Early Warning of Agglomeration in Fluidized Beds by Attractor Comparison

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An enhanced monitoring method, based on pressure fluctuation measurements, for observing nonstationarities in fluidized-bed hydrodynamics is presented. Experiments show that it can detect small changes in the particle-size distribution. Such a monitoring method is useful to give an early warning of the onset of agglomeration in a fluidized bed. In contrast to earlier methods, this method is insensitive to small changes in superficial gas velocity and can handle multiple signals, making it relevant to industrial application. By carefully choosing the measurement position, the method becomes also insensitive to small bed mass variations. It uses the attractor reconstructed from a measured pressure signal, which is a "fingerprint" of the hydrodynamics of the fluidized bed for a certain set of conditions. Using this method statistically the reconstructed attractor of a reference time series of pressure fluctuations (representing the desired fluidization behavior) is compared with that of successive time series measured during the bed operation.

Introduction

Gas-solids fluidized beds are widely applied for chemical processes, such as the catalytic production of vinyl acetate, the polymerization of olefins, the chlorination of metal oxides, and the combustion or gasification of coal, waste, and biomass. Moreover, fluidized beds are used for a large variety of physical processes, such as adsorption processes, the heating, cooling, and drying of particles, and the coating of particle surfaces. The reasons for applying fluidized beds to these processes include easy solids handling and good solids mixing, resulting in a relatively uniform bed temperature and an efficient heat exchange. Certain changes in the fluidized-bed hydrodynamics, such as agglomeration or sintering of the bed particles, are disadvantageous. Agglomeration often occurs in practice, for example, due to a too high moisture content or a too high temperature of the bed. This affects the quality of the solids mixing and may eventually lead to defluidization of parts of the bed and blockage of parts of the distributor, resulting in an uneven distribution of the gas, local hot spots, a local increase of the gas velocity, and so on. In the worst case, the fluidized state of the particles fully deteriorates due to ongoing agglomeration, resulting in a complete blockage of the distributor, so that the installation has to be shut down.

If the time scale of the defluidization process is tens of minutes or larger, it may be possible to take timely measures to prevent the bed from becoming defluidized if the agglomeration is detected early enough. In that case, agglomeration can be repelled by changing the bed temperature, by increasing the superficial gas velocity, by local gas injection through nozzles, by changing the solids feed, and so on. The early detection of the onset of agglomeration is one application of a method for monitoring the fluidized-bed hydrodynamics. Furthermore, such a method can also be applied to monitor imposed changes in the particle-size distribution, as may take place during a so-called grade change: switching from one product composition to another by changing the particle feed stream to the fluidized bed that is operated in a continuous way.

Recently, Schouten and Van den Bleek (1998) proposed a method for monitoring the fluidized-bed hydrodynamics. Their method tests in a statistical way if the short-term predictability of the fluidized-bed pressure fluctuations has

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changed. This method was a first step towards the early detection of agglomeration in industrial installations: it was shown to be sensitive to small changes in the particle-size distribution (Van Ommen et al., 1999b). However, the method is also very sensitive to small changes in the superficial gas velocity. This is a disadvantage, since small fluctuations in the gas supply often occur in industrial practice (Van Ommen et al., 1999a).

The method we presently propose is a next step in the detection of nonstationarities in fluidized-bed behavior, such as the onset of agglomeration: it is sensitive to small changes in the particle-size distribution, whereas it is insensitive to small changes in the superficial gas velocity. Moreover, we will show that this new monitoring method can easily handle multiple pressure signals. This is relevant for application to large industrial fluidized beds, since one measurement position is possibly insufficient to monitor a large bed completely.

Earlier Monitoring Methods

In literature, several methods have been proposed to monitor the fluidized-bed hydrodynamics. We will restrict ourselves to a brief overview of methods based on pressure measurements, since most other measurement techniques cannot routinely be applied in industrial installations (Werther, 1999). Moreover, a great advantage of pressure signals is that they include the effect of many different dynamic phenomena taking place in the bed, such as bubble formation, bubble coalescence, and bubble passage (Van der Schaaf et al., 1998a), which are all influenced by the particle-size distribution.

Pressure drop

The easiest way to detect a change in fluidized-bed hydrodynamics would seem to evaluate the average pressure drop over the bed. For example, if defluidization occurs, part of the bed mass will no longer be involved in the fluidization process, which will lead to a lower pressure drop. On the other hand, blocking of the distributor plate by sticky particles may lead to an increase in the average pressure drop. The question is whether the average pressure drop is a sufficiently accurate "*early* warning indicator" for changes in the hydrodynamics. Schouten and Van den Bleek (1998) showed that it is not: the agglomeration of plastic particles in a laboratory-scale fluidized bed at elevated temperature was not detected by monitoring the average pressure drop until defluidization has occurred. In that case it is often too late; a functional monitoring method should give an *early* warning such that defluidization can be *prevented.*

Intensity of pressure fluctuations

Since pressure *fluctuations* contain a lot of information about the fluidized-bed behavior, it is advantageous to base a monitoring method on these fluctuations instead of on average pressure values. The simplest property of pressure fluctuations to consider is the intensity, expressed by the standard deviation, the average absolute deviation, or the range of the signal. Indeed, several authors have proposed to monitor the fluidization state of a bed by observing the intensity of the pressure signal, such as Kai and Furusaki (1987) and Chong et al. (1987). However, this intensity strongly depends on the superficial gas velocity (Van Ommen et al., 1999a), which makes these methods inadequate for industrial application.

Spectral analysis of pressure fluctuations

Spectral analysis is often used to characterize different fluidization regimes (for example, Svensson et al., 1996), but is rarely reported in literature as being used for (on-line) monitoring of the state of fluidization in fluidized beds. A disadvantage of spectral analysis is that rather large amounts of data are needed to accurately estimate characteristics of the signal such as the spectral density function. This means that it takes a long time before changes in the hydrodynamics are detectable. Moreover, Van Ommen et al. (1999a) showed that the power spectral density of pressure signals is rather insensitive to small changes in the particle-size distribution.

Nonlinear analysis of pressure fluctuations

A third way of detecting changes in pressure fluctuations is by using techniques from nonlinear time series analysis, often referred to as chaos analysis. Monitoring methods of this type, briefly reviewed by Van Ommen et al. (1999d), are usually based on a technique called attractor reconstruction, which will be explained below.

Attractor Reconstruction. The state of a fluidized bed at a certain time can be determined by projecting all variables governing the system in a multidimensional space (the "state space"); the collection of the successive states of the system during its evolution in time is called the "attractor." However, it is practically impossible to know all governing variables of a fluidized bed. Takens (1981) proved that the dynamic state of a system can be *reconstructed* from the time series of only one *characteristic* variable (such as the local pressure in a fluidized bed). Using so-called time-delay coordinates, it is possible to convert a pressure time series (p_1, p_2, $\ldots$, p_{Np}) consisting of N_p values into a set of $N_p - m$ delay vectors P_k with m elements, where $P_k = (p_k, p_{k+1}, \ldots, p_{k+m-1})^T$. The subsequent delay vectors can be regarded as points in an m-dimensional state space yielding a reconstructed attractor, from which Takens (1981) has proven that it has the same dynamic characteristics as the "true" attractor obtained from all variables governing the system. The reconstruction of an attractor from a pressure time series is shown in Figure 1.

An advantage of the attractor reconstruction technique is that all properties of the signal are conserved, in contrast with the transformation of a signal to its power spectrum or to its probability density function, in which, respectively, phase information and time correlations are lost.

Monitoring Methods Based on Attractor Properties. Daw and Halow (1993) were the first to propose the monitoring of fluidization hydrodynamics through chaotic time series. They suggested monitoring certain time-averaged properties of the reconstructed attractor: as long as such a property remains within desired limits, the fluidization state is the desired one and no actions are necessary. However, they did not indicate how these limits had to be determined nor how sensitive such a method would be.

Wright (1995) used the Fourier transform of the probability density of a two-dimensional reconstructed attractor to characterize signals generated by nonlinear processes. He

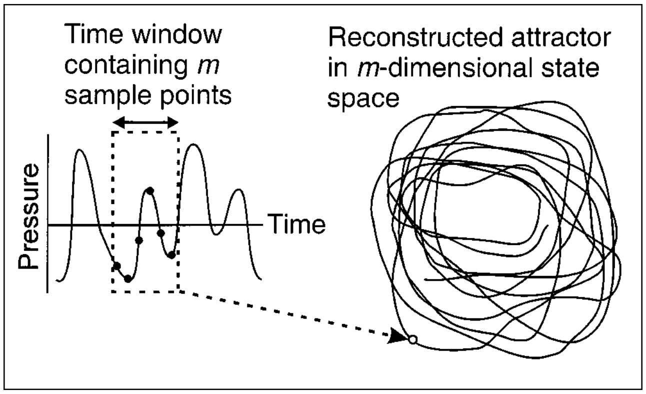

Figure 1. Reconstruction of an attractor in the *m*-dimensional state space from a pressure time series.

The *m* sample points in the time window in the pressure time series are projected onto a single point in the *m*-dimensional state space.

gave one example of discriminating between two fluidized-bed signals, but these signals were measured in two distinct fluidization regimes, and therefore easy to distinguish: also here it remains unclear how sensitive the method is. Moreover, the method does not give the standard deviation of the results: it is not clear whether observed differences are significant or not.

Kennel (1997) proposed a statistical test for detecting nonstationarity based on a nearest-neighbor-approach: for a certain point in a time series, this method looks for its nearest neighbor in the reconstructed state space. If the signal originates from a stationary process, all possible times between the two points have an equal chance to appear, but, for nonstationary processes, nearest neighbors are more often close in time. Using this method, Kennel (1997) was able to detect a significant difference in fluidization behavior with a change of 5% in superficial gas velocity in a fluidized bed.

Daw et al. (1998) and Finney et al. (1998) proposed symbol-sequence statistics as a tool for monitoring fluidization: their method observes how often a certain sequence of values is encountered in a pressure time series. It can be seen as a way of representing a coarse-grained attractor in one dimension. The method does not indicate whether observed differences are significant or not.

A problem with a number of the previously mentioned methods is that they do not give the proper statistics to decide whether a change in the fluidized state, indicated by the method, is significant or not. Moreover, the sensitivity of the methods has only been shown through experiments with a change in superficial gas velocity and not through experiments with a change in the particle-size distribution, which is much more relevant from an industrial point of view, but experimentally somewhat more difficult to carry out.

Schouten and Van den Bleek (1998) proposed to monitor the fluidized-bed hydrodynamics using the short-term predictability of pressure signals. Their method includes statistics to determine whether a change in the value yielded by that method is significant or not. Moreover, they illustrated the potential of their method by agglomeration experiments in a small laboratory-scale fluidized bed. However, as shown by Van Ommen et al. (1999a), the method is very sensitive to changes in the superficial gas velocity, because the amplitude of the pressure signal is included in the method. This is a disadvantage for industrial applications of the method, because large industrial fluidized beds will often show small variations in the gas supply. Therefore, Van Ommen et al. (1999a) proposed to remove the influence of the amplitude of the pressure signal by basing the method on the cycle time of the pressure signal; the cycle time of the signal can alternatively be seen as the orbital time of the reconstructed attractor. A disadvantage of the cycle time method is that it cannot easily be extended to a multiple-signal method, as required for application in large industrial equipment.

Monitoring Method Based on Attractor Comparison

General description of the method

We will now describe an enhanced monitoring method that is relatively insensitive to changes in the superficial gas velocity and can also handle multiple signals, which makes the method relevant for industrial application. The most promising methods up to now were based on the reconstructed attractor of the pressure signal. In these methods, the changes in a single property of the attractor (such as the distribution of nearest neighbors (Kennel, 1997), the short-term predictability (Schouten and Van den Bleek, 1998), or the orbital time (Van Ommen et al., 1999a)) are followed. We now propose a new approach by not focusing on a single attractor property, but by observing changes in the attractor *as a whole*. This makes the method more generic than the previously described methods: the different methods based on monitoring an attractor property are in fact all included in the present attractor comparison method.

The method detects if a certain change in the fluidized-bed hydrodynamics takes place, without giving information about the nature of the change. This is typical for statistical process control: it only indicates if something in the process under examination has been altered; by the use of other indicators and/or by knowledge of the process, the process operator should decide what has caused the alteration and, thereupon, which action has to be taken. However, when a monitoring method for fluidized beds is used to detect sporadic events such as agglomeration, it should not indicate changes in hydrodynamics due to a much more frequent event, such as small variations in the superficial gas velocity. The gas velocity mainly influences the standard deviation of the pressure signal; other properties of the pressure signal are hardly affected by small gas velocity changes. Therefore, the only property we exclude from comparison is the standard deviation of the pressure signals.

For the application of the attractor comparison method to pressure signals, we follow the same approach as proposed by Schouten and Van den Bleek (1998) for the method using the short-term predictability. The current method is also based on the comparison of an original time series (the *reference time series*) of pressure fluctuations with successive pressure fluctuations time series that are measured during operation of the bed; we will call these successive time series *evaluation time series*. The reference time series should reflect the required or optimum state of the fluidized-bed hydrodynamics well. The length of the reference and evaluation time series should be chosen such that a good representation of the fluidized-bed hydrodynamics can be obtained. In practice this implies that the time series should be of the order of a few minutes.

The essence of the method we currently propose is to compare in a statistical way the complete attractor reconstructed from the reference time series with the subsequent complete attractors reconstructed from the evaluation time series. This can by done by a statistical test, developed by Diks et al. (1996), that is specifically suited for comparing delay vector distributions.

Delay vector distributions

The test proposed by Diks et al. (1996) considers two sets of delay vectors (viz., two attractors) as multidimensional probability distributions, and estimates the squared distance between the two sets and the variance of the squared distance. Subsequently, the null hypothesis is tested that the two sets of vectors are drawn from the same probability distribution, viz., that the two sets are generated by the same dynamic mechanism. The Diks et al. test does not make *a priori* assumptions about the system under study: it can be applied to chaotic, regular, and noisy systems. The test is described in detail below.

Suppose we have a reference pressure time series $p_k = (p_1, p_2, \ldots, p_{Np})$ consisting of N_p values. To make the test less sensitive to the superficial gas velocity, we want to remove the influence of the standard deviation by normalizing the pressure time series. After calculating the average pressure $\bar{p}$ and the standard deviation σ_p, we can normalize p_k to x_k by

$$x_k = \frac{p_k - \bar{p}}{\sigma_p} \tag{1}$$

In the same way, we can convert an evaluation time series to a normalized evaluation time series y_k, also having a mean of zero and a standard deviation of unity. We can transform the reference time series x_k into a reference set $X_i = (x_{(i-1)m+1}, x_{(i-1)m+2}, \ldots, x_{i \cdot m})^T$ of N_x m-dimensional delay vectors with a distribution $\rho_x(X_i)$, and the evaluation time series y_k into an evaluation set Y_i of N_Y m-dimensional delay vectors with a distribution $\rho_y(Y_i)$, each distribution representing a reconstructed attractor with embedding dimension m. We will use the term "time window" for the time span described by one delay vector. This transformation from x_k to X_i is the same as the attractor reconstruction discussed in the section Earlier Monitoring Methods, except that we move with steps of size m through the time series instead of with steps of size one; this makes the total calculation procedure a factor m^2 times faster. Diks et al. (1996) used a step size of one because they mainly considered short time series (a few hundred points) and wanted to use the available data as optimally as possible. We normally use large time series (typically several ten-thousands of data points), because we want to obtain a more reliable characterization of the hydrodynamics. Since we use large time series and a rather high value of m (typically 20) in our application of the Diks et al. the test, it is worthwhile to increase the calculation speed with a factor of m^2 by using a step size m. We have found that this less optimal use of the available data only has a limited influence on the performance of the test.

The reference distribution $\rho_x(X_i)$ is converted into the smoothed reference distribution $\rho'_x(X_i)$ by convolution with a Gaussian kernel

$$\rho'_x(X_i) = \int \rho_x(R)\left(d\sqrt{2\pi}\right)^{-m} e^{-|X_i - R|^2/(2d^2)}\, dR \tag{2}$$

where $|\cdot|$ denotes the Euclidean distance in the m-dimensional state space and the band width d is a fixed length scale. The smoothed evaluation distribution $\rho'_y(Y_i)$ can be calculated in a similar way. The transformation means that the points in the state space are not represented with probability one at their exact values and zero probability elsewhere, but that they have a certain Gaussian probability distribution around their actual value. The motivation for introducing the smoothed distribution is that it is easy to give unbiased estimators for the smoothed distributions (Diks et al., 1996).

Distance between delay vector distributions

The extent to which two delay vector distributions differ can be expressed by the distance between the two distributions. The squared distance Q between the two smoothed distributions can be defined as

$$Q = \left(2d\sqrt{\pi}\right)^m \int \left[\rho'_x(R) - \rho'_y(R)\right]^2 dR \tag{3}$$

It can be proven that $Q = 0$ if and only if $\rho_x(R) = \rho_y(R)$. To calculate an estimator $\hat{Q}$ for Q, we will consider the two combined sets of delay vectors as the set Z consisting of $N_X + N_Y$ values. Moreover, we will introduce the function $h(Z_i, Z_j)$ needed to calculate the estimator $\hat{Q}$

$$h(Z_i, Z_j) = e^{-|Z_i - Z_j|^2/(4d^2)} \tag{4}$$

A crucial point in the Diks et al. test is that the dependence between the subsequent delay vectors (viz., attractor points that are close in time) is removed; this dependence would give a bias in the estimation of Q and its variance. To remove this dependence, Diks et al. divide the (i,j)-plane representing all possible pairs of Z_i and Z_j in $N \times N$ squares of size $L \times L$, with segment length L so large that pairs with $|i-j| \geq L$ are not correlated. For each square, an average value

$$h'_{pq} = \frac{1}{L^2} \sum_{i=1}^{L} \sum_{j=1}^{L} h\left(Z_{(p-1)L+i}, Z_{(q-1)L+j}\right) \tag{5}$$

is calculated. The first N_1 values of the indices p and q (ranging from 1 to N_1) refer to the reference set; the next N_2 values of the indices p and q (ranging from $N_1 + 1$ to N) refer to the evaluation set. An unbiased estimator $\hat{Q}$ can be calculated from the two sets of delay vectors by

$$\hat{Q} = \frac{2}{N_1(N_1-1)} \sum\sum_{1 \le p < q \le N_1} h'_{pq} + \frac{2}{N_2(N_2-1)} \sum\sum_{N_1+1 \le p < q \le N} h'_{pq} - \frac{2}{N_1 N_2} \sum_{p=1}^{N_1} \sum_{q=N_1+1}^{N} h'_{pq} \tag{6}$$

The larger the difference between two sets of delay vectors (each set representing a reconstructed attractor), the larger their squared distance Q will be. However, we will need to know whether a certain value of the estimator $\hat{Q}$ indicates a significant difference between the two delay vector sets, and, thus, between the two hydrodynamic states of the fluidized bed from which they originate, or whether it should be attributed to a statistical fluctuation. Therefore, we also need an estimate for the variance V_C. Diks et al. (1996) derived the following expression for the conditional variance

$$V_c(\hat{Q}) = \frac{4(N-1)(N-2)}{N_1(N_1-1)N_2(N_2-1)N(N-3)} \sum\sum_{1 \le p < q \le N} \psi_{pq}^2 \tag{7}$$

in which

$$\psi_{pq} = H_{pq} - g_p - g_q \tag{8}$$

where

$$H_{pq} = h'_{pq} - \frac{2}{N(N-1)} \sum\sum_{1 \le p' < q' \le N} h'_{p'q'} \tag{9}$$

and

$$g_p = \frac{1}{N-2} \sum_{\substack{q \\ p \ne q}} H_{pq} \tag{10}$$

We can now define a statistic S as

$$S = \frac{\hat{Q}}{\sqrt{V_C(\hat{Q})}} \tag{11}$$

which has an expectation value of zero and a standard deviation of unity under the null hypothesis of the two time series being generated by the same mechanism. Diks et al. (1996) did not prove if S has a normal distribution, but Pukkelsheim (1994) has shown that for any unimodal probability density, the probability for values larger than three times the standard deviation is smaller than 5%. We will therefore use a limiting value of $+3$; a higher S value indicates with more than 95% confidence that the null hypothesis of the two attractors being generated by the same system can be rejected. This is a conservative choice, since the test is only one-sided. Diks et al. (1996) showed by numerical simulations that the confidence level is about 98 to 99%, well above the 95% that can be strictly proven.

By calculating the S value for an evaluation pressure time series using a chosen reference time series, we can decide whether it was measured at similar hydrodynamic conditions as the reference time series or not: an S value larger than 3 indicates that a significant change has taken place in the hydrodynamic behavior of the fluidized bed. Two time series measured at the same hydrodynamic conditions should yield an S value of approximately zero.

In the remaining part of this article, a number of experiments will be described in which the hydrodynamics of a fluidized bed is altered in a controlled way by changing the particle-size distribution, the superficial gas velocity, or the bed mass. The practical application of the monitoring method using attractor comparison will be illustrated by these experiments.

Experimental Studies

To test the influence of superficial gas velocity, bed mass, and particle-size distribution on the monitoring method, five types of experiments were carried out:

(1) Experiments with stepwise changes in the superficial gas velocity

(2) Experiments with stepwise changes in the bed mass

(3) Experiments with stepwise changes in the particle-size distribution

(4) Experiments with subsequent stepwise changes in the superficial gas velocity, bed mass, and superficial gas velocity

(5) Experiments with gradual changes in the particle-size distribution.

Experimental facilities and operating conditions

In the experiments, four types of sand are used, of which the main properties are given in Table 1. The particle-size distributions of S1, S3, and S4 sand were determined by a laser diffraction system (Malvern Mastersizer S); the particle-size distribution of the S5 sand was determined by sieving.

Experiments with stepwise changes in the superficial gas velocity (type 1) were carried out in a perspex column with a circular cross-section and an internal diameter of 10 cm. The column was filled with 2.37 kg S3 sand (settled bed height 20 cm) and was operated at superficial gas velocities ranging from 0.22 to 0.38 m/s. The gas supply was controlled electronically, so that the gas velocity can be adjusted within 1%.

Experiments with stepwise changes in the bed mass (type 2) were also carried out in a perspex column with a circular cross-section and an internal diameter of 10 cm, filled with S3 sand. The bed mass was varied from 2.50 kg (settled bed

Table 1. Particle Properties of the Sands Used in the Experiments*

Sand Type	d_{10} [μm]	d_{50} [μm]	d_{90} [μm]	u_{mf} [m/s]	Geldart Type
S1	249	388	585	0.09	B
S3	356	532	760	0.14	B
S4	560	811	1,140	0.31	B−D
S5	1,710	1,920	2,290	1.0	D

*The particle diameters at 10, 50, and 90 vol. % of the size distributions, the minimum fluidization velocity, and the Geldart type are given.

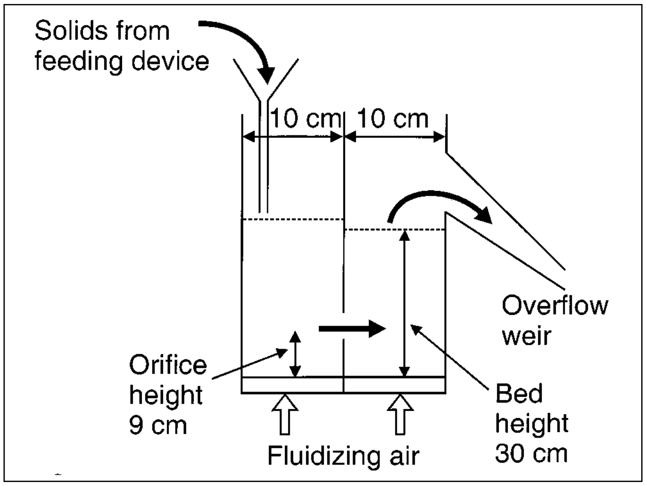

Figure 2. Overview of the twin-fluidized-bed facility.

This installation is used for experiments with a gradual change in the particle-size distribution (type 5).

height 21 cm) to 3.00 kg (settled bed height 25 cm). The superficial gas velocity was set at 0.26 m/s.

Experiments with stepwise changes in the particle-size distribution (type 3) were carried out in a perspex column with a circular cross-section and an internal diameter of 15 cm. The experiments were carried out with pure S3 sand and with S3 sand containing a mass fraction of 0.01, 0.02, 0.04, 0.08, 0.10, 0.16 and 0.32 S4 sand. The column was filled with a fixed mass of 7.83 kg (settled bed height 29 cm) and was operated at superficial gas velocities of 0.20, 0.28 and 0.36 m/s. Similar experiments with varying fractions of S5 sand were carried out at 0.28, 0.36, 0.46 and 0.60 m/s.

To illustrate the effect of changes in superficial gas velocity, bed mass, and particle-size distribution in one experiment (type 4), a 10 cm fluidized bed of S3 sand was operated for 120 min. At the start of the experiment ($t = 0$ min), the gas velocity was set at 0.30 m/s and the bed mass was 2.50 kg. From $t = 30$ min to $t = 60$ min, the gas velocity was 0.285 m/s (5% reduction); from $t = 45$ min to $t = 75$ min, the bed mass was 2.38 kg (5% reduction). From $t = 90$ min to $t = 120$ min, 5 wt. % of the bed material was replaced by S5 sand.

Experiments with a gradual change in the particle-size distribution (type 5) were carried out in a twin-fluidized bed: two compartments with a rectangular cross-sectional area of 10×10 cm, connected by a circular orifice of 20 mm diameter at 9 cm above the distributor. Figure 2 shows the twin-fluidized-bed facility. Ten minutes after the start of the experiments, a constant solids flow of 0.45 kg/min S3 sand was supplied to the first compartment, leading to a similar solids flow from the first to the second compartment through the orifice. A continuous solids flow was leaving the second compartment over an overflow weir (the opening above the weir was closed until the particle feed was started); in this way the total mass in the system remained constant. In both twin-bed compartments the superficial gas velocity was 0.30 m/s. The bed height in both compartments was about 30 cm; due to small fluctuations in the solids feed, the bed height fluctuated approximately 2 cm. Twin-bed experiments were carried out with S3 sand or S1 sand as starting material in both compartments of the twin bed. In that way, a constant particle-size distribution of S3 sand could be maintained or a "grade change" from S1 to S3 sand could be performed, respectively.

Pressure measurements and data acquisition

All types of experiments involved local pressure measurements in the bed. In the type 1 experiments, the pressure probe was located 8 cm above the distributor plate. In the type 2 and the type 4 experiments, the pressure was measured in the wind box and 4, 9, 14 and 19 cm above the distributor. In the type 3 experiments, the pressure was measured in the wind box and 1, 10, 15 and 20 cm above the distributor. In the experiments at 0.36 m/s, the pressure was also measured 30 cm above the distributor; at the lower gas velocities, this position is above the bed surface and is therefore not used. In the twin-fluidized bed (type 5 experiments) the pressure probe was located 8 cm above the distributor plate in the second compartment.

All pressure probes were 10 cm long and had an internal diameter of 4 mm; these dimensions guarantee an undisturbed transfer of the signal in the frequency range of interest (Van Ommen et al., 1999c, 2000). The end of each probe was placed flush with the fluidized-bed wall and was covered by a wire gauze to prevent particles from intruding and blocking the probe; this wire gauze has no effect on the pressure signal.

Piezoelectric pressure sensors of Kistler type 7261 connected to the probes were used to measure the pressure fluctuations. The charge from the piezo element is amplified and converted to a DC voltage signal using a Kistler amplifier type 5011. The sensitivities of amplification of the pressure signals were varied from 100 Pa/V to 300 Pa/V to utilize the amplifier output range (-10 V to $+10$ V) as optimally as possible. Due to a time constant that can be set in the amplifier, the lowest frequencies are high-pass filtered with a cut-off frequency of 0.16 Hz. By this filtering action, the fluctuations of the pressure are measured relative to the local average pressure, that is, the off-set of the signal is zero.

The signals were transferred to a Scadas II data acquisition system (Difa Measuring Systems, The Netherlands) connected to a personal computer, controlled by the software package D_TAC. The signals were low-pass filtered with a cut-off frequency of one-half or one-third of the sample frequency, satisfying the Nyquist criterion. Subsequently, 16 bits analog-to-digital conversion was applied at a sample frequency of 400 Hz. The time series were divided into parts of three minutes for evaluation by the monitoring method.

Results and Discussion

Parameter settings

The monitoring method should be as powerful as possible: it should give an S value close to 0 for two pressure time series measured at similar hydrodynamic conditions, whereas it should give a maximally high S value (at least > 3) for two pressure time series measured at different hydrodynamic conditions. The influence of four parameters on the performance of the method was investigated: (1) the time window, (2) the embedding dimension m, (3) the band width d, and (4) the segment length L. We optimized the parameter set-

Table 2. Optimal Parameter Settings for Applying the Attractor Comparison Test to the Fluidized Bed Signals

Time Window	Embedding Dimension (m)	Band Width (d)	Segment Length (L)
50 ms	20	0.5	3 s

tings by testing many different combinations of parameter settings for a large number of experiments at different superficial gas velocities and various measurements positions; the optimal parameter values are given in Table 2. These optimal values should be seen as an indication rather than exact values: limited deviations from these settings only have a minor influence on the test results.

To illustrate the influence of these four parameters, we applied the monitoring method to pressure time series measured at 1 cm above the distributor plate in the 15 cm ID column, operated at 0.20 m/s. One time series measured in a bed of pure S3 sand was used as the reference time series. Two sets of evaluation time series were used: a set of four evaluation time series measured at equal conditions as the reference time series and a set of five evaluation time series measured for a different bed composition (10 wt. % of S4 sand). All time series have a length of 180 s.

Time Window. The average cycle time was shown to be a good choice for the size of the time window when applying chaos analysis to fluidized-bed pressure signals (Schouten et al., 1994). This is due to the fact that one cycle corresponds to the physical phenomena taking place in the bed, such as bubble passage or bubble coalescence (Van der Schaaf et al., 1998a). In the experiments we present in this article, average cycle times typically range from 180 to 270 ms. In applying the attractor comparison test, a time window of about 50 ms gives the highest S values. This optimum value for the time window is considerably lower than the average cycle time.

Figure 3a shows that limited changes around the optimum time window value hardly affect the results of the test obtained for 10 wt. % S4 sand. Moreover, the S values stay well around 0 when no change in the hydrodynamic conditions has taken place (0 wt. % S4 sand). Since the average cycle time has about the same value in all currently presented experiments (15 cm bed, 10 cm bed, and twin-fluidized-bed experiments), we will use a time window of 50 ms throughout this article. However, when the method is applied to a differ-

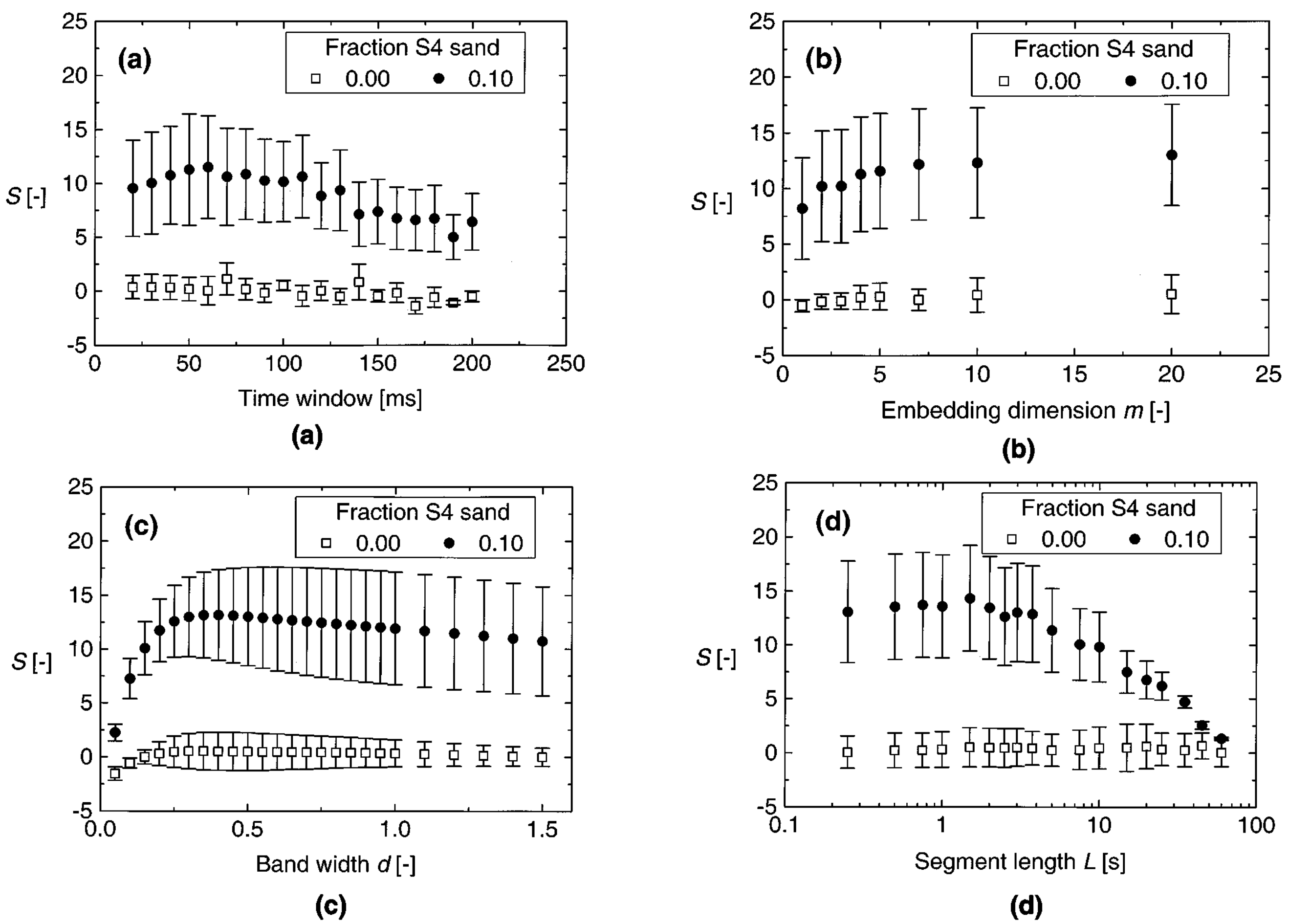

Figure 3. Influence of time window (a), embedding dimension *m* (b), bandwidth *d* (c), and segment length *L* (d) on the outcome of the attractor comparison test.

The points give the average of four or five S values, for 0 wt. % and 10 wt. % S4 sand, respectively. The error bars indicate the standard deviation. The reference time series has been measured in a bed with 0 wt. % S4 sand.

ent system with a very deviating average cycle time, the time window should be changed to about a quarter of that average cycle time as a first guess.

Embedding Dimension. When describing the signal in the optimal time window of 50 ms, we can use all measured pressure values. Since a sample frequency of 400 Hz is used, one time window contains 20 pressure values, so the embedding dimension m of the reconstructed attractor will be 20 (that is, one delay vector will contain 20 elements). Alternatively, two or more consecutive pressure values can be averaged, reducing the embedding dimension. Figure 3b shows that using all available pressure values gives the highest S values. Reducing the embedding dimension by a factor of two or three only has a limited effect on the outcome of the test, but larger reductions lead to a less powerful test. When calculation speed is important or when the data storage capacity is limited, it can be worthwhile to reduce the number of pressure values used. Moreover, when the signal is very noisy, reducing the embedding dimension by averaging a number of consecutive pressure values (at keeping the time window the same) will even increase the performance of the test, since the averaging removes a part of the noise (that is, it works as a low-pass filter).

Bandwidth. Diks et al. (1996) have shown that the choice of the bandwidth d has a considerable influence on the power of their test. By choosing it relatively small, the test will pick up local differences between the two distributions. Choosing it too small, however, leads to poor statistics. For too large a band width, the delay vector distributions are smoothed to such an extent that they become practically indistinguishable. The optimal value for the band width is expected to be found at the trade-off of these two effects (Diks et al., 1996). We have found that, for fluidized-bed pressure signals, the optimal band width lies between 0.2 and 0.7 times the standard deviation of the signal. We will use 0.5 as our standard value, but Figure 3c shows that limited deviations around this value only have a small influence on the outcome of the test.

Segment Length. The segment length L determines the number of successive values $h(Z_i, Z_j)$ over which the average value h'_{pq} is calculated (see Eq. 5). It is important to take it large enough to remove dynamic correlations between successive points in the state space. Diks et al. (1996) showed that too small a segment length leads to S values significantly different from 0 with a larger standard deviation than 1, while the null hypothesis is valid. Moreover, they state that the power of the test is near optimal as long as the number of segments is not smaller than 8. We have chosen to use a segment length of 3 s as our standard. This segment length is long enough to contain several cycles of the pressure signal, thus assuring that the different segments are not dynamically correlated anymore. On the other hand, this segment length is short enough so that one time series contains more than 8 segments. Figure 3d shows that a segment length of about 3 s yields good test results.

With these optimal parameter settings, the calculation of the S statistic is not very time consuming: on a Pentium III 550 MHz PC it takes 51 s to evaluate a time series of 3 min long, sampled with a frequency of 400 Hz. This means that the attractor comparison method can be used for on-line monitoring. The calculation time considerably increases when using a shorter time window, longer time series, or a higher sample frequency.

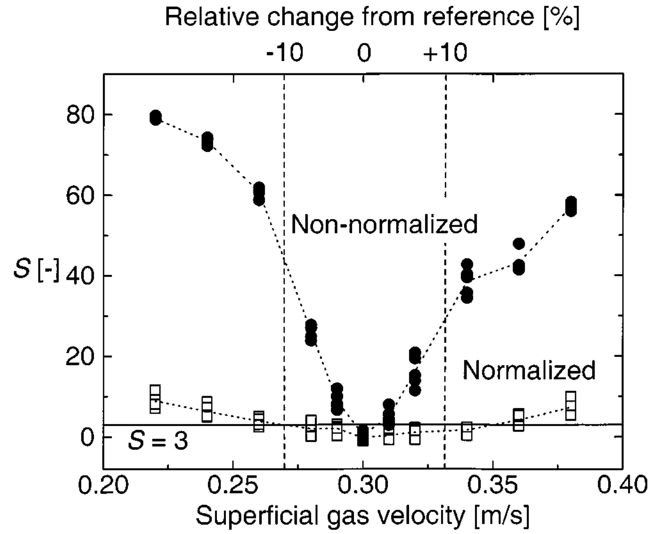

Figure 4. Influence of the superficial gas velocity on the *S* value.

The reference time series has been measured at 0.30 m/s. At each gas velocity, five time series have been evaluated with and without normalization. The solid line at $S = 3$ indicates the statistical limit above which a significant change has taken place. The dotted lines are meant to guide the eye.

Sensitivity to changes in superficial gas velocity

To reduce the sensitivity of the method to the superficial gas velocity, we normalize the pressure signal before applying the attractor comparison test, as described by Eq. 1. The influence of normalization is illustrated by experiments at various superficial gas velocities ranging from 0.22 m/s to 0.38 m/s. The S value is calculated for five time series; a time series measured at 0.30 m/s is used as the reference time series.

Figure 4 shows that without normalization the S statistic is very sensitive to changes in the superficial gas velocity. The S values calculated after normalization of the time series show a substantially reduced sensitivity to the gas velocity: for variations of the gas velocity smaller than 10% the S value stays below 3. For a reference situation at a higher superficial gas velocity, the sensitivity to the gas velocity decreases further; for a reference situation closer to minimum fluidization, the sensitivity increases.

It can be concluded that normalizing the pressure time series removes most of the influence of the superficial gas velocity. This conclusion only holds when a change in the superficial gas velocity does not lead to a hydrodynamic-regime change, such as from bubbling to slugging. In this case, many more properties of the pressure signal are affected. However, within one regime, the normalization of the pressure signal with respect to its average and standard deviation before applying the attractor comparison, is a useful way of making the monitoring method insensitive to *small* fluctuations in the superficial gas velocity.

Sensitivity to changes in bed mass

In industrial fluidized beds that are operated continuously by adding and removing particles, the bed mass will show small variations. The sensitivity of the monitoring method to the bed mass cannot be reduced as easily as the sensitivity to the gas velocity, since the bed mass does not only influence the intensity, but also the time-scale and shape of the pressure fluctuations, and thus of the attractor. However, the sensitivity to bed mass variation strongly depends on the pressure measurement position. This will be illustrated by experiments in a 10 cm ID column in which the bed mass is increased from 2.50 kg (settled bed height 21 cm) to 3.00 kg (settled bed height 25 cm) in eight steps. At every bed mass, the S value is calculated for six time series; a bed mass of 2.75 kg is used in the reference situation.

Figure 5 shows that the S value calculated for the pressure fluctuations measured at 19 cm above the distributor plate, is most sensitive to changes in bed mass. This is because this probe is closest to the bed surface. Hence, it is most influenced by the change in bed height due to an increase of the bed mass. The lower probe positions yield S values that are less sensitive to the bed mass. Especially the S value for the probe at 9 cm stays mainly below 3 for all bed mass changes $< 10\%$. It can be concluded that carefully choosing the measurement position makes the method insensitive to *small* bed mass variations.

Detection of changes in the particle-size distribution

The sensitivity of the attractor comparison method to changes in the particle-size distribution will now be illustrated by the experiments with a stepwise changing particle-size distribution (type 3 experiments). Pressure time series of 15 min have been measured in the 15 cm ID column containing mixtures of S3 sand and the coarser S4 sand at three gas velocities. The pressure time series have been divided in five parts of 3 min; for each of these partial time series, the S value has been calculated with a 3 min pressure time series measured in a pure S3 sand bed as the reference time series.

Figure 6 gives the S values as a function of the fraction S4 sand for three superficial gas velocities; the S values are calculated for the pressure time series measured at 20 cm above the distributor plate. The S statistic, comparing situations with and without coarse S4 sand, should logically increase with the fraction of S4 sand, but it is only capable to detect these changes at a sufficiently high S4 fraction. At the lowest superficial gas velocity (0.20 m/s), S already indicates a significant change in the hydrodynamics at a coarse sand fraction of 0.08. Figure 6 shows that the sensitivity of the method decreases considerably with increasing superficial gas velocity. When the method is used to detect agglomeration, this means that more or larger agglomerates may have been formed before agglomeration is detected. However, the sensitivity of the method decreases because the influence of the particle-size distribution on the hydrodynamics decreases at increasing gas velocity: close to minimum fluidization, defluidization will occur much earlier than at higher gas velocities. In other words, the "resistance" of the bed to agglomeration is larger at higher gas velocities. Therefore, we do not expect the reduced sensitivity at higher gas velocities to be a crucial problem. Figure 7 illustrates this: when an increasing amount of S5 sand (considerably larger particles than S4) is added to the bed, a significant change in the hydrodynamics is detected at much lower coarse sand fractions than for S4 sand (cf. Figure 6) up to a gas velocity of 0.60 m/s.

Still, we would like the method to be as sensitive as possible. In a previous section, we already investigated the influ-

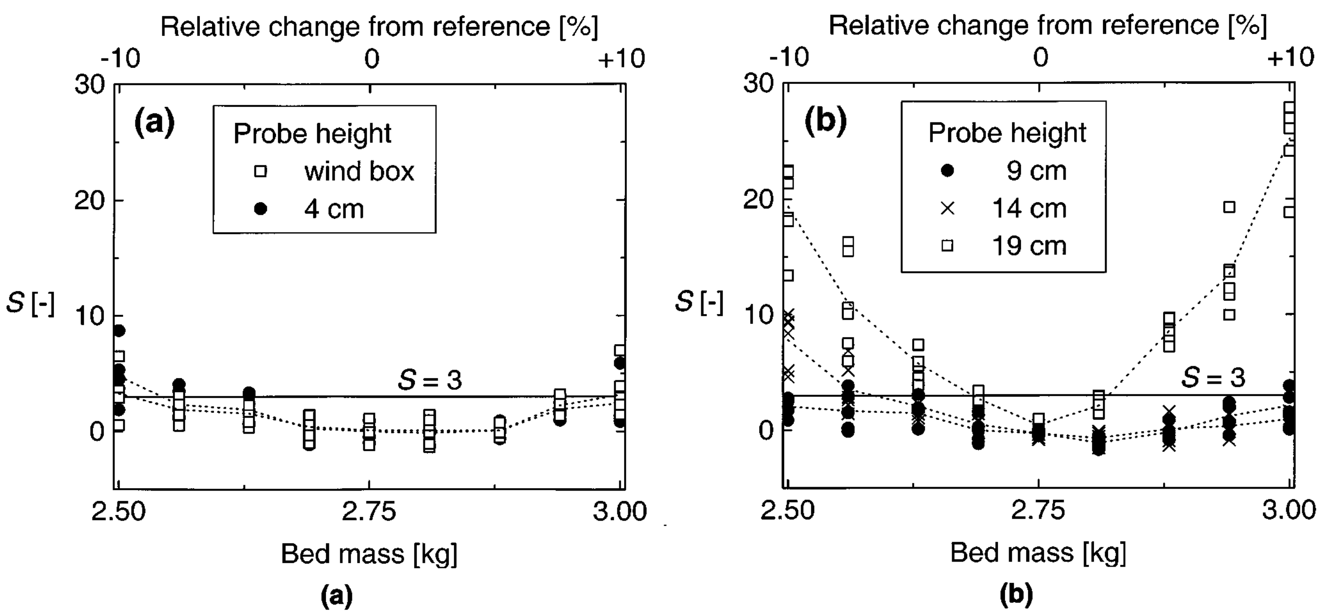

Figure 5. The influence of the bed mass on the *S* value.

The time series have been measured in the wind box and at 4 cm height (a), and at 9, 14 and 19 cm height (b). The reference time series has been measured at 2.75 kg. At every bed mass, six time series have been evaluated. The solid line at $S = 3$ indicates the statistical limit above which a significant change has taken place. The dotted lines are meant to guide the eye.

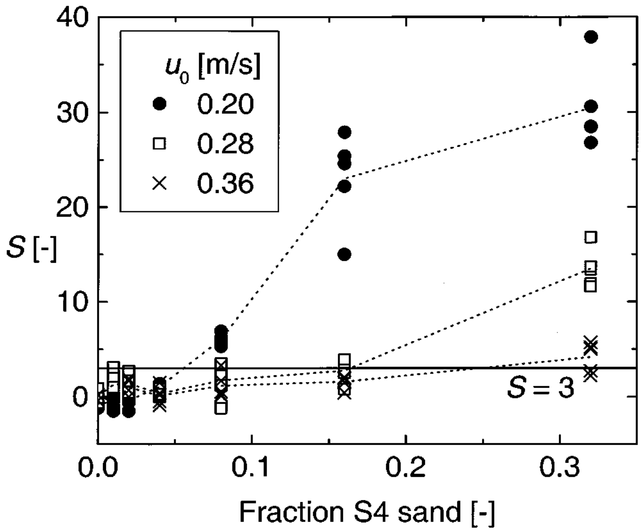

Figure 6. Influence of a change in the particle-size distribution on the S value.

At seven fractions of S4 sand and at three superficial gas velocities, five time series have been evaluated. For each gas velocity, a reference time series has been measured in a bed containing only S3 sand. The pressure time series have been measured at 20 cm above the distributor. The solid line at $S = 3$ indicates the statistical limit above which a significant change has taken place. The dotted lines are meant to guide the eye.

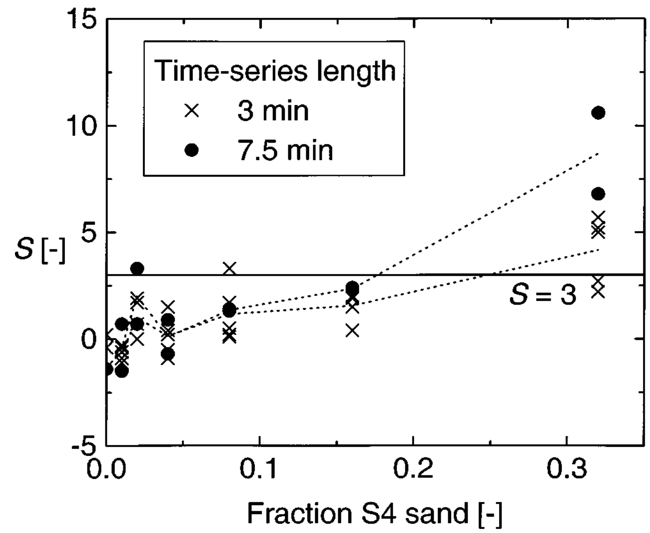

Figure 8. Influence of the time series length on the S value.

The pressure signal measured during 15 min at seven S4 sand fractions has been divided into five parts of 3 min and into two parts of 7.5 min, and subsequently evaluated. A time series measured in a bed containing only S3 sand has been used as the reference time series. The pressure time series have been measured at 20 cm above the distributor; the superficial gas velocity was 0.36 m/s. The solid line at $S = 3$ indicates the statistical limit above which a significant change has taken place. The dotted lines are meant to guide the eye.

ence of the parameter settings on the attractor comparison method; the optimal parameter settings are now used. However, there are two other ways to optimize the sensitivity of the method: increasing the length of the pressure time series and determining the optimum measurement position. The influence of the time series length is shown in Figure 8. Since the attractor comparison method is a statistical test, a larger amount of data will lead to better results (viz., a higher S value in the case of a difference between the two time series). This is indeed illustrated by Figure 8, in particular at the highest coarse sand fraction. Of course, increasing the time series length is limited. When on-line monitoring a fluidized-bed process, the use of longer time series will lead to longer times before the S value is updated. When this time becomes too long, an irrevocable change may already have taken place before the method has been able to send out a warning.

A second way to increase the sensitivity of the method is by optimizing the position of the pressure probe. This is illustrated in Figure 9, where the S value for 32 wt. % coarse sand is given as a function of the vertical probe position in the bed. For the lowest superficial gas velocity, the best position to measure the pressure is near the bottom of the bed, since segregation takes place: the larger particles are mainly located in the lower part of the bed. However, the choice of the measurement position is not crucial for this gas velocity: the change in hydrodynamics is clearly reflected at all measurement positions. At the highest superficial gas velocity, an additional measurement position at 30 cm height can be used. Whereas the pressure probe at 30 cm is above the bed surface at 0.20 m/s and at 0.28 m/s and does not give sensible results, it is under the bed surface at 0.36 m/s due to bed expansion. Figure 9 shows that applying the attractor comparison method to the pressure signals measured at this position gives the highest S values for 0.36 m/s. In other words, measuring just below the bed surface makes the monitoring

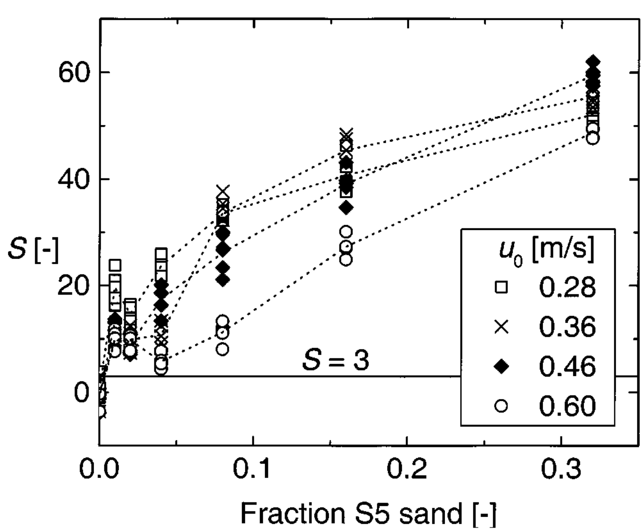

Figure 7. Influence of coarser particles and higher gas velocities on the S value.

At seven fractions S5 sand and at four superficial gas velocities, five time series have been evaluated. For each gas velocity, a reference time series has been measured in a bed containing only S3 sand. The pressure time series have been measured at 20 cm above the distributor. The solid line at $S = 3$ indicates the statistical limit above which a significant change has taken place. The dotted lines are meant to guide the eye.

method most sensitive to changes in the particle-size distribution for a superficial gas velocity of 0.36 m/s. However, it also makes the method sensitive to changes in the bed mass, as shown in the previous section.

At 0.36 m/s, S increases with increasing probe height, possibly because the changed particle-size distribution influences the bubble size. Near the bottom of the bed, the pressure signal is mainly dominated by fast-moving pressure waves, but the higher the probe position, the larger the influence of passing bubbles on the pressure signal (Van der Schaaf et al., 1998b). Another reason may be that the changing particle-size distribution leads to a change in voidage and thus in bed height. This will mostly affect the pressure fluctuations just below the bed surface. For the S values calculated for the 0.28 m/s pressure time series, no obvious trend can be seen. It is likely that at this gas velocity both the segregation observed at 0.20 m/s (decreased S with increasing probe height) and the phenomenon observed for 0.36 m/s (increased S with increasing probe height) influence the outcome of the test.

Figure 9 additionally shows that the pressure time series measured in the wind box yield about the same S values as those measured just above the distributor plate. This is relevant for industrial application of the method: when the operating conditions in the fluidized bed are such that measuring the pressure in the bed is impossible (such as, strongly oxidizing), measuring the wind box pressure can be an option. However, measuring pressure fluctuations in the wind box is only sensible if the pressure drop over the distributor is not too high; otherwise, the pressure fluctuations in the wind box will not be representative for the bed dynamics. We intend to carry out a more extensive investigation of the influence of the probe position on the test results in the near future.

Different disturbances in one experiment

To show the influence of the different disturbances that can occur in a practical situation, we carried out an experiment of 120 min duration with a temporary decrease in superficial gas velocity by 5%, followed by a temporary decrease in bed mass by 5%. Finally, 5 wt. % of the bed of pure S3 sand is replaced by the coarser S5 sand to simulate initial agglomeration. The points in time at which these changes take place are graphically depicted in Figure 10a.

The S values calculated for the five probe positions are shown as a function of time in Figures 10b and 10c; the pressure time series measured during the first 3 min of the experiment are used as the reference time series. The decrease of the gas velocity slightly influences the S value for the 19 cm probe due to the decrease in the dynamic bed height. The S values for the other measurement positions stay below 3. As expected, the decrease of the bed mass is most severely reflected by the S value for the probe at 19 cm height and to a lesser extent for the 14-cm probe; this is also due to the bed height effect. For all probe positions, the S statistic detects a significant change in the hydrodynamics almost immediately after the alteration in the particle-size distribution. Since the probe at 9 cm height (that is, around the center of the bed) is relatively insensitive to changes in the superficial gas velocity and the bed mass, and shows a high sensitivity to a change in the particle-size distribution, this probe position is the best choice for selective monitoring of the bed composition.

Ability of the method to follow a grade change

A monitoring method for fluidization hydrodynamics may not only be used to warn for agglomeration or other unwanted changes in the bed, but also to indicate if a new stationary situation has been reached after an imposed change. An example is a fluidized bed with a continuous feed and removal of solids in which the composition of the feed is changed at a certain moment to obtain a different product composition. Such a grade change is illustrated by the twin-fluidized-bed experiments (type 5). In a first experiment, both compartments are initially filled with S3 sand. Ten minutes after the start of the experiment, S3 sand is added to the first compartment: the sand is now flowing through both compartments, but the particle-size distribution remains unaltered. The first compartment is only used for the mixing of the particle feed with the bed material; in the second compartment, the pressure is measured (at 8 cm above the distributor plate) to monitor the changes in the particle-size distribution. Three minutes long pressure time series are evaluated using the attractor comparison method; the pressure time series measured during the first 3 min of the experiment serves as the reference time series. Figure 11a shows that the S value stays well below 3 during the experiment, correctly indicating that no change in the hydrodynamics takes places.

A second twin-fluidized-bed experiment was carried out in the same way as the first one, except that both compartments were initially filled with the finer S1 sand. Therefore, after ten minutes—when the particle feeder is started—the particle-size distribution begins to change. It has been shown previously (Van Ommen et al., 1999b) that a simple model for the median particle size in the twin-fluidized bed can be obtained by regarding both compartments as ideally stirred vessels for the particles. The median particle diameter as a func-

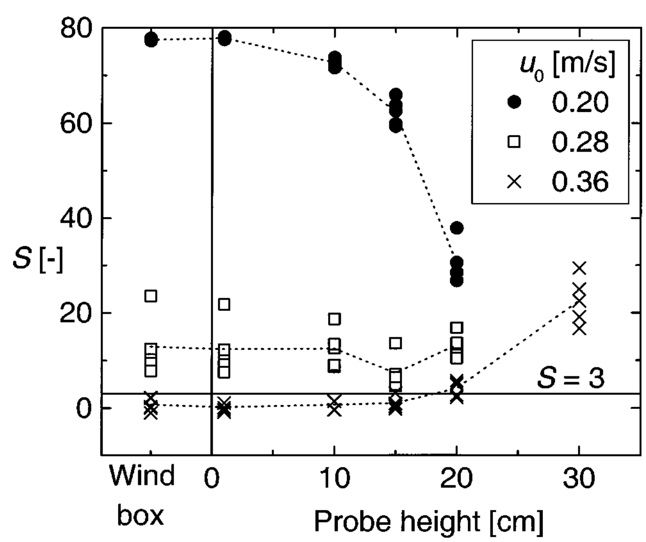

Figure 9. Influence of the measurement position on the S value.

Time series measured in a bed containing only S3 sand were used as the reference time series; time series measured in a bed containing 32 wt. % S4 sand were used as the evaluation time series. The solid line at $S = 3$ indicates the statistical limit above which a significant change has taken place. The dotted lines are meant to guide the eye.

tion of time obtained by this model is shown in Figure 11b by the dashed curve.

The solid circles in Figure 11b show the S value as a function of time when the pressure time series measured during the first three minutes of the experiment is used as the reference time series. It can be seen that the S value stays around 0 in the beginning of the experiment, but then increases as the median particle diameter in the bed increases and quickly exceeds 3: the hydrodynamics has changed significantly. The open squares in Figure 11b show the S value as a function of time when a pressure time series measured earlier for pure S3 sand (equal to the final situation of the bed) is used as the reference situation. It can be seen that in the beginning, as the bed is still filled with S1 sand, the S statistic indicates a significantly different situation (that is, $S > 3$). About 40 min after the start of the experiments—when the median particle diameter approaches its final value—the S value decreases below 3, the statistical limit for a significant difference between the reference and the evaluation time series. This shows that the monitoring method rightly indicates that the hydrodynamics has reached the desired situation, reflected by the reference time series obtained in an earlier experiment.

Extension to a multiple-signal method

For the laboratory-scale fluidized beds we used in our experiments, one probe position will be enough to monitor the hydrodynamics of the complete bed, although we already showed that the choice of this position can have a large influence on the outcome of the monitoring method. It is questionable whether one probe will also be sufficient in large, industrial installations. It is likely that by using only one probe, changes in the hydrodynamics at a distant position in the bed cannot be detected early enough; multiple probes will be needed to get a complete picture of the hydrodynamic situation. It is, of course, possible to apply the above described monitoring method to each probe separately, but it

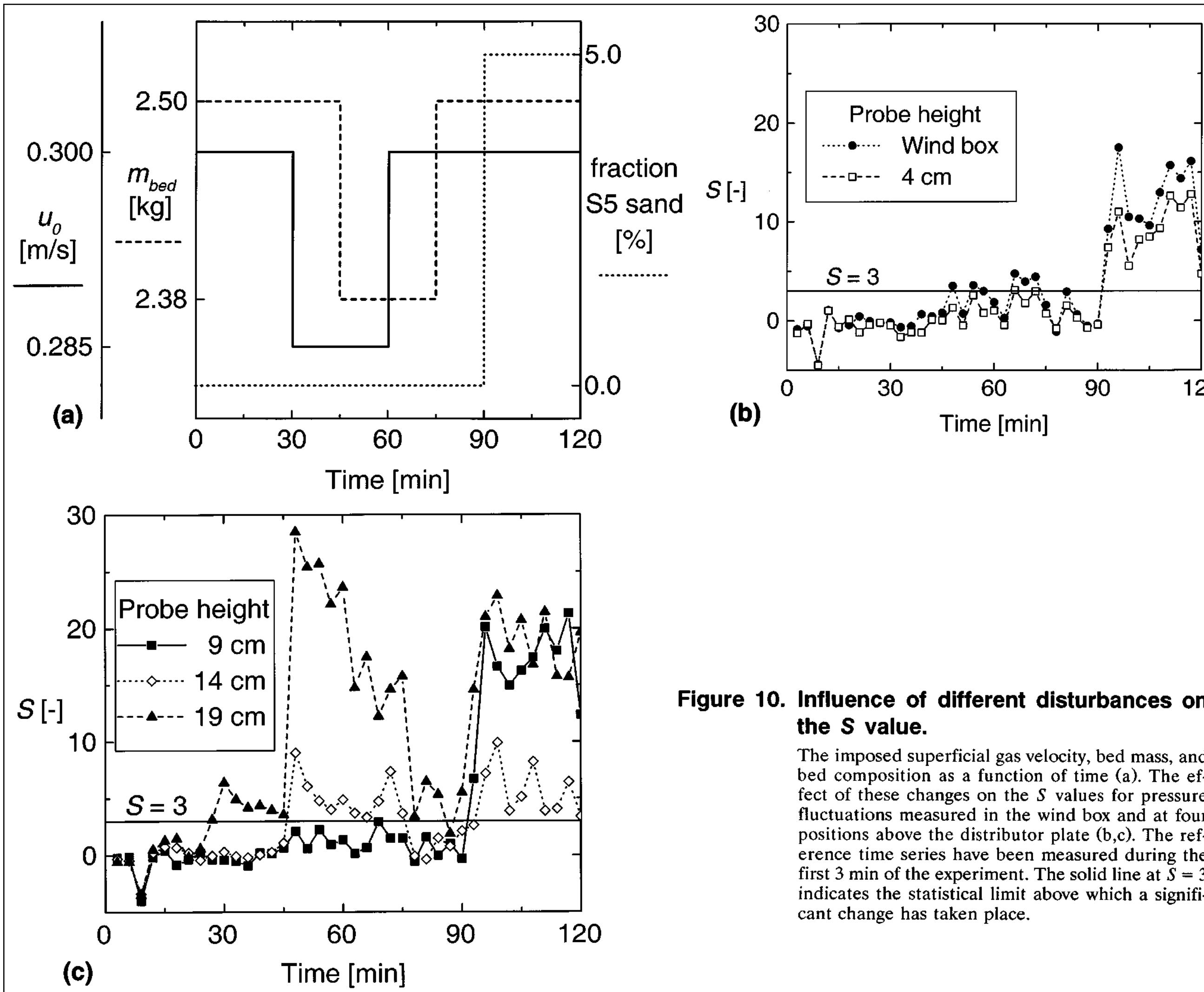

Figure 10. Influence of different disturbances on the S value.

The imposed superficial gas velocity, bed mass, and bed composition as a function of time (a). The effect of these changes on the S values for pressure fluctuations measured in the wind box and at four positions above the distributor plate (b,c). The reference time series have been measured during the first 3 min of the experiment. The solid line at $S = 3$ indicates the statistical limit above which a significant change has taken place.

would be worthwhile to incorporate all signals into one test, especially when the number of probes is large. In that case one S statistic is obtained, which serves as a characteristic for the hydrodynamics of the complete bed. Of course, it would still be possible to calculate the separate S values for each probe position representing the local hydrodynamics as well.

The extension of the previously described single-signal method to a multiple-signal variant is very straightforward. We will illustrate this method using experiments carried out in the 15 cm ID column at 0.36 m/s. Instead of using only pressure time series measured at a single probe position, four simultaneously measured pressure time series are used. We will denote these pressure time series by $p_{j,i}$, where j indicates the position and i indicates the time. After normalization (see Eq. 1) we have four time series $x_{j,i}$, which can be converted into one set of delay vectors X_i with $4 \times n$ elements, where $X_i = (x_{1,(i-1)n+1}, x_{2,(i-1)n+1}, x_{3,(i-1)n+1}, x_{4,(i-1)n+1}, x_{1,(i-1)n+2}, \ldots, x_{3,i \cdot n}, x_{4,i \cdot n})^T$. We have used $n = 5$, so that the delay vector contains 20 values, such as for the case with only a single pressure signal. To retain a time window of 50 ms, we resample the pressure signal from a sample frequency of 400 Hz to 100 Hz. Although the delay vectors contain delay coordinates from four measurement positions, they can still be treated in exactly the same way as for the single position delay vectors described in Eqs. 2 to 11.

Figure 12 presents the trends for the S values calculated for a pressure signal measured at 1, 10, 15, and 20 cm above the distributor plate as a function of the S4 mass fraction. It also shows the S values calculated with the use of the multiple-signal approach described above. The figure shows that the multiple-signal S value is not just an average of the four single-signal S values: it is about as sensitive as the most sensitive single-signal S value (the S value for the probe at 20 cm height). We expect the multiple-signal approach to show its surplus value in large (industrial) fluidized beds, where one probe position is insufficient to detect changes in the hydrodynamics over the complete bed. However, further research is needed to optimize the spatial positioning of multiple probes in a large fluidized bed.

Conclusions

(1) A method has been presented for early detection of changes in fluidization behavior. The method is based on reconstruction of the attractor from a pressure fluctuation signal measured in the fluidized bed. The technique of attractor reconstruction, originating from chaos theory, gives a representation of the dynamic state of the fluidized bed.

(2) The method compares the attractor of a reference pressure fluctuations time series reflecting a certain desired state of fluidization with the attractor of successive time series acquired during operation of the fluidized bed. The degree to which the reference time series and the successive time series differ is expressed by a statistical quantity, the so-called S value. This value is close to zero under the null hypothesis that the two hydrodynamic situations are similar; when the S value is larger than 3 the null hypothesis can be rejected with more than 95% confidence.

(3) The parameter settings for the method have been optimized to maximize the power of the method. The exact values of the parameters are not crucial: moderate variations around the optimal parameter values only have a limited influence on the outcome of the method.

(4) Normalizing the pressure time series with respect to their own average and standard deviation before attractor reconstruction makes the method insensitive to small changes in the superficial gas velocity. This is important for industrial application of the method, since a high sensitivity to the gas velocity may lead to "false alarms" caused by fluctuations in the gas supply.

(5) By measuring the pressure too close to the bed surface, the method becomes very sensitive to changes in the bed mass.

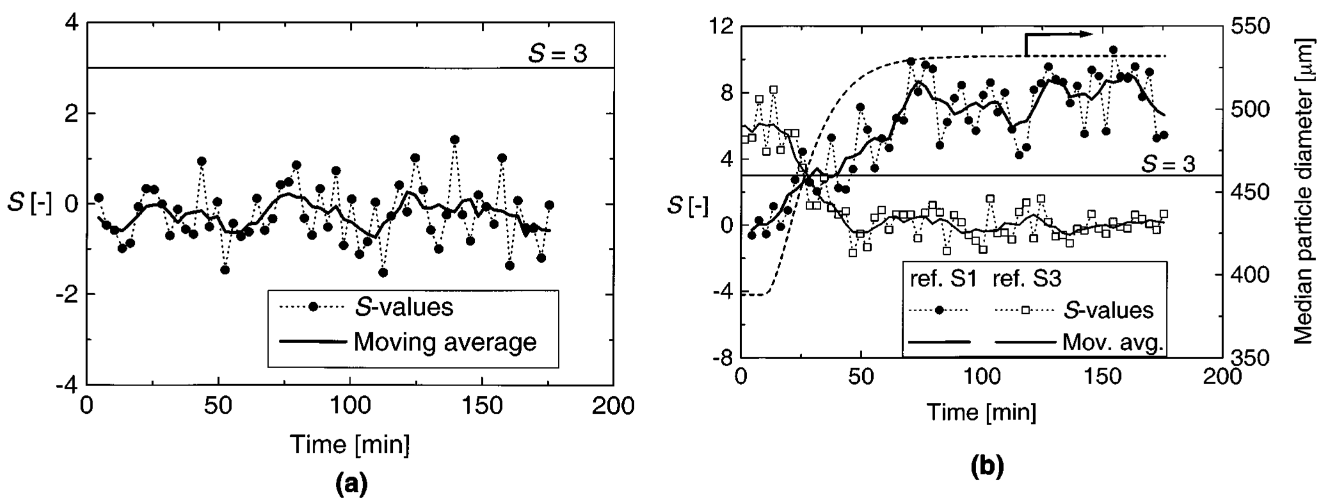

Figure 11. S value as a function of time for the twin-fluidized-bed experiments.

The S values have been calculated for a constant bed content of S3 sand (a), and for a grade change from S1 sand to S3 sand (b). In the latter case, both a time series measured with pure S1 sand and one measured with pure S3 sand have been used as reference time series. The thick lines give a moving average of the S value over 15 min (5 S values) to show the trends more clearly. The solid line at $S = 3$ indicates the statistical limit above which a significant change has taken place. In (b) the dashed curve gives the median particle diameter calculated by the two-ideally-stirred-vessels-in-series model.

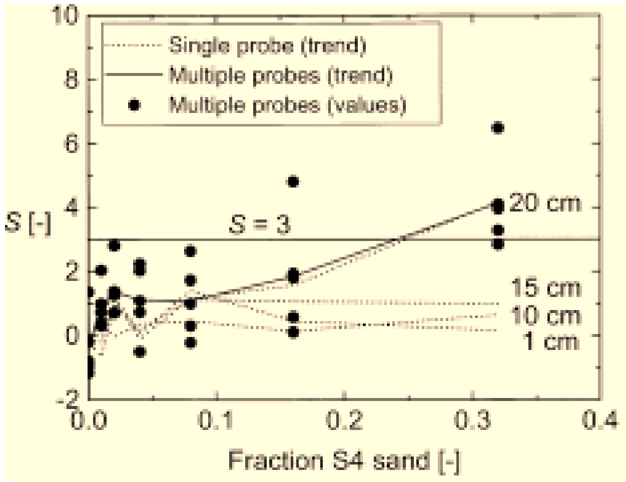

Figure 12. Single-signal method vs. multiple-signal method.

Pressure time series have been measured at 1, 10, 15, and 20 cm above the distributor; the superficial gas velocity was 0.36 m/s. For the single-signal method only the trend is given by the dotted curves; the probe heights are given near these curves. For the multiple-signal method, both the trend and the individual S values of five evaluated time series for each mass fraction are given. Time series measured in a bed containing only S3 sand have been used as the reference time series. The solid line at $S = 3$ indicates the statistical limit above which a significant change has taken place.

When the pressure is measured at a lower position (such as around the center of the bed), the method is insensitive to small bed mass variations.

(6) The method is sensitive to changes in the particle-size distribution. This makes it a useful tool for early warning in the case of incipient agglomeration. Furthermore, it can be used to indicate if a new stationary situation has been reached when performing a grade change (a change from one type of particle to another type in a through-flow type fluidized bed).

(7) The sensitivity of the method to changes in the particle-size distribution is largest at low gas velocity; this is a good property, since the ‘‘resistance’’ to agglomeration is smallest at low gas velocities. The decreased sensitivity at higher gas velocities can partially be overcome by using longer time series and by carefully choosing the probe position.

(8) The method is relatively easy from a technical point of view (pressure measurements are commonly used in fluidization practice), while the data analysis is neither time-consuming nor complicated and could be done on-line. Moreover, it has been demonstrated that the method can be easily extended to a multiple-signal variant, capable of monitoring large installations. Implementation of the monitoring method in an industrial fluidized bed is therefore considered to be realistic. However, further research is needed to optimize the spatial positioning of multiple probes in a large fluidized bed.

Acknowledgment

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Notation

d = band width for smoothing of points in the state space
d_{10} = particle diameter at 10 vol. % of the size distribution, μm
g_p = function defined in Eq. 10
$h(Z_i, Z_j)$ = function defined in Eq. 4
h'_{pq} = average value of h over $L \times L$ pairs of Z_i and Z_j
H_{pq} = function defined in Eq. 9
L = segment length
m = embedding dimension
m_{bed} = bed mass, kg
N = number of segments (of length L) in set Z
N_1 = number of segments in set Z originating from set X
N_2 = number of segments in set Z originating from set Y
N_p = number of values in pressure time series p
N_X = number of vectors in set X
N_Y = number of vectors in set Y
p_i = pressure value, Pa
$\bar{p}$ = average pressure, Pa
$\mathbf{P}_k$ = vector of pressure values
Q = squared distance between two attractors
$\hat{Q}$ = estimator for Q
$\mathbf{R}$ = point in the state space
S = estimator for the normalized squared distance between two attractors
u_0 = superficial gas velocity, m/s
u_{mf} = superficial gas velocity at minimum fluidization conditions, m/s
V_C = conditional variance
x_k = normalized pressure value in reference time series
$\mathbf{X}_i$ = vector of normalized pressure values in reference time series
y_k = normalized pressure value in evaluation time series
$\mathbf{Y}_i$ = vector of normalized pressure values in evaluation time series
Z = combined set of vectors of set X and set Y
$\rho_x(\mathbf{X}_i)$ = distribution of set X
$\rho'_x(\mathbf{X}_i)$ = smoothed distribution of set X
σ_p = standard deviation of p, Pa
ψ_{pq} = function defined in Eq. 8

Literature Cited

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

Daw, C. S., C. E. A. Finney, K. Nguyen, and J. S. Halow, ‘‘Symbol Statistics: a New Tool for Understanding Multiphase Flow Phenomena,’’ *Proc. of ASME Heat Transfer Division - 1998*, **5**, R. A. Nelson, Jr., T. Chopin, S. T. Thynell, eds., ASME, New York, 221 (1998).

Daw, C. S., and J. S. Halow, ‘‘Evaluations and Control of Fluidization Quality Through Chaotic Time Series Analysis of Pressure-Drop Measurements,’’ *AlChE Symp. Ser.*, **89**(296), 103 (1993).

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

Finney, C. E. A., K. Nguyen, C. S. Daw, and J. S. Halow, ‘‘Symbol-Sequence Statistics for Monitoring Fluidization,’’ *Proc. of ASME Heat Transfer Div. - 1998, Vol. 5*, R. A. Nelson, Jr., T. Chopin, S. T. Thynell, eds., ASME, New York, 405 (1998).

Kai, T., and S. Furusaki, ‘‘Methanation of Carbon Dioxide and Fluidization Quality in a Fluidized Bed Reactor—the Influence of a Decrease in Gas Volume,’’ *Chem. Eng. Sci.*, **42**, 335 (1987).

Kennel, M. B., ‘‘Statistical Test for Dynamical Nonstationarity in Observed Time Series Data,’’ *Phys. Rev. E*, **56**, 316 (1997).

Pukkelsheim, F., ‘‘The Three Sigma Rule,’’ *The American Statistician*, **48**, 88 (1994).

Schouten, J. C., F. Takens, and C. M. van den Bleek, ‘‘Estimation of the Dimension of a Noisy Attractor,’’ *Phys. Rev. E*, **50**, 1851 (1994).

Schouten, J. C., and C. M. van den Bleek, ''Monitoring the Quality of Fluidization Using the Short-Term Predictability of Pressure Fluctuations,'' *AIChE J.,* **44**, 48 (1998).

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, Dynamical Systems and Turbulence,* D. A. Rand and L.-S. Young, eds., Springer Verlag, Berlin, Germany, p. 366 (1981).

Van der Schaaf, J., J. C. Schouten, and C. M. van den Bleek, ''Origin, Propagation and Attenuation of Pressure Waves in Gas-Solid Fluidized Beds,'' *Powder Technol.,* **95**, 220 (1998a).

Van der Schaaf, J., J. C. Schouten, F. Johnsson, and C. M. van den Bleek, ''Decomposition of Power Spectral Density in Gas-Solids Fluidized Beds,'' *Proc. Third Int. Conf. on Multiphase Flow 98,* Lyon, France (1998b).

Van Ommen, J. R., J. C. Schouten, and C. M. van den Bleek, ''An Early-Warning-Method for Detecting Bed Agglomeration in Fluidized Bed Combustors,'' Paper No. FBC99-0150, *Proc. 15th Int. Conf. on Fluidized Bed Combustion,* R. B. Reuther, ed., ASME, New York (1999a).

Van Ommen, J. R., J. C. Schouten, and C. M. van den Bleek, ''Monitoring Fluidization Dynamics for Detection of Changes in Fluidized Bed Composition and Operating Conditions,'' *J. Fluids Eng.,* **121**, 887 (1999b).

Van Ommen, J. R., J. C. Schouten, M. L. M. vander Stappen, and C. M. van den Bleek, ''Response Characteristics of Probe-Transducer Systems for Pressure Measurements in Gas-Solid Fluidized Beds: How to Prevent Pitfalls in Dynamic Pressure Measurements,'' *Powder Technol.,* **106**, 199 (1999c).

Van Ommen, J. R., J. C. Schouten, M.-O. Coppens, and C. M. van den Bleek, ''Monitoring Fluidization by Dynamic Pressure Analysis,'' *Chem. Eng. Technol.,* **22**, 773 (1999d).

Van Ommen, J. R., J. C. Schouten, M. L. M. vander Stappen, and C. M. van den Bleek, ''Erratum to 'Response Characteristics of Probe-Transducer Systems for Pressure Measurements in Gas-Solid Fluidized Beds: How to Prevent Pitfalls in Dynamic Pressure Measurements,''' *Powder Technol.,* **106**, 199 (1999); **113**, 217 (2000).

Werther, J., ''Measurement Techniques in Fluidized Beds,'' *Powder Technol.,* **102**, 15 (1999).

Wright, J., ''Monitoring Changes in Time of Chaotic Nonlinear Systems,'' *Chaos,* **5**, 356 (1995).

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