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Development of Sorbent Manufacturing Technology by Agitation Fluidized Bed Granulator (AFBG)

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Abstract–Thousands of ppmv of hydrogen sulfide included in coal gas should be reduced to less than a hundred ppmv in the case of IGCC to prevent a gas turbine from being corroded, and few ppmv to prevent the performance of electrodes from declining in the case of MCFC. In the present paper a laboratory scale AFBG (Agitation Fluidized Bed Granulator) is made and improved. The sorbent for the removal of hydrogen sulfide is produced using an agitation fluidized bed granulator (ZnO 1.5 mole+TiO_2 1.0 mole+bentonite 5.0 wt%). The techniques for fluidizing fine particles, classified in Geldart C group, in a fluidized bed are developed by installing an agitator blade in a fluidized bed granulator. The fine particles are fluidized and granulated successfully by using the techniques. Statistical, spectral and chaos analyses with granulated sorbent (100-300 μm) are performed to investigate the hydrodynamics of granulates in a fluidized bed. The average absolute deviation, power spectral density functions, phase space trajectories, and Kolmogorov entropy obtained from pressure fluctuation are plotted as a function of fluidizing velocity. It is shown that the Kolmogorov entropy implying the rate of generation of information can be applied to the control of fluidization regimes.

Key words : Fluidized Bed Granulator, Agitator, Fine Particles, Chaos Analysis, Kolmogorov Entropy

INTRODUCTION

One of the most important problems in the de-H_2S process at high temperature and high pressure is to prepare sorbent that has high physical resistance to attrition. In the present research, agitation fluidized bed granulation, instead of conventional pan-granulation or spray drying method, is used to make the sorbent for the removal of H_2S [Choi, 1985; Watano et al., 1995].

Among various techniques, fluidized bed granulation has advantages such as reducing the powder handling costs, less contamination by dust, and the savings of time and space for the process of preparing desulfurization sorbents [Ennis, 1996]. In particular, the agitation fluidized bed granulator equipped with an agitator blade on the lower part of fluidized bed has been applied to many engineering fields. The main advantage of the process is that the size, shape and density of granules can be controlled by the rotation of the agitation blade. Therefore, the development of the agitation fluidized bed granulator is essential for the manufacturing and mass production of desulfurization sorbent [Iveson et al., 1996].

The raw materials for the agitation fluidized bed granulation were ZnO, TiO_2, and bentonite, ranging between 0.3 and 3.9 μm of average particle size. These fine particles are difficult to fluidize since interparticle forces cause agglomeration of the particles and bridging between the agglomerates [Geldart and Wong, 1984, 1985]. The agglomeration and bridging induce the formation of channels through which the gas passes, resulting in no fluidization of the fine particles. It is demonstrated that the fluidization of fine particles can be accomplished by introducing the agitator.

The dynamics of gas-solids fluidization is characterized by using statistical analysis (mean, standard deviation, skewness, etc.), spectral analysis (power spectrum density function, autocorrelation), and chaos theory. If only one variable is related to pressure fluctuation, the attractor (state-space trajectories) can be reconstructed. The objective of the characterization is to show how these analyses can be used to identify the transition regime in a fluidized bed.

The possibility of characterizing the dynamics of a gas-solids fluidized bed using deterministic chaos analysis has been reported recently [Van den Bleek and Schouten, 1993]. This nonlinear time series analysis is accomplished with time series of measured pressure signals of a fluidized bed system [Marzocchella et al., 1997]. Among various characteristic numbers in chaos analysis, Kolmogorov entropy represents the rate of generation of information. It is zero for periodic data such as sine curve and infinite for random data. The value of Kolmogorov entropy for deterministically chaotic processes is between zero and infinity. The results from deterministic chaos theory can be used to classify regimes of various types of fluidizations [Schouten et al., 1992; Van der Stappen et al., 1992].

THEORY

The average absolute deviation of a signal is expressed as $\frac{1}{n}\sum|x-\bar{x}|$, where n is the number of data and $\bar{x}$ is the average of the data.

The pressure signals of linear time-variant systems in a fluidized bed reactor are often analyzed with Fourier transform. The

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Fourier integral is expressed by the equation:

$$H(f)=\int_{-\infty}^{\infty} h(t)e^{-i2\pi ft}dt \quad (1)$$

There are various ways to define discrete Fourier transforms. In this study, the Fourier transform b_s of a list a_r of length n is expressed to be $\frac{1}{\sqrt{n}}\sum_{r=1}^{n} a_r e^{2\pi i(r-1)(s-1)/n}$.

Time series embedding projects the pressure signal time series measurements to a new coordinate system. The multidimensional points or vectors, P(t), constructed from the measured pressure signal are expressed as P(t)={p(t), p(t+k), p(t+2k), …, p(t+(m−1)k)}, where k is time steps and p(t) is the pressure at time t, m is an embedding dimension.

Average Mutual Information (AMI) between p(t) and p(t+T) chooses the value for the time delay T. The value, an integer which is the number of units of τ_s between two samples, is used to calculate other core values such as correlation dimension and Lyapunov exponent. The mutual information is based on the concept of uncertainty and related as follows:

$$I(k)=H(s)+H(s+k)-H(s, s+k) \quad (2)$$

where

$$H(s)=-\sum_i P(i)\log_2[P(i)] \quad (3)$$

and

$$H(s+k)=-\sum_i P(j)\log_2[P(j)] \quad (4)$$

$$H(s, s+k)=-\sum_{i,j} P(i, j)\log_2[P(i, j)] \quad (5)$$

Kolmogorov entropy is similar to the mutual information function except that it takes into account many pervious measurements instead of only one (mutual information function).

EXPERIMENT

A schematic diagram of the experimental apparatus and agitator is shown in Fig. 1(a) and (b). The lab-scale agitation fluidized bed granulator consists of a main bed, an agitator, a binary spray nozzle, a distributor, and an instrument for collecting the pressure signal data. The experiments are carried out in an agitation fluidized bed granulator equipped with binary spray nozzle. The main body is composed of the upper tapered cone vessel of height of 0.25 m and lower cylindrical vessel of 0.1 m in diameter and 0.25 m in depth. They are made of acrylic resin for visual confirmation. It is also designed to investigate the pressure fluctuations and flow patterns of bed materials as a function of gas fluidization velocity. A sintered plate distributor is used. Fluidizing air for the granulation was fed to the granulator from a compressor through the distributor. The agitator rotates at the center of bed to give more compaction and stirring motion to granules.

Fluctuations in dynamic pressure in the bed are measured using a differential pressure transducer (Validyne Model P24) which produces an output voltage proportional to the pressure fluctuations. The voltage-time signals are stored in an IBM PC and computed for spectral (power spectrum and autocorrelation) and chaos (average mutual information and attractor) analyses.

To reduce the error in the analyses, total acquisition time of 40.96s is chosen for each experiment. In this way, the time series with N=8,192 points are obtained.

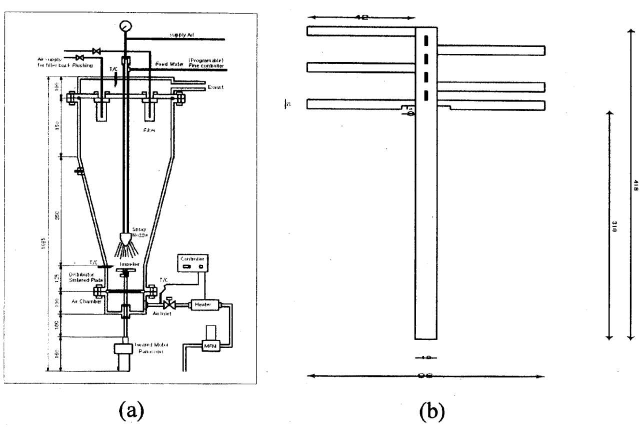

Fig. 1. A schematic diagram of (a) experimental apparatus and (b) agitator.

RESULTS AND DISCUSSION

In Table 1, average particle sizes of raw materials used in granulation are listed. Particle size distributions of these particles are shown in Fig. 2. These particles are classified in a Geldart C group. The particles in Geldart C are cohesive fine powders. The normal fluidization of the particles is extremely

Table 1. Average particle sizes of primary particles

	ZnO	TiO_2	Bentonite
Average particle size [μm]	1.3	0.3	3.9

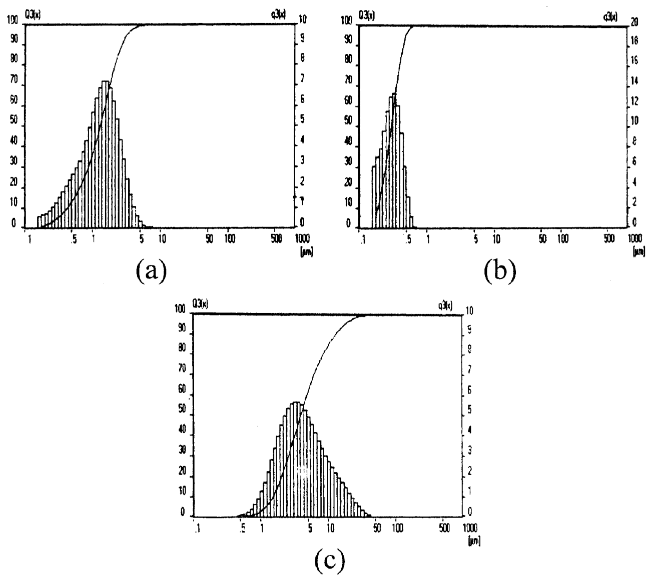

Fig. 2. Particle size distributions of (a) ZnO, (b) TiO_2, and (c) bentonite.

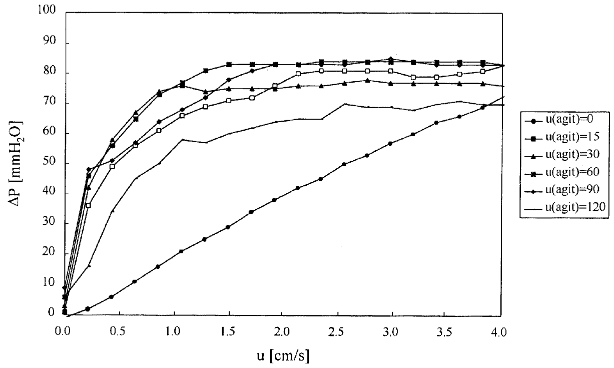

Fig. 3. Pressure drop as a function of fluidizing velocity at various agitator speeds.

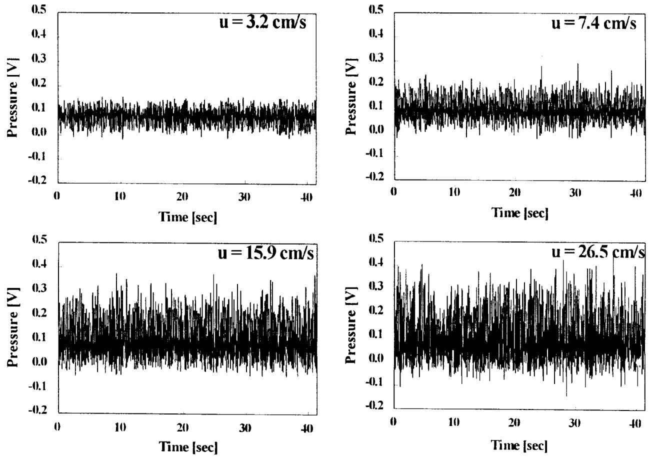

Fig. 4. Different pressure signals in terms of air velocity.

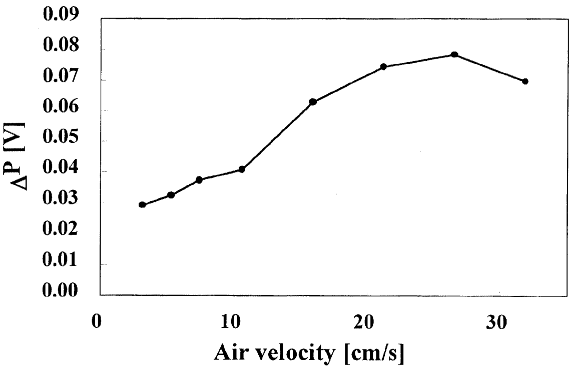

Fig. 5. Average absolute deviation as a function of air velocity.

difficult since the interparticle forces between the particles are greater than the forces by fluidizing air.

In Fig. 3, the change of pressure drop in an agitation fluidized bed is shown as a function of fluidzing velocity at various agitator speeds. Without agitation, the pressure drop increases linearly as the fluidizing velocity increases, implying that the fine particles are not fluidized even with increasing fluidizing velocity. With a rotating agitator blade (15-90 rpm), the pressure drop increases and reaches a constant as the fluidizing velocity increases, meaning that the fine particles are considered to come to a fluidization state. This is confirmed visually. At higher agitator speed (120 rpm), AFBG is in the fluidization state. However the pressure drop exhibits a little different trend from those of relatively low agitator speeds, which is presumably due to the centrifugal force by the agitator.

In Fig. 4 four different pressure signals are plotted as a function of air velocity. It is seen that the magnitude of the pressure signal increases with gas velocity. It is obvious that the increase in gas velocity gives more motion to particles and causes larger fluctuations in pressure signals. The trend for the average absolute deviation is shown in Fig. 5. The average deviation of the signal increases as the air velocity increases. As the air velocity increases further, the average deviation of the signal reaches a maximum and decreases. This is due to the change of fluidization regimes from bubbling fluidization to turbulent flui-

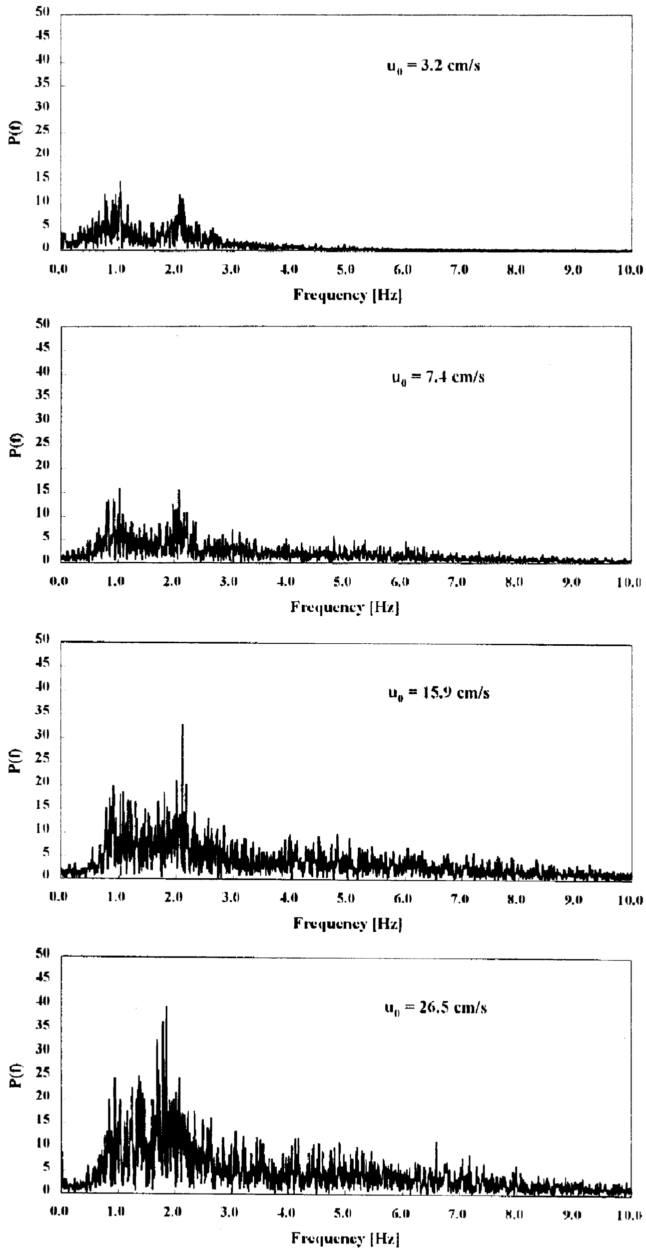

Fig. 6. Power spectral density functions of pressure drop at four selected fluidizing conditions.

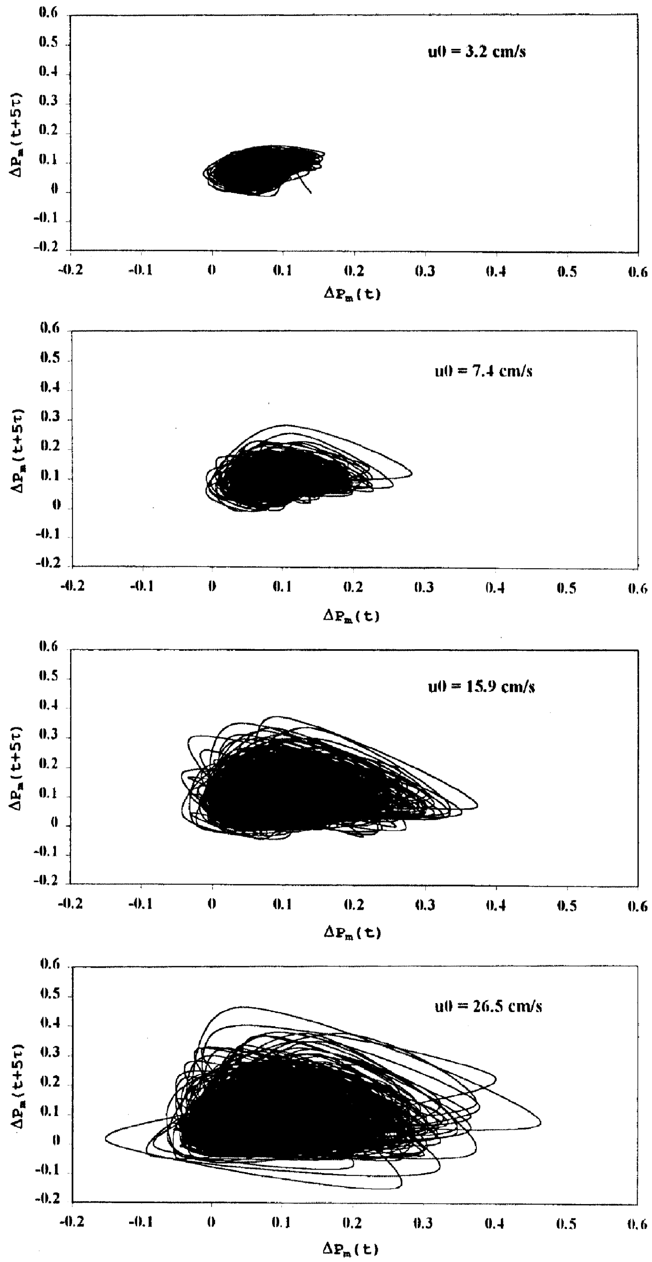

Fig. 7. Attractor plots of pressure drop at four selected fluidizing conditions.

dization.

In Fig. 6 Fourier power spectra for the pressure drop of a granulated sorbent (100-300 μm) are shown at four different fluidizing velocities. It is seen that the spectra are characterized by a couple of major frequencies between 1 and 2 Hz and a broad band around major peaks stretching up to 4-5 Hz.

The attractor projections are plotted in Fig. 7 as a function of the fluidizing velocity with the value, time delay T, calculated from average mutual information [Woo et al., 1998]. The plots are 2-dimensional projections of a higher dimensional state space on the same scale. Points are linked to guide the eye. The number of points plotted are 8,192 corresponding to 40.96 s of measuring time. The overall size of the attractor increases as fluidizing velocity increases. The pressure fluctuations increase with

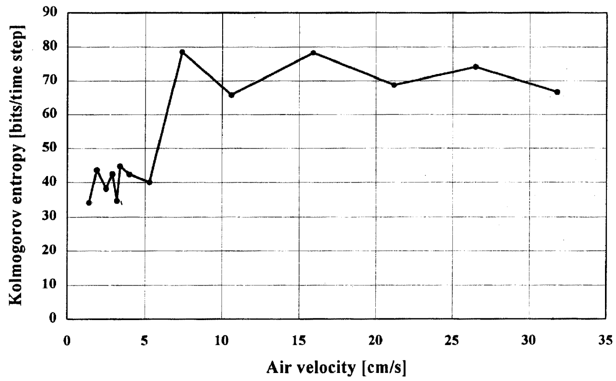

Fig. 8. Kolmogorov entropy as a function of air velocity.

increasing gas velocity leading to increasing bubble size.

In Fig. 8 typical Kolmogorov entropy in a fluidized bed is plotted as a function of air velocity. It is seen that the value of Kolmogorov entropy increases abruptly as the fludization regime changes to bubbling fluidization. It may be possible to employ the characteristic for the control of fluidized beds.

CONCLUSIONS

A technology is developed to manufacture sorbent for the removal of H_2S using a laboratory scale AFBG instead of conventional methods such as pan-granulation or spray drying. For the granulation, a method is presented for fluidized fine particles (0.3-3.9 μm in size), classified in Geldart C group, in a fluidized bed by installing an agitator blade on the lower part of a fluidized bed granulator. The ordinary fluidization of the fine particles is known to be very problematical since the interparticle forces between the particles are greater than the forces by fluidizing air.

Three sorts of time series analysis, i.e., statistical analysis, spectral analysis, and chaos analysis, of the pressure fluctuations in a fluidized bed are performed to identify the hydrodynamics of a fluidized bed. It is found that characteristic numbers in chaos analysis (carried out in the state space), especially Kolmogorov entropy representing the rate of generation of information, can be used to classify the transition regime in a fluidized bed.

NOMENCLATURE

f : frequency [1/s]
h(t) : the wave form to be decomposed into a sum of sinusoids
H(f) : the Fourier transform of h(t)
H(s) : average unconditional uncertainty
I(k) : mutual information
k : time steps
n : number of data
p(t) : pressure at time t
P(i) : the probability of the measurement falling in the *i*th bin
P(i, j) : the joint probability that measurement s falls in bin i and measurement q after k time lags falls in bin j
T : time delay
t : time [s]

u : superficial gas velocity [cm/s]
$\bar{x}$: average of data

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