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# Influence of Particle Size on the Pressure Fluctuations and Slugging in a Fluidized Bed

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In aggregative fluidization the pressure fluctuations are closely interrelated with the behavior of solids and gas in the bed. These random fluctuations of pressure can be measured by a sensitive pressure transducer, recorded and subjected to the statistical

Kang et al. (1967), Winter (1968), and Lirag (1971) appear to be early investigators who attempted to relate the statistical properties of pressure fluctuation signals to phenomena occurring in the fluidized bed. An effort was also taken to define an index of the quality of fluidization from the statistical characteristics of pressure fluctuations. The practical regime of aggregative fluidization is bounded by the beginning of bubbling at a low fluid velocity and by the onset of slugging at a high fluid velocity. For any fluidized-bed reactor there is an optimum bubbling rate providing the required mixing without excessive loss in contacting efficiency. The fluctuations of pressure drop across a fluidized bed are known to increase with an increase in the intensity of bubbling. Thus knowledge of two fundamental properties of pressure fluctuations, i.e., the frequency spectrum and pressure drop amplitudes, is a useful tool for the rational diagnosis of behavior of a fluidized-bed reactor.

The nature of pressure fluctuations in a fluidized bed is a complex function of particle properties, bed geometry, flow conditions, pressure and temperature. An effect of bed size, gas flow rate and particle properties on the pressure oscillation in slugging beds was studied by Broadhurst and Becker (1976). The authors presented dimensionless correlations for estimation of the major frequency and the peak in the frequency spectrum for group B of Geldart's classification of solids (1973). Sadasivan et al. (1980) developed empirical equations relating the characteristic frequency and maximum pressure drop fluctuations to solid properties, gas flow rate, and height of the static bed.

Aside from the influence of the operating conditions on the major frequency and amplitude of the pressure fluctuations, Fan et al. (1981) also explored the causes of the fluctuations. The authors concluded that the formation and motion of bubbles appear to be the major causes of pressure fluctuations in a fluidized bed. In our recent study (Svoboda et al., 1982) we have investigated an effect of temperature on the pressure fluctuations in the bed of different materials. We have found that the pressure fluctuations are smaller and more rapid at elevated temperatures than those at ambient temperature.

Our experience shows that no systematic investigation of the influence of particle size on the fluctuation characteristics has been conducted. Particularly, experimental data covering a wide range of gas velocities for group D of the Geldart's classification are lacking.

In this work the effect of particle size and relative gas velocity on the dominant frequency and mean pressure amplitude is investigated. Attempts have also been made to determine the onset of slugging from the power frequency spectrum, to correlate the dominant frequency of slugging with particle size and to explore the effect of excess gas velocity  $(U_f - U_{mf})$  on the mean pressure amplitude for particles of different size.

## **EXPERIMENTAL**

#### **Apparatus**

All measurements were conducted in a fluidized-bed column of inside diameter D=8.5 cm and height  $H_K=50$  cm. The fluidizing air was introduced through a perforated-plate distributor of free area  $\phi=2\%$  and orifice diameter  $d_o=0.8$  mm into the bed of particles. A special construction above the column ensured an accurate and fixed location of the vertical pressure probe  $(d_t=4\text{ mm})$  in the bed. The pressure transducer, connected to the outside opening of the tap, converted a fluctuation pressure signal to an output voltage proportional to the pressure. The output signal was amplified, digitalized and further processed on-line by a correlation analyzer and computer. Further details on the apparatus design and pressure tapping can be found elsewhere (Svoboda and Hartman, 1981; Svoboda et al., 1982).

#### **Materials**

The properties of limestone particles used in this work and their incipient fluidization velocities determined from the pressure drop-gas velocity data are summarized in Table 1. The particles were of irregular shapes, but essentially isometric. The sieved fractions investigated in this study comprized four narrow size ranges: 0.50 to 0.63 mm; 0.63 to 0.80 mm; 0.80 to 1.00 mm; and 1.00 to 1.25 mm. The particles of mean size  $\overline{D}=0.565$  mm belong to B group of particles according to Geldart (1973). The particles of mean sizes 0.715 and 0.900 mm can be classified as belonging to the boundary region between groups B and D. The largest fraction of particles conform with category D in which majority of bubbles rise more slowly than the interstitial fluidizing gas. Such a bed exhibits small expansion and low mixing rate of the particles.

#### **Procedure**

The voltage-time signals corresponding to the pressure-time signals were collected for about 60 s. The sampling time interval for analog-digital conversion was selected to be 10 ms and a total of N=6,000 values were treated.

The mean pressure drop across the bed was computed as

$$\Delta \overline{P} = \frac{1}{N} \sum_{i=1}^{N} \Delta P_i \tag{1}$$

The mean deviation of pressure fluctuations was interpreted as a mean amplitude Y of the fluctuations, i.e.

$$Y = \left[ \frac{1}{N-1} \sum_{i=1}^{N} (\Delta P_i - \Delta \overline{P})^2 \right]^{0.5}$$
 (2)

The power spectrum of frequency, which expresses the distribution of energy with frequency, was obtained by the Fourier transform of the autocorrelation function (Bendat and Piersol, 1971; Fan et al., 1981). The frequency corresponding to the maximum power in the spectrum is considered in this work the dominant or major frequency  $f_d$ . Other local maxima are classified as side frequencies  $f_{si}$ .

#### Measurement

The pressure transducer was calibrated with the aid of a laboratory manometer. Good reproducibility of the pressure transducer data required to maintain constant temperature. A pressure probe tube of  $d_t = 4$  mm with the open measuring end was employed in the work. An important factor affecting the pressure fluctuation signal is a vertical location of the probe. In accordance with previous investigations of Fan et al. (1981) and Svoboda

TABLE 1. PHYSICAL PROPERTIES OF LIMESTONE PARTICLES

Size Range (mm)	$\overline{D}_{p} \ ( ext{mm})$	$ ho_p$ $( ext{kg-m}^{-3})$	$U_{mf} \ ( ext{m-s}^{-1})$
0.50-0.63	0.565	2,220	0.244
0.63 - 0.80	0.715		0.342
0.80 - 1.00	0.900		0.425
1.00 - 1.25	1.125		0.520

et al. (1982), the maximum mean amplitudes were found in the middle of the bed. The frequency spectrum was affected by a vertical position of the tap much less than the amplitudes. No significant dependence of the frequency on the distance above the gas distributor was detected. The effect of radial position on the signal was found to be negligible at gas flow rates above  $U_f/U_{mf}=1.05$ .

All experimental measurements were performed with a static bed height  $H=17\,\mathrm{cm}$ , i.e.,  $H=2\,D$ , at ambient temperature (20–23°C). The pressure probe was located 8.5 cm above the distributor. The relative gas velocity  $U_f/U_{mf}$  ranged from 1 to 1.8.

## **Results and Discussion**

In Figures 1 and 2 are presented the dominant frequencies  $f_d$  at different relative gas velocities  $U_f/U_{mf}$ . A steep decrease of the dominant frequency, particularly illustrative for smaller particles, appears in the range of gas velocities  $1.05 < U_f/U_{mf} < 1.3$ . The occurrence of side frequencies depends considerably on the particle size. At the smallest particles  $(\overline{D}_p = 0.565 \text{ mm})$  the side frequencies with a relatively large power exist in the broad range of gas velocities  $1.0 < U_f/U_{mf} < 1.5$  as shown in Table 2. In the case of a bed with the largest particles used  $(\overline{D}_p = 1.125 \text{ mm})$ , the side frequencies of a power comparable to that of the dominant frequency were detected only at air velocities very close to incipient fluidization  $(U_f/U_{mf} < 1.03)$ .

Visual observations of the bed behavior over a broad range of gas flow rates suggested that the disappearance of higher side frequencies in a power spectrum is connected with the onset of slugging. This finding is particularly distinctive at small particles belonging to group B of Geldart's classification of powders. As gas velocity, or bed depth increase, bubble formation and motion become more ordered, giving rise to more coherent presure fluctuations and side frequencies start to disappear from the power spectra.

In the course of work we confronted a series of the power spectra with the physical picture of the fluidized bed. It was found that in the regime of well developed slugging, a single peak of the dominant frequency occurs in the power spectrum. The rest of the spectrum shows a relatively uniform distribution of power with the frequency without any significant local maximum. The ratio of powers of a side frequency and the dominant frequency,  $G_p(f_{si})/G_p(f_d)$ , is in slugging beds less than 0.1.

We have made an attempt to define the onset of slugging as the gas velocity at which the ratio  $G_p(f_{si})/G_p(f_d)$  attains a value of 0.15. Our effort to find an objective method for establishing the point of minimum slugging was also motived by the fact that its visual determination becomes increasingly difficult with larger particles of D group. The onset of slugging represents the terminal stage of bubble coalescence when the majority of bubbles are as large as the diameter of the column. With respect to irregular shapes of bubbles and chaotic behavior of the bed, the results obtained visually are affected by each observer's interpretation of the phenomena. The gas velocities at which the power ratio  $G_p(f_{si})/G_p(f_d)$  is as low as 0.15 were determined for all the fractions of particles. The results are plotted in Figure 3 along with the approximations obtained by visual observation. The trend of the data determined with the aid of the frequency spectra is

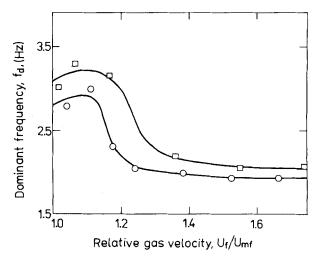


Figure 1. Dependence of the dominant frequency on the relative gas velocity: particles of limestone; □, particle size, 0.565 mm; ○, particle size, 0.715 mm.

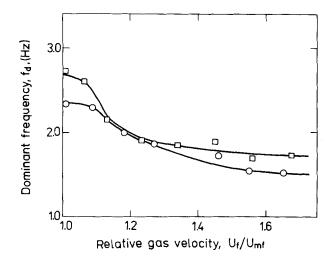


Figure 2. Dependence of the dominant frequency on the relative gas velocity: particles of limestone; □, particle size, 0.90 mm; ○, particle size, 1.125 mm.

in good agreement with the results of visual observation as well as with the values computed from the empirical correlations of Broadhurst and Becker (1975). These authors derived their correlations for  $U_{mf}$  and  $U_{ms}$  from a large number of experimental data covering widely different operating conditions.

The decrease in values of  $U_{ms}$ - $U_{mf}$  with increasing particle size is in contrast with the results in the literature (Stewart and Davidson, 1967; Geldart, 1972; Baeyens and Geldart, 1974). These authors worked with particles conforming with groups B and D of the Geldart classification and did not find any dependence of  $(U_{ms}$ - $U_{mf})$  on the diameter of particles. However, we should note that the equation of Stewart and Davidson (curve 1 in Figure 3)

$$U_{ms} = U_{mf} + 0.07 (gD)^{1/2}$$
 (3)

as well as that of Baeyens and Geldart

$$U_{ms} = U_{mf} + 0.07 (gD)^{1/2} + 1.61.10^{-3} (60D^{0.175} - H_{mf})^2$$
 (4)

(curve 2 in Figure 3) represent correlations of data from the experiments with deeper beds (H > 30 cm). Moreover, the ratio H/D was higher than that in our experimental measurements.

On the basis of the presented comparison we believe that the analysis of frequency spectra can be employed for establishing the minimum slugging point. This instrumental method is particularly useful for columns with opaque walls and operated at elevated temperatures (Svoboda and Hartman, 1981a,b).

Table 3 presents the dominant frequencies of different particle fractions determined at equal gas velocities  $(U_f - U_{mf})_s$  in the region of developed slugging. We can see that increasing particle size causes a moderate, continuous decrease in slugging frequency  $f_s$ . Good agreement of our experimental data with the empirical equation of Sadasivan et al. (1980) suggests that validity of the relation 5

TABLE 2. VARIATION OF THE DOMINANT FREQUENCY AND SIDE FREQUENCY WITH THE RELATIVE GAS VELOCITY

U <sub>f</sub> (m/s)	$U_f/U_{mf}$	$f_d$ (Hz)	f <sub>si</sub> (Hz)	$G_p(fi_{si})/G_p(f_d)$
0.261	1.07	3.30	2.94 (2.6)	0.90 (0.60)
0.285	1.17	3.17	2.28	0.59
0.332	1.36	2.19	3.30	0.51
0.379	1.55	2.03	3.30	0.15
0.427	1.75	2.11	_	< 0.1
0.522	2.14	2.03	_	<0.1

Material, limestone; particle size, 0.565 mm.

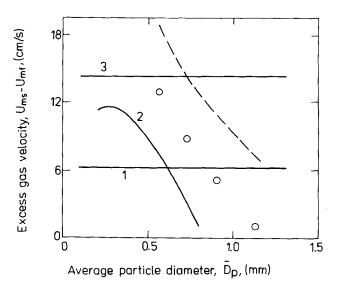


Figure 3. Influence of mean particle size on the excess gas velocity at the onset of slugging: O, experimental data points obtained from the power frequency spectra. The solid curves show the values predicted by the individual equations. The physical properties of particles summarized in Table 1 were used in computations. Curve 1: Stewart and Davidson (1967). Curve 2: Broadhurst and Becker (1975). Curve 3: Baeyens and Geldart (1974). The dashed curve outlines approximations established by visual observations of the surface of bed.

$$f_s \sim \frac{1}{D_p^{0.31}}$$
 (5)

can be also extended to the group  ${\cal D}$  of solids.

As seen in Figure 4, the mean amplitudes of pressure fluctuations increase almost linearly with the relative gas velocity for the particles of all sizes. A considerable influence of the particle size on the mean amplitudes is demonstrated for different gas velocities in Figure 5. The lower curve in this figure corresponds approximately to the bubbling regime of fluidization, while the upper one reflects developed slugging in bed. Both curves show clearly that the mean amplitude of pressure fluctuations increase with increasing particle size.

If the mean amplitude is plotted against the excess gas velocity  $(U_f - U_{mf})$  instead of the relative gas velocity  $U_f / U_{mf}$ , the effect of the particle size is somewhat less pronounced. Nevertheless, it cannot be neglected as the results shown in Figure 6 suggest. Sadasivan et al. (1980) proposed a simple equation relating the maximum pressure drop fluctuation to the bubble size

$$\delta \sim D_b^{1.213} \tag{6}$$

for the group B of materials. In light of the above findings, it appears that a similar relation between Y and  $D_b$  can be assumed. That would imply a moderate increase of bubble size with the particle diameter at constant values of the excess gas velocity  $U_f$ - $U_{mf}$ . The idea is also supported by the earlier onset of slugging in the beds of coarse particles as demonstrated in

TABLE 3. VARIATION OF THE MEAN PRESSURE AMPLITUDE AND FREQUENCY WITH PARTICLE SIZE AT A CONSTANT EXCESS GAS VELOCITY IN THE SLUGGING BED

$\overline{D}_p$		Y	$f_s$
(mm)	$U_f/U_{mf}$	(kPa)	(Hz)
0.565	1.80	0.3038	2.05
0.715	1.57	0.3430	1.95
0.900	1.46	0.3822	1.80
1.125	1.37	0.4214	1.70

Material, limestone; excess gas velocity,  $U_f$ - $U_{mf} = 0.195$  m/s.

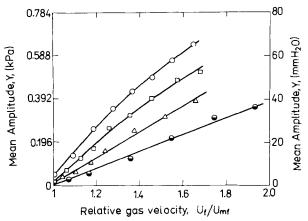


Figure 4. Dependence of the mean pressure amplitude on the relative gas velocity and particle size:  $\Theta$ , particle size, 0.565 mm;  $\Delta$ , particle size, 0.715 mm; □, particle size, 0.90 mm; ○ particle size, 1.125 mm.

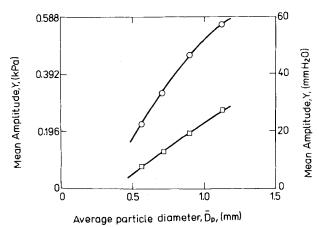


Figure 5. Dependence of the mean pressure amplitude on the particle size and relative gas velocity:  $\Box$ , relative gas velocity,  $U_l/U_{ml} = 1.2$ ;  $\bigcirc$ , relative gas velocity,  $U_l/U_{ml} = 1.55$ .

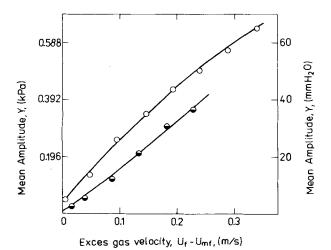


Figure 6. Dependence of the mean pressure amplitude on the excess gas velocity and particle size: ⊕, particle size, 0.565 mm; O, particle size, 1.125 mm.

Figure 3. This fact shows that at a given excess gas velocity, larger bubbles are formed in such beds near and at the point of minimum slugging. However, most of the published works (e.g., Geldart, 1972; Mori and Wen, 1975; Rowe, 1976; Darton et al., 1977) do not incorporate the particle size in correlations of the bubble size in fluidized beds.

# **NOTATION**

D= inside diameter of fluidization column, m

diameter of bubble, m mean particle diameter, m = diameter of orifice, m

= inside diameter of pressure probe tube, m = frequency of pressure drop fluctuations, Hz

= frequency with maximum power, dominant frequency,

Hz

= side frequency, Hz

= power density function,  $V^2$ = acceleration due to gravity, ms<sup>-2</sup>

Η = height of static bed, m N number of samples  $\Delta P$ = pressure drop, Pa

U= superficial gas velocity, m·s<sup>-1</sup>

= mean amplitude of pressure fluctuations, Pa

## **Greek Letters**

δ = magnitude of maximum pressure drop fluctuations

= viscosity, Pa·s μ

= density,  $kg \cdot m^{-3}$ ρ

= free area of gas plate distributor

# **Subscripts**

φ

= fluid property

= index of measurements

= property at minimum fluidization point mf

= property at minimum slugging point ms

= property of solid particle p = property in slugging regime s

sb= property of settled bed (the fixed bed obtained by vibrating a bed until no further consolidation takes place)

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# A New Integral Approximation Formula for Kinetic Analysis of Nonisothermal TGA Data

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#### INTRODUCTION

A knowledge of kinetic behavior is essential for understanding and predicting the thermal behavior of coal conversion processes (Wen and Lee, 1979; Collett and Rand, 1980), woody material thermal decomposition (Tang and Neill, 1964), and biomass pyrolysis-gasification-combustion processes (Milne, 1979). The thermal behavior of coal, wood and related biomass is frequently studied by measuring the rate of weight loss of the material as a function of time and temperature. This information, coupled with a proposed reaction mechanism, is then used to estimate activation energies and frequency factors for Arrhenius type reaction rate expressions. This note presents a new integral approximation formula for extracting these kinetic parameters from experimental data. It will be shown that this method is superior to the commonly used integral approximation formula (Coats and Redfern, 1964).

Thermogravimetric analysis (TGA) provides a semiquantative understanding of pyrolysis under well-controlled laboratory conditions. Furthermore, by the use of fine particles and small sample sizes, phenomena related to transport problems can be minimized. Consequently, the validity of TGA data for kinetic analysis of primary pyrolysis is greatly enhanced.

Thermogravimetric analysis is normally performed in either the isothermal or nonisothermal (dynamic) mode. The advantages (Freeman and Carroll, 1958) of investigating reaction kinetics by a dynamic TGA are that considerably less experimental data are required than in the isothermal method and the kinetics can be probed over the entire temperature range in a continuous manner. In addition, when a sample undergoes significant reaction in being raised to the temperature of interest, the results obtained by the isothermal method are often questionable.

Three basic approaches to analysis of nonisothermal TGA data have been discussed in the literature. The differential method (Sharp and Wentworth, 1969) and the difference-differential method (Tang, 1967; Freeman and Carroll, 1958; Sharp and Wentworth, 1969) suffer from some inherent disadvantages. The integral method (Jahnke et al. 1960; Coats and Redfern, 1964) seems to be the best method. The Coats-Redfern equation has been used as an approximation formula for the integral method, but is not as precise as desired. A new integral approximation formula is derived below that is more precise than the Coats-Redfern equation, yet is simple and easy to apply.

#### **DERIVATION OF APPROXIMATION FORMULA**

Irreversible pyrolysis of solids is typically described by the following chemical equation:

$$sS_{(s)} \rightarrow rR_{(s)} + vV_{(g)}$$

where S, R and V are original solid, final solid residue, and volatile matter, respectively.

The decomposition rate of a solid can be represented by the general rate expression shown below.

$$\frac{d\omega}{dt} = kf(\omega) \tag{1}$$

where  $\omega$  is the decomposed fraction of solid (on the decomposable basis) at time t,  $f(\omega)$  is a function of  $\omega$  depending on the reaction mechanism and k is the rate constant given by the Arrhenius equation as

$$k = A \exp(-E/RT) \tag{2}$$

where A = frequency factor, E = activation energy, R = universal gas constant, T = absolute temperature.

For a linear heating rate, say  $\beta$  K/min, the following relationship is valid

$$\beta = \frac{dT}{dt} \tag{3}$$

Combining Eqs. 1, 2 and 3 and integrating between the initial temperature,  $T_0$ , and any final temperature, T, and conversion between  $\omega_0$  and  $\omega$ , respectively results in Eq. 4.

$$\int_{\omega_0}^{\omega} \frac{d\omega}{f(\omega)} = \frac{A}{\beta} \int_{T_0}^{T} \exp(-E/RT)dT$$
 (4)

The righthand side of Eq. 4 is not analytically integrable, but can be integrated by parts to obtain the expression shown in Eq. 5.

$$\int_{T_0}^{T} \exp(-E/RT)dT$$

$$= \frac{RT^2}{E} \exp(-E/RT) \Big|_{T_0}^{T} - \int_{T_0}^{T} \frac{2RT}{E} \exp(-E/RT)dt \quad (5)$$

Rearrangement of Eq. 5 gives Eq. 6.

$$\int_{T_0}^{T} \left( 1 + \frac{2RT}{E} \right) \exp(-E/RT) dT = \frac{RT^2}{E} \exp\left( -\frac{E}{RT} \right) \Big|_{T_0}^{T}$$
 (6)

Since 2RT/E is much less than unity at moderate temperatures and