An improved model for estimating fractal structure of silica nano-agglomerates in a vibro-fluidized bed

Ali Asghar Esmailpour, Navid Mostoufi* and Reza Zarghami
Multiphase Systems Research Lab., School of Chemical Engineering, College of Engineering, University of Tehran, P.O. Box 11155/4563, Tehran, Iran

ABSTRACT

A study has been conducted to determine the effects of operating conditions such as vibration frequency, vibration amplitude on the fractal structure of silica (SiO_2) nanoparticle agglomerate in a vibro-fluidized bed. An improved model was proposed by assimilation of fractal theory, Richardson-Zaki equation and mass balance. This model has been developed to predict the properties of nanoparticle agglomerate, such as fractal dimension and its size. It has been found out the vibration intensity increase leads to a slight reduction in fractal dimension of agglomerate. This Paper is also indicated that the size of agglomerate has the same behavior as fractal dimension with respect to vibration intensity changes. This study demonstrated that the fractal dimension of Silica nanoparticle agglomerate is in the range of 2.61 to 2.69 and the number of primary particles in the agglomerate is in the order of 10^{10}. The vibration frequency is more impressive than its amplitude on agglomerate size reduction. Calculated Minimum fluidization velocity by applying predicted agglomerate sizes and experimental data are acceptable fitted.

Keywords: Fluidization, Agglomerates, Fractal dimension, Nanoparticles, Vibration

NOMENCLATURE

A	vibration amplitude, m
d_a	agglomerate diameter, m
D_f	fractal dimension
F	vibration frequency, Hz
g	acceleration of gravity, m/s^2
g_{eff}	effective gravity acceleration, m/s^2
H	bed height, m
H_0	initial bed height, m
k_g	pre-factor
m_a	mass of an agglomerate, kg
N	number of primary particles in an agglomerate
n	Richardson-Zaki exponent
R_a	agglomerate radius, m

*Corresponding Author: E-mail: mostoufi@ut.ac.ir

R_g gyration radius, m
R_h hydrodynamic radius, m
R_p particle radius, m
Re_p particle Reynolds number
S bed cross section area, m^2
u_{mf} minimum fluidization velocity, m/s
V_a agglomerate volume, m^3
V_p particle volume, m^3
v_a settling velocity of a single agglomerate, m/s
v_g superficial gas velocity, m/s
v_{p_0} settling velocity of a single particle, m/s
v_s settling velocity, m/s

Greek Symbols
ε total bed voidage
ε_a agglomerate voidage
ε_0 bed voidage at rest
ε_{mf} bed voidage at minimum fluidization
Γ vibration intensity
μ_g gas viscosity, Pa.s
φ volume fraction of particles in bed
φ_a agglomerates volume fraction in bed
φ_s solid volume fraction
ρ_a agglomerate density, kg/m^3
ρ_{a0} initial agglomerate density, kg/m^3
ρ_b bulk density, kg/m^3
ρ_p particle density, kg/m^3
ρ_g gas density, kg/m^3
ψ_s particle sphericity

1. INTRODUCTION

In recent years, there has been growing interest in nanoparticle fluidization. Agglomeration is a common phenomenon in nanoparticles fluidization as result of strong interparticle force [1–4]. Several attempts have been made to determine the aspects of nanoparticle fluidization [3, 5–9].

Previous studies have revealed that the fluidization of nanoparticles can be classified into two categories, including agglomerate particulate fluidization (APF) and agglomerate bubbling fluidization (ABF). The APF shows homogeneous (bubble-less) fluidization, high bed expansion ratio by increasing the gas velocity and behaves as fluid-like fluidization [10]. In contrast, the ABF has large bubbles and low bed expansion ratio [10].

Bed expansion of nanoparticles in the APF fluidization can be described by Richardson-Zaki (R-Z) equation [4, 6, 10, 12–13] and modified R-Z equation [14]. Valverde and Castellanos [14] revealed that the modified R-Z equation is more accurate than the R-Z equation.

Due to strong cohesive forces, the nanoparticles tend to stick to each other and produce a large porous agglomerate. This leads to decrease in small size and high specific surface area of nanoparticles. Hence, external forces such as mechanical vibration [13, 15–17], sound waves [18–19], magnetic fields [20] and centrifugal fields [7, 21], are applied to improve fluidization quality by decreasing the size of agglomerates. Vibration can help to remove channels, break up agglomerates into smaller ones and overcome the interparticle

forces [22]. Indeed, using the vibration leads to increased external forces (repulsive forces). Therefore, the cohesive forces and repulsive forces reach equilibrium at smaller diameter of agglomerate [7].

Fractal theory has been used for description of agglomerates [23]. Fractal is an object that exhibits self-similar pattern. The structure of an agglomerate is characterized by its fractal dimension.

The fractal dimension is one of the most important properties of an agglomerate which illustrates compactness of the agglomerate. The maximum theoretical amount of fractal dimension is 3 [24].

Nanoparticles agglomerates have porous and fragile structure and they can have significant effect on fluidization behavior. Therefore, most studies have focused on estimating agglomerate size, which is difficult to measure. The proposed models are classified into three types, including force balance [3], energy balance [5] and the use of R-Z equation and modified R-Z equation [13]. Chaouki et al. [3] and Morooka et al. [5] were one of the pioneers in this field that presented a model based on force balance and energy balance to predict the agglomerate size. Afterwards, Zhu et al. [6] and Yao et al. [10] suggested a model using the R-Z equation and the terminal velocity of agglomerate in the Stokes regime to calculate the agglomerate size. A model was developed by Nam et al. [13] to estimate both the fractal dimension and the size of agglomerate by measuring the bed height ratio and pressure drop in fluidized bed.

The main purpose of this study was to develop an improved model for predicting the fractal dimension and the size of agglomerate varying different operating parameters such as vibration frequency and vibration amplitude and the properties of gas and solids in the vibro-fluidized bed. Changes in fractal dimension, the number of primary particles in an agglomerate and minimum fluidization velocity due to vibration were also investigated.

2. THEORY

Richardson-Zaki equation can be used to describe the relationship between the settling velocity (v_s) and the solids volume fraction (φ_s) in a homogeneous fluidization of a non-cohesive spherical powder:

$$\frac{v_s}{v_{p_0}} = \left(1-\phi_s\right)^n \tag{1}$$

where v_{p_0} is terminal velocity of a single particle and n is the R-Z exponent. The Stokes terminal velocity of a single particle in the creeping flow regime (Re < 1) is as follows:

$$v_{p_0} = \frac{2\left(\rho_p-\rho_g\right)R_p^2 g_{eff}}{9\mu_g} \tag{2}$$

where ρ_p, R_p, ρ_g, μ_g and g_{eff} are primary particle density, primary particle radius, gas density, gas viscosity and effective gravity acceleration, respectively. The R-Z equation can be used for estimating the bed expansion of nanoparticles fluidization [4, 6, 10, 12–13]. To achieve this aim, it was suggested to use the terminal velocity of particles (v_{p_0}) and the primary particles volume fraction (φ_s) by the settling velocity of the agglomerate (v_a) and the agglomerates volume fraction (φ_a), respectively [4, 6, 10, 12–13]. The error caused by using the superficial gas velocity instead of the terminal velocity is negligible and the superficial

gas velocity is approximately equal to the initial terminal velocity in the APF regime [25]. Therefore, eqn. (2) can be written as:

$$\frac{v_g}{v_a} = \left(1-\phi_a\right)^n \tag{3}$$

in which the velocity of a single agglomerate in the creeping flow can be defined by:

$$v_a = \frac{2\left(\rho_a - \rho_g\right)R_a^2 g_{eff}}{9\mu_g} \tag{4}$$

Here, ρ_a and R_a are the agglomerate density and agglomerate radius, respectively. Agglomerate radius is the radius of the sphere circumscribed on the agglomerate.

The effective acceleration in vibro-fluidized bed can be obtained from [26]:

$$g_{eff} = g\left(1+\frac{A\left(2\pi f\right)^2}{g}\right) \tag{5}$$

where f is vibration frequency, A is vibration amplitude and g is gravity acceleration. The second term in the parentheses is defined as the vibration intensity, Γ.

Fractal agglomerates can be evaluated by the following equation [27–30]:

$$N = k_g\left(\frac{R_g}{R_p}\right)^{D_f} \tag{6}$$

where k_g is the fractal pre-factor, D_f is the fractal dimension, R_g is the gyration radius, R_p is the primary particle radius and N is the number of primary particles in an agglomerate. In order to determine the structure of agglomerates, both the fractal dimension and the pre-factor must be previous studies have reported that both the fractal dimension and the pre-factor must be properly specified [30–32].

Chaouki et al. [3] and Yao et al. [10] supposed that:

$$\varepsilon = 1-\frac{H_0}{H} \tag{7}$$

where ε is bed voidage and H and H_0 are the bed height, initial bed height, respectively. This hypothesis is incorrect because the voidage becomes zero at $H = H_0$ which is impossible. Nam et al. [13] utilized agglomerate density and particle density ratio to overcome this problem which need an supposition for agglomerate density such as Wang et al. [33] assumption a correlation between agglomerate density and bulk density (ρ_b) that $\rho_a = 1.15\rho_b$. Nam et al. [13] assumed that the hydrodynamic radius (R_h) is equal to gyration radius. Hydrodynamic radius of agglomerate which is the radius of an impermeable sphere of the same mass, having the same dynamic properties as the real agglomerate. They used the agglomerate radius instead of the gyration radius in eqn. (6) [13]. This assumption is correct in an ideal state that the agglomerates are compact ($D_f = 3$). Figure 1 illustrates a comparison between these radiuses. They also supposed that the pre-factor is equal to unity [13]. To solve

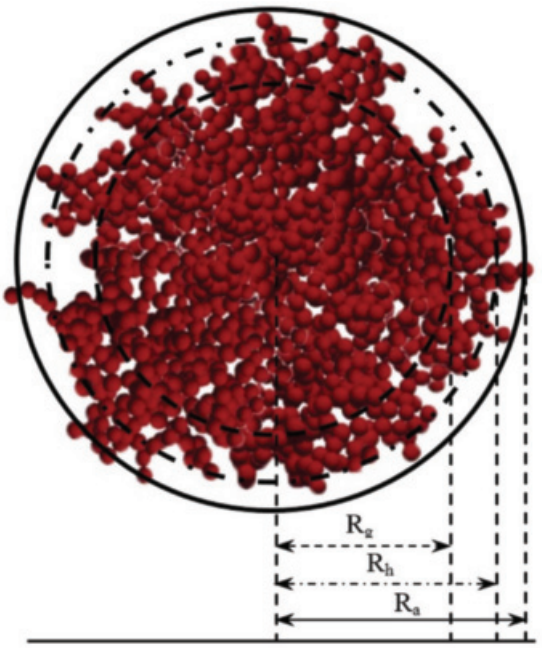

Figure 1: Comparison of agglomerate radius (R_a), hydrodynamic radius (R_h) and gyration radius (R_g)

the above problems and achieve a proper voidage, the initial bed voidage (ε_0) was utilized in this model. Moreover, some correlations between agglomerate radius and gyration radius are employed instead of Nam et al. [13] supposition.

The overall mass balance for nanoparticles can be given by:

$$\rho_a (1-\varepsilon) HS = \rho_b H_0 S = \rho_{a_0} (1-\varepsilon_0) H_0 S \tag{8}$$

where ρ_{a_0} is initial agglomerate density and S is bed cross section area. Assuming that the hydrodynamic conditions does not affect agglomerate density:

$$\rho_a \approx \rho_{a_0} \tag{9}$$

Thus, eqn. (8) can be defined as:

$$\varepsilon = 1 - \frac{H_0}{H}(1-\varepsilon_0) \tag{10}$$

It can be assumed that agglomerates are spherical, their mass can be described by:

$$m_a = \frac{4}{3}\pi R_a^3 \rho_a = N \frac{4}{3} R_p^3 \rho_p \tag{11}$$

The gyration radius can be related to the agglomerate radius by the following correlation [34–35]:

$$R_g = R_a \sqrt{\frac{D_f}{D_f + 2}} \tag{12}$$

The combination of eqns. (11) and (12) is obtained as:

$$\frac{\rho_a}{\rho_p} = N \left(\frac{D_f}{D_f + 2} \right)^{\frac{3}{2}} \left(\frac{R_p}{R_g} \right)^3 \tag{13}$$

Insertion of eqn. (6) into eqn. (13) yields:

$$\frac{\rho_a}{\rho_p} = N^{\left(1 - \frac{3}{D_f}\right)} k_g^{\frac{3}{D_f}} \left(\frac{D_f}{D_f + 2} \right)^{\frac{3}{2}} \tag{14}$$

To simplify the eqn. (4), the gas density is considered negligible compared to the particle density, eqn. (4) turns into:

$$v_a = \frac{2 g_{eff} \rho_p R_p^2}{9 \mu_g} \left(\frac{\rho_a}{\rho_p} \right) \left(\frac{R_a}{R_p} \right)^2 = v_{p_0} \left(\frac{\rho_a}{\rho_p} \right) \left(\frac{R_a}{R_p} \right)^2 \tag{15}$$

The volume fraction of agglomerates can be given by:

$$\phi_a = \phi \frac{\rho_p}{\rho_a} \tag{16}$$

where φ denote the volume fraction of primary particles in the bed. Thus, the R-Z equation can be expressed as:

$$\frac{v_g}{v_{p_0}} = N^{\left(1 - \frac{1}{D_f}\right)} \left(\frac{D_f + 2}{D_f} \right)^{\frac{-1}{2}} k_g^{\frac{1}{D_f}} \left[1 - \phi N^{\left(\frac{3}{D_f} - 1\right)} \left(\frac{D_f + 2}{D_f} \right)^{\frac{3}{2}} k_g^{\frac{-3}{D_f}} \right]^n \tag{17}$$

Eqn. (17) can be written as:

$$\left(\frac{v_g}{v_{p_0}} \right)^{\frac{1}{n}} = \left(F\left(D_f, N\right) \right)^{\frac{1}{n}} \left[1 - \phi G\left(D_f, N\right) \right] \tag{18}$$

in which:

$$F = N^{\left(1 - \frac{1}{D_f}\right)} k_g^{\frac{1}{D_f}} \left(\frac{D_f + 2}{D_f} \right)^{\frac{-1}{2}} \tag{19}$$

$$G = N^{\left(\frac{3}{D_f}-1\right)} k_g^{\frac{-3}{D_f}} \left(\frac{D_f+2}{D_f}\right)^{\frac{3}{2}} \tag{20}$$

By considering eqn. (18), a linear relationship can be observed between $(v_g/v_{p_0})^{1/n}$ and φ. For each value of the superficial gas velocity, the primary particles volume fraction (φ) can be ascertained from eqns. (10) and (16). By plotting $(v_g/v_{p_0})^{1/n}$ versus φ for a specified value of the R-Z exponent (n) and calculating intercept and slope of the line, the fractal dimension and the number of primary particles in agglomerate can be obtained. Then, the agglomerate radius can be evaluated by combining of eqns. (12) and (6):

$$R_a = R_p \cdot \sqrt{\frac{D_f+2}{D_f}} \left(\frac{N}{k_g}\right)^{\frac{1}{D_f}} \tag{21}$$

According to Gmachowski [35], the pre-factor can be represented as:

$$k_g = \left[\sqrt{1.56 - \left(1.728 - \frac{D_f}{2}\right)^2} - 0.228\right]^{D_f} \left(\frac{D_f+2}{D_f}\right)^{\frac{D_f}{2}} \tag{22}$$

3. RESULTS AND DISCUSSION

Experimental results of Yang et al. [15], were used to verify the model presented in this work. Properties of SiO_2 nanoparticles used in their experiments are listed in Table 1. Yang et al. [15] utilized the vibration frequency in the range of 0 Hz to 40 Hz and the vibration amplitude from 0.5 mm to 3 mm. Moreover, experimental data of Zhu et al. [6] were employed to investigate the accuracy of the present model [36].

3.1. FRACTAL PROPERTIES OF AGGLOMERATE

The R-Z exponent was obtained from Garside and Al-Dibouni [37] correlation.

$$\frac{5.1-n}{n-2.7} = 0.1\ Re_p^{0.9} \tag{23}$$

where Re_p denotes particle Reynolds number. The R-Z exponent reaches to 5.1 for silica nanoparticles at small values of particle Reynolds number based on eqn. (23). As regards n = 5.1 and using the experimental data of Yang et al. [15], a linear plot of $(v_g/v_{p_0})^{1/n}$ vs. φ can be drawn. Estimation of the bed volume fraction by eqn. (10) requires initial bed voidage. This can be computed by eqn. (10) and the R-Z equation:

Table 1: Properties of nanoparticles [15]

Nanoparticles	Primary particle size (nm)	Bulk density (kg/m^3)	Primary particle density (kg/m^3)
SiO_2	30	85	2560

$$v_g^{1/n} = v_a^{1/n} - \frac{H_0}{H}\left(1 - H_0\right) v_a^{1/n} \tag{24}$$

Considering $n = 5.1$, the initial bed voidage can be calculated from a curve of $v_g^{1/n}$ versus H_0/H via linear regression which is approximately 0.35 for SiO_2. Then, the plot of $(v_g/v_{p_0})^{1/n}$ against φ can be drawn according to eqn. (18) for each values of vibration frequency and vibration amplitude such as Fig. 2 for SiO_2 nanoparticles at frequency of 30 Hz and amplitude of 3 mm.

Slope and intercept of eqn. (18) are both a function of fractal dimension and number of primary particles in an agglomerate, as seen in eqns. (19) and (20). By simultaneous solution of these two equations, D_f and N can be calculated. The Results of these calculations for silica nanoparticles are illustrated in Figs. 3 and 4. As shown in these figures, the vibration intensity increase from 0 to 20 leads to a slight reduction in fractal dimension of agglomerate, while the number of primary particles in the agglomerate reduces dramatically. The decrease in fractal dimension and the number of primary particles in an agglomerate can be attributed to additional kinetic energy to the bed (in form of pressure fluctuations) by means of vibration. This leads to decreasing the impact of the relative adhesive force and improves the fluidization quality by segmenting the agglomerate. Thus, higher vibration intensity leads to formation of a more delicate structure (less fractal dimension) with fewer particles.

As can be seen in Fig. 4, the number of primary particles in an agglomerate reduces with increasing the vibration intensity and varies insignificantly at high vibration intensities. Indeed, the effect of vibration intensities on the number of primary particles in agglomerate at high vibration intensities is less strong than that at low vibration intensities. This can be attributed to the direct relationship between the repulsive force and the vibration intensity.

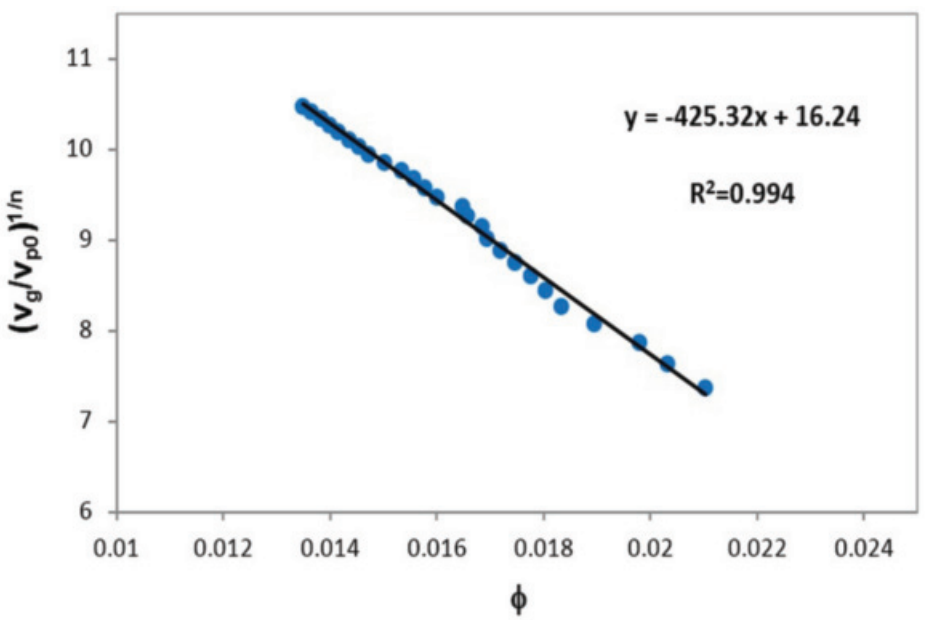

Figure 2: $(v_g/v_{p_0})^{1/n}$ vs. φ for SiO_2 at vibration frequency of 30 Hz and vibration amplitude of 3 mm

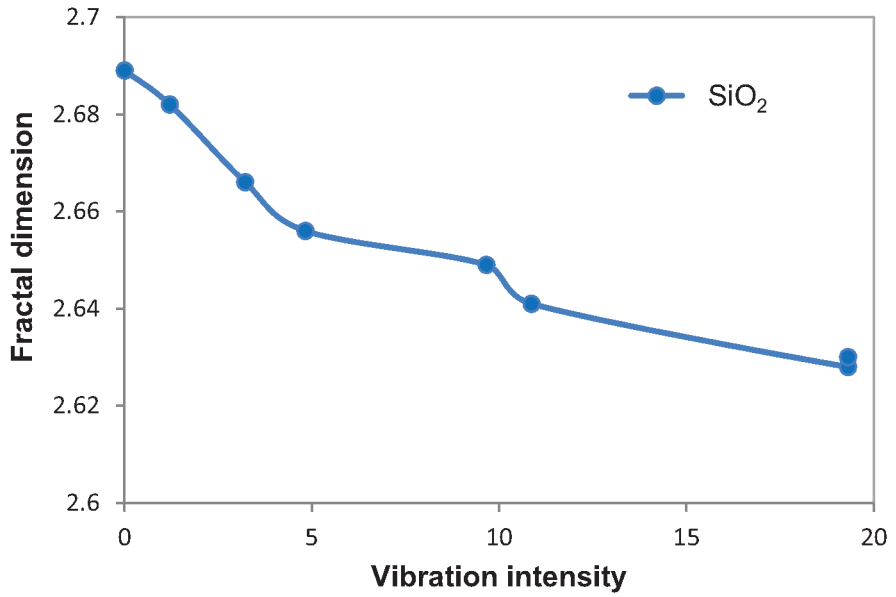

Figure 3: Fractal dimension at various vibration intensities

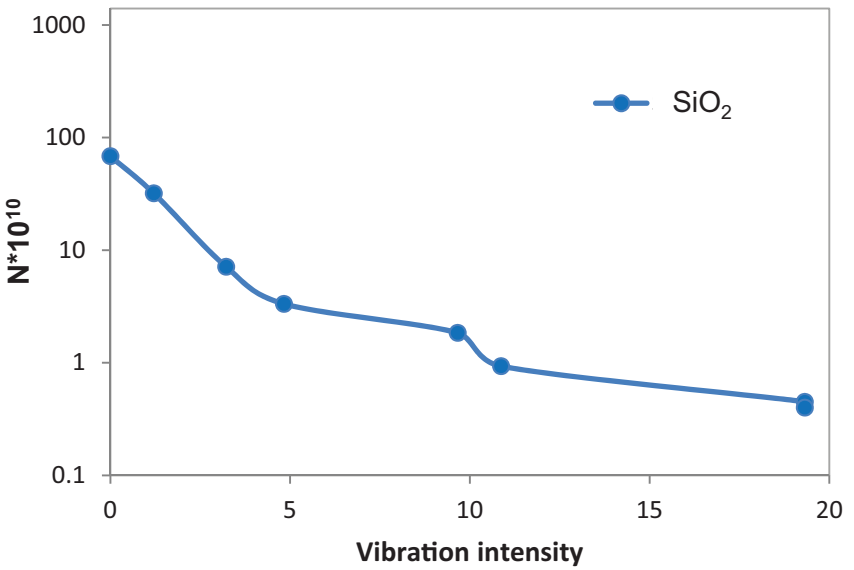

Figure 4: Number of primary particles in agglomerate at various vibration intensities

As pointed above, agglomeration is a dynamic phenomenon. By increasing the vibration intensity and repulsive forces, the primary particles are separated from the agglomerate. Particles that separate from the agglomerate are more than the particles that join to it. This process continues until the cohesive forces become such that number of particles adding the agglomerate becomes equal to the number of segregated ones.

3.2. AGGLOMERATE SIZE

After determining the fractal dimension and the number of primary particles in an agglomerate, the agglomerate size can be calculated from eqn. (21). Variations of the estimated size of silica agglomerate at various vibration frequencies at 3 mm vibration amplitude are shown in Fig. 5. In addition, Fig. 6 illustrates calculated mean agglomerate size against vibration amplitude at constant frequency of 40 Hz. These figures indicate that the size of silica agglomerates reduces with increasing either frequency or amplitude of vibration. As mentioned above, large agglomerates are broken into smaller ones through the mechanical vibration [2]. External forces (such as mechanical vibration) can appear in the role of repulsive forces [4, 36] which act in opposite side of adhesive forces. Repulsive forces augment dramatically with increasing vibration frequency and amplitude whereas cohesive forces do not vary. In order to retain a balance between repulsive and cohesive forces, size of agglomerate is decreased to omit the influence of vibration frequency and amplitude increase.

By comparing Fig. 5 and 6, it can be found out that the vibration frequency is more impressive than its amplitude on agglomerate size reduction. This can be explained by the fact that the relation between vibration intensity and vibration amplitude is weaker than vibration intensity and vibration frequency.

3.3. MINIMUM FLUIDIZATION VELOCITY

The minimum fluidization velocity in APF fluidization (u_{mf}) is given by [38]:

$$u_{mf} = \frac{\left(\psi_s d_a\right)^2}{150\mu_g}\left(\rho_a - \rho_g\right)\left(\frac{\varepsilon_{mf}^3}{1-\varepsilon_{mf}}\right)g \tag{25}$$

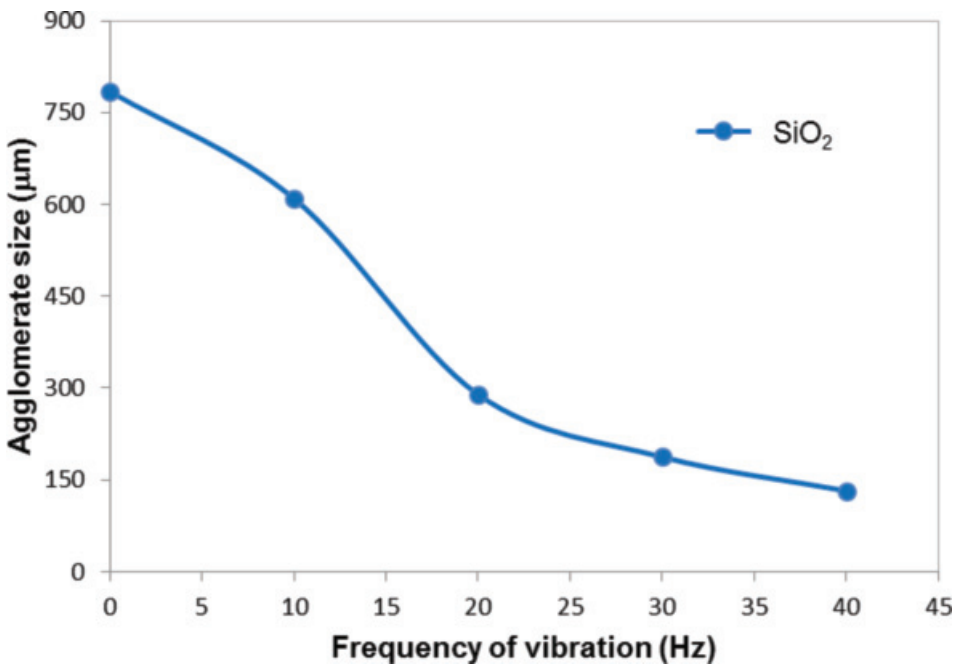

Figure 5: Mean agglomerate sizes with different vibration frequency at constant amplitude of 3 mm

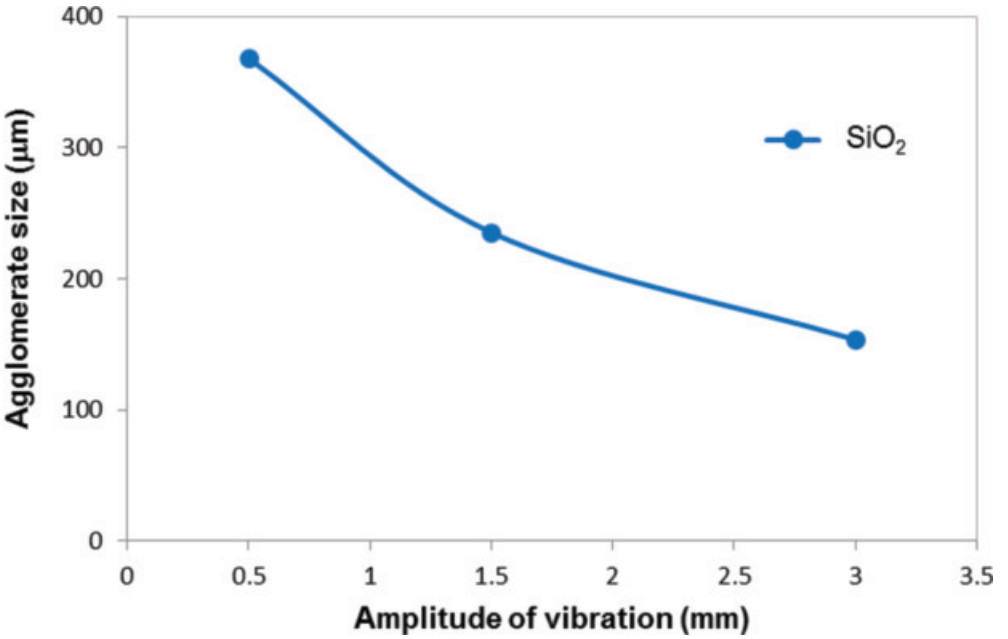

Figure 6: Mean agglomerate sizes with different vibration amplitude at constant frequency of 40 Hz

where ψ_s is the particle sphericity and ε_{mf} is the bed voidage at minimum fluidization velocity. Value of ε_{mf} can be estimated from plot of the bed height ratio against superficial gas velocity at minimum fluidization. It was assumed that the sphericity of agglomerate is equal to unity. Using the agglomerate diameter calculated from eqn. (21), the minimum fluidization velocity was obtained from eqn. (25).

Experimental results of Yang et al. [15] and estimated minimum fluidization velocity of silica agglomerate by the present model in the frequency range of 0 Hz to 40 Hz at a constant amplitude of 3 mm are compared in Fig. 7. As shown in this figure, estimated minimum fluidization velocities by applying predicted agglomerate sizes and experimental data are acceptable fitted, with 0.995 correlation coefficients. Figure 8 also shows that a good agreement exists between experimental and calculated values of minimum fluidization velocities in the range of vibration amplitude from 0.5 mm to 3 mm at a constant vibration frequency of 40 Hz. Average relative errors is 4.6%.

To investigate the precision of the model presented in this work, results of this model, in terms of agglomerate size and minimum fluidization velocity, were also compared to the results of models suggested by Nam et al. [13], Zhu et al. [6] and Wang et al. [33] based on the experimental values reported by Zhu et al. [6] (see Table 2). This comparison shows that the present model provides closest amounts to the experimental ones.

3.4. EFFECT OF THE R-Z EXPONENT

In order to examine the sensitivity of various agglomerate characteristics to the R-Z exponent, calculations in the previous sections were repeated for six different values of n. The calculation results for SiO_2 nanoparticles at vibration frequency of 40 Hz and vibration amplitude of 0.5 mm are represented in Table 3. The experimental minimum fluidization velocity was 0.04 m/s at this condition based on Yang et al. [15] experimental work.

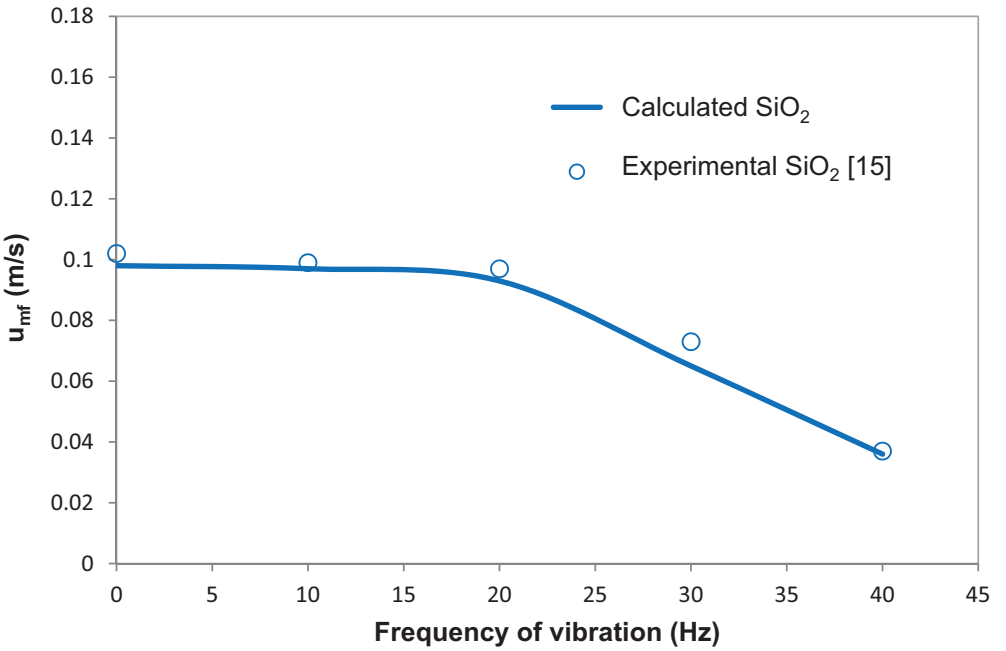

Figure 7: Comparison of estimated and experimental minimum fluidization velocity as a function of vibration frequency at a constant amplitude of 3 mm

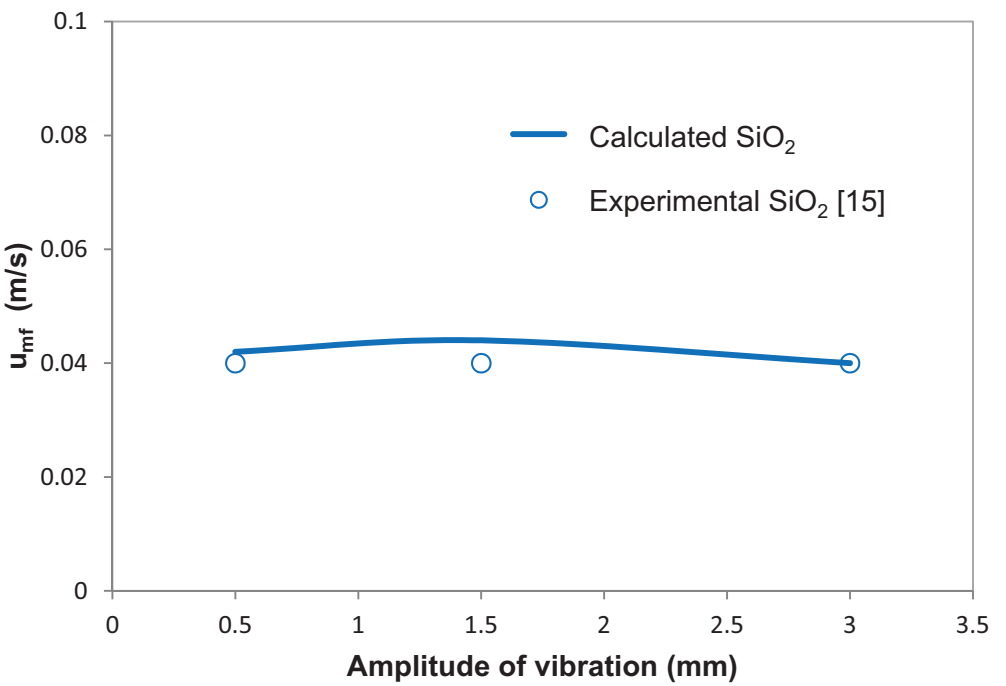

Figure 8: Comparison of estimated and experimental minimum fluidization velocity as a function of vibration amplitude at a constant frequency of 40 Hz

Table 2: Comparison of models used to estimate the mean agglomerate size of R974 and R711 nanoparticles

Nanoparticles	Parameter	Experimental value [6]	Present work	Nam et al. [13]	Zhu et al. [6]	Wang et al. [33]
R974	d_a (μm)	315	285	262	211	214
	u_{mf} (m/s)	0.23	0.21	0.19	0.17	0.18
R711	d_a (μm)	274	256	230	207	227
	u_{mf} (m/s)	0.38	0.39	0.33	0.31	0.32

Table 3: Agglomerate properties of SiO_2 for different Richardson-Zaki exponents at vibration frequency of 40 Hz and vibration amplitude of 0.5 mm based on Yang et al. [15] experimental data

N	D_f	N	k_g	d_a (μm)	u_{mf} (m/s)
5	2.663	7.02×10^{10}	1.874	361	0.037
5.1	2.665	7.122×10^{10}	1.875	368	0.042
5.3	2.666	7.41×10^{10}	1.875	373	0.045
5.5	2.667	7.81×10^{10}	1.876	380	0.051
5.7	2.668	8.14×10^{10}	1.877	386	0.057
6	2.669	8.68×10^{10}	1.878	390	0.067

This table indicates that properties of the agglomerate structure are insensitive to the value of the R-Z exponent. Whereas, variations in the minimum fluidization velocity with respect to n is substantial with a difference of 2.5% to 70%. The best results for the minimum fluidization velocity were achieved at n = 5.1. Therefore, Garside and Al-Dibouni [37] equation was selected in calculations of the previous sections.

4. CONCLUSION

An improved model is proposed to determine size and fractal characteristics of nanoparticle agglomerates in vibro-fluidized beds. This model is based on the fractal analysis, R-Z equation and mass balance. The findings indicate that the fractal dimension of silica agglomerates is in the range of 2.61 to 2.69, which is lower than the theoretical maximum value of 3. The number of primary particles in the agglomerate was in the order of 10^{10}. It was found that both the fractal dimension and the number of primary particles in an agglomerate reduces with increasing the vibration intensity. At high vibration intensities, the effect of vibration intensity on the number of primary particles reduces significantly and it changes little. The results has clearly shown that the vibration frequency has a greater impact on decreasing the agglomerate size than the vibration amplitude. Compared to the existing models, the model developed in this work estimates the minimum fluidization velocity of silica agglomerates closer to the experimental value.

The calculated minimum fluidization velocity are broadly consistent with experimental values which have shown that the predicted agglomerate size is close to the real one. The results show that characteristics of the agglomerate structure are insensitive to the R-Z exponent, whereas the minimum fluidization velocity is sensitive to this exponent.

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