COMPARISON OF A COMPUTATIONAL MODEL OF SINGLE BUBBLE COLLECTION EFFICIENCY IN A HALLIMOND TUBE

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ABSTRACT

The paper reports how a CFD model compares to a set of experimental results to address single bubble collection efficiency (η). The particles were mono-dispersed (100-1000 nm) spherical nanoparticle. The experiment was performed in a Hallimond Tube (HT), one bubble at a time was produced at the bottom and was passed through water suspended with the nanoparticle. The main focus of this research is to solve the nanoparticle as a convection-diffusion model (CDM) and discrete particle model (DPM) and compare the results with the experimental collection efficiency. Prediction of the flow around a solid sphere and the CDM were compared with the pressure coefficient and theoretical collection efficiency respectively. The diffusivity of nanoparticles was calculated using the Einstein equation. It was concluded that neither CDM nor DPM predicts the collection efficiency well. This approach of transport of nanoparticles is a fundamental approach as it deals with the accurate solution around a rising bubble and well-established Brownian diffusion model (BDM).

NOMENCLATURE

k_B	Boltzmann constant	J/K
T	Temperature	K
h	Collector-particle distance	m
H	$H = h/R_p$	-
d_b	Diameter of the bubble	m
d_p	Diameter of the particle	m
R_p	Radius of the particle	m
C_c	Cunningham slip factor	-
F_{bm}	Brownian motion force	N
F_g	Gravity force	N
λ	Molecule mean free	m
PNC	No of particle/volume	m^{-3}
Pe	Peclet number	-
η	Collection efficiency	-

INTRODUCTION

The validation of a computer code of the collection of nanoparticle has industrial and research interest. First, this would allow directing the research of single bubble collection efficiency, η, with surface force modelling. Second, the fate of a nanoparticle in the environment (Blum, 2006) and in human lung (Park, et al., 2007, Robinson, et al., 2006) could be well-predicted.

The interaction of nanoparticle with bubble is very complex phenomena compared to the intermediate (10-100 μm) particle size range (Gaudin, 1957, Rulyov, 2001a, Trahar, et al., 1976). Nanoparticles are affected by Brownian diffusion (Kim, et al., 2006, Tan, et al., 2008), in absence of it, the particles would just follow the streamlines around a rising bubble. The collision efficiency should decrease with a decrease in particle size in the absence of Brownian motion, but the opposite occurs. Brownian motion is random motion of small particles, a schematic diagram is shown in Figure 1. Detachment of a nanoparticle is not significant, however, it is significant for coarse particles with less hydrophobicity (Pyke, et al., 2003). The experiments (Tan, et al., 2009) performed recently also confirms that there is no visible detachment of the nanoparticle (personal communication).

The current investigation is totally different from collection efficiency of swam of bubbles (Ramirez, et al., 2000, Reay, et al., 1973, Rulyov, 2001b, Yang, et al., 1995). In this part of the research, a portion of the experimental data is presented where the hydrophobic force is not significant (zero contact angle of the particle). The bubble-particle interaction experiment considered was under well-controlled conditions. The key parameter to determine whether particle diffusion is important or not in η is the $Pe = \frac{Ud_p}{D}$ number, which determines the ratio of convective transport to the diffusive transport. Here, D is the diffusivity. Using D (eq. (4)), the collection efficiency is compared with a tubular flow field (Imdakm, et al., 1991) using CDM. Most of the HT experiments were performed for a large particle bigger than nanoparticles (Hicyilmaz, et al., 2005, Markowski, et al., 2004). For a large d_p, uniform distribution of the particle in the HT is difficult to maintain because of the gravity effect. Bigger bubbles experience shape deformation (Bozzano, et al., 2001) and zigzag motion (Hassan, et al., 2008, Veldhuis, et al., 2008) and these introduce many other physical complications in terms of particle collection. Considering 1 mm bubble has minimised both of the issues. There is no 'tuning' of any computational parameters to match the experimental data. Later in a set of series of the

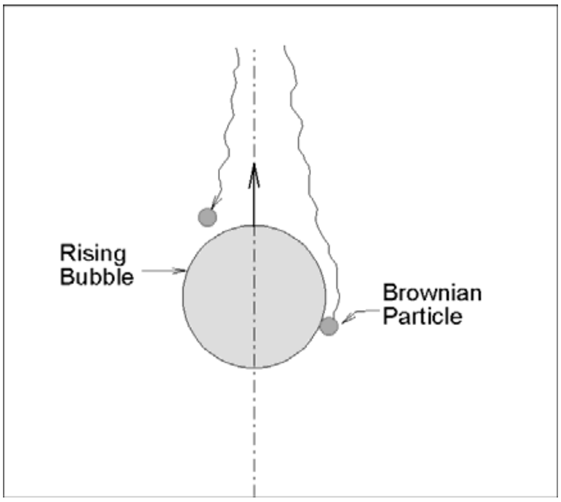

Figure 1: Schematic diagram of a rising bubble encountering a nanoparticle with Brownian motion.

publications, CFD validation of the calculation of η in the presence of some surface forces will be addressed. In this research, a steady state Navier-Stokes equation is solved using commercial software Fluent, a DPM and CDM were solved with the initial condition of the steady solution around a rising bubble. The particle diffusivity is taken from Einstein (Finlay, 2001). This is a proven approach applied and validated by many (Lee, et al., 2002, Lee, et al., 2000, Shi, et al., 2004, Zhang, et al., 2005) in other applications of nanoparticle deposition. Standard DPM model was solved, and the default Brownian motion does not work in Fluent (Longest, et al., 2008). A user defined Brownian diffusion model (BDM) of (Li, et al., 1992) is under development which should predict the η more accurately.

EXPERIMENTAL SETUP

The detailed experimental methods are available in previous publication (Tan, et al., 2009). Fluorescent core-plain shell silica particles of various sizes were prepared by the modified Stöber method (Blaaderen, et al., 1992). The morphology and size of the synthesized particles were characterized by scanning electron microscopy (SEM) (Cambridge Stereoscan, Camscan 2-90FE). A Hallimond tube (HT), (Figure 2) has two parts, the top part is clean and separated by a three-way valve. The bottom part is concentrated with suspended particles. Experiments were conducted using a dilute particle suspension (0.01 wt %). The experimental efficiency, $\eta_{s-\exp}$ was determined by dividing the number of particles recovered per bubble (N_{pf}) by the number of particles along the path of the rising bubble as given by eq. (1) (Dai, 1998):

$$\eta_{\exp t} = \frac{N_{pf}}{PNC\dfrac{\pi h_s\left(d_p + d_b\right)^2}{4}} \quad \text{.......................}\mathbf{1}$$

where h_s is the height of the column from the needle to the suspension-free zone (260 mm). The experiments were repeated in triplicate and the average is calculated. The experiments were based on the modified HT used by (Stearnes, 2001) as shown in Figure 2.

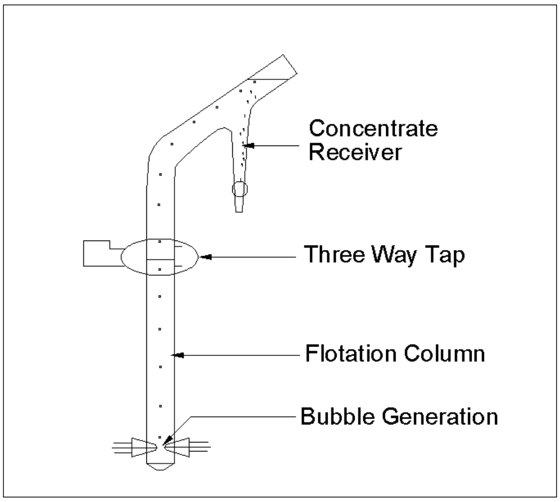

Figure 2: A schematic diagram of a HT used to measure the single bubble collection efficiency.

Table 1 shows experimental data and results of the collection efficiency and standard deviation (σ_{std}) of it. Mass of one particle (m_p) and the total number of particles (N_{tot}) in the suspension are shown Table 2. The particle concentration (*PNC*) and total number of particle collected by each bubble (N_{pf}) are shown in Table 3.

Table 1: Test cases of collection efficiency (η) for various particle size (d_p)

d_p (nm)	η	σ_{std}
100	2.30E-02	7.97E-04
200	1.78E-02	8.54E-04
500	5.97E-03	7.08E-05
650	5.80E-03	5.03E-04
1000	5.55E-03	3.43E-04

Table 2: Experimental data of particle mass (m_p) and total number (N_{tot})

d_p (nm)	m_p (kg)	N_{tot}
100	1.15E-18	5.30E+12
200	9.22E-18	6.62E+11
500	1.44E-16	4.24E+10
650	3.16E-16	1.93E+10
1000	1.15E-15	5.30E+09

Table 3: Experimental data of particle concentration (*PNC*) and number collected per bubble (N_{tot}).

d_p (nm)	$PNC(\#/m^3)$	N_{pf} (#/b)
100	9.63E+16	4.52E+08
200	1.20E+16	4.38E+07
500	7.70E+14	9.39E+05
650	3.51E+14	4.16E+05
1000	9.63E+13	1.09E+05

PNC decreases with the increase of d_p as the total mass of the particle is kept constant (0.0061 mg). The m_p or N_{tot} or *PNC* is proportional to d_p^{-3}, i. e., is if the d_p is doubled, m_p or N_{tot} or *PNC* will reduce by 8 times.

MODEL DESCRIPTION

Assuming a steady state flow around a rising bubble (laminar, isothermal), the continuity and momentum equations are solved. Conservation of nanoparticle is governed by CVM eq (2):

$$\frac{\partial C}{\partial t} + \nabla . \vec{J} = 0 \quad \text{……… 2}$$

$\vec{J}$, mass flux of nanoparticles is given by eq. (3):

$$\vec{J} = C\vec{U} - D.\nabla C \quad \text{… …3}$$

D in eq. (3) is the particle diffusivity as defined by Einstein (Finlay, 2001) equation by eq. (4):

$$D = \frac{kTC_c}{3\pi\mu d_p} \quad \text{……4}$$

Where k is the Boltzmann constant, d_p is the particle diameter.

In DPM, the model equation for the particle is defined as eq. (5):

$$\frac{du_{p,i}}{dt} = \frac{\rho}{\rho_p}\frac{Du_i}{Dt} + \frac{f}{\tau_p}\left(u_i - u_{p,i}\right) + f_{bf,i} \quad \text{....5}$$

Where f is the drag factor. On the bubble surface, a mobile boundary condition is considered for the liquid water (assuming bubble surface is clean) and Fick's law (Shi, et al., 2004) with zero particle concentration for CDM and a trap boundary condition for DPM.

RESULTS

Figure 3 shows pressure coefficient, C_p, around a solid sphere for Re=100 vs. the angular position of the surface. First, the convergence criteria (10^{-6}) is sufficient as beyond this there is no further change in C_p (not shown here for clarity). Secondly, around 200 divisions are needed around the periphery of the solid sphere to attain grid independent solution. These statements need further checking during CVM and DPM.

This part of the research is carried out to improve confidence in the CFD prediction on the bubble surface. The Re for a case of 1 mm rising bubble (with 0.82 m/s rising velocity) is 816.

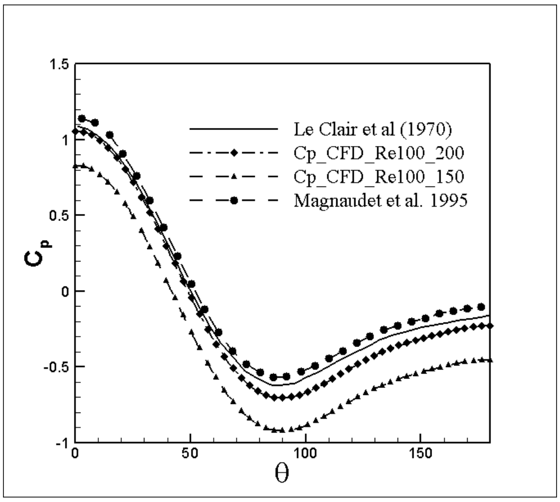

Figure 3: Pressure coefficient comparison for flow around a spherical solid

It is assumed that the mesh of this quality would produce the same level of accuracy for the case of Re=816. All the CFD cases presented are with higher order discretization. A higher order of discretization required extra computational costs and does cause stability in the convergence.

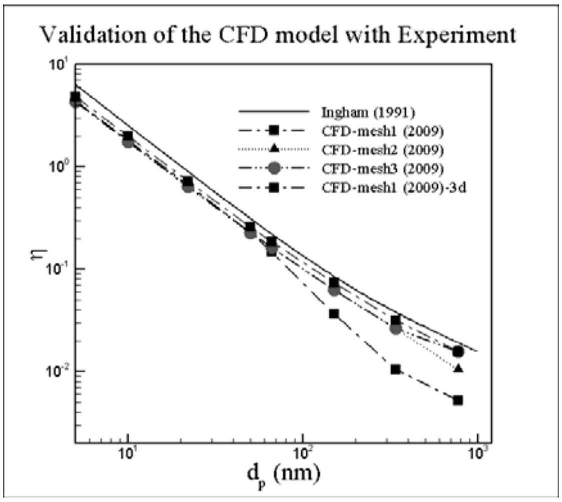

Figure 4: Validation of the current CFD model (convection-diffusion of (Ingham, 1991))

The CDM is validated with the analytical solution (Imdakm, et al., 1991). A user defined scalar of concentration is solved as shown by eq. (3) in addition to the continuity and momentum equation to calculate the flow inside a pipe of 56 mm length and collection of nanoparticles by the tube wall. Nanoparticles of size 2-1000 nm were injected and some of them were collected by the wall of the pipe. The results are compared and shown in Figure 4. The diffusivity was given by eq. (4), the Cunningham correction factor, C_c, is given by (Allen, et al., 1985). For all these cases, higher order discretization and strict convergence criteria (10^{-20} for C) were imposed. As shown in Figure 4, mesh3 provides a more accurate solution. Apparently, for the bigger particle size collection efficiency is more influenced by the grid size. The mesh size near the wall were 50 μm, 27 μm and 7.3 μm for the mesh1, mesh2 and mesh3 respectively. So as the mesh is refined, the CFD solution moves towards the theoretical prediction of Imdakm and Sahimi, 1991. This mesh size knowledge has been applied to case of the rising bubble simulation when nanoparticle considered as a diffusion problem (CDM). It is worth noting that, for the case in Figure 4, the particle inertia is not considered and so the solution does vary (Longest, et al., 2007) from the case when inertia is considered.

Figure 5 shows the CFD prediction of collection efficiency compared to the experimental collection efficiency. There are four different sets of CFD results shown. From the clean bubble surface towards the final surface condition, and from the zero velocity to 0.82 m/s, the rate of collection would be different. In CFD, only a steady bubble velocity and steady particle collection rate were considered. The lower the velocity, the higher the collection efficient for the DPM and the opposite phenomena happens for the case of CDM consideration. The CDM ignores the inertia; on the other hand the DPM ignores the effect of Brownian motion in the standard case shown in Figure 5. The CDM collection efficiency is much smaller than the experimental η which indicates that this is not a convection-diffusion phenomenon. On the other hand DPM η is very high compared to CDM. Note that the η_{dpm} increases rapidly because of the inertia above the 1000 nm which is not shown here.

These comparisons show that neither the standard DPM nor CDM ($Pe >> 1$) would accurately predict the collection of nanoparticle by the rising bubble. The consideration of one-way coupling (d_b/d_p =10000 to 1000) or two way coupling did not alter the collection (in DPM) as particle inertia is very negligible. Now an UDF for the Brownian diffusion model (BDM) is under development to investigate the effect of Brownian diffusion, based on an established theory (Li, et al., 1992), to predict the collection. The effect of Brownian motion depends on how long the bubble takes to rise to the top of HT as the total distance traveled by diffusion is $\sqrt{2Dt}$.

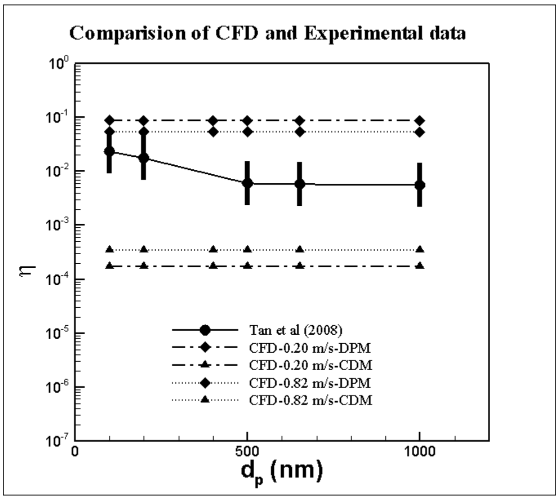

Figure 5: Comparison of current CFD and experimental data for the collection efficiency of a rising bubble in a HT.

It should be noted that most of the cases studied by others, the collection efficiency prediction is one order magnitude lower than the experimental data.

There are some key differences to mention. The single bubble experiment was performed in a HT, velocity was raised to 0.82 m/s from zero. Need further investigations to answer this: for the unsteady nature of the rising bubble does the collection happens at the same rate? Still need to quantify the collection rate for the unsteady nature. Another key difference from the HT is that, a 0.82 m/s is set across the radius of the HT even though the bubble was set to be stationary. Will that alter to collection as the dynamics is different? In the case of bubble, as it collects the particles, the dynamics around it changes and there is mostly likely particle-particle attraction which is yet to be determined by experiment. The hydrophobic force model is poor in this region. The presence of the particle modifies the smoothness of the surface of the bubble and so is the condition around the rising bubble.

It is interesting to note that the $\eta_{\exp t}$ decreases from 100 nm to 500 nm and then it remain constant. More interestingly, the error bar is quite high for lower particle size. The experimental data needs to be analysed more to attain further insight for this case presented. Nevertheless, this is a fundamental approach to look at the interaction of a rising bubble with nanoparticle.

CONCLUSIONS

Validation of the flow around a solid sphere is performed with great level of confidence (for Re=100), so the accuracy of the primary flow is very accurate for Re=816. From the experience of the flow around a solid sphere, the flow around a complete mobile surface is predicted by solving full Navier-Stoke equation for an axi-symmetric case. Further knowledge was gained by validation of a CDM of nanoparticle flow inside a pipe.

For DPM, any particle touching a virtual boundary is considered to be collected by the rising bubble. The virtual boundary was created by a UDF which is basically a trap (sink) around the rising bubble perpendicularly away from the surface of r_p distance.

The rising of a bubble from zero is difficult to model in terms of accumulation of the collected particles.

Single bubble experiments in a HT is a fundamental approach in terms of validating the computational model, rather than tuning the model parameters and comparing the collection efficiency data from a bubble swam experiment.

The experimental data has been used to test CFD collection efficiency. This would allow to model surface forces to incorporate the particle attachment process or develop a new model. The surface of bubble is a complex phenomenon in terms of its mobility, hydrophobicity and interaction with nanoparticle.

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