

# Chaotic Behavior of Gas-Solids Flow in the Riser of a Laboratory-Scale Circulating Fluidized Bed

Antonio Marzocchella

Dipt. di Ingegneria Chimica, Università degli Studi di Napoli "Federico II,"  
Istituto di Ricerche sulla Combustione-CNR, 80125 Naples, Italy

Robert C. Zijerveld, Jaap C. Schouten, and Cor M. van den Bleek

Faculty of Chemical Technology and Materials Science, Delft University of Technology, 2628 BL Delft, The Netherlands

*A cold model of a circulating fluidized bed with a 0.030-m-ID, 2.77-m-high riser was operated in a wide range of operating conditions. Several solids were tested, from 57  $\mu\text{m}$  to 1,830  $\mu\text{m}$  in size and from 1,100  $\text{kg}/\text{m}^3$  to 2,540  $\text{kg}/\text{m}^3$  in density. Pressure fluctuations were measured at several points along the riser, and time series were processed to evaluate chaotic invariants (Kolmogorov entropy and correlation dimension). Axial profiles of average values of pressure and voidage were also evaluated. At fixed operating conditions, the Kolmogorov entropy changed along the riser, which appeared to be a function of the local voidage and showed a minimum when voidage decreases from 1.00 until about 0.90. Changes of the Kolmogorov entropy with local voidage were interpreted based on interactions among solids and gas turbulence structures. Three regions were identified in the voidage range investigated: particles-controlled region, clusters-controlled region, and bottom-bed-controlled region.*

## Introduction

The dynamics of gas-solids suspensions in the riser of a circulating fluidized bed (CFB) are particularly important in the control, design, and scale-up of efficient new reactors. In the past, researchers have focused their attention on the time-averaged behavior of gas-solids suspensions in the riser and have shown that a considerable solids segregation exists both in axial and radial direction (Kunii and Levenspiel, 1991). Only in recent years, have they studied the time-dependent behavior of these suspensions, evaluating characteristic dynamics (Dry and Christensen, 1988; Schnitzlein and Weinstein, 1988; Arena et al., 1989, 1992; Marzocchella, 1992). In particular, researchers have identified flow structures that alternate at a nearly constant frequency, provided that a fast fluidization regime was established in the riser. In addition, some authors have developed models that consider the dynamic nature of the two-phase suspension flow. Tsuo and Gidaspow (1990) have developed a model that describes the hydrodynamic behavior of a riser operated under a wide range of conditions and have pointed out the dynamic behav-

ior of this system. Others have developed models considering the role of the interactions between particles and gas turbulent structures (Zethraeus et al., 1992; Bolio and Sinclair, 1995). Latter models dealt with dilute suspensions of small particles for which the damping effect on the turbulence is known (Hetsroni, 1989; Gore and Crowe, 1989). Unfortunately, information about the effect of concentration of small particles on the turbulence level is available up to a limited value of concentration (such as up to volumetric fractions of 0.005). This is due to the fact that reliable diagnostic techniques adopted to carry out this kind of investigation are not intrusive (such as the LDV technique), and generally at higher solids concentration these measurement apparatus are obscured by the amount of solids present in the pipe. Therefore, models cannot be verified over the wide range of flow regimes under which CFB risers may be operated.

Stringer (1989) suggested that a fluidized bed might be interpreted as a *chaotic* system, that is, as a system governed by nonlinear interactions between the system variables. Due to this nonlinearity, these deterministic systems are sensitive to small changes in initial conditions and are, therefore, charac-

Correspondence concerning this article should be addressed to A. Marzocchella.

terized by a limited predictability. The peculiarity of chaotic systems is that their dynamics are fully represented by the attractor in the phase space (Moon, 1992; Hilborn, 1994). The characteristics of an attractor can be estimated from the time series of only one of the system's characteristic variables (such as pressure fluctuations in gas-solids fluidized beds) via a technique called (attractor) reconstruction (see also van den Bleek and Schouten, 1993). Following Stringer's suggestion, several studies have been carried out to characterize the dynamic behavior of gas-solids suspensions operated under captive regimes of fluidization by means of chaotic invariants (Daw and Halow, 1991; Schouten et al., 1992; Hay et al., 1995). These studies have shown that chaos analysis can be used for a quantitative characterization of transitions between different states of fluidization. Some of these studies have shown that the Kolmogorov entropy (a chaotic invariant that gives a measure of the predictability of the system and is large for very irregular dynamic behavior, small in the case of more regular, periodic-like behavior and zero for completely periodic systems) is a dependent variable that is sensitive to changes in operating conditions. Only recently, chaotic analysis has been applied to characterize gas-solids suspensions operated under transport regimes of fluidization (van der Stappen et al., 1993b; Marzocchella et al., 1994; Bai et al., 1996; Zijerveld et al., 1996).

The physical meaning of the Kolmogorov entropy has been discussed in the case of single-phase systems. For example, Gaspard and Wang (1993) have demonstrated that the Kolmogorov entropy provides a unified quantitative measure of the dynamical state of a system in both chaotic as well as stochastic processes. These authors have pointed out that an entropy estimation provides a quantity to detect transitions between dynamical states of different degrees of chaos or randomness when a parameter of the system is varied. Further, Gaspard and Wang have shown that the entropy may be used to characterize different regimes of hydrodynamic turbulence. The relationship between the Kolmogorov entropy and flow order may also be inferred from data reported by Haucke et al. (1985). These authors have calculated the Kolmogorov entropy of the single-phase flow in a Rayleigh-Bernard cell by measuring the time series of the temperature: it appeared that the entropy increases moving from periodic to turbulent flow. Some attempts have been made to relate the Kolmogorov entropy (evaluated from the time series of pressure fluctuations, solids concentration, and so on) to hydrodynamic behavior of fluidized beds operated under captive regimes. It has been shown that the Kolmogorov entropy changes when the quality or state of the fluidization changes (Fuller et al., 1993; Skrzyzke et al., 1993; van der Stappen et al., 1993a, 1995). In particular, it was found that the Kolmogorov entropy has a minimum when the bed is operated under slugging regime, that is, when a regular behavior establishes.

## Objective of This Work

The objective of this work is to investigate and to provide a first attempt to interpret the chaotic behavior of gas-solids suspensions in the riser of a laboratory-scale CFB operated in a wide range of experimental conditions. The solids distribution in the riser has been characterized in terms of axial

profiles of time-averaged voidage. The main assumptions made in the present investigation are:

- Pressure fluctuations at a certain level along the riser are not affected by far dynamics and are constant throughout the riser section, that is, pressure fluctuations measured at the riser wall are considered to be characteristic of the suspension dynamic present in a limited riser length (generally about 0.50 m) around the measurement point;
- Radial solids distribution has been neglected, that is, the gas-solids suspension has been characterized by means of voidage averaged over both time and limited length of the riser (indicated in this article as *voidage*).

Under the above assumptions, the variation of the Kolmogorov entropy with the voidage in the range of operating conditions tested has been interpreted on the basis of known effects of particles on gas turbulence in vertical pipe flows. This study has been focused on the comparison of the observed variation of Kolmogorov entropy with known relationships between turbulence related variables and voidage, rather than on the meaning of the change of the Kolmogorov entropy in relation to its definition of predictability of dynamical state of the system. In the next section a short summary of particles effects on gas turbulence is reported (for more thorough reviews, see Hetsroni, 1989; Tsuji, 1991).

## Note on particles effects on gas turbulence

The influence of particles on turbulent fluid flow depends on particle characteristics, operating conditions, and pipe size. Depending on the values of these variables, particles may behave as a tracer, damp turbulence, or even enhance it.

A particle behaves as a tracer when it moves at the local fluid velocity without appreciable relative motion. According to Clift and Gauvin (1970), tracer particles should fulfill the criteria

$$\frac{d_p}{\eta} \ll 1 \quad (1)$$

$$Re'_p = \frac{d_p u'}{\nu_g} \ll 1 \quad (2)$$

$$\vartheta_p \ll \vartheta_k \quad (3)$$

where  $d_p$  is the particle diameter (m),  $\eta$  the Kolmogorov length scale of turbulence (m),  $u'$  the instantaneous component of velocity fluctuation in average flow direction (m/s),  $\nu_g$  the gas kinematic viscosity ( $\text{m}^2/\text{s}$ ),  $Re'_p$  the particle Reynolds number based on  $u'$ ,  $\vartheta_p$  the particle relaxation time (s) (the time taken by a particle to adjust to a change in gas velocity), and  $\vartheta_k$  the Kolmogorov time (s).

Larger particles do not behave as a tracer, but interact with flow fluctuations. The turbulence level in a vertical pipe flow is damped by the presence of "small" particles when particles characterized by  $\vartheta_p \leq \vartheta_k$  have also low particle Reynolds number based on the slip velocity ( $Re_p$ ) (Hetsroni, 1989). In particular, Eq. 4 should hold

$$Re_p = \frac{d_p(u_g - u_p)}{\nu_g} \leq 110 \quad (4)$$

where  $u_g$  and  $u_p$  are the effective fluid velocity and particle velocity (m/s), respectively. Under this condition, the gas velocity fluctuations are decreased by the particles in the ratio (Owen, 1969)

$$\left[ 1 + \frac{\rho_s(1-\epsilon)}{\rho_g} \frac{\vartheta_k}{\vartheta_p} \right]^{-1/2} \quad (5)$$

$\rho_g$  is the gas density (kg/m<sup>3</sup>). Larger particles on the other hand enhance turbulence and Hetsroni (1989) proposed  $Re_p = 400$  as a lower limit for this kind of behavior. Under these circumstances the intensity of turbulence scales up with the ratio

$$\beta \frac{(1-\epsilon)}{\epsilon} \quad (6)$$

that is, with increasing solids concentration in the two-phase flow, where the  $\beta$  factor is an unknown function of the particles size.

A rough parameter to establish whether particles damp or enhance turbulence was given by Gore and Crowe (1989). The ratio between the particle diameter and the characteristic length of the large eddies,  $\ell_e$  (m) was found to be a critical parameter to estimate whether the relative turbulent intensity of the fluid flow is increased or decreased by particles addition. Provided that particles are large enough to interact with flow fluctuations, particles damp turbulence if Eq. 7 holds

$$\frac{d_p}{\ell_e} < 0.1 \quad (7)$$

Two-phase particles flow systems characterized by a value of the ratio higher than 0.1 show turbulence enhancement.

The phenomenology of particle-turbulence interactions suggests that knowledge of length and time scales in a turbulent system is essential to predict how the particles affect a turbulent flow. In the following, some relationships to evaluate relevant variables are mentioned.

Hutchinson et al. (1971) have found that the ratio between the size of the large eddies and the pipe diameter  $D_r$  (m) was almost constant throughout the pipe section under a wide range of operating conditions. They found

$$\frac{\ell_e}{D_r} = 0.11 \quad (8)$$

The Kolmogorov length scale  $\eta$  (m) is related to the kinematic viscosity of the fluid and to the dissipation rate per unit mass  $\alpha$  (m<sup>2</sup>/s<sup>3</sup>) and it can be evaluated by means of the following relationship set (Tennekes and Lumley, 1974)

$$\eta = \left( \frac{\nu_g^3}{\alpha} \right)^{1/4} \quad (9)$$

$$\alpha = \frac{u_*^3}{\ell_e} \quad (10)$$

where  $u_*$  (m/s) is the *friction velocity* that may be evaluated, as a first approximation, by means of

$$\frac{U_g}{u_*} = 2.5 \ln \left( \frac{D_r u_*}{2 \nu_g} \right) + 1.5 \quad (11)$$

In the present work, the values of both particle relaxation time and Kolmogorov time are estimated by means of the equations proposed by Owen (1969) to be coherent with the limit he found between particles that damp and enhance turbulence. The relaxation time for small particles may be evaluated assuming Stokes' law of resistance

$$\vartheta_p = \frac{\rho_s d_p^2}{18 \rho_g \nu_g} \quad (12)$$

For larger particles,  $Re_p > 1$ , Eq. 12 overestimates the relaxation time. Nevertheless, it has been used for the particles tested in the present investigation. Following the approximation of Owen (1969), the Kolmogorov time is of the order of

$$\vartheta_k \approx \frac{D_r}{u_*} \quad (13)$$

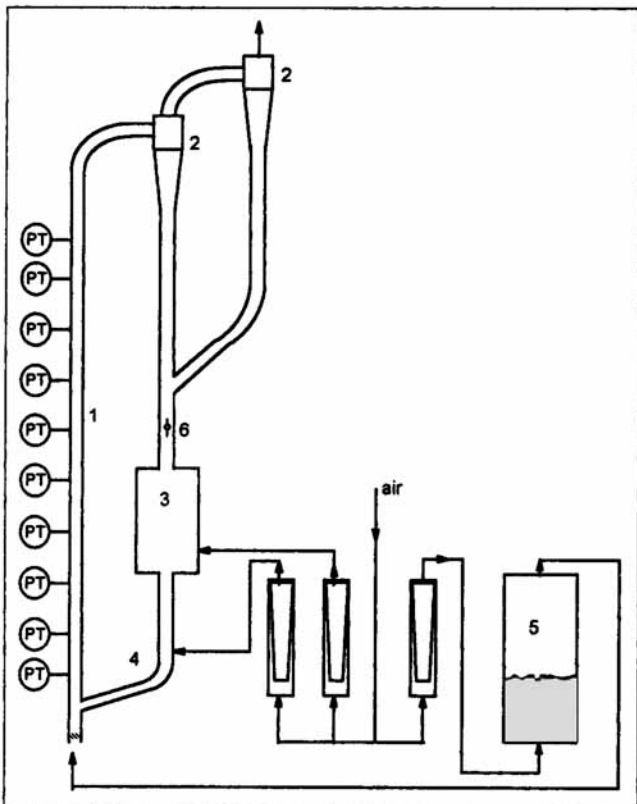
## Experimental

### Apparatus

A circulating fluidized-bed glass model, located at Delft University of Technology was used. Characteristics of the loop (Figure 1) are listed in Table 1. Basically, it consists of a 0.030-m-ID, 2.77-m-high riser, a solids collecting system, a 0.105 m standpipe and an L-valve. Gas and solids were separated in the collecting section by means of a set of two cyclones: the first at medium efficiency and the second at high efficiency. Air fed at the standpipe bottom helped in maintaining the descending solids in a state of gentle fluidization. Solids were reinjected into the riser by means of a 0.020 m pipe and the flow rate was controlled by means of the L-valve. Air distributors at the bottom of both the riser and the recirculation column consisted of porous plates. Air forced at the bottom of the riser was properly humidified.

Solids flow rates were measured by means of a butterfly valve located in the pipe above the recirculation column (Figure 1). When the valve is open, the solids fall freely in the recirculation column; when the valve is closed, the solids accumulate in the pipe. The solids mass flux ( $G_s$ ) (kg/sm<sup>2</sup>) was evaluated by means of the relationship  $G_s = h_1 \rho_s (1 - \epsilon_b) / t_m$  where  $\rho_s$  is the particle density (kg/m<sup>3</sup>),  $t_m$  is the valve closing time (s),  $\epsilon_b$  is the voidage of solids accumulated above the valve, and  $h_1$  is the level of solids in the pipe above the valve at the time  $t_m$ . The value of  $\epsilon_b$  has been measured during calibration tests and errors of solids mass flux due to possible fluctuations around the average value of  $\epsilon_b$  are within 5%.

Pressure taps along the riser were spaced uniformly at 0.25 m. Probes to measure pressure at the riser wall were made of 4-mm-ID, 25-cm-long tube equipped at the tip with a 54  $\mu$ m mesh. The tube length has been chosen to be short enough to prevent time delays; moreover, experiments carried out with



**Figure 1. Circulating fluidized-bed unit.**

(1) 0.030 m ID riser; (2) cyclones; (3) recirculation column; (4) solids control L-valve; (5) humidifier; (6) butterfly valve to measure solids mass flux. PT—pressure taps.

different tube lengths showed that the signal does not change significantly (vander Stappen, 1996). The size of the mesh was small enough to prevent probe blocking by particles and large enough to avoid damping of pressure fluctuations. Water manometers equipped with a damping system were used to measure time-averaged pressure profiles along the riser. Piezoelectric pressure transducers (KISTLER mod. 5011), used to measure pressure fluctuations with respect to their average value, were interfaced with a DIFA 16 bits data acquisition system. Sampling frequency was between 800 and 1,000 Hz and the low-pass frequency was always set at values lower than the Nyquist frequency. Transducer drift did not affect the results since a high-pass filter at 0.03 Hz was used.

### Operating conditions

Properties of the materials tested are reported in Table 2. The Geldart classification group, the Archimedes number of

**Table 1. Characteristics of CFB Loop**

Riser	
Cross-section shape	Circular
Cross-section size	0.030 m ID
Height, m	2.77
Recirculation Column	
ID, m	0.105
Solids Reinjection Pipe	
ID, m	0.020
Solids Circulation Control Device	
	L valve

**Table 2. Characteristics of Solids Used**

Material	Ballotini	FCC	Sand	Polystyrene
Particle-size range, $\mu\text{m}$	40–90	30–190	50–500	420–2,500
Particle Sauter mean dia., $\mu\text{m}$	57	85	260	1,830
Particle density, $\text{kg}/\text{m}^3$	2,540	1,690	2,500	1,100
Terminal velocity, m/s	0.20	0.32	1.9	6.1
$Re_t$	0.9	1.8	37	800
Min. fluidization velocity, m/s	0.004	0.006	0.06	0.57
Archimedes no.	18	41	1,800	264,000
Geldart classification group	A	A	B	D
Particle relaxation time, s	$3 \times 10^{-2}$	$4 \times 10^{-2}$	$5 \times 10^{-1}$	$1 \times 10^1$

particles, the particle terminal velocity ( $V_t$ ), the Reynolds number based on the particle terminal velocity  $Re_t$ , and the particle relaxation time (the time taken by a particle to adjust to a change in gas velocity) are also indicated in the table.

Table 3 reports operating conditions at which runs were carried out. Experiments with sand and with polystyrene were carried out at superficial gas velocity ( $U_g$ ) (m/s) equal to 3 and 7 m/s, respectively, since at lower velocities the turbulent regime established.

The Reynolds number based on the riser diameter  $D_r$ ,  $Re_r = \rho_g D_r U_g / \mu_g$ , was  $4.3 \times 10^3$ ,  $6.5 \times 10^3$  and  $1.5 \times 10^4$  at  $U_g$  equal to 2, 3, and 7 m/s, respectively. Therefore, turbulent flow was established in the riser at least under the condition  $G_s = 0$ . Table 4 reports values of variables characteristic for the turbulence level in the riser. The Kolmogorov length scale was evaluated by means of Eqs. 9, 10 and 11. The Reynolds number based on velocity fluctuations was estimated by assuming that the instantaneous component of the velocity fluctuation  $u'$  equals to the friction velocity  $u_*$ . The Kolmogorov time  $\vartheta_k$  is also reported. The large eddy size, evaluated by means of Eq. 8, is 3.3 mm.

### Experimental procedure

Time-averaged voidages  $\epsilon$  were determined by means of pressure drops  $\Delta P$  (Pa) measured at successive pressure taps. Acceleration effects and friction effects at the riser walls were neglected. The relationship  $\Delta P = \rho_s g(1 - \epsilon)\Delta z$  was used with  $g$  and  $z$  being the acceleration due to gravity ( $\text{m}/\text{s}^2$ ) and the riser axial coordinate (m), respectively. It should be pointed out that when the riser diameter is as small as that of the Delft University facility and the plant is operated at a high gas velocity with large and heavy particles, voidages are underestimated since wall friction effects are not completely negligible (Arena et al., 1988). However, errors may be con-

**Table 3. Operating Conditions**

Material	$U_g$ m/s	$U^*$	$G_s$ $\text{kg}/\text{sm}^2$	Solids Inventory kg
FCC—85 $\mu\text{m}$	2	3.4	3–32	1.5
	3	4.1	8, 20	
Ballotini—57 $\mu\text{m}$	2	3.1	15–120	2.0
	3	4.7	15–110	
Sand—260 $\mu\text{m}$	3	4.6	20–100	2.0
Polystyrene—1,830 $\mu\text{m}$	7	12	22, 75	1.2

**Table 4. Variables Related to Turbulence Level in Riser**

$U_g$ m/s	$u_*$ m/s	$Re'$				$\eta$ $\mu\text{m}$	$\vartheta_k$ s
		Ballotini	FCC	Sand	Polystyrene		
2	0.14	0.57	0.85	—	—	250	$1 \times 10^{-1}$
3	0.21	0.86	1.3	3.9	—	180	$7 \times 10^{-2}$
7	0.42	—	—	—	54	100	$4 \times 10^{-2}$

sidered negligible when the riser is operated at superficial gas velocities as low as 2 and 3 m/s.

A rough estimation of the air-polystyrene suspension voidage in the riser was carried out by using the relationship

$$G_s = \rho_s(1 - \epsilon)V_s \quad (14)$$

where the solids velocity  $V_s$  (m/s) was assumed as an approximation to be

$$V_s = U_g - V_t \quad (15)$$

Pressure fluctuations were processed by means of the RRCHAOS software package (Schouten and van den Bleek, 1993) to estimate chaotic invariants. This package computes correlation dimension and Kolmogorov entropy based on maximum-likelihood estimation methods (Schouten et al., 1994a,b) and it calculates various statistical values as well. In this article, results of correlation dimension ( $D_2$ ), Kolmogorov entropy ( $K$ ) (bits/s), and average absolute deviation ( $\Delta$ ) are reported. The latter statistical variable is a robust estimator of the data's width around the mean and is computed from:

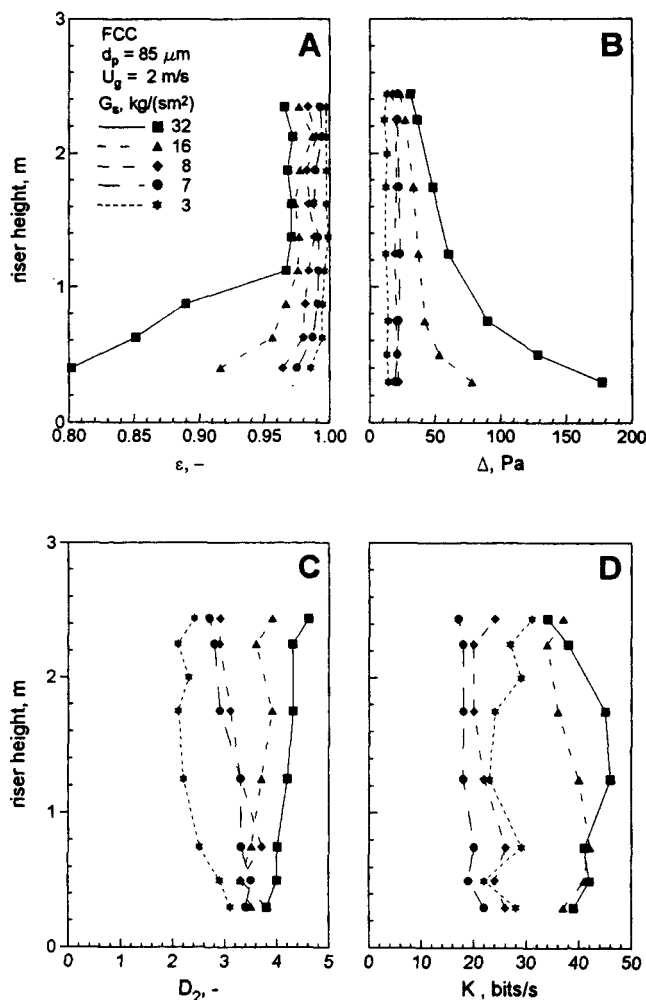
$$\Delta = \left( \sum_{i=1}^N |x_i - \bar{x}| \right) / N,$$

where  $\bar{x}$  is the mean of the  $x_i$  time series. ( $\bar{x} = (\sum_{i=1}^N x_i) / N$ )  $N$  is the number of data in a time series. Power spectral density of the pressure fluctuations time series were evaluated by means of the DIFA acquisition system.

For each set of operating conditions (material,  $G_s$  and  $U_g$ ), axial profiles of pressure and voidage as well as pressure fluctuations were measured along the riser. Generally, two runs were carried out with each material at each value of the superficial gas velocity and solid mass flux. During each run pressure fluctuations were measured at four different taps along the riser. Chaotic invariants were evaluated from four successive parts of a time series, each part 5 min long. Within each run, the variation of chaotic invariants evaluated at each level was below 8% with respect to the mean values.

## Results and Discussion

The investigations on the 0.030 m ID riser were conducted to point out the dynamic behavior of gas-solids suspensions operated in a relatively wide range of hydrodynamic flow regimes. Ballotini, FCC, and sand could be operated from the dilute transport up to the onset of the fast fluidization regime. Large polystyrene particles were operated only under



**Figure 2. Axial profiles of time-averaged voidage (A); average absolute deviation (B); correlation dimension (C); and Kolmogorov entropy (D).**

dilute transport regimes. The interest in the latter solids was to examine peculiar dynamic characteristics of the two-phase flow with large particles. Results of the investigations are reported in the following; their interpretation is discussed on the basis of the solids-turbulent structures interactions.

Data reported in Figure 2 refer to 85- $\mu\text{m}$  FCC operated at  $U_g = 2 \text{ m/s}$ . Figure 2a shows axial profiles of time-averaged voidages at different solids mass fluxes. As expected, the higher  $G_s$  the lower the voidage along the riser. In particular, at the highest solids mass flux the onset of an S-shaped voidage axial profile, which is typical of the fast fluidization regime (Kunii and Levenspiel, 1991), can be recognized. Figures 2b, 2c, and 2d report axial profiles of the average absolute deviation, the correlation dimension and the Kolmogorov entropy, respectively. The average absolute deviation decreased upon a decrease of the solids mass flux and moving upwards along the riser. The correlation dimension changed between 2 and 5, that is, more than one order of magnitude lower than the embedding dimension used to evaluate the chaotic invariants (van den Bleek and Schouten, 1993). Values of  $D_2$  are in the same range as that evaluated by vander Stappen et al. (1993b) operating a large-scale CFB

riser ( $0.8 \times 1.2$  m) at a superficial gas velocity equals to 3.3 and 4.5 m/s and at low solids mass fluxes ( $< 12$  kg/sm<sup>2</sup>) with the same 260  $\mu$ m sand used in the present work. Observing the overall change in the Kolmogorov entropy in Figure 2d, it appears that when the solids mass flux increased the Kolmogorov entropy first decreased and then increased. Values of Kolmogorov entropy are higher than those found by van der Stappen et al. (1993b). They have found that the Kolmogorov entropy decreased throughout the riser when the solids mass flux increased in the range of operating conditions tested.

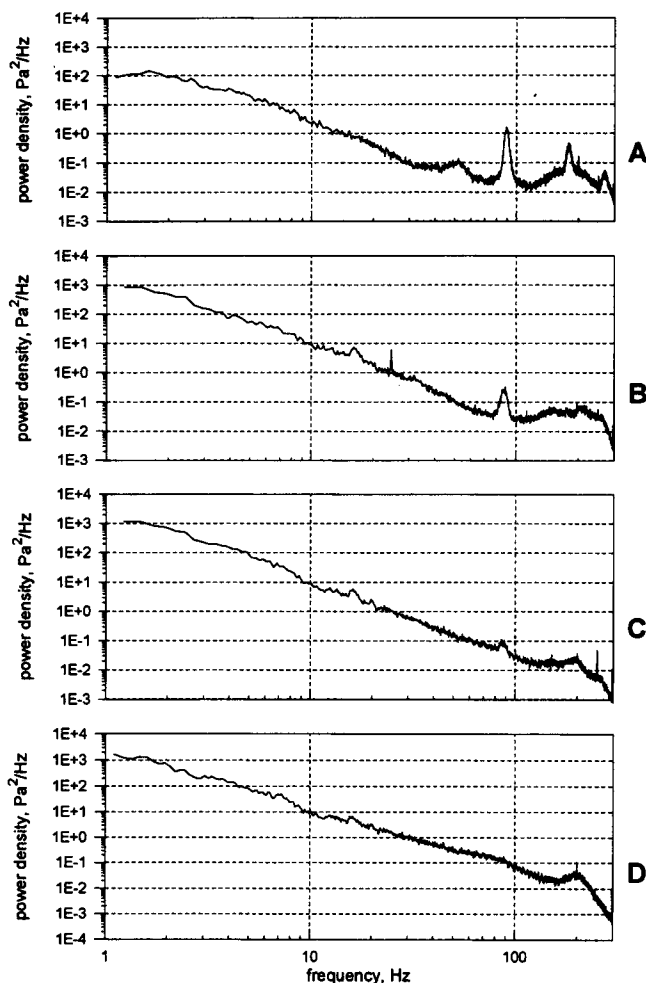
Results obtained with Ballotini and sand at different operating conditions (that is, at superficial gas velocity and solids mass flux) were similar to those described for 85  $\mu$ m FCC operated at  $U_g = 2$  m/s.

The average voidage in the riser during runs carried out with polystyrene was about 0.98 and 0.93 at  $G_s = 22$  and 75 kg/sm<sup>2</sup>, respectively. Measurements of pressure fluctuations provided a time series by means of which it was not possible to evaluate reliable values of the Kolmogorov entropy since a too low number of points per average cycle was found (of the order of 10 points per average cycle). The value of the Kolmogorov entropy was very high ( $> 200$ –300 bits/s), which is an indication that the time series is very erratic and random-like. This is confirmed by the value of the correlation dimension which was of the order of magnitude of the embedding dimension used.

Power spectral density of pressure fluctuations are reported in Figure 3. They were evaluated during runs with Ballotini at  $U_g = 3$  m/s and refer to different operating conditions. At low solids concentrations (Figures 3a, 3b, and 3c), the increase of the power density at about 90 Hz and 180 Hz may be due to resonance phenomena (fundamental and second harmonic, respectively) of the Delft University facility. It can be seen that its power density decreased when solids concentration increased. Analyzing this figure, it appears that the power density of high frequencies decreased progressively upon an increase of solids concentration. At higher values of solids concentration (Figure 3d), the power density increased throughout the frequency spectrum.

Figure 4 shows the average absolute deviation, correlation dimension and Kolmogorov entropy as a function of the local voidage measured under the same operating conditions of Figure 2. Riser levels at which the data were evaluated are also indicated. The average absolute deviation (Figure 4a) increased monotonously when the voidage decreased whatever the riser level is. The correlation dimension (Figure 4b) increased when voidage decreased and appeared to level off at a value of about 4 at lower voidages independently from the riser level. The Kolmogorov entropy decreased when the voidage decreased, reached a minimum, and then increased. Decreasing the voidage even further, the Kolmogorov entropy leveled off at about 42 bits/s; the value of voidage at which this occurred depended on the riser level.

The diagrams reported in Figures 5 through 7 are similar to those reported in Figure 4 and refer to runs carried out with Ballotini (Figures 5 and 6) and sand (Figure 7). The average absolute deviation and the correlation dimension appeared to change when the voidage changed in a way similar to that measured during runs carried out with FCC. Generally, the correlation dimension increased upon a decrease in



**Figure 3. Power spectral density evaluated from time series measured during runs with Ballotini at  $U_g = 3$  m/s.**

Frequency analysis parameters: 8,192 points per spectrum; sampling frequency 1,000 Hz; low-pass frequency 333 Hz; Hanning window; number of spectra averaged 40. (A)  $G_s = 10$  kg/(sm<sup>2</sup>),  $z = 1.8$  m,  $\epsilon = 0.99$ ,  $K = 51$  bits/s; (B)  $G_s = 13$  kg/(sm<sup>2</sup>),  $z = 2.3$  m,  $\epsilon = 0.98$ ,  $K = 36$  bits/s; (C)  $G_s = 13$  kg/(sm<sup>2</sup>),  $z = 1.8$  m,  $\epsilon = 0.97$ ,  $K = 17$  bits/s; (D)  $G_s = 13$  kg/(sm<sup>2</sup>),  $z = 0.50$  m,  $\epsilon = 0.96$ ,  $K = 18$  bits/s.

voidage and leveled off at a value between 4 and 5 for all the operating conditions tested. The dependency of the Kolmogorov entropy upon a decrease in voidage appeared to be the same as that described for measurements carried out with FCC at  $U_g = 2$  m/s. The minimum value of the Kolmogorov entropy  $K_{\min}$  is about 19 bits/s under all operating conditions tested and the voidage at which this occurs  $\epsilon_{K_{\min}}$  changes when the operating conditions change. Table 5 reports values of the  $\epsilon_{K_{\min}}$  as well as the limit value of the Kolmogorov entropy  $K_{\text{const}}$  (bits/s) upon a further decrease in voidage.

The trend of the average absolute deviation when voidage changed is as expected. Upon a decrease of voidage, the amount of solids in the riser increased and therefore pressure fluctuations, which are related to the solids holdup, increased as well. The values of average absolute deviation measured during runs carried out with FCC were lower with respect to those measured with other materials since at con-

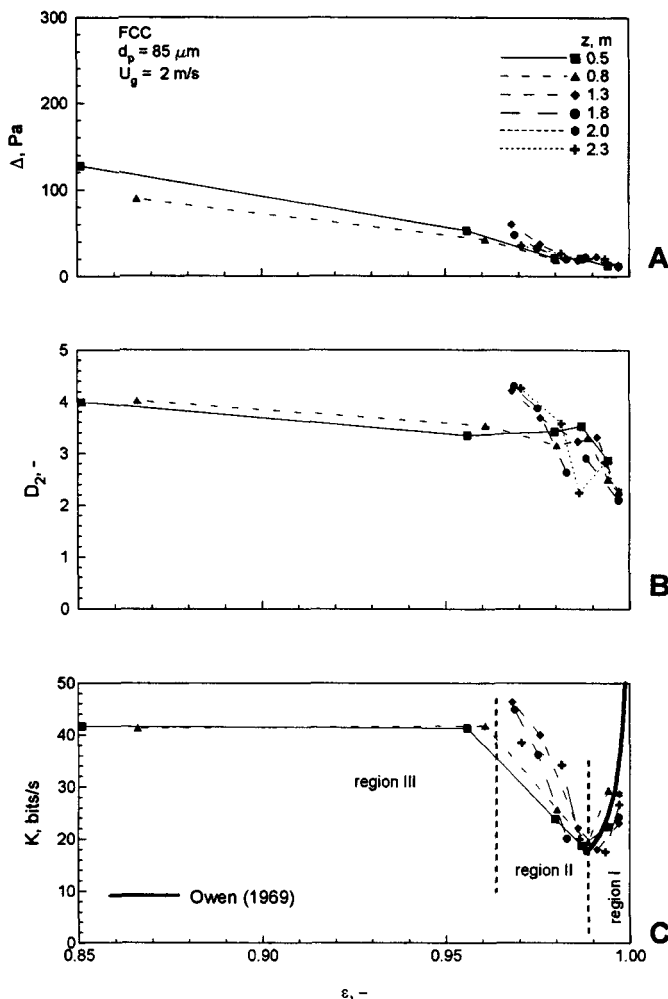


Figure 4. Average absolute deviation (A); correlation dimension (B); and Kolmogorov entropy (C) as a function of voidage.

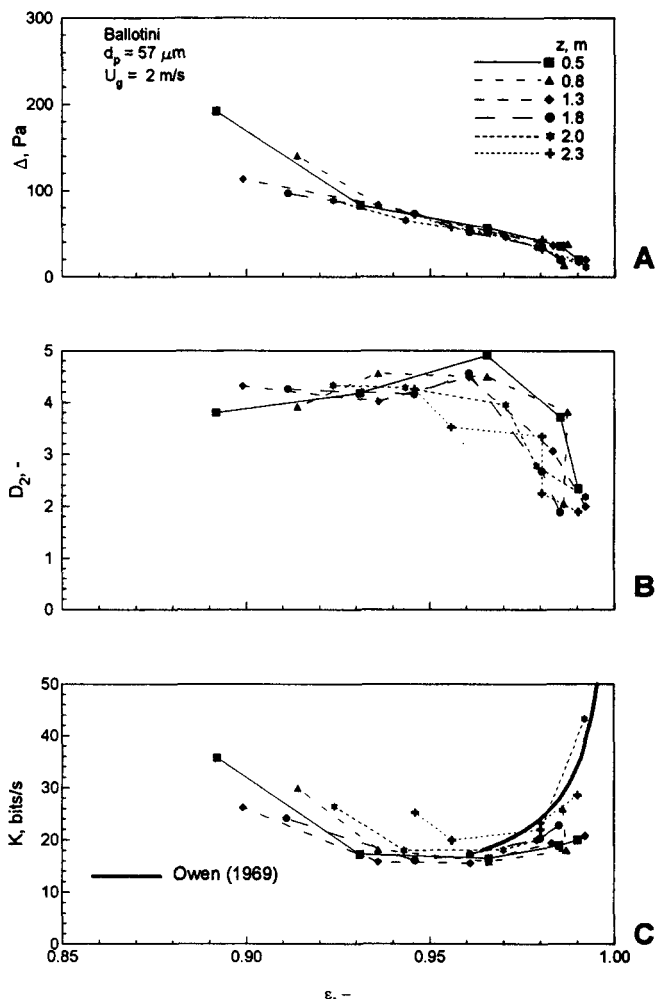


Figure 5. Average absolute deviation (A); correlation dimension (B); and Kolmogorov entropy (C) as a function of voidage.

stant voidage the solids mass holdup in the riser, and then the pressure, increase with particles density.

Comparing in each set of runs (material and  $U_g$  fixed) the trend of the average absolute deviation and of  $K$  upon a voidage decrease, it appears that the more complex shape of the  $K$  vs. voidage diagram is unrelated to the trend of the average absolute deviation vs. voidage diagrams. It can be inferred that the dynamics order level in the riser is not a monotonous function of the amount of solids in the suspension and that at  $\epsilon > \epsilon_{K_{\min}}$  an increase of solids holdup corresponds to a more ordered flow (low Kolmogorov entropy). Analysis of figures 4c, 5c, 6c, and 7c suggests that the Kolmogorov entropy should be a function of both the local voidage and the particles characteristics at fixed superficial gas velocity, and does not depend on the voidage profile along the riser provided that fast fluidization regime was not established. This means that far dynamics, such as injection of gas and solids at the riser bottom, are not able to affect strongly the local gas-solids suspension behavior. In addition, it can be assumed that the suspension flow in the riser is a spatially-extended system, that is, the dynamics of the suspension in the riser cannot be simply described by the value of

the Kolmogorov entropy measured in whatever point along the riser (Grassberger et al., 1991; van der Schaaf et al., 1997).

As a result of the previous observations, it appears that the Kolmogorov entropy is able to point out changes of the dynamics of the suspension in the riser. Since the Kolmogorov entropy is a measure of the predictability of the system and it may be related to the dynamical state (Gaspard and Wang, 1993) or turbulence level (Haucke et al., 1985) of the flow, it could be used as a tool to identify different dynamical states experienced under operating conditions tested.

The study of the dynamical level of a flow, as already discussed in the introduction should be spatially resolved. However, it should be stressed that a higher spatial resolution is not available yet for the higher solids concentration experienced. Therefore, the present approach, although limited, may provide useful information on changes of the dynamics of the suspension flow when particles concentration changes.

The variation in the Kolmogorov entropy as a function of the voidage suggests that it is possible to have the same value of Kolmogorov entropy under two widely differing flow conditions. Therefore, there is no guarantee that a flow is unchanged when the Kolmogorov entropy is kept constant and

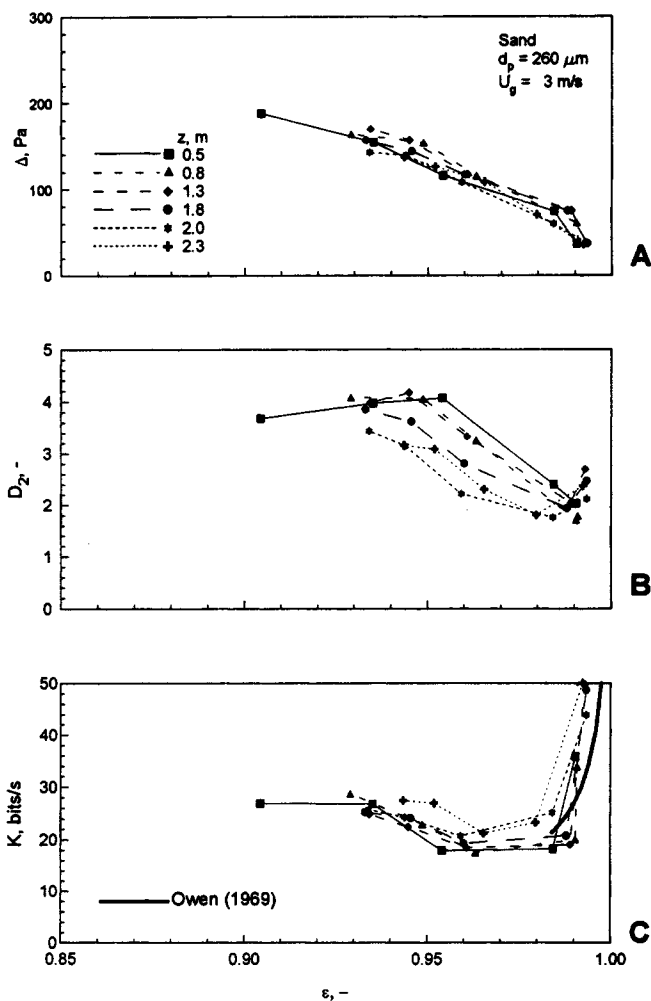
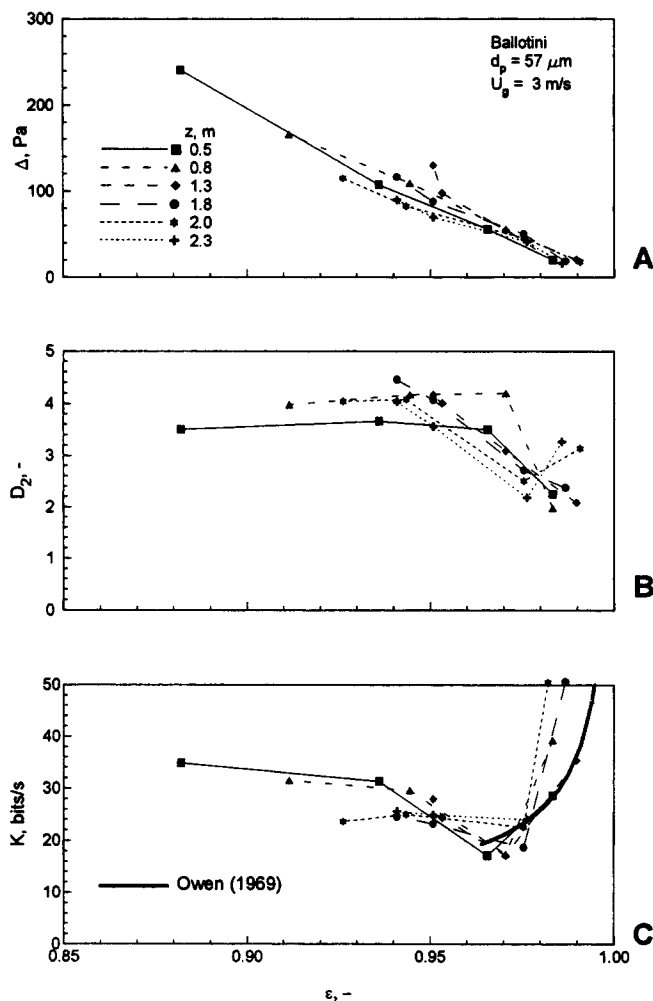


Figure 6. Average absolute deviation (A); correlation dimension (B); and Kolmogorov entropy (C) as a function of voidage.

Figure 7. Average absolute deviation (A); correlation dimension (B); and Kolmogorov entropy (C) as a function of voidage.

other invariants (such as dimension) should be used in addition to Kolmogorov entropy then. In any case, Figures 4c, 5c, 6c, and 7c show that it is possible to identify three regions in the voidage range investigated (indicated in Figure 5c): Region (I) from  $\epsilon = 1$  to the value  $\epsilon_{K_{\min}}$  at which the Kolmogorov entropy is assumed to be at its minimum value; Region (II) from  $\epsilon_{K_{\min}}$  to the value  $\epsilon_{K_{\text{const}}}$  below which the Kolmogorov entropy becomes constant; Region (III) below  $\epsilon_{K_{\text{const}}}$ . As described above, the value of  $\epsilon_{K_{\text{const}}}$  depends on the riser level and therefore the value reported in Figure 4c is only an indicative boundary of region III. It is generally possible to identify regions I and II, while region III is clearly present in the diagrams referring to runs carried out with FCC and Ballotini.

### Region I: particles-controlled region

The decrease of the Kolmogorov entropy as a function of voidage in region I may be interpreted by considering the damping effect of small particles on the turbulence level of the gas flow. It should be pointed out that although the turbulence is not uniform throughout the riser section (radial

profiles of velocity fluctuations are not flat), it is reasonable to assume that the turbulent pressure fluctuations are constant throughout the section. As a consequence, the Kolmogorov entropy evaluated by means of pressure fluctuations may be considered constant throughout the riser section in agreement with the results of van der Stappen et al. (1993b) showing that radial profiles of pressure fluctuations and Kolmogorov entropy were flat in a much larger riser ( $0.8 \times 1.2 \text{ m}$  riser section).

Data reported in Table 4 indicate that under the operating conditions tested Eqs. 1, 2, and 3 are not satisfied. These conditions mean that particles used in the present study were large enough to interact with the gas turbulence, that is, particles did not behave as tracer. On the other hand, the com-

Table 5. Values of  $\epsilon_{K_{\min}}$  and of  $K_{\text{const}}$  Under Operating Conditions Tested

Material	FCC	Ballotini	Sand
$U_g$ , m/s	2	2	3
$\epsilon_{K_{\min}}$	0.99	0.968	0.963
$K_{\text{const}}$ , bits/s	42	27	28–29



parison of particles sizes with the larger eddy size (3.3 mm) indicates that the relationship (Eq. 7) is always fulfilled for Ballotini, FCC, and sand while it is not for polystyrene. Moreover, assuming that the particle-gas relative velocity is equal to the particle terminal velocity it also results that, at least at high voidage, Ballotini, FCC and sand are small enough to damp turbulence ( $Re_p \leq 110$ ) while polystyrene large enough to enhance it ( $Re_p > 400$ ).

A result of the damping effect of small particles on gas turbulence is that velocity fluctuations, and other variables related to them, should scale down by the ratio of expression (Eq. 5). To this end, this relationship (Eq. 5) was plotted in Figures 4c, 5c, 6c, and 7c and it was forced to pass the point ( $\epsilon_{K_{\min}}, K_{\min}$ ), that is, the point where the Kolmogorov entropy is at its minimum value. The agreement of the trend of the relationship with that of  $K$  vs.  $\epsilon$  is fairly good in region I suggesting that the phenomena that induce the change of Kolmogorov entropy could be related to the turbulence level in the suspension. Moreover, the higher values of the Kolmogorov entropy measured during runs carried out at  $U_g = 3$  m/s with respect to those measured at  $U_g = 2$  m/s are in agreement with the proposed interpretation since the higher  $U_g$  is, the higher the turbulence level is.

The above proposed relationship between the value of the Kolmogorov entropy and the flow turbulent level is also supported by the measurements carried out during runs with 1,830  $\mu\text{m}$  polystyrene. The indicative value of the Kolmogorov entropy ( $> 200$ – $300$  bits/s) measured during these runs was as high as that in the empty riser ( $K > 250$  bits/s). This finding is in agreement with the proposed interpretation of the Kolmogorov entropy since these particles are large enough to enhance turbulence, and, therefore, the indicative value of the Kolmogorov entropy should be as high as that measured in the empty riser.

It is worth noting the order of magnitude of pressure fluctuations related to the phenomena studied. Assuming that the local pressure fluctuations in a turbulent flow are of order of magnitude of  $\rho_f(u')^2$ , where  $\rho_f$  and  $u'$  are the suspension density ( $\text{kg}/\text{m}^3$ ) and the velocity fluctuations, respectively, their amplitude results to the order of  $10^{-1}$  Pa in the empty riser. Therefore, the pressure fluctuations due only to the gas turbulence are lower than the minimum sensitivity of the KISTLER transducers used ( $\approx 1$  Pa), but become measurable by means of these transducers as soon as solids are added to the gas since  $\rho_f$  increases orders of magnitude due to solids contribution. It should be noted that the values of the average absolute deviation reported in Figures 4a, 5a, 6a, and 7a depend on the solids holdup in the riser and that pressure fluctuations due to the turbulence are superimposed on the main oscillations.

A further support to the relationship between the value of the Kolmogorov entropy and the suspension turbulent behavior may be obtained by analyzing the power spectrum density of pressure fluctuations. Decreasing the voidage in region I, the power density of high frequencies, that is, those involved by small-scale turbulent behavior, decreases as found by Tsuji et al. (1984) for small particles in the same frequency range ( $10^2$ – $10^3$  Hz). This means that under these operating conditions the reduction of the Kolmogorov entropy is in a certain way related to the damping of high frequency components of the pressure fluctuations. The role of the Kolmogorov en-

tropy in highlighting the reduction of the high frequencies amplitude had also been examined by evaluating its value based on time series filtered with a low-pass filter of 40 Hz. The value of the Kolmogorov entropy was always lower than that measured without low-pass filtering. Therefore, high frequency fluctuations appear to be relevant for the high value of the Kolmogorov entropy at low solids concentrations.

### Region II: clusters-controlled region

The trend of the Kolmogorov entropy upon a decrease of voidage below  $\epsilon_{K_{\min}}$  may be interpreted taking into account that at these values of voidage solids are not present as single particles but form clusters (Arena et al., 1989; Ishii et al., 1989; Horio and Kuroki, 1994; Soong et al., 1995). From the point of view of particles-turbulence interactions, clusters may be seen as large particles which are characterized by a  $Re_p$  higher than 400 and, therefore, clusters enhance turbulence. Assuming a cluster voidage equal to about the minimum fluidization value (0.5), the dimension of a cluster to enhance turbulence should be larger than 2 mm for the small particles tested (Ballotini, FCC and sand) and experimental observations report cluster size even larger than the reported value. This interpretation was also given by Gore and Crowe (1989) analyzing turbulent velocity fluctuations data obtained by experiments with small glass beads which tended to agglomerate under electrostatic charging: turbulence was not damped, as the authors expected, but enhanced.

The increase of the Kolmogorov entropy in region II is also consistent with the scale-up relationship (Eq. 6). However, this relationship was not plotted in Figures 4c, 5c, 6c, and 7c since it is not able to take into account the role of particle size on turbulence enhancement. The lack of this information is particularly noteworthy, because in a relatively concentrated gas-solids suspension the "particle" size ranges from single particle to cluster diameters. Moreover, the turbulence level is affected by the particle-size distribution and by the relative amount of each size as well (Yarin and Hetsroni, 1994). This last aspect plays a relevant role in high concentrated two-phase flow since the cluster size not only ranges over a wide interval with a wide concentration distribution, but both the interval and the distribution change when operating conditions change.

Under operating conditions typical of region II, it is possible to note an increase of the power density of low frequencies (Figure 3) and this effect amplifies as voidage decreases. A power density increase of low frequencies was also found by Tsuji et al. (1984) fluidizing large particles. This suggests that under operating conditions typical of region II, the solids structures relevant in the interaction with the gas turbulence should have a size as large as the coarse particles used by Tsuji et al. (1984).

The slope of the diagrams of the Kolmogorov entropy vs. voidage in region II at a fixed  $U_g$  appears to be in agreement with the tendency of particles to form clusters. As this tendency increases in particles tested, such as sand, Ballotini and then FCC (Marzocchella et al., 1991; Marzocchella, 1992), the diagram becomes steeper. This should suggest that when suspensions of particles inclined to form long life clusters are transported, the turbulence level, and then the Kolmogorov entropy, increases more steeply in region II.

An interesting result may be obtained by comparing the Kolmogorov entropy in the regions I and II with the variation of transport phenomena coefficients, such as of gas dispersion and heat transfer, induced by voidage changes. One of the first researchers on turbulent diffusion coefficients was conducted by Kada and Hanratty (1960). They pointed out that the turbulent diffusion coefficient showed a minimum when voidage decreased from 1.0 down to 0.975. It was supposed that the sudden increase of the turbulent diffusion coefficient, upon an increase of solids concentration, was related to the presence of groups of particles which "tended to move in unison." The voidage depending variation of the radial gas dispersion coefficient in a gas-solids suspension, flowing in a CFB riser, has also been interpreted on the basis of particle effects on gas turbulence. Researchers have found that the coefficient decreases when the voidage decreases down to 0.98 (Adams, 1988) and that it increases when the voidage decreases between 0.98 and 0.90 (Yang et al., 1983). This trend has also been confirmed by a set of measurements covering a wide range of voidage (Zheng et al., 1992), and it is believed that particles-turbulence interaction is responsible for it. The wall-to-suspension heat-transfer rate also shows a minimum upon a voidage decrease, and Al Taweel and Landau (1978) interpreted the reduction of the rate at high voidages considering the damping effects of particles on turbulence.

The analysis of the above described transport phenomena suggests that the variations of the Kolmogorov entropy in regions I and II appear to be related to the same phenomena that control the variations of these transports in the same voidage range. Furthermore, Bremhorst and Bullock (1973) have showed that in a fully developed pipe flow the transport phenomena, as that of the heat, are identical to those of the turbulent momentum. As a consequence of the above points, it may be inferred again that the Kolmogorov entropy is sensitive to a change in turbulence level in dilute gas-solids flows.

### Region III: bottom-bed-controlled region

The values of Kolmogorov entropy along the riser appear to level off as soon as a bottom bed is established in the riser. It can be supposed that the almost constant value of the Kolmogorov entropy measured throughout the riser was due to a strong phenomenon that was established at the riser bottom and that it was able to effect the dynamics of the suspension everywhere in the riser. This interpretation is further supported by results obtained with larger laboratory units (0.083 and 0.12 m ID risers) under circulating conditions with a bottom bed (Zijerveld et al., 1996). Those results show that the Kolmogorov entropy becomes constant with increasing riser solids holdup, provided that a bottom bed is established.

### Conclusion

Chaos analysis was used to quantify the dynamics of gas-solids suspensions in the riser of a laboratory circulating fluidized bed operated under a wide range of conditions. Results show that the Kolmogorov entropy, a chaotic invariant, is a function of the *local* voidage for a fixed material and superficial gas velocity. This means that it is a spatially extended system in which suspension dynamics cannot be

described by measuring the Kolmogorov entropy only in whatever single point along the riser.

By analyzing the Kolmogorov entropy vs. voidage diagrams, it is possible to identify three regions: the particles-controlled region (I), the Kolmogorov entropy decreases upon a decrease of the voidage up to  $\epsilon_{K_{min}}$ ; the clusters-controlled region (II), the Kolmogorov entropy increases when voidage decreases down to  $\epsilon_{K_{const}}$ , below which the Kolmogorov entropy is leveled off; the bottom-bed-controlled region (III), below  $\epsilon_{K_{const}}$ .

The presence of the minimum of the Kolmogorov entropy between regions I and II has been interpreted by considering the interactions between particles and turbulence structures. At high voidages, particles damp turbulence and the flow becomes more ordered. At voidages below  $\epsilon_{K_{min}}$ , clusters enhance turbulence.

As a result of the above findings, it is possible to infer that the Kolmogorov entropy could be considered a tool to characterize fluidization state and to quantify the turbulence level change of a suspension in the riser of a CFB. In its turn, this information plays a relevant role in the studies of mass- and heat-transfer rate. Further work is necessary to determine if the Kolmogorov entropy may also be used as a quantitative measure of the dynamic state of gas-solids flow. The answer to this point needs investigations on several CFB plants of different size and geometry.

### Acknowledgment

This study was mainly supported by the Nederlandse Organisatie voor Wetenschappelijk Onderzoek (NWO), project NLS 61-227. In particular, NWO supported A. Marzocchella for a five-month research period at Delft University and R. C. Zijerveld for his PhD study. This support is greatly appreciated.

### Notation

- $Ar = d_p^3 [\rho_g (\rho_s - \rho_g) g / \mu_g^2]$ , Archimedes number  
 $U_{mf}$  = minimum fluidization velocity, m/s  
 $U_t$  = particle terminal velocity, m/s  
 $U^* = U_g [\rho_g^2 / (\rho_s - \rho_g) \mu_g]^{1/3}$  dimensionless superficial gas velocity  
 $\epsilon_{K_{const}}$  = voidage below which the Kolmogorov entropy is constant  
 $\mu_g$  = gas viscosity, N/sm<sup>2</sup>

### Literature Cited

- Adams, C. F., "Gas Mixing in Fast Fluidized Beds," *Circulating Fluidized Bed Technology (II)*, J. F. Large and P. Basu, eds., Pergamon Press, Oxford, p. 299 (1988).  
 Al Taweel, A. M., and J. Landau, "The Effect of Turbulence Modulation on the Rate of Heat Transfer to Gas-Solids Suspensions," *J. Eng. Sci.*, **4**, 97 (1978).  
 Arena, U., A. Cammarota, L. Massimilla, and D. Pirozzi, "The Hydrodynamic Behaviour of Two Circulating Fluidized Bed Units of Different Sizes," *Circulating Fluidized Bed Technology (II)*, J. F. Large and P. Basu, eds., Pergamon Press, Oxford, p. 223 (1988).  
 Arena, U., A. Cammarota, A. Marzocchella, and L. Massimilla, "Solids Flow Structures in a Two-Dimensional Riser of a Circulating Fluidized Bed," *J. Chem. Eng. Japan*, **22**, 31 (1989).  
 Arena, U., A. Marzocchella, L. Massimilla, and A. Malandrino, "Hydrodynamics of Circulating Fluidized Beds with Risers of Different Shape and Size," *Powder Technol.*, **70**, 237; **71**, 116 (1992).  
 Bai, D., E. Shibuya, Y. Masuda, N. Nakagawa, and K. Kato, "Flow Structure in a Fast Fluidized Bed," *Chem. Eng. Sci.*, **51**, 957 (1996).  
 Bolio, E. J., and J. L. Sinclair, "Dilute Turbulent Gas-Solid Flow in Risers with Particle-Particle Interactions and Turbulence Modulation," *Preprints Fluidization VIII*, C. Laguerie and J. L. Large, eds., Engineering Foundation, New York, p. 515 (1995).

- Bremhorst, K., and K. J. Bullock, "Spectral Measurements of Turbulent Heat and Momentum Transfer in Fully Developed Pipe Flow," *Int. J. Heat Mass Transfer*, **16**, 2141 (1973).
- Clift, R., and W. H. Gauvin, "The Motion of Particles in Turbulent Gas Streams," *CHEMECA*, Butterworth, Australia, p. 14 (1970).
- Daw, C. S., and J. S. Halow, "Characterization of Voidage and Pressure Signals from Fluidized Beds using Deterministic Chaos Theory," *FBC Conference. Clean Energy for the World*, E. J. Anthony, ed., ASME, New York, p. 777 (1991).
- Dry, J. R., and I. N. Christensen, "Periodic Density-Inversion Effects in a High-Velocity Fluidized Bed," *Chem. Eng. Sci.*, **43**, 731 (1988).
- Fuller, T. A., T. J. Flynn, C. S. Daw, and J. F. Halow, "Interpretation of Pilot-Scale, Fluidized Bed Behaviour Using Chaotic Time Series Analysis," *FBC's Role in the World Energy Mix*, L. N. Rubow and G. Commonwealth, eds., No. I0344A, ASME, New York, p. 141 (1993).
- Gaspard, P., and X. Wang, "Noise, Chaos, and  $(\epsilon, \tau)$ -Entropy per Unit Time," *Physics Reports*, **235**, 291 (1993).
- Gore, R. A., and C. T. Crowe, "Effect of Particle Size on Modulating Turbulent Intensity," *Int. J. Multiphase Flow*, **15**, 279 (1989).
- Grassberger, P., T. Schreiber, and C. Schaffrath, "Nonlinear Time Sequence Analysis," *Int. J. Bifurcation Chaos*, **1**, 521 (1991).
- Haucke, H., R. E. Ecke, and J. C. Wheatley, "Dimension and Entropy for Quasiperiodic and Chaotic Convection," *Dimension and Entropies in Chaotic Systems*, G. Mayer-Kress ed., Springer Series, Berlin, p. 198 (1985).
- Hay, J. M., B. H. Nelson, C. L. Briens, and M. Bergougnou, "The Calculation of the Characteristics of a Chaotic Attractor in a Gas-Solid Fluidized Bed," *Chem. Eng. Sci.*, **50**, 373 (1995).
- Hetsroni, G., "Particles-Turbulence Interaction," *Int. J. Multiphase Flow*, **15**, 735 (1989).
- Hilborn, R. C., *Chaos and Nonlinear Dynamics*, Oxford University Press, New York (1994).
- Hinze, J. O., *Turbulence*, 2nd ed., McGraw-Hill, New York, p. 460 (1975).
- Horio, M., and H. Kuroki, "Three-Dimensional Flow Visualization of Dilutely Dispersed Solids in Bubbling and Circulating Fluidized Beds," *Chem. Eng. Sci.*, **49**, 2413 (1994).
- Hutchinson, P., G. F. Hewitt, and A. E. Dukler, "Deposition of Liquid or Solid Dispersions from Turbulent Gas Streams: A Stochastic Model," *Chem. Eng. Sci.*, **26**, 419 (1971).
- Ishii, H., T. Nakajima, and M. Horio, "The Clustering Annular Flow Model of Circulating Fluidized Beds," *J. Chem. Eng. Japan*, **22**, 484 (1989).
- Kada, H., and T. J. Hanratty, "Effects of Solids on Turbulence in a Fluid," *AIChE J.*, **6**, 625 (1960).
- Kunii, D., and O. Levenspiel, *Fluidization Engineering*, 2nd ed., Butterworth-Heinemann, Boston (1991).
- Marzocchella, A., "Fluidodinamica di Sistemi Solido-gas nelle 'Colonne Veloci' di Letti Fluidi Circolanti," PhD Thesis, Univ. of "Federico II," Naples, Italy (1992).
- Marzocchella, A., R. C. Zijerveld, J. C. Schouten, and C. M. van den Bleek, "Chaotic Analysis of a Circulating Fluidized Bed of Laboratory Scale," *Proc. Fourth Int. Conf. on Multiphase Flow Dynamics in Ind. Plants*, Offset Grafica, Pisa, p. 124 (1994).
- Marzocchella, A., U. Arena, A. Cammarota, and L. Massimilla, "Break Up of Cylindrical Clusters of Solids Particles under Gravity Flow in a Two-Dimensional Column," *Powder Technol.*, **65**, 453 (1991).
- Moon, F. C., *Chaotic and Fractal Dynamics*, Wiley, New York (1992).
- Owen, P. R., "Pneumatic Transport," *J. Fluid Mech.*, **39**, 407 (1969).
- Schnitzlein, M., and H. Weinstein, "Flow Characterization in High-Velocity Fluidized Beds Using Pressure Fluctuations," *Chem. Eng. Sci.*, **43**, 2605 (1988).
- Schouten, J. C., M. L. M. van der Stappen, and C. M. van den Bleek, "Deterministic Chaos Analysis of Gas-Solids Fluidization," *Fluidization VII*, O. E. Potter and D. J. Nicklin, eds., Engineering Foundation, New York, p. 103 (1992).
- Schouten, J. C., and C. M. van den Bleek, "RRCHAOS, A Menu Driven Software Package for Chaotic Time Series Analysis," Reactor Research Foundation, Delft, The Netherlands (1993).
- Schouten, J. C., F. Takens, and C. M. van den Bleek, "Maximum-Likelihood Estimation of the Entropy of an Attractor," *Phys. Rev. E*, **49**, 126 (1994a).
- Schouten, J. C., F. Takens, and C. M. van den Bleek, "Estimation of the Dimension of a Noisy Attractor," *Phys. Rev. E*, **50**, 1851 (1994b).
- Skrzyzke, D. P., K. Nguyen, and C. S. Daw, "Characterisation of the Fluidization Behavior of Different Solid Types Based on Chaotic Time Series Analysis of Pressure Signals," *FBC Conf. FBC's Role in the World Energy Mix*, L. N. Rubow and G. Commonwealth, eds., ASME, New York, p. 155 (1993).
- Soong, C. H., K. Tuzia, and C. J. Chen, "Experimental Determination of Cluster Size and Velocity in Circulating Fluidized Bed," *Preprints Fluidization VIII*, C. Laguerie and J. F. Large, eds., Engineering Foundation, New York, p. 1 (1995).
- Stringer, J., "Is a Fluidized Bed a Chaotic Dynamic System?," *FBC Conference. Technology for Today*, A. M. Manaker, ed., ASME, New York, p. 265 (1989).
- Tennekes, H., and J. L. Lumley, *A First Course in Technology*, MIT Press, Cambridge, MA (1974).
- Tsuji, Y., Y. Morikawa, and H. Shiomi, "LDV Measurements of an Air-Solid Two-Phase Flow in a Vertical Pipe," *J. Fluid Mech.*, **139**, 417 (1984).
- Tsuji, Y., "Review: Turbulence Modification in Fluid-Solid Flow," *Turbulence Modification in Multiphase Flows*, E. E. Michaelides, ed., ASME, New York, p. 1 (1991).
- Tsuo, Y. P., and D. Gidaspo, "Computation of Flow Patterns in Circulating Fluidized Beds," *AIChE J.*, **36**, 885 (1990).
- van den Bleek, C. M., and J. C. Schouten, "Deterministic Chaos. A New Tool in Fluidized Bed Design and Operation," *Chem. Eng. J.*, **53**, 75 (1993).
- van der Schaaf, J., J. C. Schouten, F. Takens, and C. M. van den Bleek, "Correlation Entropy in Fluidized Beds," *Phys. Rev. Lett.*, in press (June, 1997).
- vander Stappen, M. L. M., J. C. Schouten, and C. M. van den Bleek, "Deterministic Chaos Analysis of the Dynamical Behaviour of Slugging and Bubbling Fluidized Beds," *FBC Conference, FBC's Role in the World Energy Mix*, L. N. Rubow and G. Commonwealth, eds., ASME, New York, p. 129 (1993a).
- vander Stappen, M. L. M., J. C. Schouten, and C. M. van den Bleek, "Application of Deterministic Analysis to Pressure Fluctuation Measurement in a 0.96 m<sup>2</sup> CFB Riser," *4th Int. Conf. on Circulating Fluidized Beds*, Somerset, PA, A. Avidan, ed., AIChE, New York, p. 55 (1993b).
- vander Stappen, M. L. M., J. C. Schouten, and C. M. van den Bleek, "Chaotic Hydrodynamics and Scale-up of Gas-Solids Fluidized Beds using the Kolmogorov Entropy for Quantification," *Preprints Fluidization VIII*, C. Laguerie and J. F. Large, eds., Engineering Foundation, New York, p. 625 (1995).
- vander Stappen, M. L. M., "Chaotic Hydrodynamics of Fluidized Beds," PhD Thesis, Delft Univ. Press, The Netherlands (1996).
- Yang, G., Z. Huang, and L. Zhao, "Radial Gas Dispersion in a Fast Fluidized Bed," *Fluidization IV*, D. Kunii and R. Toei, eds., Engineering Foundation, New York, p. 145 (1983).
- Yarin, L. P., and G. Hetsroni, "Turbulence Intensity in Dilute Two-Phase Flows—1," *Int. J. Multiphase Flow*, **20**, 1 (1994).
- Zethraeus, B., C. Adams, and N. Berge, "A Simple Model for Turbulent Gas Mixing in CFB Reactor," *Powder Technol.*, **69**, 101 (1992).
- Zheng, Q., W. Xing, and F. Lou, "Experimental Study on Radial Gas Dispersion and its Enhancement in Circulating Fluidized Beds," *Fluidization VII*, O. E. Potter and D. J. Nicklin, eds., Engineering Foundation, New York, p. 285 (1992).
- Zijerveld, R. C., F. Johnsson, A. Marzocchella, J. C. Schouten, and C. M. van den Bleek, "Chaotic Hydrodynamics in the Riser Bottom Zone of Circulating Fluidized Beds of Different Scale," *Preprints of Fifth Int. Conf. on Circulating Fluidized Bed Technol.*, Beijing, China, p. DT2 (May 28–31, 1996).

Manuscript received Apr. 22, 1996, and revision received Dec. 27, 1996.