Monitoring the Quality of Fluidization Using the Short-Term Predictability of Pressure Fluctuations

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In various industrial applications of bubbling fluidized beds, defluidizing parts of the bed or even of the complete bed can occur as a result of agglomeration and sintering of particles due to unintentional maloperation of the bed, changes in operating conditions, or variations in gas or solids feed. Defluidization may be prevented by increasing the gas velocity or changing the solids feed, if the change in the "quality" of the fluidized state of particles is detected early enough. An analysis method is proposed that uses the short-term predictability of time series of (local) pressure fluctuations in the fluidized bed to detect changes early in the hydrodynamic state of the bed. It is based on the comparison of an original time series of pressure fluctuations with successive time series measured during operation of a fluidized bed. The comparison is based on a discriminating statistic computed for the original time series as well as for each successive series. The null hypothesis that the original and successive time series are similar is rejected if the discriminating statistics of both time series significantly differ. Experimental application of the method is illustrated for fluidization at elevated temperatures (ca. 120°C) of agglomerating plastic particles in a 5-cm-ID laboratory fluidized bed. The method recognizes the change in the hydrodynamics due to the incipient agglomeration of the particles. In this particular case the time period between the moment of detection of a significant change in the hydrodynamics and the end of the experiment when the bed becomes defluidized seems sufficiently large to take preventive measures. The average bed-pressure drop is not a sensitive quantity to detect changes early in the fluidization behavior.

Fluidization Quality

Gas–solid fluidized beds are among the most important reactor systems in the chemical industry, because they offer excellent possibilities for dealing with chemical reactants in reaction systems in which good fluid mixing, high heat and mass-transfer rates, and low pressure drops are required. Typical examples of industrial applications of gas–solid fluidized beds include synthesis and catalytic reactions, chlorination of metal oxides, catalyst regeneration, and combustion and gasification of coal. Fluidized beds are also used for physical processes in which the excellent heat and mass-transfer characteristics of these reactors are exploited. Typical examples include drying of particles, cooling of particles, coating of particle surfaces, granulation, and filtration processes.

A good mixing of gas and particles is crucial in all these chemical and physical applications. In practice, however, there are numerous practical reasons why the mixing of the particles may become worse during operation of the fluidized bed (see, e.g., Baeyens and Geldart, 1986). For example, in continuous processes the properties of the incoming solids may change over time. Depending on the powder properties (e.g., presence of moisture or binding agents) and the fluidization conditions (e.g., temperature of fluidizing gas), this may lead to softening or sintering of the particles. This may cause the particles to agglomerate, which will subsequently lead to defluidization of parts of the bed and blockage of

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parts of the distributor, resulting in maldistribution of the gas, local hot spots, local increase of the gas velocity, larger bubble sizes, increased bypass of gas, lower conversions, and so forth. In the worst situation, the fluidized state of the particles may fully deteriorate due to ongoing agglomeration, leading to complete blockage of the distributor, which finally necessitates shutting down the fluidized bed.

Operation of the fluidized bed at too low a gas velocity may also lead to deterioration of the fluidization quality of the gas–solid suspension due to too low a pressure drop over the distributor compared to the pressure drop over the fluidized bed. This may lead to the presence of local nonfluidized zones of particles on the distributor, which can cause hot spots that through a sequence of particle sintering and agglomeration can lead to gas entry-point blockage and eventually distributor failure (Song et al., 1984).

In industrial practice it has been found that these defluidization processes can occur at very different time scales, which means within a few days to even within a few minutes. In the latter case it will be clear that the occurrence of defluidization can only be prevented by carefully choosing the operation conditions. However, if the defluidization process takes place on the time scale of a few tens of minutes or longer, then it may be possible to prevent the ongoing defluidization process by changing the solids feed or by (locally) increasing the gas velocity through which the particle momentum is increased, which may lead to a decrease of the agglomeration tendency.

In order to be able to intervene in the fluidization process at the right moment, it is crucial that one can detect in an early stage that the "quality" of the gas–solid fluidized state is changing. Here the *fluidization quality* is defined as the state of the fluidized bed that leads to an optimal mixing of the gas and the solids throughout the bed, with easy handling of bed material, a steady in-bed temperature distribution, and a stable average bed-pressure drop.

The easiest way to detect a change in fluidization quality seems to be the recording and evaluation of the average bed-pressure drop. If defluidization occurs, part of the bed mass will no longer be involved in the fluidization process, which will lead to a lower bed-pressure drop. However, the question is whether the bed-pressure drop may serve as a sufficiently accurate "*early* warning indicator" of (initially small) changes in the quality of the fluidization. In this article, we demonstrate the use of a different "indicator" next to the measurement of the (static) bed-pressure drop. This indicator is based on the measurement and analysis of the (chaotic) *fluctuations* of the local or differential pressure in the bed. We will show that this dynamic indicator can detect at an early stage (small) changes in the fluidization state.

Pressure Fluctuations

Measurement of pressure fluctuations within fluidized beds has been frequently used to study the bed hydrodynamics (see Yates and Simons (1994) for a review of pressure measurement techniques and results). Nowadays pressure sensors with high accuracy and high response frequency are commercially available for a wide range of operating conditions (including high temperature and high pressure) at which fluidized beds are operated. The techniques to measure average differential pressure are well developed and are commonly used in industrial practice, for example, to estimate the bed height in a fluidized-bed reactor.

A great advantage of the use of pressure signals to quantify the hydrodynamic state of the fluidized bed is that they include the effects of many different (dynamical) phenomena taking place in the fluidized bed, such as gas turbulence, gas bubble formation, and passage and eruption of gas bubbles. This is illustrated in a recent study by Van der Schaaf et al. (1997) showing that pressure fluctuations in bubbling fluidized beds are a result of slow and fast propagating pressure *waves*. Pressure waves with high propagation velocities (i.e., larger than 20 m/s) are unambiguously identified as compression waves, which move upward and downward. Upward moving compression waves coincide with gas-bubble formation. The amplitude of the pressure wave is linearly dependent on the bed depth. Downward moving compression waves are caused by gas-bubble eruptions at the fluidized-bed surface and by local changes in bed voidage. In this case the pressure-wave amplitude is independent of bed depth and is neither attenuated nor amplified. Pressure waves with propagation velocities of less than 2 m/s are caused by rising gas bubbles. These pressure waves move upward only, with an amplitude that is proportional to the bubble size. It was found that the average wave-propagation velocity measured in freely bubbling beds is lower than predicted from the pseudo-homogeneous compressible wave theory due to the presence of the slowly rising gas bubbles.

The invariant properties of a time series of pressure fluctuations can be evaluated in different ways. In principle, three types of time series analyses are available: (1) statistical analysis, which is done in the time domain; (2) spectral analysis, which is done in the frequency or Fourier domain; and (3) chaos analysis, which is done in the state space of the system. These different analyses give specific invariant characteristics of the time series that are all in some way a measure of the underlying dynamic behavior of the fluidized bed. These characteristics can be used for different purposes, for example, fluidization regime identification or monitoring and control of the dynamical state of the fluidized bed.

The first type of analysis—*statistical analysis* (e.g., Clarke and Disney, 1970)—is reported mostly in the literature as being used for (on-line) diagnosis or control of the state of fluidization of a (bubbling) gas–solids fluidized bed. Statistical analysis is concerned with the probability density distribution of the data points in the time series from which various invariants can be calculated, such as the time series' average, its standard deviation or variance, and its higher-order moments like the skewness. Also other invariant measures are used that are based on the ones just mentioned.

For example, Kai and Furusaki (1987) used the average deviation of the amplitude of fluctuations in differential pressure as a measure of the state of fluidization in a laboratory-scale fluidized-bed reactor of 0.081-m ID in which methanation of carbon monoxide was carried out. They found a relationship between the average amplitude of the deviation of the pressure fluctuations and the extent of carbon dioxide conversions. The amplitude gave different states of fluidization at different degrees of conversions in which the fluidization quality changed significantly, leading to partial defluidization, slugging, and entrainment of catalyst particles. The fluidization quality and contacting efficiency could be improved by such devices as baffle internals and two-stage spargers. Again the average amplitude of the deviation of the pressure fluctuations was used to quantify the improved fluidization quality.

Chong et al. (1987) used the variance of differential pressure fluctuations as part of a computer control scheme to maintain the quality of fluidization in a tall fluidized bed very close to minimum fluidization throughout the length of the column. This was done by bleeding gas from the fluidized bed at several different levels to prevent bubbling caused by expansion of the gas due to the pressure decrease from taking place higher up in the column. A simple proportional feedback control strategy was used based on the measurement of differential pressure fluctuations, of which the variance is used as the operational setpoint.

Clough and Gyure (1984) used differential pressure across some portion of a fluidized bed to control its state of fluidization. The fluctuations of differential pressure were modeled as a time series (viz., described by an autoregressive model) and related to the degree of mixing of gas and solids within the bed. Clough and Gyure defined this degree of mixing in qualitative terms, that is, they related a small variance of the pressure fluctuations to a mild, quiescent state of fluidization and a large variance to bubbly fluidization or slug flow. Mean values of the estimated model parameters changed regularly with fluidizing velocity and, consequently, with the degree of mixing within the bed (i.e., fluidized state). Control of the fluidized state was inferred through closed loop-control of the model parameters.

Song et al. (1984) showed that fluctuations in local or differential pressure are a sensitive indicator of the malfunction of the distributor of a fluidized bed. They suggested pressure fluctuations be exploited for monitoring and controlling the fluidized-bed performance. Specifically they found that the 80% fluctuating interval of the pressure time series and the standard deviation of the fluctuations can be used to detect distributor faults.

The second type of time series analysis—*spectral analysis* (e.g., Priestley, 1981)—includes the estimation of auto- or cross-correlation functions and spectral density functions that give information about the characteristic time scales or frequencies that are present in the time series. Spectral analysis is often used to characterize different fluidization regimes (see, for example, Svensson et al., 1996), but is rarely reported in the literature as being used for (on-line) monitoring of the state of fluidization in fluidized beds. One reason may be that *on-line* determination of frequency spectra is not yet common practice in either experimental studies of fluidized-bed behavior or in industrial applications due to the more stringent data-acquisition requirements compared to statistical analysis, while statistical measures such as the time series' variance are better known and easier determined. Moreover, a disadvantage of frequency spectra may be that quantification of the time series' dynamics through some invariant is not always straightforward. For example, one could take the frequency with the largest power in the spectrum or, alternatively, the relative power in the spectrum below or above some characteristic frequency.

The third type of time-series analysis—*chaos analysis* (e.g., Moon, 1992)—comprises a set of analytic tools that specifically focuses on the topological structure of the attractor of the dynamical system in its *state space*. The term "chaos" generally refers to dynamical phenomena that can be characterized by typical features such as a short-term predictability and a high sensitivity to small changes in system parameters. On the one hand, nonlinearity is often presumed as the prerequisite for chaos to take place while, on the other hand, chaos is also seen as the generic term for all dynamical phenomena that are not completely predictable over time nor completely random. For that reason chaos can be considered as the generic term for all dynamical systems showing *erratic behavior* with a *limited predictability.* Limited predictability means that future states of the system are predictable from previous states by some deterministic model where the accuracy of the predictions of the future states becomes less accurate the longer the prediction time is. Among the analytic chaos tools are the estimations of invariant properties of the attractor such as (1) its dimension, namely, a measure of the number of degrees of freedom of the system, and (2) the Kolmogorov entropy, namely, a measure of the degree of predictability (which should not be confused with the thermodynamical entropy) (Schouten et al., 1994a,b).

Since Stringer had suggested in 1989 that the gas–solid fluidized bed's dynamic behavior might be interpreted as being chaotic, several studies have been carried out to characterize this dynamic behavior by means of chaos invariants. In 1990, Daw et al. published the first attractors that were reconstructed from pressure time series from a fluidized bed, operated in different fluidization regimes, together with some indicative values of chaos invariants, such as the correlation dimension and the maximum Lyapunov exponent. In later work, Daw et al. (1995) showed that fluidized beds in the slugging regime exhibit low-dimensional bulk behavior that is typical for chaotic systems. In an extensive study, Vander Stappen (1996) showed that other fluidization regimes (i.e., bubbling and circulating regimes) also exhibit typical characteristics of apparently chaotic behavior. Schouten et al. (1996) used the Kolmogorov entropy in engineering-type correlations for scale-up of the chaotic hydrodynamics of bubbling fluidized reactors. Marzocchella et al. (1997) showed that Kolmogorov entropy may serve as a quantitative measure of the interactions among particles and gas turbulence in a wide range of fluidized flow regimes. An overview of the methodologies used in the analysis of chaotic time series from fluidized beds has been given by Van den Bleek and Schouten (1993). A detailed description of the analysis methods and techniques can also be found in Vander Stappen (1996).

Daw and Halow (1993) were the first authors to recognize the potential of chaos analysis in the evaluation of fluidization quality through analysis of chaotic time series of pressure-drop measurements. In their article they discuss various tools for the analysis of chaotic time series that, as they suggest, might also be used for evaluation and monitoring purposes. Among others they mention monitoring of the Kolmogorov entropy, the principal component eigenvalues, or generally, the appearance of the visualized trajectory in the state space. However, no specific quantitative method for the monitoring of fluidization quality based on chaos analysis was implemented or demonstrated. Therefore, in this article, we further pursue the idea of using a characteristic feature of chaotic systems (i.e., the short-term predictability) to develop a quantitative tool for fluidized bed monitoring.

Method to Monitor Fluidization Quality

General description of method

Our method is based on the comparison of an original time series of pressure fluctuations with successive time series of pressure fluctuations that are measured during the operation of the fluidized bed. The original time series should well reflect the required or optimum state of fluidization of the bed. The length of the original and successive time series should be chosen such that a good representation of the fluidization dynamics can be obtained. In practice this implies that the time series should be of the order of tens of seconds which, in case of a bubbling bed, corresponds to the passage of tens or even up to hundreds of bubbles through the bed.

The idea is now to compare in a *statistical* manner the original time series with each successive time series in order to observe a possible change in the dynamics. The comparison is therefore based on some discriminating statistic that is computed for the original time series as well as for each of the successive series. This discriminating statistic should provide a sensitive measure of the fluidized-bed dynamical state. It can be some single invariant of the data sets, for example, one that is based on a quantification of the time series' short-term predictability in the state space. The reason for choosing the time series' short-term predictability instead of other invariant properties, such as a measure of the intensity of the signal (e.g., standard deviation) or a measure of the time scale of the signal (e.g., average frequency), is that the calculation of the short-term predictability as we adopt it here also already includes a measure of the signal's intensity (viz., average absolute deviation) as well as of a typical time scale (viz., average cycle time). This is explained below.

The following step is to formulate a null hypothesis that can be rejected or approved, depending on the outcome of the discriminating statistic. Here, we use as the null hypothesis that the original and the successive time series are similar, which means that it is hypothesized that the pressure fluctuations in both time series are from the same underlying distribution function (i.e., the same underlying (chaotic) fluidization phenomenon). This null hypothesis is rejected if the discriminating statistic obtained for the original time series is (statistically) significantly different from the one obtained from the successive series. The difference is expressed in a number of standard deviations, usually indicated as the Z-value. In the case of a normally distributed discriminating statistic with zero mean and unit variance, an absolute Z-value larger than 2.33 would lead to disproval of the null hypothesis at the 99% confidence level. Here we use the Mann–Whitney rank sum test as a statistical measure of the difference between the original and the successive time series, just as was done by Kennel and Isabelle (1992) in the case of a search for nonlinearities in a time series. The Mann–Whitney test is a so-called nonparametric test that is independent of the distribution functions of the populations considered (i.e., the original and successive time series that are compared; see Spiegel (1988)).

In fact, this proposed test method is another variant of different statistical methods that recently have been given in the literature to test for the null hypothesis that two time series have been generated by the same mechanism. A short overview of these different methods is given by Diks et al. (1996). For example, Albano et al. (1996) compare delay vector distributions by comparing the corresponding correlation integrals with the Kolmogorov–Smirnov test. Kantz (1995) uses a method for comparing the delay vector distributions of two time series. Furthermore Diks et al. (1996) have developed a test for the null hypothesis that two sets of vectors are drawn from the same multidimensional probability distribution. The typical feature of the test method that we propose and apply in this article is that it not only considers delay vector distributions but also focuses on the short-term predictability. A detailed description of the test method is given in the following subsections.

Pressure time series

We start with a stationary, continuous, scalar time series of measured pressure data points p_i, $(p_1, p_2, \ldots, p_N)$, where N is the number of points in the time series. In fact, the succession of the measured pressure data points as such can already be seen as the most simple way to represent the quality of the fluidization dynamics in the time covered by the time series. In a more advanced way the quality of the dynamics is represented by the so-called attractor in the state space of the fluidized bed. The attractor can be seen as the collection of successive states of the fluidized bed during its evolution in time. We can represent one state, at some time t, by a vector that corresponds to one point on the attractor in the state space. Using Takens' reconstruction theorem (Takens, 1981), such a state vector $\boldsymbol{P}_t$ is set up using so-called time-delayed coordinates, namely, $\boldsymbol{P}_t = (p_t, p_{t+1}, \ldots, p_{t+m-1})^T$, where m is the number of elements of the vector, which is defined as the embedding dimension of the state space. In this way the time series is transformed into a collection of points (i.e., the attractor) in state space.

Two choices have to be made for the reconstruction of these points (or state vectors) in state space: (1) the time delay (viz., the time step between two successive vector elements), and (2) the number of elements of the state vector (viz., the embedding dimension m or length of the state vector). We have found it workable (see Schouten et al., 1994b; Vander Stappen, 1996) to relate the length of the reconstructed vector to a specific time scale T in the time series. This time scale, or embedding time window, is a segment $[t, t+T]$ of the time series with length in time $T = m\tau_s$ where τ_s is the sample time step. It is our experience that the average cycle time T_c provides a robust and characteristic measure for the length of the time window T. The average cycle time is defined as the average time that is needed to complete a full cycle after the first passage through the average of the time signal:

$$T_c = \frac{\text{length of time series (units of time)}}{(\text{no. of crossings of average of time series})/2}. \quad (1)$$

This choice is predominantly based on the assumption that the structural information needed to reconstruct a point (i.e., the state vector) in state space is mainly contained in the data sampled during one average time period of length T_c. Generally, this period of time can be compared to an average orbital period on the attractor. The value of T_c is unambiguous for each given time series and can be readily calculated by counting the number of crossings of the time series' average, Eq. 1.

Independent of the choice of the length of the time window is the choice of the number of measurement points to be contained in the time window. This number of points m determines the number of elements of the state vector $\boldsymbol{P}$ in the reconstructed state space. This number of points can also be considered as the embedding dimension of the state space that should be chosen in such a way that a faithful and smooth embedding is obtained to have the data points sufficiently open up the details of the attractor. As soon as m has been chosen, the sampling frequency automatically follows from

$$f_s = \frac{m}{T_c}. \tag{2}$$

We have found (see Schouten et al., 1994b; Vander Stappen, 1996) that m should be of the order of 100 points per time window in order to sufficiently accommodate the attractor in its state space. This immediately fixes the required sampling frequency f_s at approximately 100 times the average cycle frequency f_c:$f_s = mf_c$, with $f_c = 1/T_c$ and $m \simeq 100$.

Short-term predictability

The discriminating statistic that we use here is based on a comparison of the growth of the distance in state space between points (i.e., delay vectors) that were initially very close together. These points are followed for some fixed period of time during which the growth of the distance between the points is measured by the so-called supremum norm. Two points are considered to be initially close together when the distance between these points is smaller than some distance d_o, which we typically take equal to the average absolute deviation of the original time series. The average absolute deviation is defined as

$$d_o = \frac{1}{N}\sum_{i=1}^{N} |p_i - \bar{p}|, \tag{3}$$

where the average of the time series is obtained from

$$\bar{p} = \frac{1}{N}\sum_{i=1}^{N} p_i. \tag{4}$$

We have chosen to use the average absolute deviation because it is generally a robust estimator of the time series' width around the mean, and it is very convenient in combination with the supremum norm to calculate interpoint distances in state space.

We now demand that the (supremum) distance between two reconstruction vectors $\boldsymbol{P}_i = (p_i, p_{i+1}, \ldots, p_{i+m-1})^T$ and $\boldsymbol{P}_j = (p_j, p_{j+1}, \ldots, p_{j+m-1})^T$, which means between two points on the attractor, is smaller than the distance d_o, or

$$\max_{0 \le s \le m-1} |p_{i+s} - p_{j+s}| \le d_o, \tag{5}$$

where m is thus of the order of 100, and d_o is the average absolute deviation of the original time series. Then we measure the maximum size of the distance between these reconstruction vectors, using the supremum norm, during a subsequent period of time of 10% of the average cycle time. We define this period of time as a period of divergence, τ_d. We choose this period of divergence relatively small compared to a complete orbital period, because we focus on the *initial* growth of the distance between the points. This growth of the distance gives information about the short-term predictability of the time series. For example, the short-term predictability will be small if the two points rapidly diverge within the small period of time of $0.1T_c$, while the short-term predictability is large when the two points remain close together in state space and do not diverge. So we first demand the reconstruction vectors to have a distance smaller than the average absolute deviation, Eq. 5, and then we estimate

$$d_{i,j} = \max_{0 \le s \le m+m_d-1} |p_{i+s} - p_{j+s}|, \tag{6}$$

where the number of points in the period of divergence, m_d, is 10% of m; so in practice, m_d will be of the order of ten sample time steps. To avoid any spurious effects of short-term, dynamical correlations (Theiler, 1991), we require the reconstruction vectors to be at least one average cycle period of length T_c in time apart, namely, $|i-j| > m$.

We now compare the set of distances $d_{i,j}$ of the original time series to subsequent sets that are obtained from the time series measured during the further evolution of the fluidization process. Here we take the original time series of say, 4000 data points (viz., a time series of approximately 40 s). From this time series, we randomly draw 100 pairs of reconstruction vectors $\{\boldsymbol{P}_i, \boldsymbol{P}_j\}$ that are all within distance d_o according to Eq. 5. This gives us a set of $N_A = 100$ distances $d_{i,j}$, which we will denote as A. In the same way we randomly draw a set B of $N_B = 100$ distances from the successive time series.

Discriminating statistic

With the set A of distances of the original time series, and the set B of distances of the successive time series, the Mann–Whitney rank-sum statistic is formed as

$$U = \sum_{i=1}^{N_A}\sum_{j=1}^{N_B} \Theta(A_i - B_j), \tag{7}$$

where Θ is the Heaviside step function, which is defined as $\Theta(x) = 1$ for $x > 0$, and $\Theta(x) = 0$ for $x \le 0$. The statistic U may also be calculated from the rank sums of the sets A and B, respectively, R_A and R_B (note that the ranks are assigned to the sample values for the *combined* sample consisting of sets A and B):

$$U = N_A N_B + \frac{N_A(N_A+1)}{2} - R_B, \tag{8}$$

or, alternatively, if U is based on $\Theta(B_j - A_i)$,

$$U = N_A N_B + \frac{N_B(N_B+1)}{2} - R_A. \tag{9}$$

The sampling distribution of U is symmetrical and has a mean and variance given, respectively, by the formulas (Spiegel, 1984)

$$\mu_U = \frac{N_A N_B}{2}, \tag{10}$$

and

$$\sigma^2_U = \frac{N_A N_B (N_A + N_B + 1)}{12}. \tag{11}$$

If N_A and N_B are large enough, namely, at least equal to 8 (Spiegel, 1984) (but here we use 100), the quantity

$$Z = \frac{U - \mu_U}{\sigma_U} = \frac{U - \dfrac{N_A N_B}{2}}{\sqrt{\dfrac{1}{12} N_A N_B (N_A + N_B + 1)}} \tag{12}$$

is normally distributed with zero mean and unit variance, under the null hypothesis that the two observed samples A and B came from the same distribution.

We repeat the calculation of the Z-value of Eq. 12 ten times, using the same set A of distances in the original time series. When the Z-value is calculated repeatedly, a lower limit on $|Z|$ is required than 2.33 to disprove the null hypothesis at the 99% confidence level. Alternatively, with 10 repetitions of the calculation of the Z-value, we here typically require the $|Z|$-value to be larger than 3 to reject the null hypothesis at more than the 99% confidence level. This means that as soon as the $|Z|$-value becomes larger than 3, it can be stated with more than 99% confidence that the pressure fluctuations in the original and successive time series did not come from the same underlying distribution function. Consequently, a change in hydrodynamic behavior has occurred that caused the pressure distribution functions to differ. Obviously this test method does not give information about what the *nature* of the change is, but it indicates that *some* statistically significant change has occurred.

Experimental Example

Particles, measurement equipment, and operation conditions

In this section we illustrate the use of the test method to diagnose the state of fluidization during agglomeration of particles in a small fluidized bed. Hereto we have fluidized plastic particles of the Geldart B type that are slightly porous. The particles contain up to 10 wt. % of an organic fluid that is present in the pores of the particles as well as in a small film layer on the particles. This small layer makes the particles sticky. The particles are fluidized at elevated temperatures (i.e., about 120°C), at which the particles may also soften a little; together with the stickiness of the particles, this may eventually lead to agglomeration and the formation of large clumps of particles that cause defluidization and finally blockage of the bed.

Particles. Experiments were carried out with spherical plastic particles of the Geldart B type (see Table 1). The particle-size distribution was determined with the Malvern 2600D particle sizer. The particle density was measured by suspending the particles in an ethanol solution and measuring the increase in volume. The minimum fluidization velocity was determined from pressure-drop measurements in a 5-cm-ID glass column at room temperature (i.e., 18°C) and atmospheric pressure.

Table 1. Size Distribution, Density, and Minimum Fluidization Velocity of Plastic Particles Used in Fluidization Experiments

D10 (mm)	D50 (mm)	D90 (mm)	Density* (kg/m³)	U_{mf}^{**} (m/s)	Geldart Type
0.41	0.48	0.67	910	0.06	B

*Density of porous particles including approximately 10 wt. % of organic liquid.
**Measured at ambient conditions (18°C, 1 atm).

Fluidized-Bed Setup. Fluidization experiments were carried out in a cylindrical 5-cm-ID glass column (see Figure 1) equipped with a distributor made of sintered glass with a pore size of 100 to 160 μm. The pressure drop over the distributor is 45 kPa at a superficial gas velocity of 0.40 m/s; this pressure drop is sufficiently large (about 40 to 50 times the pressure drop over the bed) to ensure an even gas distribution. The glass column is placed in a circular electrical heater. The fluidization air is preheated up to the temperature of the bed. The temperature used for controlling the heater is measured in the bed at 1 cm below the settled bed height (i.e., 19 cm above the distributor). Independently the bed temperature is also measured at 1 cm above the distributor. In general, a temperature difference of about 2°C is measured between the top and bottom of the bed.

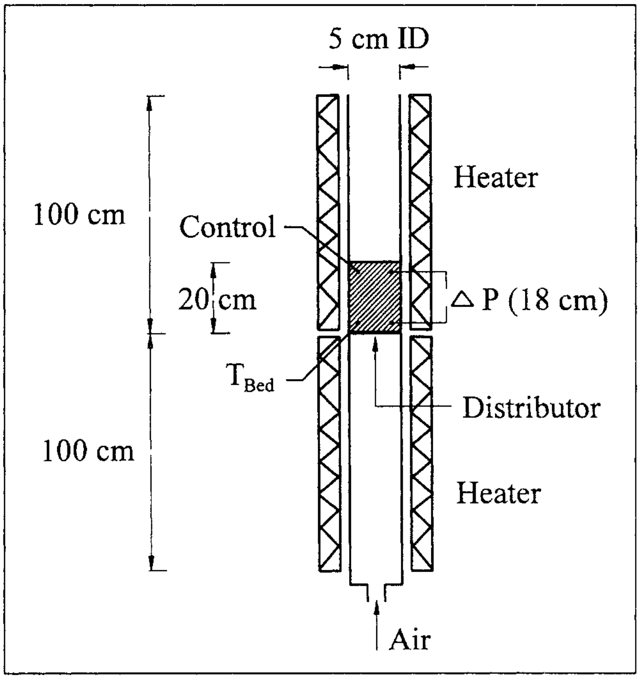

Figure 1. Fluidized bed setup.

Cylindrical 5-cm-ID glass column equipped with a distributor of sintered glass with a pore size of 100 to 160 μm. The glass column is placed in a circular electrical heater. The fluidization air is preheated up to the temperature of the bed (about 120°C).

Experimental Conditions. The bed is operated at a superficial gas velocity of 0.40 m/s. In all experiments, a settled bed height of 20 cm is used. The experiments are carried out at constant bed temperature (in the range of 117°C to 127°C). At the start of each experiment the bed is filled at room temperature with a new batch of particles. Subsequently the bed is heated up from room temperature to the required bed temperature in about 40 minutes to one hour. The acquisition of pressure data is started as soon as the bed temperature is constant (see Figure 2, in which a typical example is given). The temperature is continuously measured and recorded.

Pressure Measurement and Analysis. Bed-pressure drop was measured with a differential pressure transducer (Validyne DP103; diaphragm for pressure range 0–1,400 Pa); the pressure drop was measured over a bed slice of 18 cm high, namely, between 1 cm above the distributor and 1 cm below the settled bed height (see Figure 1).

A piezoelectric pressure transducer (Kistler type 7261) was used to measure local pressure fluctuations (viz., with respect to the average local pressure) at 1 cm below the settled bed height, or 19 cm above the distributor. The charge from the piezo element is amplified and converted to a dc voltage signal by means of an amplifier (Kistler type 5011). The differential range of the pressure transducer is adjustable from 1000 Pa to 10 bar; the most sensitive range used is 100 Pa/V; the accuracy is $\pm 1\%$ full scale, or approximately ± 1 Pa. Due to a time constant that can be set in the amplifier, the lowest frequencies are high-pass filtered by the amplifier. Typically this high-pass filter is set at 0.16 Hz. The advantage of this filtering action is that only the fluctuations of the pressure are measured relative to the average pressure (here "average" means all fluctuations in pressure at frequencies lower than the cutoff frequency of 0.16 Hz). The response frequency of the Kistler pressure transducer is sufficiently high (viz., of the order of 1 kHz) to cover the frequencies of interest in the pressure signal (i.e., frequencies typically below 50 Hz).

The pressure sensors were connected to hollow metal tubes of 4-mm ID and 6-mm OD. The end of the tube was covered with a wire gauze whose pores were typically 280-μm. The length of the tubes was about 1.2 m. The frequency response of the tubes in this type of pressure measurement setup has been tested by Vander Stappen (1996); no significant damping or resonance effects are measured for frequencies below 50 Hz, which are typical for bubbling fluidized beds.

The pressure data were acquired through the SCADAS II data-acquisition system by DIFA Measuring Systems BV. In this system, the pressure signal is filtered by a separate analog filter and subsequently, via a sample-and-hold multiplexer, the analog-to-digital conversion is done in a 16-bit ADC. The low-pass filter is of the elliptic (i.e., inverted Chebyshev) type. The roll-off rate is 80 dB/oct, with 0.01 dB ripple. The so-called filter efficiency (useful bandwidth divided by 50% of the sample frequency) is 65%. In the experiments the sampling frequency was set at 200 Hz in accordance with the requirement of approximately 100 points per average cycle. The data were low-pass filtered at 200/3 Hz (viz., sufficiently lower than the Nyquist frequency). The sampling frequency was kept constant during the experiment, which implies that the number of approximately 100 points per average cycle changed when the time series characteristics changed over time because of the agglomeration. Time series of the bed-pressure drop and of the local pressure fluctuations were stored on a computer hard disk, and subsequently analyzed off-line.

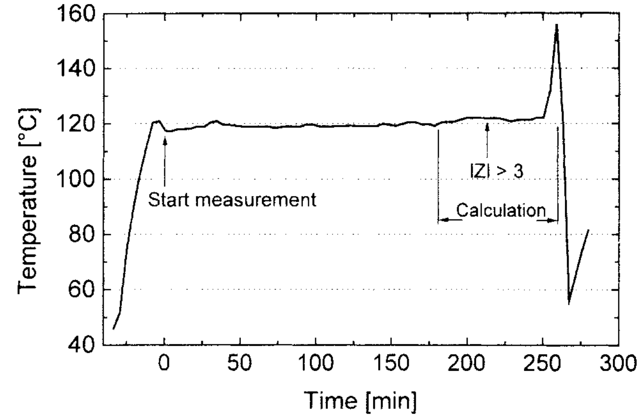

Figure 2. Typical example of the temperature of the bed that is continuously measured and recorded as a function of time (experiment 1, Table 2).

At the start of the experiment the bed is filled at room temperature with a new batch of particles. The bed is heated to the required bed temperature of 120°C. The recording of the pressure fluctuations is started as soon as the bed temperature is relatively constant (here indicated with "Start measurement"). The off-line calculation of the $|Z|$ value (cf. Eq. 12) is started approximately 70 min before the end of the experiment (here indicated with "Calculation"). The moment at which the moving-average $|Z|$ value becomes larger than 3 is also marked.

The detailed description of the pressure measurement system and the data-acquisition method that were used in this study, can be found in Vander Stappen (1996).

Experimental results

Bed Temperature. An overview of a typical experiment is shown in Figure 2 (see Table 2, experiment 1). At ambient

Table 2. Overview of Experimental Results

Exp.	Bed Temp. [°C]	Time between Moment of $\|Z\| > 3$ and End of Exp. [min]	Rel. Change in Bed-Pres. Drop between Start of Exp. and Time at $\|Z\| > 3$ [%]	Rel. Change in Bed-Pres. Drop between Start and End of Exp. [%]
1	121	38	14	50
2	120	37	24	45
3	118	80	19	36
4	117	53	13	18
5	120	72	9	16
6	127	55	3	8

conditions the column is filled with a bed of plastic particles with a settled bed height of 20 cm. The temperature of the bed is raised in approximately 40 min to the operation temperature of about 120°C. As soon as the temperature is constant, the acquisition of the pressure data is started and the time is set at $t = 0$ min. In Figure 2 it is observed that the bed temperature remains constant at approximately 121°C up to $t = 250$ min, after which the bed temperature increases sharply in about 10 min up to approximately 160°C. The reason for this sharp increase is that a clump of agglomerated particles is formed around the thermocouple that is used to control the bed temperature. The heat transfer to the thermocouple is now significantly reduced due to the "insulating" clump of plastic plasticles. This results in an action of the temperature-controlling unit to heat up the bed that is recorded by the thermocouple at the bottom of the bed, as is shown in Figure 1. After the sharp increase in temperature, the heater is switched off by hand and the bed cools off.

Time Series. Figures 3a to 3c show typical examples of time tracers of 20 s that were measured in experiment 1 (see Table 2) at three different times, namely, at $t = 180$ min, at $t = 212$ min (at which the average $|Z|$-value becomes for the first time larger than 3), and at $t = 250$ min (at which the bed temperature and the bed-pressure drop suddenly increase and decrease, respectively). It is visually observed that the time trace at $t = 250$ min is very different from the other ones: the pressure fluctuations have a significantly smaller amplitude due to the nearly complete defluidized state of the bed. Visually, only a small difference exists between the time traces at $t = 180$ min and at $t = 212$ min; the latter time sequence has a somewhat smaller average amplitude; however, the average $|Z|$-value already indicates a significant change in the dynamics.

Bed-Pressure Drop. In Figure 4a it is observed that the bed-pressure drop in experiment 1 (Table 2) is constantly decreasing. The bed-pressure drop reduces about 22% from the start of the pressure acquisition at $t = 0$ ($\Delta P \approx 400$ Pa) until the sudden decrease at about $t = 250$ min ($\Delta P \approx 310$ Pa). The sudden decrease is related to the formation of one or more large "clumps" of particles that do not take part in the fluidization that results in the lower pressure drop. Figure 5 shows an example of a big clump of particles that has been formed in the bed; this clump was collected from the column after the bed cooled down.

Experiment 1 was repeated five times. The corresponding graphs of the bed-pressure drop are shown in Figures 4b–4f. The decrease in bed-pressure drop is very different among the six repeated experiments, which is an indication that the agglomeration process can be very different at similar operating conditions. Experiment 6 was performed at the highest temperature (i.e., 127°C), but it is remarkable that the decrease in bed-pressure drop for this experiment is the smallest. Moreover the decrease in bed-pressure drop is very gradual and does not give a clear indication of a significant change in fluidization behavior. Furthermore, the total drop in pressure at the end of the experiment is only 8%, while in the other cases this total drop varies between 16% and 50%.

The change in bed-pressure drop in experiment 6 over time is quite similar to experiment 5 where a very gradual decrease without a distinct change that could be indicative of some change in fluidization behavior is also observed. In experiment 2 a clear decrease in bed-pressure drop is found after approximately 40 min; however, this decrease is followed by a very gradual decrease in bed-pressure drop over the subsequent period of about 100 min. Probably after 40 min a small part of the bed does not take part anymore in the

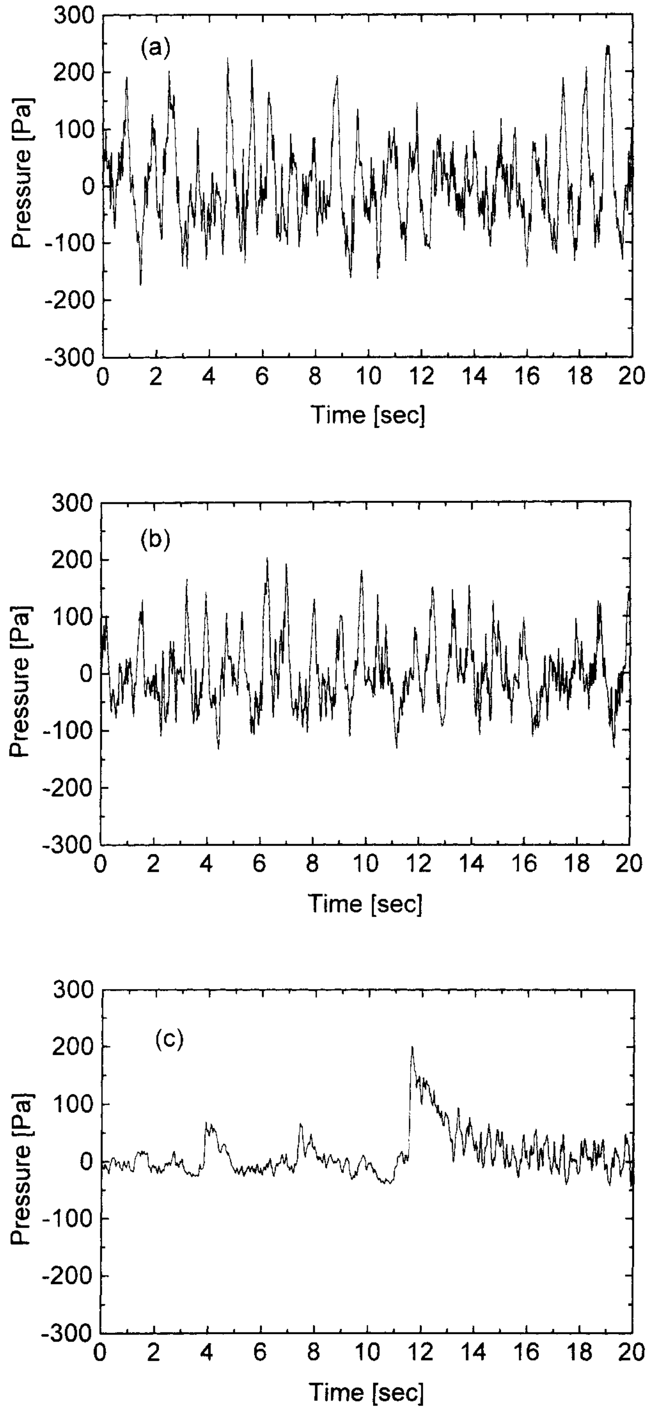

Figure 3. Typical examples of three time series of 20 s of measured pressure fluctuations (experiment 1, Table 2) at three different moments after the start of the experiment (cf. Figure 2): (a) $t = 180$ min; (b) $t = 212$ min (at which $|Z| > 3$); and (c) $t = 250$ min (end of experiment).

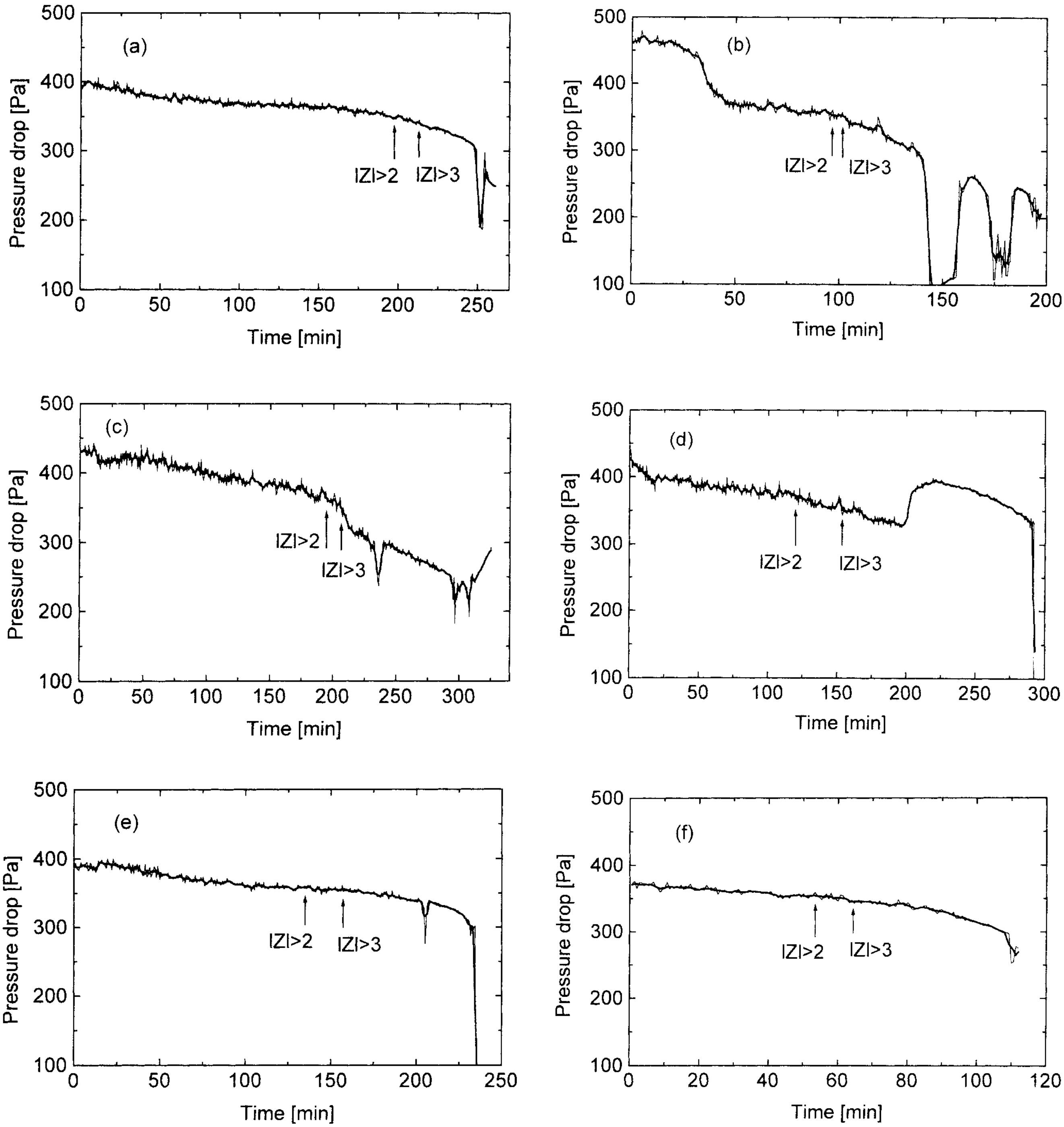

Figure 4. Pressure drop over the bed as a function of time.

(a) Experiment 1; (b) experiment 2; (c) experiment 3; (d) experiment 4; (e) experiment 5; (f) experiment 6. The pressure drop is measured over a bed slice 18-cm high between 1 cm above the distributor and 1 cm below the settled-bed height. The moments at which the moving-average $|Z|$ values become larger than 2 and 3, respectively, have been marked. Thin line (—): average bed-pressure drop for each successive time series of 40 s. Thick line (——): moving-average value of bed-pressure drop based on ten successive time series.

fluidization due to agglomeration. The main effect is, however, observed only after 140 min.

Experiment 3 shows a clear change in the decrease of the bed-pressure drop after about 200 min. The decrease in bed-pressure drop is suddenly much steeper with a significant jump at about 230 min, which is, however, restored after about 10 min; most probably a clump of agglomerated particles is broken up again to take further part in the fluidization. Experiment 4 shows a notable phenomenon: after about 200 min the pressure drop of the bed increases significantly, after which it decreases gradually until the end of the experiment after about 290 min. Here also a large clump of agglomerated particles is probably broken up and the particles take part in the fluidization again.

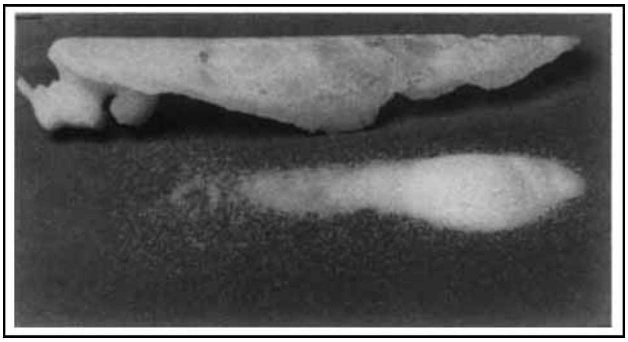

Figure 5. Plastic particles used in agglomeration experiments and a clump of agglomerated particles formed in the fluidized bed as collected from the column after cooling down of the bed.

Summarizing, the graphs in Figures 4a–4f illustrate that it is not simple to use the bed-pressure drop as an "early warning indicator" of changes in the fluidization behavior in these experiments. Different types of phenomena can take place in the bed that may all affect the bed-pressure drop in a different manner such that no unique criterion can be defined for early warning detection of changes in fluidization quality based on bed-pressure drop alone.

Fluidization Quality: $|Z|$-Value. The calculated $|Z|$ values for the six experiments are shown in Figures 6a–6f. The single $|Z|$-values for the separate successive time series of 40 s fluctuate considerably while incidentally a single $|Z|$ value may be larger than 3, indicating a statistically significant different time series compared to the initial series. It is observed, however, that the $|Z|$-value of a subsequent time series of 40 s may again be well below 3, statistically indicating a similarity between the time series. For that reason the single $|Z|$-values may not be directly used as a quantitative early warning indicator of changes in the fluidization behavior. In all cases, it is observed, however, that the moving-average value of $|Z|$ gives a better and more adequate indication of the changing fluidization quality. The moving-average value is based on 10 successive single values of $|Z|$, corresponding to a time period of 400 s or 6.7 min. The moving-average $|Z|$ gradually increases from values well below 3 (generally of the order of 1 to 2) to values significantly larger than 3 close to the end of the experiment where the bed becomes defluidized as is represented by a significant decrease in bed-pressure drop (cf. Figures 4a–4f).

At a specific moment in time, the moving-average $|Z|$ passes the value of 3 at which the null hypothesis of similar time series can be rejected at more than the 99% confidence level. At that point the fluidization quality has become statistically significantly different from that in the initial time series. These points at which the moving-average $|Z|$ becomes larger than 3 have also been indicated in the graphs of the bed-pressure drop in Figures 4a–4f. It is observed that no clear relation exists between the moment that $|Z|$ becomes larger than 3 and the decrease in the bed-pressure drop. In all cases it is found that the criterion of $|Z| = 3$ can very well serve as an early warning detector for changes in the fluidization behavior: the time period between the moment that $|Z|$ is larger than 3 and the end of the experiment when the bed becomes defluidized is relatively large (i.e., ranging from 37 min (experiment 2) to 80 min (experiment 3), see Table 2). In a practical situation this period of time may be sufficient to intervene in the fluidization process to prevent further agglomeration and defluidization.

The less strict criterion of $|Z| = 2$ may also be used as the early warning detector; in that case, the warning is given about 10 min to even up to 50 min earlier (see Figures 4a–4f) than when $|Z| = 3$ is used. The choice of what criterion should be used will generally be determined by the actual practical situation. A lower value of $|Z|$, of course, corresponds to a lower level of confidence at which the null hypothesis of similar time series is rejected.

Discussion

Relevance of Test Method. The relevance of the proposed method is that it actually combines and integrates three types of time series analyses: statistical, spectral or Fourier, and chaos analysis.

1. First, in the test method the average absolute deviation is chosen to select sets of two points in the state space that are close together, see Eqs. 3 and 5. In fact, the average absolute deviation is a statistical measure of the width of the probability distribution function of the measured pressure fluctuations. In our test method the distance between points in the state space is measured by the supremum norm, so the maximum distance between points (i.e., the size of the attractor) is directly related to the maximum width of the probability distribution function of the measured pressure fluctuations. The average absolute deviation is mostly about 10 to 20% of this maximum width and, therefore, it is an adequate measure for locating points that are close together in state space. Consequently, if for some reason the average absolute deviation changes over time, the selection of points in state space also changes. If this change is statistically significant, it will therefore be detected by the test method.

2. Second, in the test method the length of the reconstructed state vectors is based on the average cycle time (see Eq. 1). The average cycle time is related to the average cycle frequency and, therefore, it actually refers to a characteristic time scale in the time series. Obviously this average cycle frequency is related to the frequencies in the frequency spectrum that are obtained through Fourier transformation of the time series. For example, it is found for pressure-fluctuation time series measured in fluidized beds that the average cycle frequency is typically 50 to 300% larger than the peak frequency (frequency with largest amplitude in frequency spectrum) (see Vander Stappen, 1996). If for some reason the average cycle time changes over time, the selection of points in state space will also change. If this change is statistically significant, it will therefore also be detected by the test method.

3. Third, the comparison of the distributions of distances in state space through the degrees of *predictability* is, of course, a typical feature of chaos analysis.

The test method will thus be able to detect a change in the dynamic behavior of the system when this change is reflected in the average absolute deviation, in the average cycle time, or in the degree of predictability. This is a great advantage

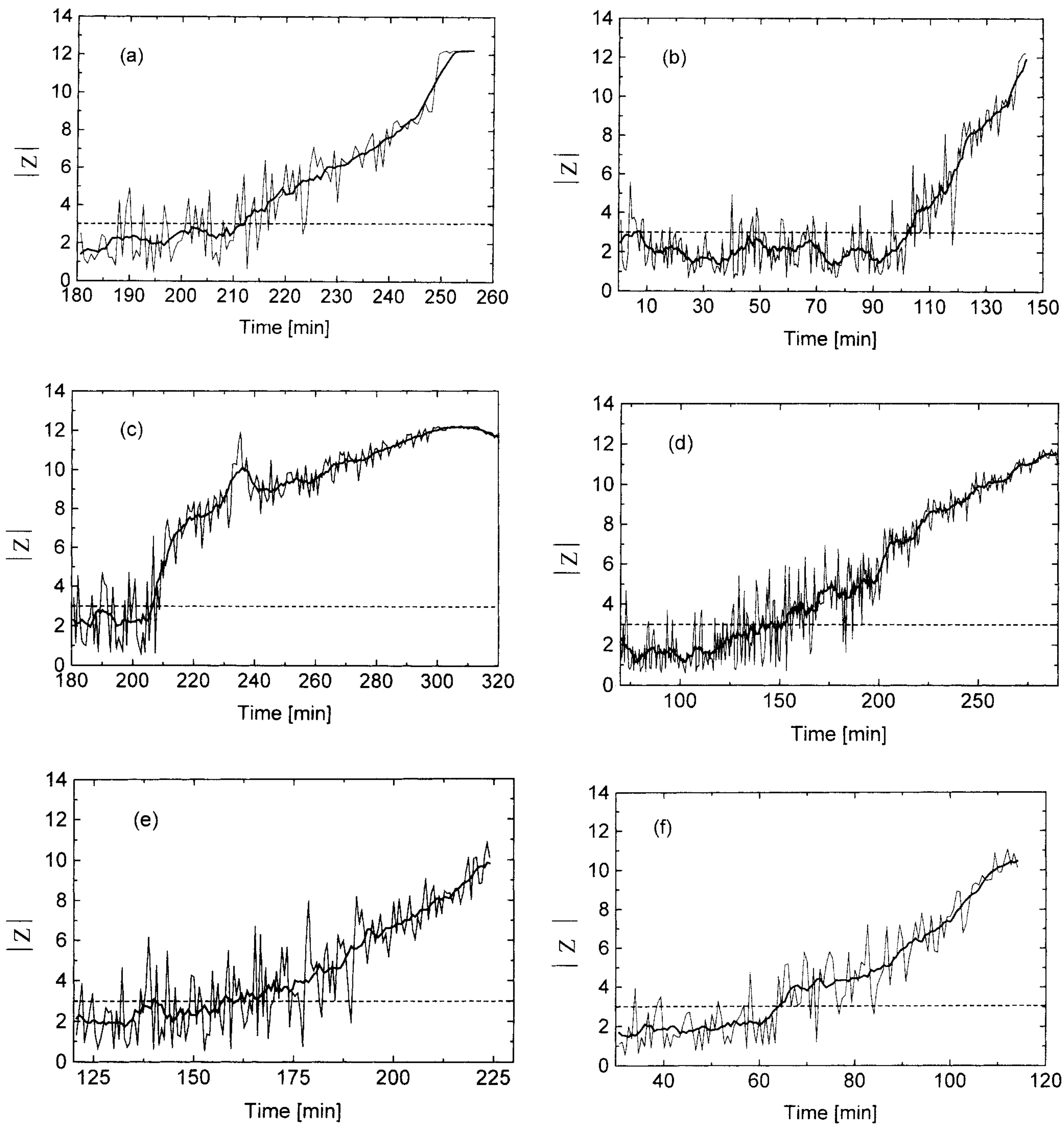

Figure 6. Values of $|Z|$ (Eq. 12) as a function of time.

(a) Experiment 1; (b) experiment 2: (c) experiment 3; (d) experiment 4; (e) experiment 5; (f) experiment 6. Thin line (—): single $|Z|$ value for separate successive time series of 40 s. Thick line (——): moving-average value of $|Z|$ based on ten successive time series.

over test methods that would only focus on one of the three. This is illustrated by Figures 7a–7b in which the average absolute deviation (Figure 7a) and the average cycle time (Figure 7b) of experiment 1 are given, respectively. It is seen that the average cycle time does not notably change over time during the whole experiment; however, the average absolute deviation clearly decreases during the experiment. So in this case no early warning detection was obtained when only the average cycle time would have been used as indicator. However, the decrease of the average absolute deviation in combination with the probable change in the time series' predictability now showed a significant change in dynamics through the $|Z|$ value (cf. Figure 6a).

Obviously the present method can be made even more specific by including more information about the distribution in the state space of the pairs of points that are close together.

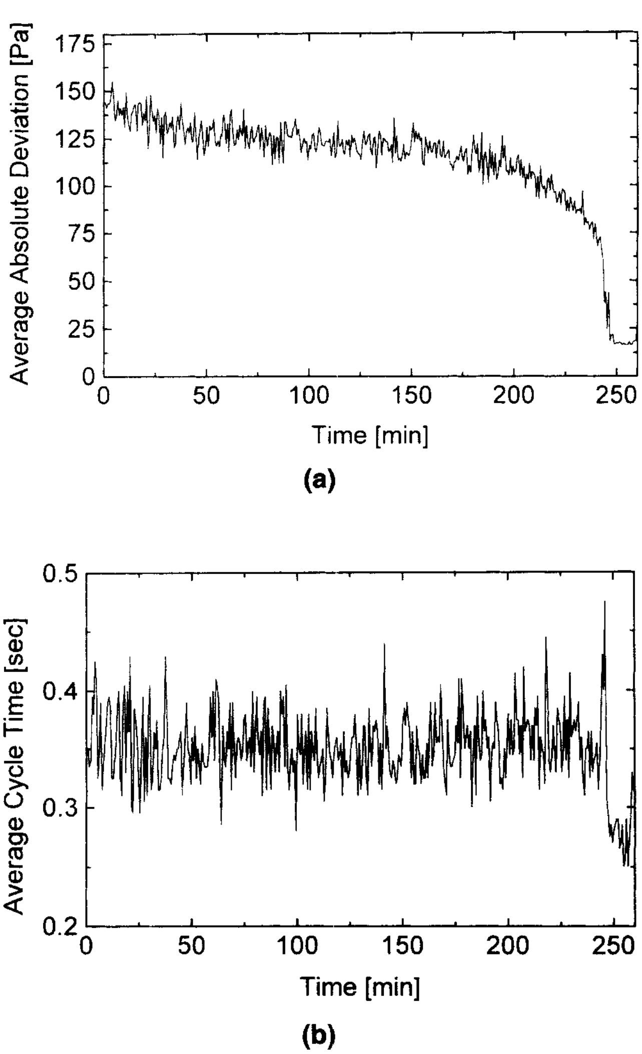

Figure 7. Experiment 1: (a) average absolute deviation as a function of time; (b) average cycle time as a function of time.

Average values for each successive time series of 40 s.

In this way, for example, groups of pairs of points that have a different (local) predictability may be identified in state space. The way in which the predictability of each of these groups changes over time can similarly be used to detect changes in the time series' dynamics.

Industrial Application of Method. The proposed method can readily be implemented in an industrial type of fluidized-bed reactor. The method is based on pressure measurements that are most often relatively easy to perform in industrial practice. Although in this study pressure sensors have been used with a high accuracy and a relatively fast response, in industrial applications it is foreseen that less expensive types of sensors will also be suitable. Most probably the pressure measurements have to be carried out at different locations in the bed to obtain a complete overview of the dynamic behavior. Each time signal could be analyzed on-line with the proposed method, although it most probably would be better to develop an advanced version of the method, in which multiple signals are analyzed simultaneously through a combined state-space representation. This type of further development of the analysis method is currently being carried out at Delft University of Technology.

Conclusions

1. A method is presented to detect changes in fluidization behavior early based on the measurement and analysis of time series of the (chaotic) fluctuations of local pressure in a fluidized bed. The method combines and integrates features of three types of time series analysis: (1) statistical analysis (carried out in the time domain); (2) spectral analysis (carried out in the frequency domain); and (3) chaos analysis (carried out in the state space). The method focuses on the quantification of the short-term predictability of the time series of pressure fluctuations that is a typical property of chaotic systems. The short-term predictability is determined in the state space by measurement of the growth of initially close distances between points on the attractor.

2. The method compares an original time series that reflects the required or optimum state of fluidization with successive pressure time series acquired during operation of the fluidized bed. The degree to which the original time series and the successive time series differ is expressed by a statistical quantity, the so-called Z value (Eq. 12). It is found that the best result is obtained if this quantity is based on a moving-average value through which time series of approximately 7 min in length are compared. This period of time corresponds to approximately 200 to 300 average cycles of the fluctuating pressure, which would indicate an approximately similar number of bubble passages at the location of the pressure probe. This number of cycles therefore seems to be a minimum to accurately pinpoint the moment at which the fluidization behavior starts to change. If agglomeration processes take place on a smaller time scale, the present method will not be able to accurately detect this.

3. It is found that in the particular case of agglomerating plastic particles as presented in this article the application of the method provides a time span of a few tens of minutes up to more than one hour to intervene in the fluidization process to prevent further agglomeration. Of course, in a different experimental setup (larger bed diameter, higher bed, etc.), this time span may be different. For each case this should be investigated separately.

4. The average bed-pressure drop is not an adequate measure to detect changes in fluidization behavior early. It is not reproducible due to the randomlike nature of the agglomeration process. No clear relation exists between the decrease in bed-pressure drop and the moment at which the Z value indicates a statistically significant change in hydrodynamic behavior. For that reason the Z value is preferred as an early warning detector of changes in the quality of fluidization.

5. The method is relatively easy from a technical point of view (pressure measurements are commonly used in fluidization practice), while the data analysis is not time-consuming or complicated and could be done on-line. Implementation of this method in an industrial type of fluidized bed for monitoring and/or control purposes is therefore considered to be realistic. However, a multiple-signal variant of the method will have to be developed.

Literature Cited

Albano, A. M., P. E. Rapp, and A. Passamante, "Kolmogorov-Smirnov Test Distinguishes Attractors with Similar Distances," *Phys. Rev. E*, **52**, 196 (1995).

Baeyens, J., and D. Geldart, "Solids Mixing," *Gas Fluidization Technology*, Chap. 5, D. Geldart, ed., Wiley, New York, p. 97 (1986).

Chong, Y. O., D. P. O'Dea, E. T. White, P. L. Lee, and L. S. Leung, "Control of the Quality of Fluidization in a Tall Bed Using the Variance of Pressure Fluctuations," *Powder Technol.*, **53**, 237 (1987).

Clarke, A. B., and R. L. Disney, *Probability and Random Processes for Engineers and Scientists*, Wiley, New York (1970).

Clough, D. E., and D. C. Gyure, "Stochastic Inferential Control of the Fluidized State Via Differential Pressure Measurements," *Chem. Eng. Commun.*, **28**, 59 (1984).

Daw, C. S., W. F. Lawkins, D. J. Downing, and N. E. Clapp, Jr., "Chaotic Characteristics of a Complex Gas-Solids Flow," *Phys. Rev. A*, **41**(2), 1179 (1990).

Daw, C. S., and J. S. Halow, "Evaluation and Control of Fluidization Quality Through Chaotic Time Series Analysis of Pressure Drop Measurements," *AIChE Symp. Ser.*, **89**(296), 103 (1993).

Daw, C. S., C. E. A. Finney, M. Vasudevan, N. A. van Goor, K. Nguyen, D. D. Bruns, E. J. Kostelich, C. Grebogi, E. Ott, and J. A. Yorke, "Self Organization and Chaos in a Fluidized Bed," *Phys. Rev. Lett.*, **75**(12), 2308 (1995).

Diks, C., W. R. van Zwet, F. Takens, and J. DeGoede, "Detecting Differences Between Delay Vector Distributions," *Phys. Rev. E*, **53**, 2169 (1996).

Kai, T., and S. Furusaki, "Methanation of Carbon Dioxide and Fluidization Quality in a Fluid Bed Reactor—The Influence of a Decrease in Gas Volume," *Chem. Eng. Sci.*, **42**(2), 335 (1987).

Kantz, H., "Quantifying the Closeness of Fractal Measures," *Phys. Rev. E*, **49**, 5091 (1994).

Kennel, M. B., and S. Isabelle, "Method to Distinguish Possible Chaos from Colored Noise and to Determine Embedding Parameters," *Phys. Rev. E*, **46**(6), 3111 (1992).

Marzocchella, A., R. C. Zijerveld, J. C. Schouten, and C. M. van den Bleek, "Chaotic Behavior of Gas-Solids Flow in the Riser of a Laboratory-Scale Circulating Fluidized Bed," *AIChE J.*, **43**(6), 1458 (1997).

Moon, F. C., *Chaotic and Fractal Dynamics*, Wiley, New York (1992).

Priestley, M. B., *Spectral Analysis and Time Series*, Academic Press, London (1981).

Schouten, J. C., F. Takens, and C. M. van den Bleek, "Maximum Likelihood Estimation of the Entropy of an Attractor," *Phys. Rev. E*, **49**(1), 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**(3), 1851 (1994b).

Schouten, J. C., M. L. M. vander Stappen, and C. M. van den Bleek, "Scale-Up of Chaotic Fluidized Bed Hydrodynamics," *Chem. Eng. Sci.*, **51**(10), 1991 (1996).

Song, J. C., L. T. Fan, and N. Yutani, "Fault Detection of the Fluidized Bed Distributor by Pressure Fluctuation Signal," *Chem. Eng. Commun.*, **25**, 105 (1984).

Spiegel, M. R., *Schaum's Outline of Theory and Problems of Statistics*, Schaum's Outline Series, McGraw-Hill, New York (1988).

Stringer, J., "Is a Fluidized Bed a Chaotic Dynamic System," L. Rubow and G. Commonwealth, eds., *Proc. Int. Conf. on Fluidized Bed Combustion*, Amer. Soc. Mech. Eng., New York, p. 265 (1989).

Svensson, A., F. Johnsson, and B. Leckner, "Fluidization Regimes in Non-Slugging Fluidized Beds: The Influence of Pressure Drop Across the Air Distributor," *Powder Technol.*, **86**, 299 (1996).

Takens, F., "Detecting Strange Attractors in Turbulence," *Lecture Notes in Mathematics*, Vol. 898, D. A. Rand and L.-S. Young, eds., Springer-Verlag, Berlin, p. 366 (1981).

Theiler, J., "Some Comments on the Correlation Dimension of $1/f^{\alpha}$ Noise," *Phys. Rev. Lett.*, **135**(8,9), 480 (1991).

Van den Bleek, C. M., and J. C. Schouten, "The Fluidized Bed as a Deterministic Chaotic System," *Chem. Eng. J.*, **53**, 75 (1993).

Van der Schaaf, J., J. C. Schouten, and C. M. van den Bleek, "Origin, Propagation and Attenuation of Pressure Waves in Gas-Solids Fluidized Beds," *Powder Technol.* (1997).

Vander Stappen, M. L. M., *Chaotic Hydrodynamics of Fluidized Beds*, PhD Thesis, Delft Univ. of Technology, Delft Univ. Press, The Netherlands (1996).

Yates, J. G., and J. R. Simons, "Experimental Methods in Fluidization Research," *Int. J. Multiphase Flow*, **20**(Suppl.), 297 (1994).

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