Penerbit Akademia Baru

Journal of Advanced Research in Numerical Heat Transfer

Journal homepage:
http://www.akademiabaru.com/submit/index.php/arnht/index
ISSN: 2735-0142

Study of Self Diffusion of Nanoparticle Using Dissipative Particle Dynamics

Jian Hong Tan[1], Toru Yamada[2], Yutaka Asako[1,*], Lit Ken Tan[1], Nor Azwadi Che Sidik[1]

[1] Department of Mechanical Precision Engineering, Malaysia-Japan International Institute of Technology, University Technology Malaysia, Jalan Sultan Yahya Petra, Kuala Lumpur, 54100, Malaysia
[2] Nagoya Institute of Technology, Gokisocho, Showa-ku, Nagoya 466-8555, Japan

ABSTRACT

Nanofluids have been proven to exhibit enhanced thermal properties in heat transfer applications. To further improvise the thermal enhancement, it made sense to investigate the self-diffusion of the nanoparticles in the base fluid. However, the numerical study on the diffusivity of the nanoparticle is very limited. In this study, the diffusivity of nanoparticles with diameters of 17nm and 500nm was investigated numerically. Only the collision force between DPD and nanoparticles as well as drag force by the surrounding fluid is considered. The alternative expression of Boltzmann temperature proposed in the previous publication was applied to the current DPD (Dissipative Particle Dynamics) model for nanofluids. The volume concentration of the nanoparticles was fixed at 4% with water as the base fluid. Four test cases with different testing parameters were reported. The developed DPD model successfully captured the Brownian Motion of the nanoparticles. Other than that, the obtained diffusivity of the nanoparticles also agreed well with the past experimental results and Einstein's correlation. The present study obtained a percentage difference of approximately 3% to 30% compared to both past experimental results and Einstein's correlation. However, it is worth noting that a higher number of nanoparticles reduced the diffusivity of nanoparticles.

Keywords:
DPD; Dissipative Particle Dynamics; Self-diffusion; nanofluids; nanoparticle;

Received: 23 May 2022 **Revised: 12 August 2022** **Accepted: 18 August 2022** **Published: 8 September 2022**

1. Introduction

When Masuda *et. al* [1] first discovered that the thermal conductivity of water increased with the addition of nanoparticles, nanofluids began to attract a lot of interest. Numerous studies were carried out to discover the factors affecting the thermal enhancement of nanofluids. However, due to high experimental costs and time consumption, numerical approaches became important. By developing a relevant mathematical model, the behavior and properties of nanofluids can be studied. For example, Al Mahmud and Ismail [2] employed a type of mathematical model called Mixture and Volume of Fluid to study the effect of particle concentration and Reynolds number on the heat transfer enhancement of nanofluids. Good research usually validates its findings with both experimental and numerical results. Mahian [3] also mentioned that different CFD (Computational Fluid Dynamics) techniques are capable of studying different factors affecting the thermal

* *Corresponding author.*
E-mail address: y.asako@utm.my

enhancement of nanofluids. However, each method has its limitations. Hoogerbrugge and Koelman [4] introduced Dissipative Particle Dynamics (DPD) as a mesoscale, coarse-grained model of the Molecular Dynamics (MD) method. DPD can achieve larger time and length scales at a lower computational load when compared to the MD method. Later, DPD with energy conservation (DPDe) was introduced and used to study the suspension of nanoparticles in the base fluid.

Scientists have identified several factors that affect the heat transfer enhancement of nanofluids. The majority of the numerical and experimental studies focus on the type of nanoparticles, volume fractions, nanoparticle concentration, and Reynolds number. While Angayarkanni and Philip [5] also discovered that Brownian motion and self-diffusion affect the heat transfer enhancement of nanofluids, the related numerical studies are very limited. The self-diffusion coefficient of nanoparticles was predicted by Litvinov *et. al* [6] using the Smoothed DPD method. Groot and Warren [7] also derived an approximate expression for the self-diffusion coefficient by ignoring the conservative forces. Siddiqa *et. al* [8] also discovered that increasing the diffusivity ratio reduces the thermal enhancement of dusty nanofluids.

Since self-diffusion has been proven to affect the thermal enhancement of nanofluids, it can be used to validate the numerical model. This paper aimed to propose a modified DPD method with energy conservation (DPDe) capable of predicting the diffusion and Brownian motion of nanoparticles. The diffusion coefficient obtained numerically will be compared with Einstein's correlation and several experimental results.

2. Governing Equation

2.1 Isothermal Nanofluid

The governing equations for DPD are well documented by Tan *et. al* [9]. In this paper, the DPD model for nanofluids will be presented. The vibration of nanoparticles in the nanofluid within a fixed location is called lattice vibration. Such a vibrational method ensures zero thermal motion. In that case, it is safe to assume that the forces acting on the nanoparticles are only collision force, $\vec{f}_{ij}^{\ C}$, and drag force by the surrounding fluid, $\vec{f}_{ij}^{\ D}$. Also, only the random force between DPD particles is considered. External forces such as gravity will not be considered. With these, the time evolution of a nanoparticle is given by Newton's second law of motion as

$$m_{np}\frac{d\vec{v}_i}{dt} = \sum_{j\neq i}\left(\vec{f}_{ij}^{\ C} + \vec{f}_{ij}^{\ D}\right) \tag{1}$$

where m_{np} is the mass of the nanoparticle. The collision between a DPD particle and a nanoparticle during the time Δt is assumed to be an elastic collision. Since water is the base fluid in this research, a weighting function W^N is added to the collision force expression.

$$\vec{f}^C = \frac{2m_{np}m_{DPD}}{m_{np}+m_{DPD}}\frac{(\vec{v}_{DPD}-\vec{v}_{np})}{\Delta t}W^N/{N_m}^{\kappa} \tag{2}$$

N_m is the coarse-graining parameter. For low Reynolds number flow, Munson et. al. [10] suggested that the drag force by the surrounding fluid which is acting on the nanoparticle can be expressed as Eq. (3) where μ is the dynamic viscosity of the fluid and d_{np} is the diameter of the nanoparticle. Also, assuming the nanofluid is diluted, the collision between nanoparticles is considered rare. Therefore, for the interaction between nanoparticles, $\vec{f}_{ij}^{\ C} = 0$ and $\vec{f}_{ij}^{\ D} = 0$.

$$\vec{f}_{ij}^{\,D} = -3\pi\mu d_{np}\vec{v}_{np} \tag{3}$$

2.2 Dimensionless Form

The following dimensionless variables are defined so that the dimensionless form of the governing equations can be formed.

$$M_i = \frac{(m_{np})_i}{m_{DPD}}, R = \frac{r}{r_c}, \tau = \frac{t}{r_c/u_{DPD}}, m_{DPD} = \left(\frac{M}{N_A}\right) N_m, u_{DPD} = \sqrt{d(k_BT)_{DPD}/m_{DPD}} \tag{4}$$

By using the dimensionless variables from Eq. (4), the dimensionless form of Eq. (1) can then be expressed as

$$M_i \frac{d\vec{V}_i}{d\tau} = \sum_{j\neq i}\left(\vec{F}_{ij}^{\,C} + \vec{F}_{ij}^{\,D}\right) \tag{5}$$

The interaction between DPD particles can be expressed as

$$\vec{F}_{ij}^{\,C} = a^* W^C R_{ij}\vec{e}_{ij} \tag{6}$$

$$\vec{F}_{ij}^{\,D} = -\gamma^* W^D(R_{ij})(\vec{e}_{ij}\cdot\vec{V}_{ij})\vec{e}_{ij} \tag{7}$$

where a^* and γ^* are the controlling parameters that are expressed as

$$a^* = \left[75(\rho_{DPD})^{1/3}\right]/d \tag{8}$$

$$\gamma^* = \frac{\gamma r_c}{(m_{DPD})^{1/2} d^{1/2} (k_BT)_{DPD}{}^{1/2}}, \gamma = \frac{1575}{2\pi}\frac{\mu {N_m}^{1/3}}{n^{1/3}{\rho_{DPD}}^{5/3}} \tag{9}$$

The cell size is $r_c = (\rho_{DPD}N_m/n)^{1/3}$ and the number density of a fluid is $n = \rho/(M/N_A)$. M is the molecular weight, N_A is the Avogadro number, and ρ is the density of the fluid. In this paper, the water temperature is set at 300 K. Hence, $n = 3.33114 \times 10^{28}$. The Boltzmann temperature of the DPD particle is $(k_BT)_{DPD} = 0.83 k_B T {N_m}^{1/3}$ as proposed by Tan *et. al* [9].

As for the interaction between a DPD particle and a nanoparticle, the forces are expressed as

$$\vec{F}_{ij}^{\,C} = \frac{u_{ref}}{\sqrt{d(k_BT)_{DPD}m_{DPD}}}\frac{2m_{np}m_{DPD}}{m_{np}+m_{DPD}}\frac{(\vec{V}_{DPD}-\vec{V}_{np})}{\Delta\tau} W^N/{N_m}^{\kappa} \tag{10}$$

$$\vec{F}_{ij}^{\,D} = -\frac{-3\pi\mu d_{np} r_c}{\sqrt{d(k_BT)_{DPD}m_{DPD}}}\vec{V}_{np} \tag{11}$$

The weighting functions $W^C(R_{ij})$, $W^D(R_{ij}) = [W^C(R_{ij})]^2$, and $W^N(R_{ij}) = [W^C(R_{ij})]^6$ are expressed as Eq. (12) where $R_{ij} = r_{ij}/r_c$. Also, $k_B = 1.38065 \times 10^{-23}\, J/K$, $N_A = 6.02214 \times 10^{26} kmol^{-1}$, $M = 18.0153\, kg/kmol$, $\rho = (1.38951 \times 10^{-5}T^3 - 1.69792 \times 10^{-2}T^2 + 6.16839T - 298.957)\, kg/m^3$, and $\mu = 10^{0.000048T^2 - 0.038522T + 4.1678}\, Pa.s$.

$$W^C(R_{ij}) = \begin{pmatrix} (1-R_{ij}) & (R_{ij}<1) \\ 0 & (R_{ij}\geq 1) \end{pmatrix}, \tag{12}$$

3. Diffusivity of Nanoparticle

3.1 Simulation Parameters

In this paper, the diffusivity of nanoparticles with diameters of 17nm and 500nm is investigated. Table 1 shows the parameters of each test case. The temperature T will be set at 300 K for all cases.

Table 1
Parameters of test cases for different d_{np}

Test case	1	2	3	4
$d_{np}\ (nm)$	500	500	17	17
N_m	10^9	10^8	10^5	10^4
κ	0.4	0.4	0.6	0.6
ρ_{DPD}	10	10	4	4
N_{np}	10	4	10	4
$L \times M$	10×10	20×20	10×10	20×20
Volume Concentration (%)	4.16	4.16	4.09	4.09
r_c	6.6958×10^{-7}	3.1079×10^{-7}	2.2899×10^{-7}	1.0629×10^{-7}
m_{DPD}	2.9915×10^{-17}	2.9915×10^{-18}	2.9915×10^{-21}	2.9915×10^{-22}
$(k_B T)_{DPD}$	3.4378×10^{-18}	1.5957×10^{-18}	1.5957×10^{-19}	7.4066×10^{-20}
a^*	80.791	80.791	59.528	59.528
γ^*	66.858	66.858	226.85	226.85

3.2 Brownian Motion

The random movements of a nanoparticle in the nanofluid are well-known as the Brownian Motion. Figure 1 (a) and (b) show the unpredictable trajectories of the nanoparticles in the base fluid for $d_{np} = 500nm$ and $d_{np} = 17nm$ respectively. It can be seen that within $3 \times 10^6 s$, the position of three particles with the size of $d_{np} = 17nm$ randomly changes. Notice how the particles marked as red, blue, and black in Fig. 1 (a) crossed paths with each other and then went off in different directions. Notice how the particles marked as red, blue, and black in Fig. 1 (a) crossed paths with each other and then went off in different directions. Similar behavior can be seen in Fig. 1 (b).

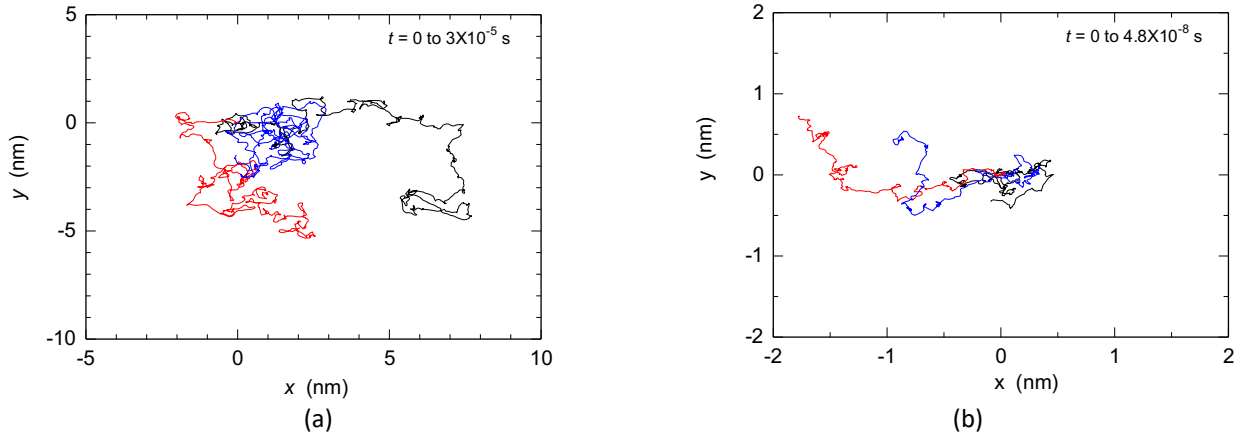

Fig. 1. Trajectories of nanoparticles (a) $d_{np} = 500nm$ (b) $d_{np} = 17nm$

3.3 Diffusivity of Nanoparticle

Albert Einstein [11] proposed that the diffusivity of a particle travels as Brownian Motion and it can be expressed as an equation. The theory was then translated into a book in 1956 [12]. The equation is rewritten into Eq. (13). Later, Guasto and Breuer [13], Sancataldo et. al. [14], and Dzakpasu and Axelrod [15] discovered the diffusivity of the nanoparticle experimentally. The percentage difference between the diffusivity obtained experimentally and theoretically was about 12% to 18%.

$$D = \frac{k_B T}{3\pi d_{np} \mu} \tag{13}$$

In this paper, the diffusivity of the nanoparticle can be obtained with Eq. (15).

$$D = \frac{S^2}{2t} \tag{14}$$

where $S^2 = \frac{1}{N_p}\sum_{i=1}^{N_p}[r_i(t) - r_i(0)]^2$ is the mean-square displacement that can be calculated from the computation, t is the time, and S^2 increases linearly with time. As such, the diffusivity obtained will become almost constant. Also, $r_i(t)$ is the location of the nanoparticle at time t while N_p is the number of nanoparticles.

Table 2
Diffusivity D (m^2/s) of nanoparticle

d_{np} (nm)	500		17	
Eq. (13)	1.03×10^{-12}		3.03×10^{-11}	
Guasto and Breuer [13]	-		3.57×10^{-11}	
Sancataldo *et. al.* [14]	9.00×10^{-13}		-	
Dzakpasu and Axelrod [15]	8.75×10^{-13}		-	
Eq. (14)	Test case 1	Test case 2	Test case 3	Test case 4
	5.60×10^{-13}	1.00×10^{-12}	2.60×10^{-12}	2.70×10^{-11}

Table 2 shows the diffusivity of the test cases 1 to 4. In Table 2, the computational results are tabulated along with the past experimental results and Einstein's correlation as in Eq. (13). From Table 2, it can be observed that for $d_{np} = 500\,nm$, test case 2 showed the best result when compared to both Einstein's correlation and experimental results. The percentage difference is only 2.91% and 14.29% respectively. The experimental results of Sancataldo *et. al* [14] and Dzakpasu and Axelrod [15] scored a percentage difference of 12.62% and 15.05% respectively when compared to Einstein's correlation. This means that the computational results agree very well with both experimental results and Einstein's correlation. As for $d_{np} = 17\,nm$, test case 4 showed the best result with a percentage difference of 10.89% and 32.22% respectively. Guasto and Breuer's [13] experimental result is approximately 17.82% different when compared to Einstein's correlation [11]. Test case 4 showed better agreement towards Einstein's correlation compared to the experimental result. Other than that, it has been discovered that $\mathrm{N_{np}}$ affects the diffusivity of nanoparticles more than other parameters.

4. Conclusions

As mentioned earlier, the numerical study of the self-diffusion of nanoparticles is very limited. In this paper, it has been proven that the DPD method can be used to predict the diffusivity of nanoparticles. Good agreement between 3% to 30% can be obtained when compared to both past experimental results and Einstein's correlation. Other than that, the DPD method also managed to correctly simulate the Brownian motion of nanoparticles suspended in the base fluid. It is also worth mentioning that a higher number of nanoparticles will generally reduce diffusivity. Upcoming works are being carried out with more test cases with different parameters so that the accuracy of the simulation can be improved.

Acknowledgement

The authors would like to express their deepest gratitude to Takasago Thermal Engineering Co, Ltd., Japan, for providing financial support for this work through Takasago grant (Vote No: 4B631).

References

[1] Hidetoshi Masuda, Akira Ebata, and Kazumari Teramae. "Alteration of thermal conductivity and viscosity of liquid by dispersing ultra-fine particles. Dispersion of Al2O3, SiO2 and TiO2 ultra-fine particles." (1993): https://doi.org/10.2963/jjtp.7.227

[2] Suaib Al MahmudAhmad Faris Ismail. "Multiphase CFD Investigation on Convective Heat Transfer Enhancement for Turbulent Flow of Water-Al2O3 Nanofluid." *CFD Letters* 13, no. 10 (2021): 11-24. https://doi.org/10.37934/cfdl.13.10.1124

[3] O. Mahian, L. Kolsi, M. Amani, Patrice Estellé, G. Ahmadi, C. Kleinstreuer, J.S. Marshall, R.A. Taylor, E. Abu-Nada, S. Rashidi, H. Niazmand, S. Wongwises, T. Hayat, A. Kasaeian, and I. Pop. "Recent advances in modeling and simulation of nanofluid flows—Part II Applications." *Physics Reports* 791 (2019): 1-59. https://doi.org/10.1016/j.physrep.2018.11.003

[4] PJ HoogerbruggeJMVA Koelman. "Simulating microscopic hydrodynamic phenomena with dissipative particle dynamics." *EPL (Europhysics Letters)* 19, no. 3 (1992): 155. https://doi.org/10.1209/0295-5075/19/3/001

[5] SA AngayarkanniJohn Philip. "Review on thermal properties of nanofluids: Recent developments." *Advances in colloid and interface science* 225 (2015): 146-176. https://doi.org/10.1016/j.cis.2015.08.014

[6] Sergey Litvinov, Marco Ellero, Xiangyu Hu, and Nikolaus A Adams. "Self-diffusion coefficient in smoothed dissipative particle dynamics." *The Journal of chemical physics* 130, no. 2 (2009): 021101. https://doi.org/10.1063/1.3058437

[7] Robert D GrootPatrick B Warren. "Dissipative particle dynamics: Bridging the gap between atomistic and mesoscopic simulation." *The Journal of chemical physics* 107, no. 11 (1997): 4423-4435. https://doi.org/10.1063/1.474784

[8] Sadia Siddiqa, Naheed Begum, MA Hossain, Rama Subba Reddy Gorla, and Abdullah AAA Al-Rashed. "Two-phase natural convection dusty nanofluid flow." *International Journal of Heat and Mass Transfer* 118 (2018): 66-74. https://doi.org/10.1016/j.ijheatmasstransfer.2017.10.067

[9] Jian Hong Tan, Toru Yamada, Yutaka Asako, Lit Ken Tan, and Nor Azwadi Bin Che Sidik. "Alternative expression for Boltzmann temperature of dissipative-particle-dynamics particles." *Numerical Heat Transfer, Part A: Applications* (2022): 1-17. https://doi.org/10.1080/10407782.2022.2066864

[10] T.H. Okiishi B.R. Munson, W.W. Huebsch and A.P. Rothmayer, *Fundamentals of Fluid Mechanics 7th edition*. 2013: John Wiley & Sons, Inc., New Jersey, .

[11] Albert Einstein. *Annual Physic (Berlin)* 17 (1905): 549-560.

[12] Albert Einstein, *Investigations on the Theory of the Brownian Movement*. 1956: Courier Corporation.

[13] Jeffrey GuastoKenneth Breuer. *Micro-velocimetry using time-resolved measurements of quantum dots in a microchannel*. in *ECI International Conference on Heat Ttransfer and Fluid Flow in Microscale, Whistler*. 2008.

[14] Giuseppe Sancataldo, Lorenzo Scipioni, Tiziana Ravasenga, Luca Lanzanò, Alberto Diaspro, Andrea Barberis, and Martí Duocastella. "Three-dimensional multiple-particle tracking with nanometric precision over tunable axial ranges." *Optica* 4, no. 3 (2017): 367-373. https://doi.org/10.1364/optica.4.000367

[15] Rhonda DzakpasuDaniel Axelrod. "Dynamic light scattering microscopy. A novel optical technique to image submicroscopic motions. II: Experimental applications." *Biophysical journal* 87, no. 2 (2004): 1288-1297. https://doi.org/10.1529/biophysj.104.041400