Hydrodynamic Correlations with Experimental Results from Cold Mockup Spouted Beds for Nuclear Fuel Particle Coating

***Jiandong Zhou*[1], *Duane D. Bruns*[1], *Charles E. A. Finney*[2], *C. Stuart Daw*[2] and *Sreekanth Pannala*[3], *David L. McCollum*[4]**

(1) Chemical Engineering Department, University of Tennessee, Knoxville, 419 Dougherty Hall, Knoxville, TN 37996.
(2) Engineering Science and Technology Division, Oak Ridge National Laboratory, Oak Ridge, TN 37831,
(3) Computational Science and Mathematics Division, Oak Ridge National Laboratory, Oak Ridge, TN 37831,
(4) Institute of Transportation Studies, University of California, Davis, CA 95616.

Abstract

A cold mockup of a nuclear particle spouted bed coater is systematically studied over an experimental matrix of four particle radii, three cone angles, several gas (humidified air) flow rates, and four particle inventories. Spherical ZrO_2 particles of 300, 400, 500 and 650μm with a tight distributed size are selected as surrogates for nuclear fuel particles due to their similar physical properties. Three spouted beds with 45, 60, and 75-degree cone angles are used. The gas flow rate spans the fully spouted condition to minimum spouted condition. The bed inventories are H/D_c of 0.50, 0.55, 0.60 and 0.65. The column diameter is 5 cm with an inlet throat diameter of 0.04 cm. The U_{mf} measurements closely follow the Wen and Yue correlation. Due to the high density of our particles a new U_{ms} correlation is needed. Our dimensionless U_{ms} correlation for ZrO2 particles is presented as a function of Re number at U_{ms}, particle size (D_p), cone angle of the spouted bed (γ) and static particle height (H_0/D_c). A new quantitative method for U_{ms} evaluation is presented. Another dimensionless correlation for average pressure drop across the spouted bed covering the experimental matrix is given in terms of U/U_{ms}, γ, H_0/D_c, and D_p/D_c. The first four moments and the power spectral density of the pressure time series are calculated with trends described. The main frequency peak of the pressure drop signal shifts from a high to a low value as the air velocity drops from fully spouted velocity to near minimum spouting velocity. As the air flow rate is lowered to 1.1~1.2 U_{ms}, the main frequency peak increases sharply. Main frequency peak of the pressure signal increases as particle size, static particle height or cone angle increase. The 0.5mm particles have the sharpest main frequency peak plot while the other plots become wider as D_p increases or decreases. The standard deviation of the pressure signal for the 0.3mm, 0.4mm and 0.65mm particles tend to increase as gas flow rate is lowered to U_{ms} while that of the 0.5mm particles tends to decreases. The skewness also increases as gas flow rate is lowered to U_{ms} except for 0.65mm particles. In general, the kurtosis also increases as gas flow rate approaches U_{ms}.

1. Introduction

Coating nuclear fuel particles can improve safety and flexibility during their storage, movement, and usage [1]. The spouted bed is a good candidate for coating nuclear fuel particles due to good mixing affects and uniform exposure time of the particles to the CVD (Chemical Vapor Deposition) gases so that the different layers coat the fuel particles uniformly.

The TRISCO coated uranium fuel particle has been developed. It is coated by 4 layers: an amorphous (soft) carbon layer, then a pyrolytic (hard) carbon layer, then Silicon Carbide (SiC), and finally another pyrolytic carbon layer. This coating process uses a CVD spouted bed running at temperatures typically from 1200 to 1800 degree C. In order to produce large quantities of fuel to support the option to use AGRs, the spouted bed coater process must be scaled-up from the present experimental 5-cm diameter coaters. There is no universal way to scale-up fluidized beds even for hydrodynamics [2]. Spouted fluidized beds are more considered more complex than fluidized beds. In addition, in the high temperature coaters, it is nearly impossible to optically observe and monitor the particles due to the opaque carbon wall of the bed and dense carbon soot inside the bed. In general, it is also very expensive to set up and run a high-temperature surrogate coater. Given the above constraints and in consideration of the fact that the state-of-the-art in fluidized bed design and scale-up is to establish the hydrodynamics from cold bed studies without reaction, the ambient room-temperature spouted bed coater will be studied first utilizing both experiment and simulation. Only experimental results are reported in this paper. Other members of the research team are using the experimental results to calibrate and evaluate simulation code. Initial interaction between experimental and simulation campaigns is published elsewhere [16].

The experiments that are necessary for validating the simulations [3] are focused on exploring the hydrodynamics of surrogate particles (ZrO_2) in cold mockup spouted beds. For this purpose the bed pressure drop signal is studied first including its variation characteristics. Beyond the average pressure, the dynamic pressure characteristics dictate the behavior of fluidized and spouted fluidized beds. Thus, it is critical that simulations capture this dynamic behavior. It is also one of the most easily obtained process signals from mockup coaters and hot coaters. By comparing the hot pressure signal with preliminary experimental results from the cold mockup we are getting insight into the running status of the hot spouted bed coaters. Although there are many papers analyzing the pressure signals in fluidized beds [4-6], most of them deal with the particles with densities lower than or around 3 grams/cc. These included glass, polymer materials, wheat, beans etc. The density of the surrogate nuclear fuel particle, ZrO_2, is 6 grams/cc, which is much higher than the normally studied and reported materials. We need to investigate the behavior of higher density particles in order to provide solid data for validating the computer models being developed for uranium particle coaters.

In this paper the gas pressure drop across the spouted bed at stable spouting conditions ($1 \leq U/U_{ms} \leq 1.9$) is studied extensively. The Ums and average pressure drop prediction equations are developed, which is a function of particle size, cone angle, gas flow rate, and static solid particle height inside the spouted bed. The high frequency variations in the pressure signals are analyzed with statistical methods and in the frequency domain.

2. Experiments

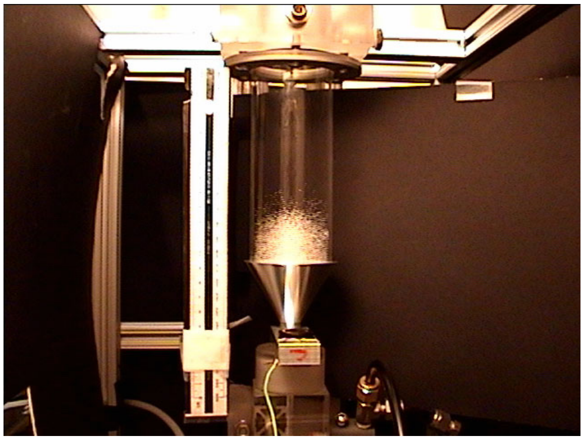

Figure 1. Photo of experimental spouted bed, γ=60 degree.

Three spouted beds with 45, 60, and 75-degree cone angles are used in this research. Figure 1 shows the 60 degree spouted bed. The diameter of the columns are all equal to 0.05m. The inlet throat diameter is 0.004m. The conical base is made of Aluminum for regularity while the cylindrical column is made out of quartz to provide optical access. The pressure drop across the spouted bed is measured with MKS pressure sensors whose ranges are 0~ 1,333Pa and 0-13,332Pa.

The data is collected by means of a National Instrument data acquisition system. Typically the pressure time series is collected at 1,000Hz for one minute. Then the mean pressure drop is calculated. The gas flow rate is controlled with a MKS mass flow controller (MFC) with a range of 0~0.2m^3/min. The MFC is periodically calibrated to guarantee its reliability and accuracy. As a result, the gas flow rate is controlled very accurately and precisely.

Spherical ZrO_2 particles of 300, 400, 500, and 650μm with tight distributed size are selected as surrogates for nuclear fuel particles due to their similar physical properties to uranium particles. The ZrO_2 particles have a density of 6,050 kg/m^3. Compressed air is humidified and used as the fluidizing gas. The air temperature is between 70-79 ˚F and ambient pressure is around 740mmHg.

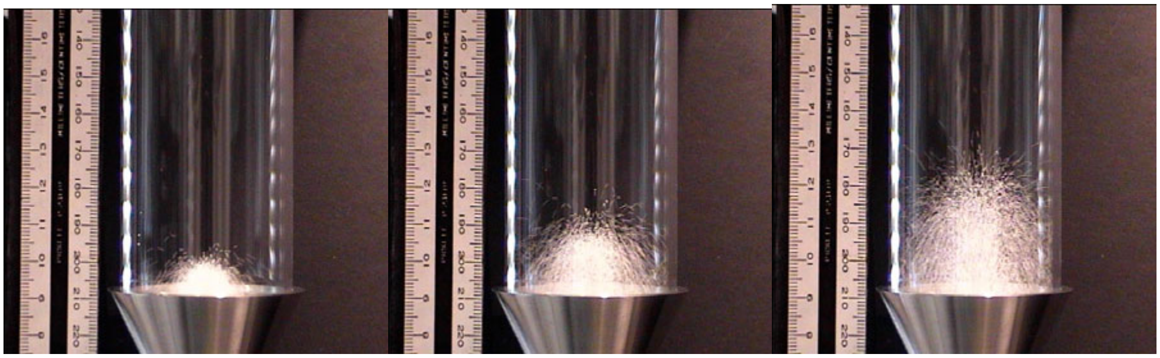

Figure 2. Spouting bed with 54.5 grams of 0.5mm ZrO_2 particles in 60 degree spouted bed with gas flow rates U/Ums being 1.8, 2.1, 2.4 from left to right.

The experimental matrix includes particle diameter, gas flow rate, cone angle, and particle inventories (H/Dc). Gas flow rate varies from 1.0 U/U_{ms} to 1.9 U/U_{ms} where the spouted bed is running at stable spouting conditions. Below 1 U/U_{ms}, the particles inside will stop spouting. Above 2 U/U_{ms}, the particles are spouted unstably and tend to be carried out of the spouted bed. The particles are classified as group B by Geldart classification [7, 8].

3. Results and Discussions

3.1 Measurement of U_{mf}.

Umf is one key parameter for computer simulation and fluidized bed design. The classical method [9] is used to measure the U_{mf}, minimum fluidizing velocity.

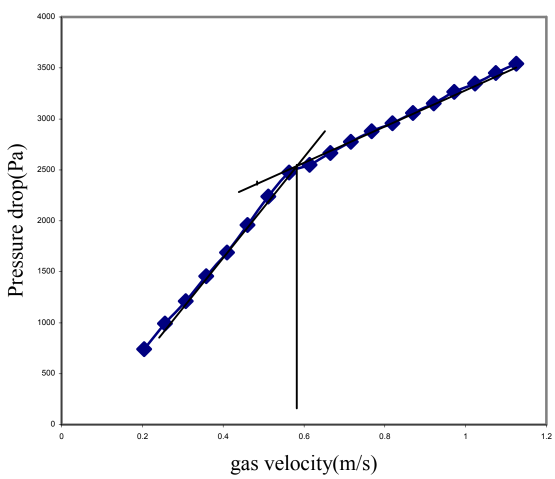

Figure 3. Measurement of U_{mf} of 0.65mm ZrO_2.

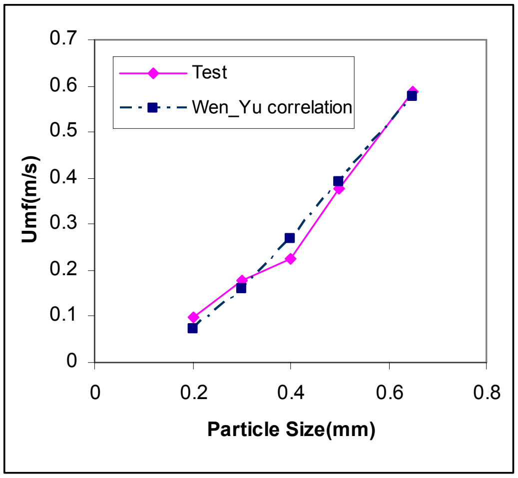

Figure 4. Comparison of measured U_{mf} with Wen-Yu [8] correlation predicted U_{mf} for different D_p.

Figure 3 shows that Umf is measured with little scatter in the data. Figure 4 indicates that the U_{mf} of ZrO_2 particles used in this research closely follows the Wen and Yue correlation.

3.2 Measurement of U_{ms}

The minimum spouting velocity, U_{ms}, which corresponds to the minimum gas velocity maintaining the spouting condition of the spouted bed, plays an important role in dimensionless analysis of the hydrodynamic of the spouted bed and design and operation of gas-solid spouted beds [10]. Although there are quite a few correlations predicting U_{ms} [11, 12], most of them are based on Ums of particles with density lower than or around 3,000 kg/m^3. It is necessary to measure Ums for the experimental matrix used to investigate gas pressure drop (ΔP) and study the hydrodynamics further as well as design and sale-up spouted beds in future. Ums is a function of particle size (D_p), cone angle (γ), and static particle height (H). It is measured in a way similar to measure the minimum fluidized velocity, Umf. The gas velocity is raised up first to get full spouting condition. Then lower the gas velocity step by step and the corresponding ΔP keeps decreasing at first. At $U=U_{ms}$, it reaches the minimum value and increases sharply as gas velocity is just lower than Ums, as shown in Figure 5.

Quantitative evaluation of Ums using the ΔP *vs* gas flow rate plot is new. The HMI (Human-Machine-Interface) allows the gas flow rate to be decreased in small increments accurately without any overshoot in the flow rate. This may be one aspect of identifying the new technique. Previously minimum spouting velocity was determined by carefully watching for particle movement on the bed surface visually. Using the minimum in the plots as, Figure 5, corresponds closely to our visual observations, generally, the particles appear to have stopped moving at a slightly higher gas velocity than the minimum in the figure. This method is more quantitative and reproducible than visual observations.

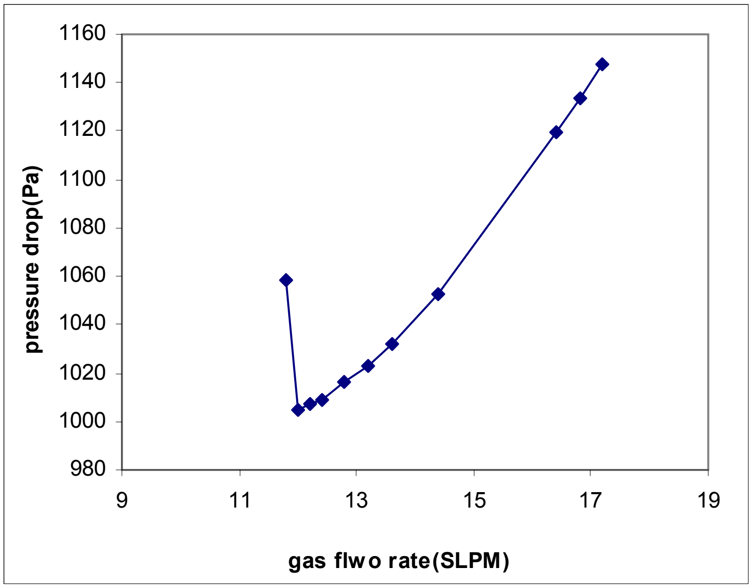

Figure 5. Measurement of U_{ms} for 53.9 grams 0.5mm ZrO_2 in 60 degree spouted bed. U_{ms} = 12 liter/min.

The Ums correlation is developed as below with the regression coefficient being 0.98.

$$\mathrm{Re}_{ms} = 0.0015 Ar^{0.86} \left(\frac{H_0}{D_c}\right)^{1.59} \tan\left(\frac{\gamma}{2}\right)^{0.87} \quad (1)$$

Table 1 Comparison of measured U_{ms} with predicted U_{ms} values. Dp=0.5mm, γ=45 degree.

H_0/Dc	Measured Value(m/s)	Equation (1)(m/s)	Method 1(m/s)	Method 2(m/s)	Method 3(m/s)	Method 4(m/s)	Method 5(m/s)
0.5	**0.0450**	0.0393	7.43	14.99	3.78	10.61	0.072
0.6	**0.0586**	0.0525	9.6	18.8	3.9	12.7	0.0869
0.8	**0.0874**	0.0829	14.7	26.8	4.2	17	0.1159
1	**0.1290**	0.1182	20.7	35.4	4.4	21.2	0.1448

Among methods 1 to 5, method 5 is the best and its predicted error is between 12%~60%. The prediction error of correlation (1) is 8~13%. Method 1, 2, 3, 4, and 5 are from [12], [13], [14], [15] and [10] respectively. Other correlations have higher errors and are not listed here. Some correlations do not take into account the cone angle and are not compared here either. Most of the correlations are based on particles with a density lower than or around 3,000 kg/m^3 while the density of ZrO_2 is 6,050kg/m^3. In addition, some of them are more suitable for predicting Ums at H_0/D_c greater than 1 while equation (1) works at H_0/D_c equal to or less than 1.

The obtained U_{ms} values are used to specify the gas flow rate for measuring the gas pressure drop at stable spouting conditions at which U/U_{ms} is between 1 and 1.9 for this case.

3.3 Average gas pressure drop correlation

At stable spouting conditions, the total gas pressure drop across the spouted bed is fluctuated around its mean value, as shown in Figure 6. The average ΔP for different combinations of U/U_{ms}, D_p/D_c, γ, and H_o/D_c is based on the data collected at 1,000Hz for one minute and then averaged.

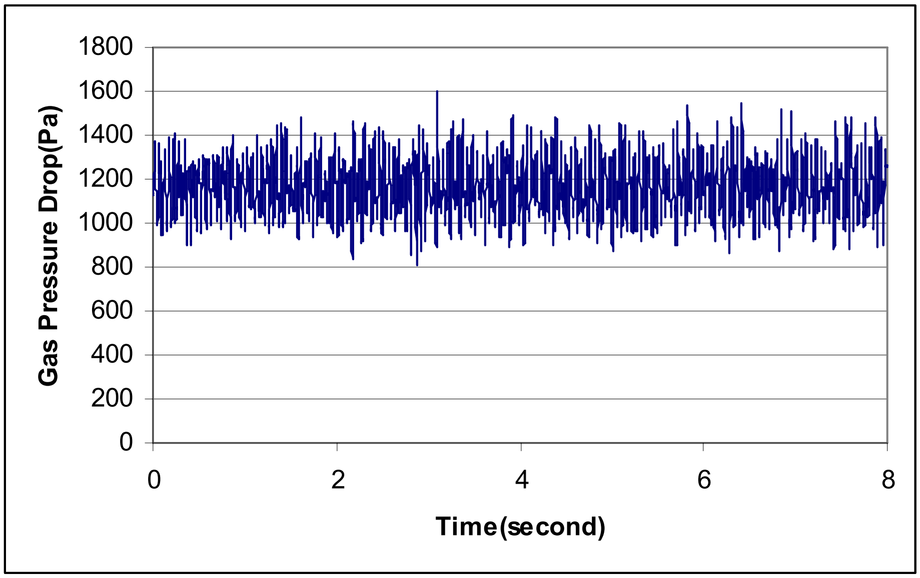

Figure 6. Total gas pressure drops in time domain for 53.9 grams 0.5mm ZrO_2 in 60 degree spouted bed.

3.3.1 ΔP versus U/U_{ms}

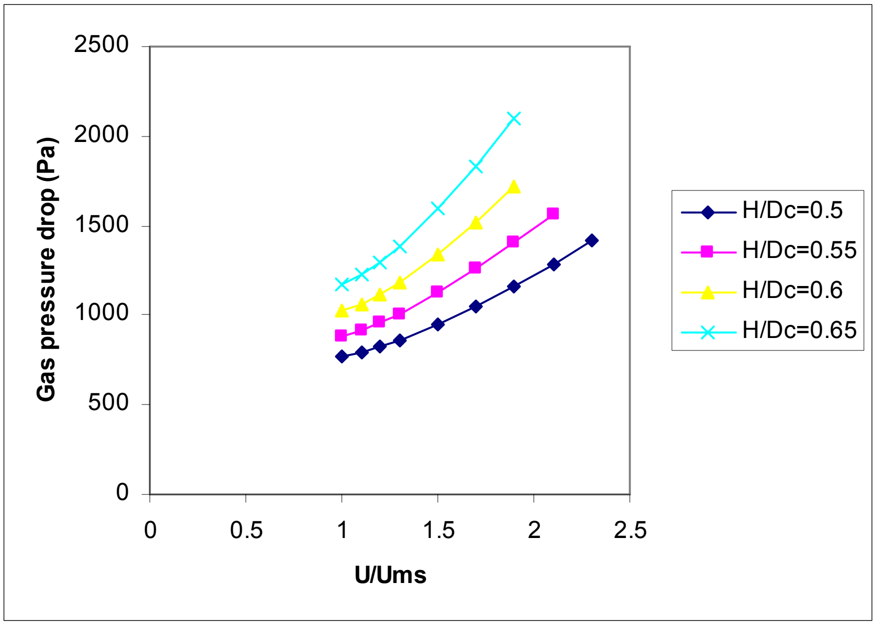

Figure 7. **ΔP versus U/Ums for 53.9 grams 0.5mm ZrO_2 in 60 degree spouted bed.**

At constant Dp, H/D_c, and γ, ΔP is 2nd or 3rd order of polynomial function of U/Ums as shown in Figure 7. (1≤U/Ums≤1.9).

3.3.2 ΔP versus tan(γ/2)

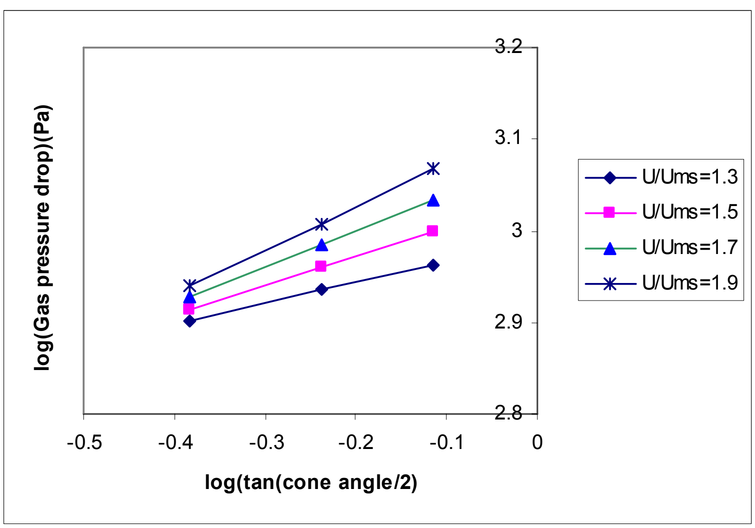

Figure 8. Log(ΔP) *vs.* log(tan(γ/2)) at Dp=0.5mm, H/Dc=0.6, and different U/U_{ms}.

In order to make angle dimensionless, a tangent transformation of angle is used. Log(ΔP) linearly increases as log(tan(γ/2)) increases at constant Dp, H/D_c and U/U_{ms}. Ums increase as cone angle increases. As a result, the absolute gas flow rate increases. So pressure drop increases.

3.3.3 ΔP versus H/Dc

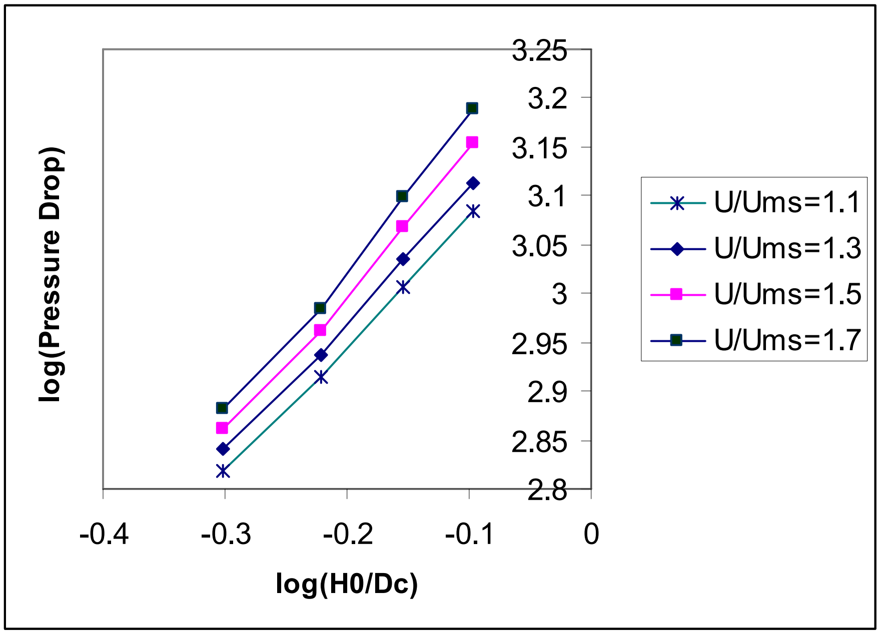

Figure 9. Log(ΔP) *vs.* log(H_0/D_c) at Dp=0.5mm, γ=60 degree and different U/Ums.

As static particle height increases, the pressure drop increases exponentially.

3.3.4 ΔP versus D_p

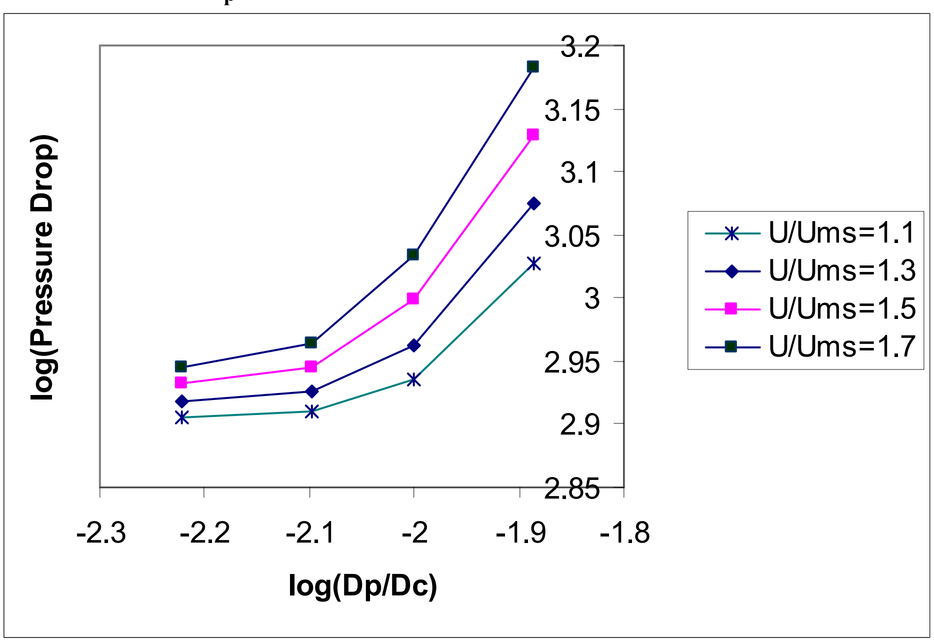

Figure 10. Log(ΔP) *vs.* log(Dp/Dc) at H_0/Dc=0.7, γ=75 degree and different U/U_{ms}.

Pressure drop increases as Dp increases while other factors are kept constant. Linear relationship approximation between log (ΔP) and log (Dp/Dc) is taken to fit the data. The effect of Dp/Dc on ΔP is much smaller than the effect of H_0/D_c, as shown in figure 5 and figure 6. It is safe to do this approximation.

3.3.5 Average ΔP prediction equation

$$\frac{\Delta P}{\rho_b H_0 g} = \left[56.38(\frac{U}{U_{ms}})^3 - 179.6(\frac{U}{U_{ms}})^2 + 110.01\frac{U}{U_{ms}} - 74.35\right] \times [(\tan\frac{\gamma}{2})^{-0.235+0.454\frac{U}{U_{ms}}}(\frac{H_0}{D_c})^{-0.081+0.378\frac{U}{U_{ms}}}(\frac{D_p}{D_c})^{-0.304+0.516\frac{U}{U_{ms}}}] \quad (2)$$

Equation (2) is based on 273 data points and r^2 is 0.93. The prediction error is less than 15%. For lower value of U/U_{ms}, the error is even less than 5%.

No correlation equation has been found in literature, which use U/U_{ms}, γ, H_0/D_c, and D_p/D_c as predictors to estimate the stable gas pressure drop across the spouted bed.

3.4 Analyze pressure drop in time and frequency domain

The gas pressure drop fluctuations are analyzed in the time domain by calculating the first four moments of the time series: average or mean, standard deviation, skewness and Kurtosis. Fourier transformation is used to calculate the PSD (power spectral density) for the time series. The characterization of experimental results in these two domains has been effective in validating the computer model [16].

3.4.1 Main frequency peak *vs.* gas flow rate

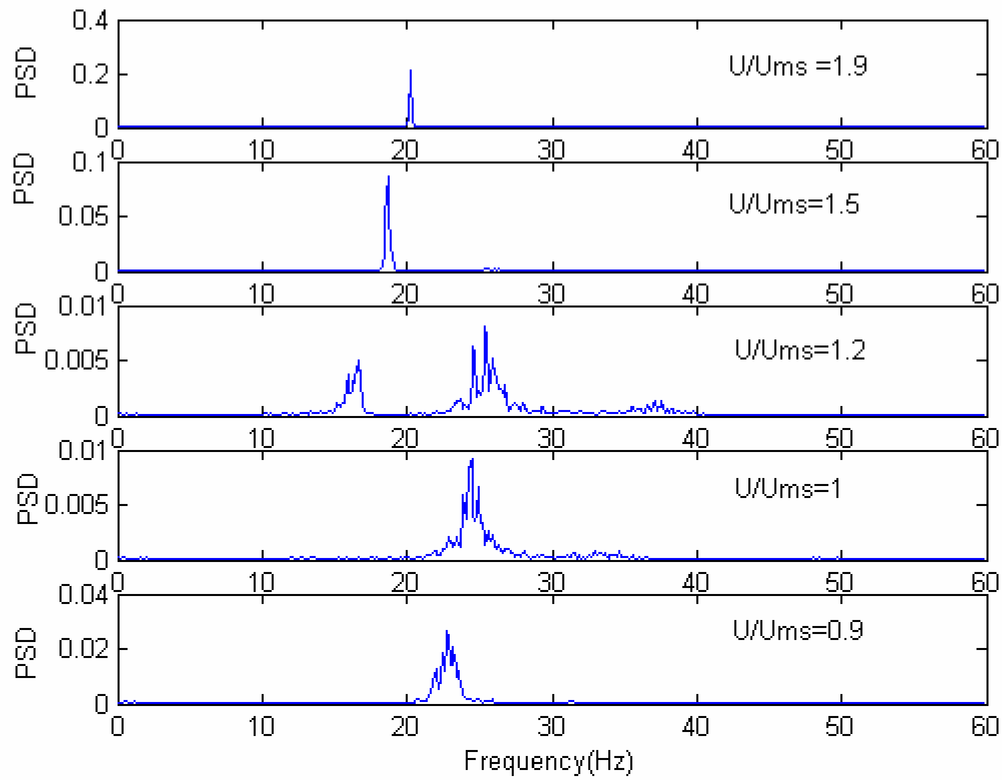

Figure 11. Main frequency peak at different gas flow rate U/U_{ms} for 53.9 grams 0.5mm ZrO_2 in 60 degree spouted bed with Dc 0.05m and Di 0.004m.

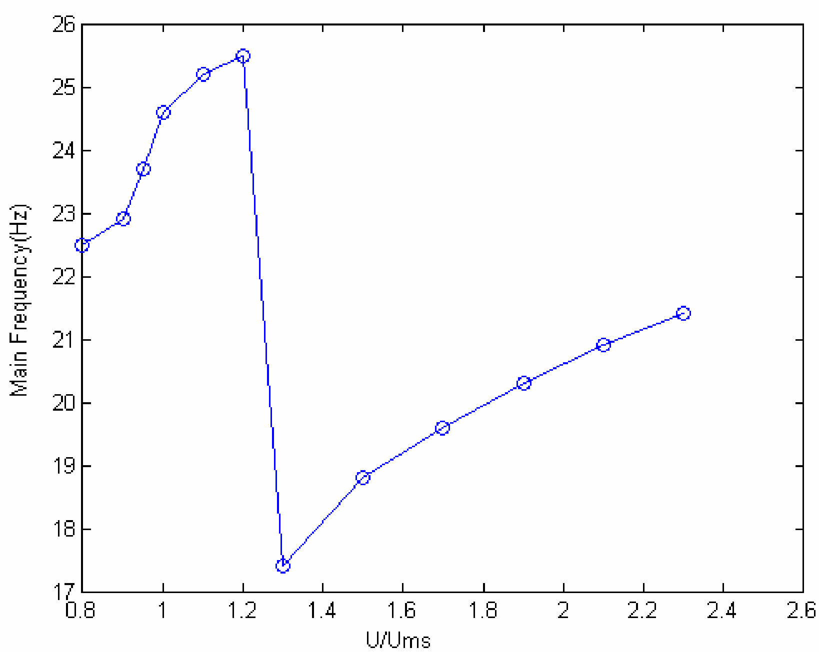

Figure 12. Main frequency peak *vs.* gas flow rate for 53.9 grams 0.5mm ZrO_2 in 60 degree spouted bed.

The main frequency peak of the pressure drop represents for the formation rate of the bubble inside the spouted bed. It shifts from 21.4Hz to 17.4Hz as air velocity drops slowly from fully spouted velocity 2.3Ums to low 1.3Ums, which means the bubble formation rate is lowering as gas flow rate decreases. The gas passing through the spout region dominates the particle behavior. At U/Ums = 1.2, the main frequency peak splits into two peaks. The smaller frequency peak corresponds to the bubble formation frequency. The higher frequency peak reflects the vibration of the annulus. The void fraction of the solid particles is lowered so much that the vibration of the annulus tends to dominate. The variation frequency peak is as high as 25.3 Hz and then shifts down slowly as air velocity keep lowering until the particles fall out of the spouted bed. As U/Ums is equal to or less than 1, the spout is diminishing. And the whole gas passes slowly through whole particles inside the spouted bed. And the vibration of the whole particles dominates. It can be eyeballed for lower static particle height (H_0/Dc) for 0.5mm ZrO_2. It can also be observed even for high H_0/Dc for 0.3mm ZrO_2.

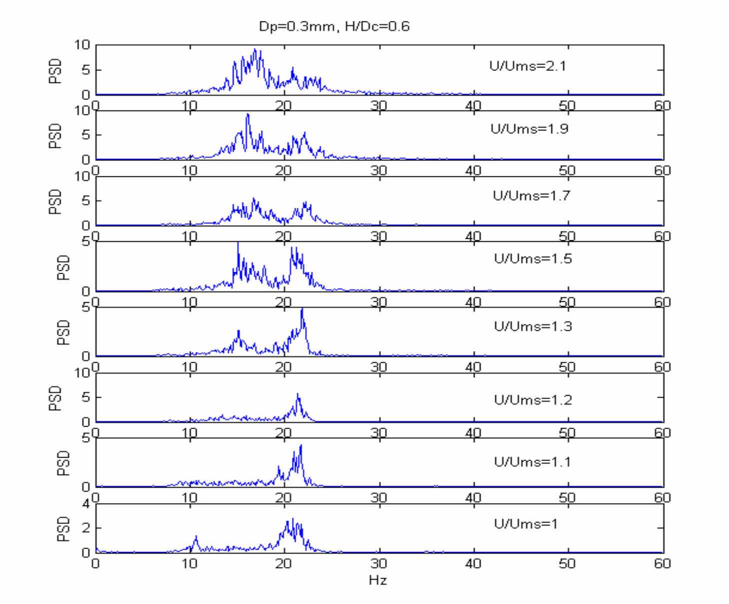

Figure 13. Main frequency peak *vs.* gas flow rate for 53.9 grams 0.3mm ZrO_2 in 60 degree spouted bed.

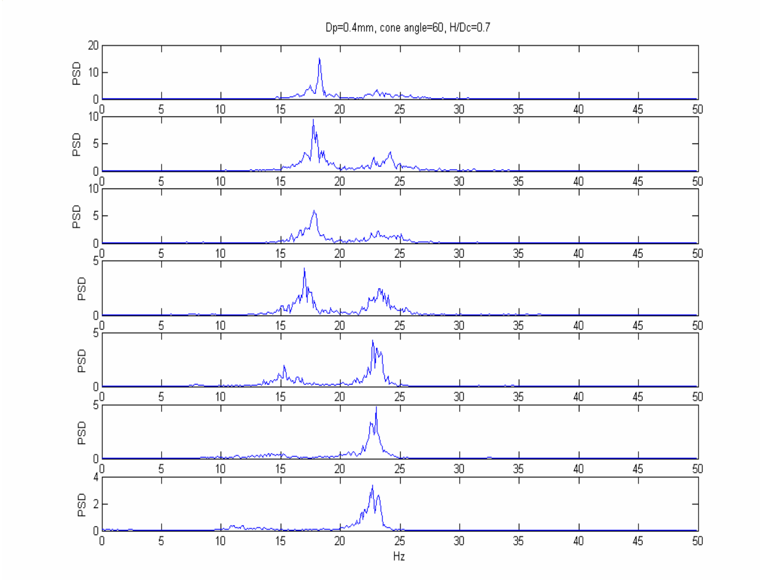

Figure 14. Main frequency peak *vs.* gas flow rate for 53.9 grams 0.4mm ZrO_2 in 60 degree spouted bed.

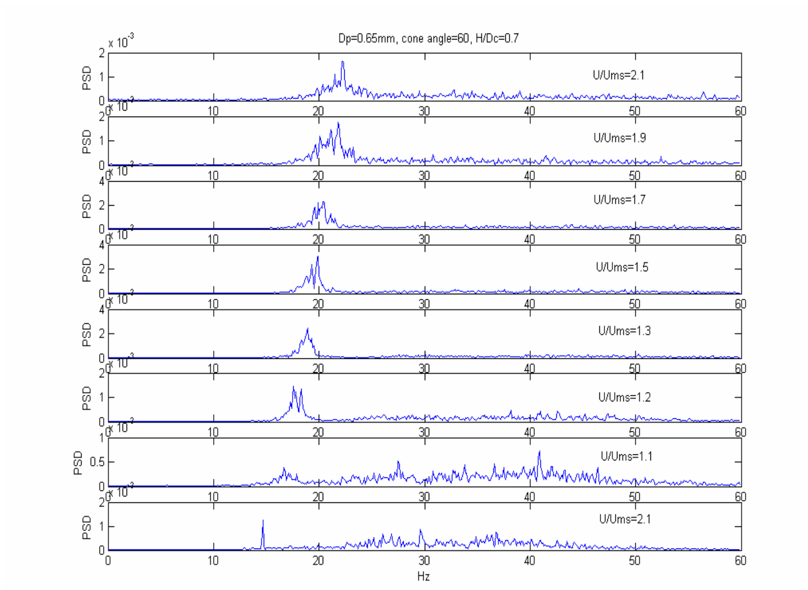

Figure 15. Main frequency peak *vs.* gas flow rate for 53.9 grams 0.65mm ZrO_2 in 60 degree spouted bed.

The main frequencies of 0.4mm and 0.65mm particles show the same tendency as gas flow rate changes from high to low. The tendency of 0.3 mm particle is not obvious.

3.4.2 Main frequency peak *vs.* particle size

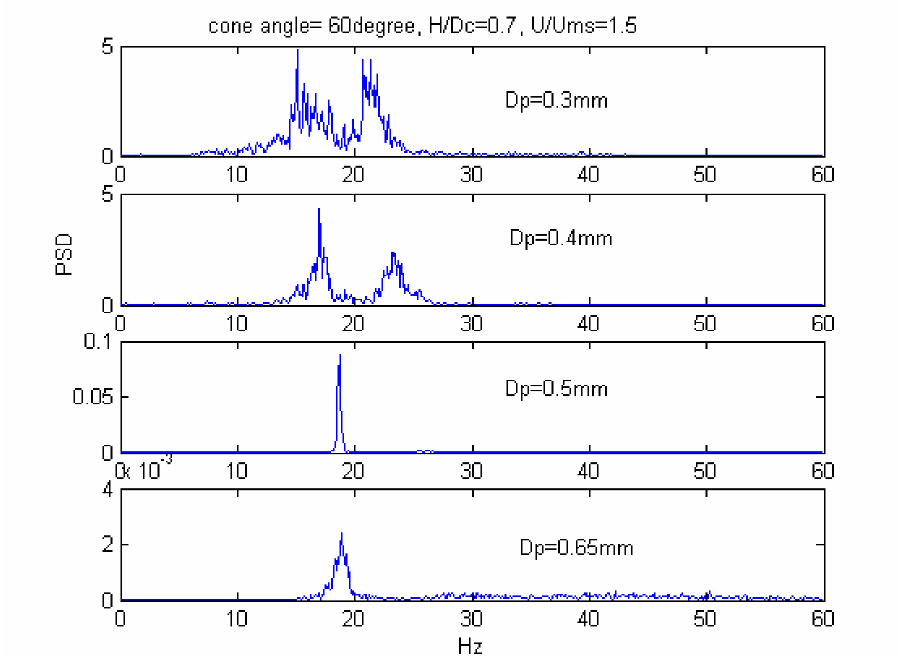

Figure 16. Main frequency peak *vs.* particle size for 53.9 grams ZrO_2 in 60 degree spouted bed.

As particle size increases, the main frequency peak increases slowly. 0.5mm particle has sharpest main frequency peak while others become wider as Dp increases or decreases.

3.4.3 Main frequency peak *vs.* cone angle

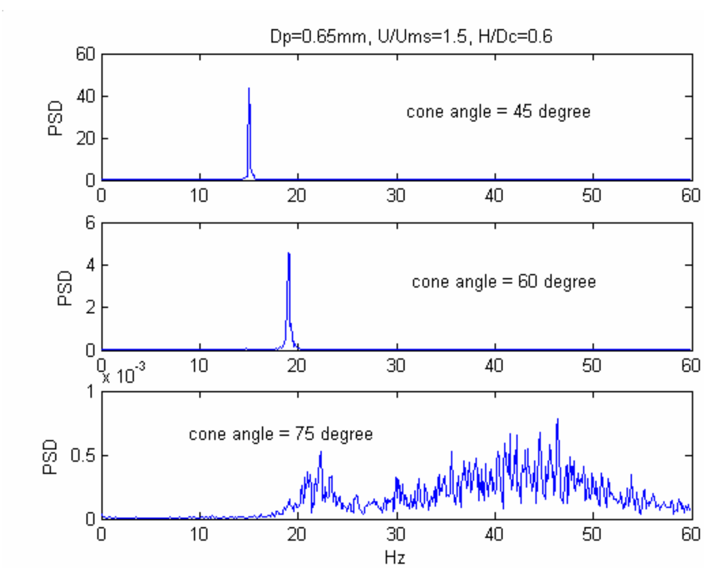

Figure 17. Main frequency peak *vs.* cone angle at U/Ums=1.5, H_0/Dc=0.6 and Dp=0.65mm.

As cone angle increases, the main frequency peak increases and its width increases.

3.4.4 Main frequency peak *vs.* H_0/Dc

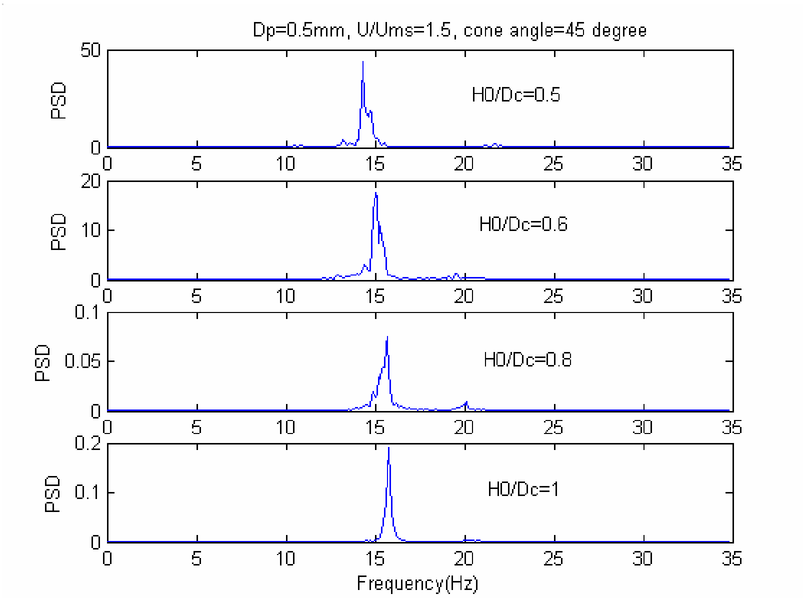

Figure 18. Main frequency peak *vs.* H_0/Dc at U/Ums=1.5, cone angle=45 and Dp=0.65mm.

As H_0/Dc increases, main frequency peak increases.

3.4.5 Analyze pressure drop statistically

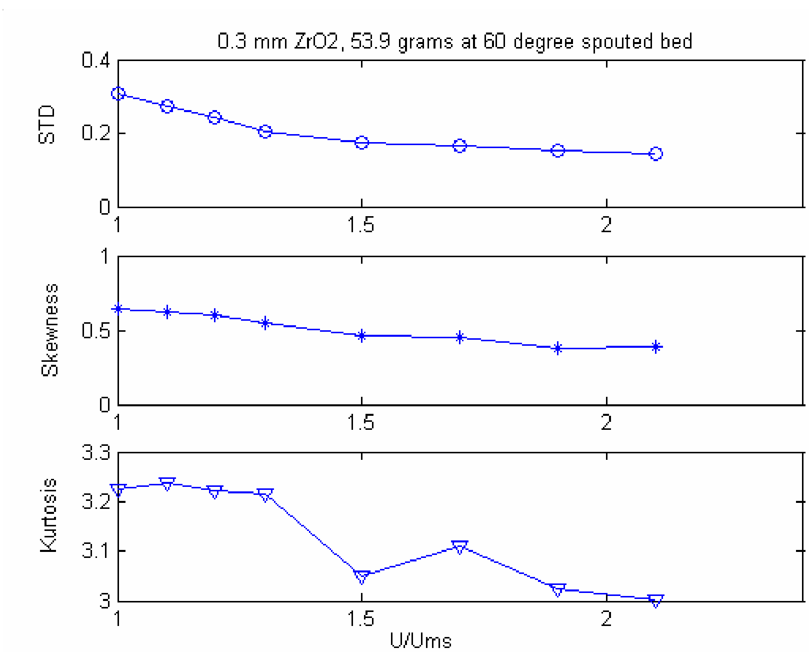

Figure 19. Standard deviation, skewness and Kurtosis vs. U/Ums for 53.9 grams 0.3mm ZrO_2 at 60 degree spouted bed.

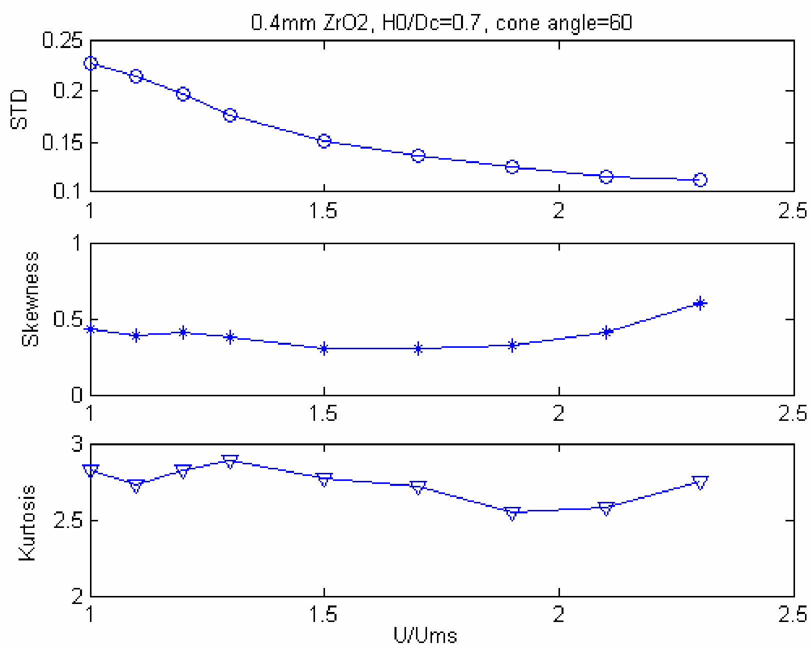

Figure 20. Standard deviation, skewness and Kurtosis vs. U/Ums for 53.9 grams 0.4mm ZrO_2 at 60 degree spouted bed.

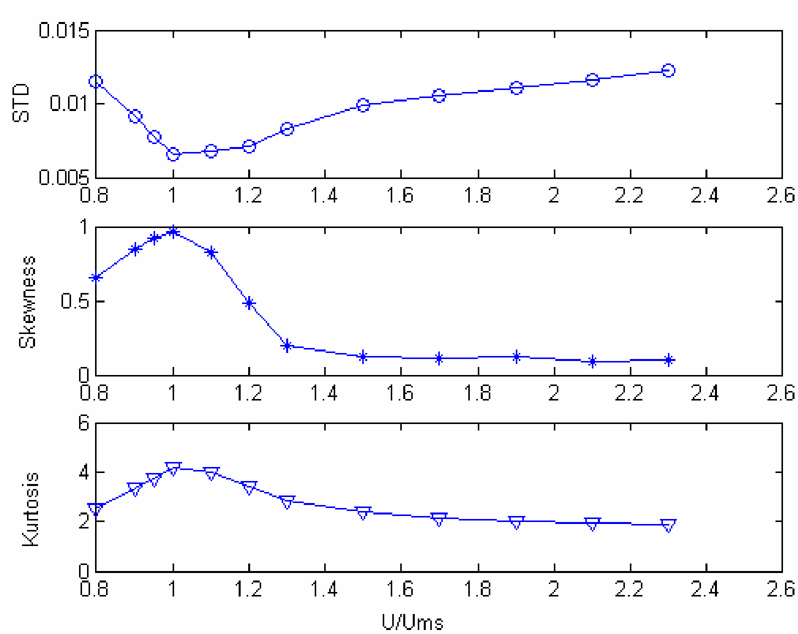

Figure 21. Standard deviation, skewness and Kurtosis vs. U/Ums for 53.9 grams 0.5mm ZrO_2 at 60 degree spouted bed.

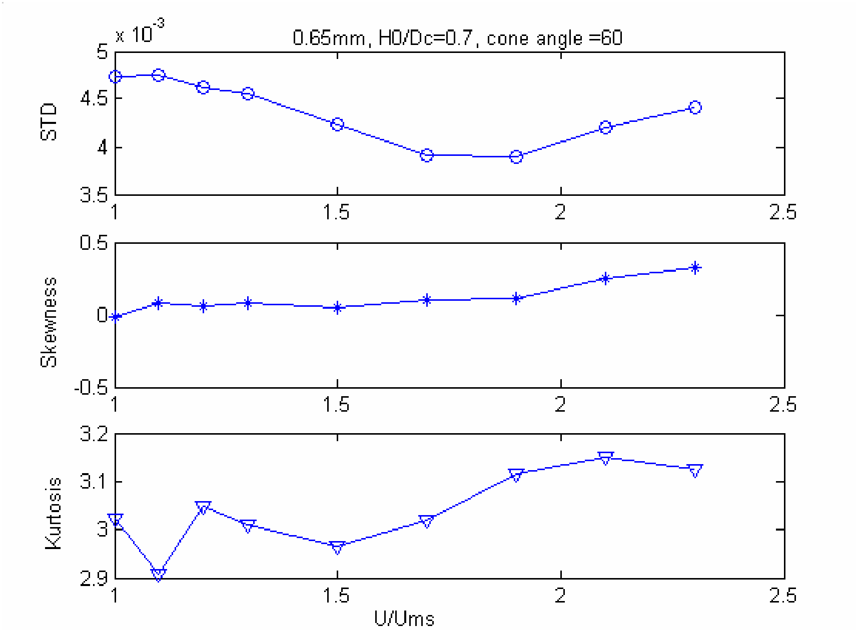

Figure 22. STD, skewness and Kurtosis vs. U/Ums for 53.9 grams 0.65mm ZrO_2 at 60 degree spouted bed.

The changes of standard deviation, skewness and kurtosis vs. U/Ums are shown above. All STD increases as gas flow rate is lowered to Ums. The skewness also increases except 0.65mm particles. In general, kurtosis also increases as gas flow rate is lowered to Ums.

4. Conclusions

A quantitative method is used to measure Ums of ZrO2 particles. And an effective correlation to predict Ums of ZrO2 particles is developed.

An effective dimensionless equation to predict the gas pressure drop across the spouted bed, ΔP, at stable spouting conditions $1 \le U/U_{ms} \le 1.9$ is developed. The relationship between log(ΔP) and log(tan(γ/2)) or $\log(H_0/D_c)$ is linear while other factors are constant. ΔP is a third order polynomial function of U/Ums. And the effect of tan(γ/2) and H_0/D_c as well as Dp/Dc is depended on the U/Ums. The larger the U/Ums is, the larger the effects they exert.

The main frequency peak of the pressure drop signal shifts from high to lower value as gas flow rate is lowered from higher value corresponding to full spouting condition to lower value. As gas flow rate is lowered to 1.1~1.2Ums, the main frequency peak increases sharply. Main frequency peak of the pressure signal increases as particle size or static particle height inside the spouted bed or cone angle increase. 0.5mm particle has sharpest main frequency peak while others become wider as Dp increases or decreases. The standard deviation of the pressure signal of 0.3mm, 0.4mm and 0.65mm particles tend to increase as gas flow rate is lowered to Ums while the one of 0.5mm particles tends to decreases. The skewness also increases except 0.65mm particles as gas flow rate is lowered to Ums. In general, kurtosis also increases as gas flow rate is close to Ums.

5. Acknowledgements
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6. References

[1] R. L. Beatty, F. A. Carlsen Jr., J. L. Cook, Pyrolytic-carbon coating on ceramic fuel particles. Nucl. Appl. 1, 560, 1965.

[2] T. M. Knowlton, S. B. R. Karri, A. Issangya, Scale-up of fluidized-bed hydrodynamics, Powder Technology, 150(2005), 72-77

[3] Joseph A. Sholtis, Jr. Ronald J. Lipinski, and Mohamed S. El-Genk, Coated Particles for Radioisotope Power Systems and Radioisotope Units, Space Technology and Applications International Forum-1999, 1378-1384.

[4]Jian Xu, Xiaojun Bao, Weisheng Wei, etc., Statistical and frequency analysis of pressure fluctuations in spouted beds, Powder Technology, 140, 141-154, 2004.

[5] Lii-Ping Leu, Chih-Nan Wu, Prediction of pressure fluctuations and minimum fluidization velocity of binary mixtures of Geldart Group B particles in bubbling fluidized beds, The Canadian Journal of Chemical Engineering, Vol. 78, 578-585, 2000.

[6] M. Olazar, M. J. San Jose, T. Aguayo, J. M. Arandes and J. Bilbao, Pressure drop in conical spouted beds, The Chemical Engineering Journal, 51, 53-60, 1993.

[7] D. Geldart, Gas Fluidization Technology, Wiley, New York, 1986.

[8] Daizo Kunii, Octave Levenspiel, Fluidization Engineering, Butterworth-Heinemann, 1991.

[9] J. F. Dadiason, R. Clift, D. Harrison, Fluidization, Academic Press, 1985.

[10] S. Aravinth, T. Murugesan, Bioprocess Engineering, 16, 289-293, 1997.

[11] H. T. Bi, A. Macchi, J. Chaouki, and R. Legros, The Canadian J. of Chemical Engineering, 460-465, Vol 75, 1997.

[12] San Jose, M. J. Olazar, M. Aguayo, A. T. Arandes, J. M. Bilbao, Fluidization VII, Engineering Foundation, 1992.

[13] M. Z. Tsvik, M. N. Nabiev, N. U. Rizaev et al., Uzb. Khim, Zh. 11(2), 50-59, 1967.

[14] A. E. Gorshtein, I .P. Muknlenov, Zh. Prikl. Khim., 37, 1887-1893, 1964

[15] I. P. Mukhlenov, A. E. Gorshtein, Khim. Prom. 41(6), 443-446, 1965.

[16] S. Pannala, C. S. Daw, C. E. A. Finney, D. Boyalakuntla, D. Bruns and J. Zhou, "ORNL FY04 Process Modeling Summary Report for the Advanced Gas Reactor Fuel Development and Qualification Program," Oak Ridge National Laboratory Technical Report ORNL/CF-04/11, September 2004.

Nomenclature

Ar: Archimedes number = $gd_p^3(\rho_p-\rho_g)/\ \mu_g^2$
D_c: diameter of the column of the spouted bed, m
Dp: particle size, mm,
Di: gas inlet diameter, m,
g: gravitational acceleration, m/s^2
H_0: static particle height, m,
ΔP: gas pressure drop across the spouted bed, Pa,
Re_{ms}: Reynolds number at $U_{ms} = \rho_g U_{ms} d_p/\mu_g$
U: gas flow rate, m^3/s
U_{ms}: minimum spouting velocity, m^3/s
ρ_b: bulk density of the particle, kg/m^3
γ: cone angle of the spouted bed, degree