, consulting in the reduction of nanoparticle motion. It could be verified from the Brownian velocity comparison of 20 nm TiO₂ and SiO₂ under the same solid loading: the particle number of TiO₂ was less than that of SiO₂ due to the difference of two molecular densities ($\rho_{\text{TiO}_2} > \rho_{\text{SiO}_2}$); thus, the distance of TiO₂ was longer than that of SiO₂ and the Brownian velocity of TiO₂ was greater than that of SiO₂.

It could be concluded that the Brownian motion velocity was influenced by the diameter and solid loading of nanoparticles. When the solid concentration was from 0.4 to 0.8 g/l, the Brownian motion velocity was smaller for smaller particle sizes under the same operating condition. However, some other things need to be further studied for the effects of solid loadings on Brownian motion velocity: both Brownian motion velocities for 10 nm TiO₂ and 20 nm SiO₂ at a solid loading of 1.0 g/l increased.

4.2 Mass transfer coefficient measurements

At the initial time, the pressure on both sides of gas and liquid is the same. Due to the chemical reaction between oxygen and sodium sulfite, the pressure of the liquid side was greater than that of the oxygen side as oxygen was

Figure 7: Images of sample at 0 and 30 min.

Figure 8: Diagram of gas-liquid interface in micro-groove.

absorbed. Thus, the liquid column moved to the oxygen side until the pressure is balanced on both sides of the liquid column. The volume of oxygen absorbed by nanofluids of sodium sulfite was equal to the volume passed by the liquid column and could be calculated by the migration length of the gas-liquid interface. The images of samples at 0 and 30 min are shown in Figure 7. Comparing the coordinates of the gas-liquid interface position at 0 and 30 min using Micro-Manager software, the distance traveled by the gas-liquid interface could be calculated. The curved shape of the gas-liquid interface is shown in Figure 8, which was affected by the interfacial tension. In this paper, the curved shape was assumed to be constant in 30 min because the interfacial tension change of nanofluids could be negligible.

Five corresponding positions on the gas-liquid interface of images were selected at 0 and 30 min equidistantly, and the distances between them were calculated. The average value is regarded as the distance traveled by the gas-liquid interface. The volume of oxygen absorbed by nanofluids is given by

$$V_0 = \frac{\pi d^2 l}{8}, \quad (4)$$

where d is the inner diameter of liquid micro-groove and l is the distance traveled by the gas-liquid interface. The

mass transfer of oxygen is calculated according to the following equation:

$$J = \frac{273V_0}{V_m T t} = \frac{34,125\pi d^2 l}{V_m T t}, \quad (5)$$

where V_m is gas molar volume under standard conditions, T is experimental temperature, which is 25°C, and t is the time for mass transfer. According to the definition of the mass transfer coefficient of gas and liquid, it could be expressed as follows:

$$k_L = \frac{J}{a(c_A^* - c_A)}, \quad (6)$$

where k_L represent the mass transfer coefficient of liquid; c_A^* and c_A are oxygen concentrations at the gas-liquid interface and another boundary of nanofluid, respectively; and a is cross-sectional area for mass transfer, which is measured multiple times and averaged through the contact angle measuring instrument. c_A is assumed to be 0 due to the very slow oxygen diffusion rate. c_A^* can be given according to Henry's law:

$$c_A^* = p / H, \quad (7)$$

where p is oxygen pressure and its value is 101,325 Pa and H is Henry's constant, which is 7.813×10^7 Pa · l/mol. Here, a is given by

$$a = \frac{(\pi - 2\theta)d^2}{8\cos^2\theta}, \quad (8)$$

where θ is the contact angle between nanofluid and oxygen, as shown in Figure 8. The mass transfer coefficients in the quasi-static liquid micro-groove enhanced by nanoparticles under various conditions are shown in Figure 9.

Figure 9: Mass transfer coefficients in nanofluids for various nanoparticle loadings.

Figure 10: Sherwood number in nanofluids for various nanoparticle loadings.

4.3 Theoretical analysis

Liquid creeping flow [27] could be caused by nanoparticle Brownian motion, and the thickness of the moving hydrodynamic boundary layer around the nanoparticles is larger than the distance between the nanoparticles even for very low particle loadings. Therefore, the flow field around particles can interact and form micro-convection, which increases the velocity gradients of liquid infinitesimal to generate additional mass diffusion called “micro-convective mass transfer.” Considering various factors affecting mass transfer, such as nanofluid properties, nanoparticle loadings, diffusion coefficient, and Brownian motion velocity, dimensionless analysis is used to study micro-convective mass transfer, which mainly includes the Sherwood number (Sh_p), Reynolds number (Re_p), and Schmidt number (Sc_p) [28].

4.3.1 Sherwood number

Assuming that the distribution structures of particles in the nanofluids are the same and the additional mass diffusion coefficients are isotropic, the micro-convective mass transfer caused by nanoparticle Brownian motion could be expressed as

$$J_i = -D_i \frac{\partial c}{\partial i}, \quad (9)$$

where J is the mass transfer; D represents the diffusion coefficient; c is the gas concentration; and i represents x , y , and z directions, respectively.

The effective diffusion coefficient in nanofluids can be considered as the sum of the diffusion coefficient in the pure solutions and the additional mass diffusion coefficient [29]. A modified Sherwood number (Sh_p) is introduced to characterize the ratio of micro-convection and diffusion mass transfer based on macroscopic convective mass transfer theory. The effective diffusion coefficient is given by

$$D_{\text{eff}} = D_0 + D_p = D_0(1 + Sh_p), \quad (10)$$

where D_0 and D_p are the diffusion coefficient in the pure solutions and the additional mass diffusion coefficient, respectively. The value of D_0 is determined by the Stokes-Einstein equation, which is $1.128 \times 10^{-9} \text{ m}^2/\text{s}$.

Therefore, the mass transfer enhancement by nanoparticles in this experiment could be analyzed by Sh_p , which could be calculated by the following equation:

$$Sh_p = \frac{k_L d_p}{D_0}, \quad (11)$$

where k_L is the mass transfer coefficient calculated in Section 4.2 and d_p is the diameter of nanoparticles. The calculated results for the Sh_p of the two nanoparticles (20 nm TiO₂, 20 nm SiO₂) are shown in Figure 10 with various nanoparticle loadings (0.2–1 g/l).

As can be seen from Figure 10, the Sh_p rises with the increase of nanoparticle loading when the nanoparticle loading is $< 0.6 \text{ g/l}$, but decreases with the increase of solid content when the nanoparticle loading is $> 0.6 \text{ g/l}$, showing that there is an optimum solid content for nanoparticles in this quasi-static gas-liquid mass transfer experiment, similar with Bahmanyar et al. [30] and Moghiman et al. [31]. For SiO₂ nanoparticles, the Sh_p is obviously weaker

Figure 11: Reynolds number in nanofluids for various nanoparticle loadings.

than that of TiO₂ nanoparticles because of the different adhesion force of the two kinds of particles.

4.3.2 Reynolds number

In addition to the modified Sherwood number, the Reynolds number characterizes the influence of Brownian motion on the moving hydrodynamic boundary layer around the nanoparticles and the expression of Re_p in the local area affected by nanoparticle motion is as follows:

$$Re_p = \frac{\rho_{nf} v d_p}{\mu_{nf}}, \quad (12)$$

where v is the nanoparticle Brownian motion velocity that could be calculated through the above measurement, and ρ_{nf} and μ_{nf} are density and dynamic viscosity of nanofluids, respectively.

Figure 11 shows the variations in the Re_p number for TiO₂-Na₂SO₃ and SiO₂-Na₂SO₃ nanofluids; the nanoparticle loading differs from 0.2 to 1.0 g/l at a same diameter of 20 nm. It can be obviously seen that the variation trend is consistent with the velocity change of particles shown in Figure 6, as the type and loading of nanoparticles have a weak influence on the density and dynamic viscosity of nanofluids in the TIRF experiment. The Re_p for both kinds of TiO₂ and SiO₂ is very close. It could verify that there is a similarity in Brownian motion for the different nanoparticles with the same diameter and temperature condition [32].

4.3.3 Schmidt number

The Schmidt number (Sc_p) is a non-dimensional parameter defined as the ratio of the kinetic viscosity and the

diffusion coefficient, shown in Eq. (13), which is used to describe the effect of viscosity on the mass diffusion coefficient:

$$Sc_p = \frac{\nu_{nf}}{D_0} = \frac{\mu_{nf}}{\rho_{nf} D_0}, \quad (13)$$

where ν_{nf} is the kinetic viscosity coefficient, D_0 is the diffusion coefficient in the pure solutions, and ρ_{nf} is the density of nanofluids, obtained using Eq. (14). μ_{nf} is the dynamic viscosity of nanofluids, which is experimentally measured. The experimental data on ρ_{nf} and μ_{nf} of 20 nm TiO₂ and SiO₂ nanofluids with different solid contents are listed in Table 1:

$$\rho_{nf} = \varepsilon \rho_p + (1 - \varepsilon) \rho_w, \quad (14)$$

where ε is volume fraction of nanoparticles, and ρ_p and ρ_w are the density of nanoparticles and base fluid, respectively.

As Figure 12 shows, the values of Sc_p for TiO₂-Na₂SO₃ and SiO₂-Na₂SO₃ nanofluids are equal and increase with the increase of nanoparticle loadings gradually, almost

Table 1: Experimental data on ρ_{nf} and μ_{nf} of 20 nm TiO₂ and SiO₂ nanofluids with different solid contents.

Solid content (g/l)	0.2	0.4	0.6	0.8	1.0
TiO ₂					
ρ_{nf} (kg/m ³)	1038.28	1038.43	1038.58	1038.72	1038.87
μ_{nf} (mPa · s)	1.400	1.413	1.452	1.481	1.499
SiO ₂					
ρ_{nf} (kg/m ³)	1038.25	1038.37	1038.48	1038.60	1038.72
μ_{nf} (mPa · s)	1.382	1.414	1.449	1.474	1.504

Figure 12: Schmidt number in nanofluids for various nanoparticle loadings.

linearly related. This is mainly because the addition of the two nanoparticles would affect the density of the solution slightly but their addition could affect the dynamic viscosity of nanofluids significantly. Moreover, the Sc_p numbers are all $>1.15 \times 10^3$, which are in conformity with the result of the calculation of the Sherwood number and the Reynolds number.

5 Conclusions

A novel method was proposed to study the Brownian motion effect on mass transfer enhancement by using the TIRF microscope. The images with high SNR were recorded by using EMCCD and processed to calculate nanoparticle Brownian motion velocity using a self-made procedure based on the particle tracking velocimetry algorithm. The Brownian motion velocity calculated in the experiment was significantly influenced by nanoparticle loading, and it was bigger with a smaller particle size under the same operating condition. Moreover, the mass transfer coefficients in nanofluids were calculated using Micro-Manager software. Three non-dimensional numbers (Sh_p , Re_p , and Sc_p) for the mass transfer theory were studied. The nanoparticle motion made the effect of convection a thousand times greater than that of diffusion in the mass transfer process. Compared with the blank solution, the introduction of the nanoparticles caused the nanofluid dynamic viscosity to increase and linearly vary with nanoparticle loadings.

Although the flow field around nanoparticles could interact and form micro-convection and the mass transfer in micro-groove was a process with low Reynolds number, the motion of nanoparticles could obviously generate additional mass diffusion called “micro-convective mass transfer” due to the Brown effect.

Acknowledgment: This work was supported by the National Natural Science Foundation of China (51606065), Funder Id: 10.13039/501100001809.

References

- [1] Raja M, Vijayan R, Dineshkumar P, Venkatesan M. Review on nanofluids characterization, heat transfer characteristics and applications. *Renew. Sustain. Energy Rev.* 2016, 64, 163–173.
- [2] Krishnamurthy S, Lhattacharya P, Phelan PE, Prasher RS. Enhanced mass transport in nanofluids. *Nano Lett.* 2006, 6, 419–423.
- [3] Koo J, Kleinstreuer C. Impact analysis of nanoparticle motion mechanisms on the thermal conductivity of nanofluids. *Int. Commun. Heat Mass Transf.* 2005, 32, 1111–1118.
- [4] Buongiorno J. Convective transport in nanofluids. *J. Heat Transf.-Trans. ASME* 2006, 128, 240–250.
- [5] Fisenko SP, Khodyko YA. Thermophoresis and the Brownian diffusion of nanoparticles in a flow reactor. *Tech. Phys.* 2012, 57, 336–343.
- [6] Rossi di Schio E, Celli M, Barletta A. Effects of Brownian diffusion and thermophoresis on the laminar forced convection of a nanofluid in a channel. *J. Heat Transf.-Trans. ASME* 2014, 136.
- [7] Prasher R, Phelan PE, Bhattacharya P. Effect of aggregation kinetics on the thermal conductivity of nanoscale colloidal solutions (nanofluid). *Nano Lett.* 2006, 6, 1529–1534.
- [8] Gupta A, Kumar R. Role of Brownian motion on the thermal conductivity enhancement of nanofluids. *Appl. Phys. Lett.* 2007, 91, 3102–3105.
- [9] Ho CJ, Chen DS, Yan WM, Mahian O. Rayleigh-Benard convection of Al_2O_3 /water nanofluids in a cavity considering sedimentation, thermophoresis, and Brownian motion. *Int. Commun. Heat Mass Transf.* 2014, 57, 22–26.
- [10] Michaelides EE. Brownian movement and thermophoresis of nanoparticles in liquids. *Int. J. Heat Mass Transf.* 2015, 81, 179–187.
- [11] Pinto RV, Fiorelli FAS. Review of the mechanisms responsible for heat transfer enhancement using nanofluids. *Appl. Therm. Eng.* 2016, 108, 720–739.
- [12] Xiao BQ, Yang Y, Chen LX. Developing a novel form of thermal conductivity of nanofluids with Brownian motion effect by means of fractal geometry. *Powder Technol.* 2013, 239, 409–414.
- [13] Turanov AN, Tolmachev YV. Heat- and mass-transport in aqueous silica nanofluids. *Heat Mass Transf.* 2009, 45, 1583–1588.
- [14] Nagy E, Feczko T, Koroknai B. Enhancement of oxygen mass transfer rate in the presence of nanosized particles. *Chem. Eng. Sci.* 2007, 62, 7391–7398.
- [15] Goodarzi HH, Esfahany MN. Experimental investigation of the effects of the hydrophilic silica nanoparticles on mass transfer and hydrodynamics of single drop extraction. *Sep. Purif. Technol.* 2016, 170, 130–137.
- [16] Kim JH, Jung CW, Kang YT. Mass transfer enhancement during CO_2 absorption process in methanol/ Al_2O_3 nanofluids. *Int. J. Heat Mass Transf.* 2014, 76, 484–491.
- [17] Linek V, Kordac M, Soni M. Mechanism of gas absorption enhancement in presence of fine solid particles in mechanically agitated gas-liquid dispersion. Effect of molecular diffusivity. *Chem. Eng. Sci.* 2008, 63, 5120–5128.
- [18] Veilleux J, Coulombe S. Mass diffusion coefficient measurements at the microscale: imaging a transient concentration profile using TIRF microscopy. *Int. J. Heat Mass Transf.* 2010, 53, 5321–5329.
- [19] Komati S, Suresh AK. Anomalous enhancement of interphase transport rates by nanoparticles: effect of magnetic iron oxide on gas-liquid mass transfer. *Indust. Eng. Chem. Res.* 2010, 49, 390–405.
- [20] Nie DM, Lin JZ. A fluctuating lattice-Boltzmann model for direct numerical simulation of particle Brownian motion. *Particuology* 2009, 7, 501–506.
- [21] Veilleux J, Coulombe S. A dispersion model of enhanced mass diffusion in nanofluids. *Chem. Eng. Sci.* 2011, 66, 2377–2384.

- [22] Yoon S, Chung JT, Kang YT. The particle hydrodynamic effect on the mass transfer in a buoyant CO₂-bubble through the experimental and computational studies. *Int. J. Heat Mass Transf.* 2014, 73, 399–409.
- [23] Dunkel J, Hanggi P. Relativistic Brownian motion. *Phys. Rep. - Rev. Sect. Phys. Lett.* 2009, 471, 1–73.
- [24] Pang C, Lee JW, Kang YT. Review on combined heat and mass transfer characteristics in nanofluids. *Int. J. Therm. Sci.* 2015, 87, 49–67.
- [25] Li TC, Kheifets S, Medellin D, Raizen MG. Measurement of the instantaneous velocity of a Brownian particle. *Science* 2010, 328, 1673–1675.
- [26] Doerr A, Hardt S, Masoud H, Stone HA. Drag and diffusion coefficients of a spherical particle attached to a fluid-fluid interface. *J. Fluid Mech.* 2016, 790, 607–618.
- [27] Bird RB. Five decades of transport phenomena. *Aiche J.* 2004, 50, 273–287.
- [28] Xu ZF, Khoo BC, Wijesundera NE. Mass transfer across the falling film: simulations and experiments. *Chem. Eng. Sci.* 2008, 63, 2559–2575.
- [29] Mu D, Liu ZS, Huang C, Djilali N. Determination of the effective diffusion coefficient in porous media including Knudsen effects. *Microfluid. Nanofluid.* 2008, 4, 257–260.
- [30] Bahmanyar A, Khoobi N, Moharrer MMA, Bahmanyar H. Mass transfer from nanofluid drops in a pulsed liquid-liquid extraction column. *Chem. Eng. Res. Des.* 2014, 92, 2313–2323.
- [31] Moghiman M, Aslani B. Influence of nanoparticles on reducing and enhancing evaporation mass transfer and its efficiency. *Int. J. Heat Mass Transf.* 2013, 61, 114–118.
- [32] Ovcharenko FD, Mikhailovskii VN. Theory of Brownian motion in nondilute suspensions. Influence of the indirect interaction between particles due to the disturbance of the medium during their motion. *Phys. Chem.* 1987, 292, 4–6.