Monitoring of Fluidized Beds Hydrodynamics Using Recurrence Quantification Analysis

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*A new method is presented for on-line monitoring of fluidized beds hydrodynamics using pressure fluctuations signal by recurrence quantification analysis. The experiments were carried out at different gas velocities and sand types. A 95% confidence interval was computed for determinism (*Det*) of signals obtained from reference state as well as other operating conditions named as unideal states.* Det *of unideal states was compared with* Det *of the reference state to reject the null hypothesis that all the signals have been generated from the reference state. It was shown that* Det *is sensitive to small change in particles size whereas it is not sensitive to minor superficial gas velocity variations, indicating its ability for hydrodynamic on-line monitoring. Furthermore, in this method it is no need for time series embedding, long-term data sampling and time-consuming numerical algorithms.* © 2012 American Institute of Chemical Engineers *AIChE J*, 59: 399–406, 2013
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Introduction

Gas-solids fluidized beds are extensively used in various multiphase flow industries such as pharmaceutical, food, agricultural, mineral, catalytic cracking, and gas-phase polymer production. The wide application of fluidized beds is due to their thermal homogeneity, efficient contact between fluid and particulate phases, high mixing ability and high heat, and mass transfer rates.

In many large scale applications, continuous processes are more efficient than batch processes, making it necessary to implement effective monitoring strategies. For example, in pharmaceutical industries, fluidized beds are widely applied in mixing, granulation, coating, and drying processes. As Food and Drug Administration guidance on process analytical technology suggests the drug manufacturers to upgrade the products quality using continuous operations, efficient monitoring methods find special importance.[1] In these applications of fluidized beds, unwanted changes in fluidization parameters, such as particle size, and gas velocity and temperature, should be detected quickly to prevent undesirable situations (for example, agglomeration and defluidization) leading to inappropriate product quality. The hydrodynamics of fluidization are determined by its parameters. Therefore, the fluidization parameters can be monitored by on-line characterization of the fluidization hydrodynamics. On-line monitoring of fluidized beds hydrodynamics is achieved through time series evaluation of measured signals, such as local pressure fluctuations or void fraction of the bed. Because

pressure fluctuations are easy to measure in both industrial and laboratory units[2] and includes the effect of various hydrodynamic phenomena taking place in the bed, such as gas turbulence and bubbles hydrodynamics,[3] it is applied in this work for the monitoring of small changes in fluidized bed hydrodynamics.

Pressure measurement-based monitoring methods can be divided into two categories: linear and nonlinear methods. Many researchers have applied linear methods to monitor fluidized beds hydrodynamics.[4] However, as the hydrodynamics of gas-solids fluidized bed are governed by complex nonlinear dynamic relationships and are mainly controlled by different dynamic phenomena occurring in the bed (such as bubble formation, bubble coalescence, and splitting, bubble passage, bubble eruption as well as particles behaviors), a proper understanding of the state of the fluidized bed at a certain time could not be determined by linear methods. In addition, in many cases (e.g., monitoring of the bed hydrodynamics for early detection of agglomeration), linear methods are not applicable to the industrial applications because they are sensitive to superficial gas velocity, which fluctuates in many industrial applications.[4] Nonlinear methods, such as short-term predictability proposed by Schouten and Van den Bleek[5] and attractor comparison in the state space,[6] can investigate intrinsic nonlinear dynamics of fluidized bed more accurate than linear methods. However, these methods are accompanied with some difficulties such as long-term data sampling required in the analysis, time-consuming numerical algorithms, and uncertainty in the determination of embedding parameters.[7]

In this work, a monitoring method based on the recurrence plot (RP)[8] and recurrence quantification analysis (RQA)[9] was developed for detecting small changes in the fluidized bed particles size. Using this method, the problems

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associated with long-term data sampling which is required in typical nonlinear methods are solved. Furthermore, while embedding is required for reconstruction of attractor in state space, RP may be constructed without embedding. All information contained in the embedded RP can be attained in the nonembedded one.[10]

Theory

The RP, introduced first by Eckmann et al.,[8] is a powerful tool to analyze nonlinear time series. RP can visualize the structures relating to the dynamics of the system. Although embedding is mandatory for reconstruction of attractor in state space, RP may be constructed without embedding.[10] All information contained in the embedded RP can be attained in the nonembedded one. While in the state space, attractors with dimensions more than three cannot be visualized, any phase space trajectory can be represented in a two-dimensional (2-D) plot using RP. Moreover, the remarkable properties of RP are its ability to evaluate nonstationary and short-term data, in which it can be used in on-line monitoring of fluidization. These features make RP a very potent tool to study fluidized bed hydrodynamics and eliminates needs for time consuming and difficult long-term data sampling required in typical nonlinear methods.

Mathematical definition of RP

RP is a 2-D plot that is defined as

$$R_{i,j} = \Theta(\varepsilon - \|x_i - x_j\|) \qquad i,j = 1, 2, 3, ..., N \qquad (1)$$

where x_i, $x_j \in R^d$ represent the i-th and j-th points of the d-dimensional state space trajectory, ε is radius threshold, $\| \ \|$ represents the norm (magnitude of the vector), and $\Theta(.)$ is the Heaviside function. The Heaviside function compares any two points of the trajectory. If the norm is $<\varepsilon$, it is considered as a recurrence point and appears as a black spot ($R_{i,j}$), otherwise, it forms a white spot. In fact, the closed points throughout trajectory can be visualized through a 2-D matrix. In other words, RP shows recurrence states in the phase space. Since, March et al.[10] showed that RP can be constructed without embedding, thus, it was thought desirable to choose the embedding parameters of 1.

RP construction

The RP is based on the computation of the distance matrix (DM) between the reconstructed points in the phase space. This produces an array of distances in an $N \times N$ DM, where N is the number of points under study. The DM is a matrix whose elements $a_{i,j}$ are equal to the difference of points x_i, x_j. This matrix is converted to RP using radius threshold. The elements of DM, which are smaller than ε are considered as recurrence points and form black spots, otherwise they form white spots within RP. Before conversion of DM to RP, the DM may be normalized through dividing each DM element by the mean value of the whole matrix elements or by standard deviation of the time series. Normalizing eliminates the effect of the standard deviation variations of the signal. In this work, both normalized and non-normalized DMs were applied. Normalizing is carried out using standard deviation of the time series.

Recurrence quantification analysis

The RQA is a method to quantify RP structures. Determinism (*Det*) measures the fraction of repeated points forming diagonal lines. Mathematically *Det* is defined as

$$Det = \frac{\sum_{l=2}^{N} l.P(l)}{\sum_{l=1}^{N} l.P(l)} \qquad (2)$$

$\sum l.P(l)$ represents the number of black spots forming diagonal lines. Determinism is related to predictability of the system. For example, *Det* is high for a periodic system and is low for a stochastic system. In monitoring applications, determinism of the pressure fluctuations can be computed in consecutive time windows (epochs) along time.

Experiments

Experiments were carried out in a gas-solid fluidized bed made of Plexiglas. The column was 15 cm in inner diameter and 2 m in height. During the experiments, air at ambient temperature entered the column through a perforated plate distributor with 435 holes of 7 mm arranged in a triangular pitch. A cyclone was used to separate particles from air at high superficial gas velocities. Sand particles (Geldart B) with mean sizes of 150 and 490 μm and a particle density of 2640 kg/m^3 were used in the experiments. The experiments were carried out at gas velocities ranging from 0.1 to 0.7 m/s.

Absolute pressure fluctuations were recorded through a probe of 50 mm length and 4 mm diameter with a fine mesh net on its tip, located at the side facing of the fluidized bed, 15 cm above the distributor. The Piezoresistive transducer (Kobold, SEN-3248 B075) used in the experiments had a response time of <1 ms. Van Ommen et al.[11] showed that the model of Bergh and Tijdeman[12] provides reliable predictions of the frequency response characteristics of probe-transducer system over a wide range of probe length and diameter. The model for transducer with assumed inner space volume of 1500 mm^3 predicts the first resonance frequency of 679 Hz and the amplitude ratio of 1.1 at 200 Hz (Nyquist frequency). This shows that the measuring technique is suitable for gathering dynamic information from pressure signals in the expected range of frequencies, typically less than 20 Hz in a fluidized bed.[13]

The measured signals were band-pass filtered (hardware) at lower cut-off frequency of 0.1 Hz to remove the bias value of the pressure fluctuations and upper cut-off Nyquist frequency (200 Hz). The filtered signals were then amplified by a gain of 100. The pressure transducer was connected to a 16 bit data acquisition board (Advantech 1712L). The sampling frequency for pressure fluctuations was 400 Hz, satisfying the Nyquist criterion. This sampling frequency is also in accordance with criterion of 50 to 100 times the average cycle frequency (ACF) (typically between 100 and 600 Hz) which is required for nonlinear evaluation of the pressure fluctuations in bubbling fluidized beds.[13–15]

Results and Discussion

In this work, the pressure fluctuations signal obtained from the bed, operating at superficial gas velocity of 0.5 m/s, particles size of 150 μm and bed aspect ratio of $L/D = 1$, is considered as reference time series. Using the mean and the standard deviation of *Det* values of the signals, a 95% confidence interval of *Det* (±2.06 standard deviations from the mean) is determined for the signals obtained from different operating conditions (unideal states) as well as the reference signal. If *Det* of the signals is not within the confidence interval of *Det* of the reference state then the null hypothesis, fluidization hydrodynamics has remained unchanged, is rejected.
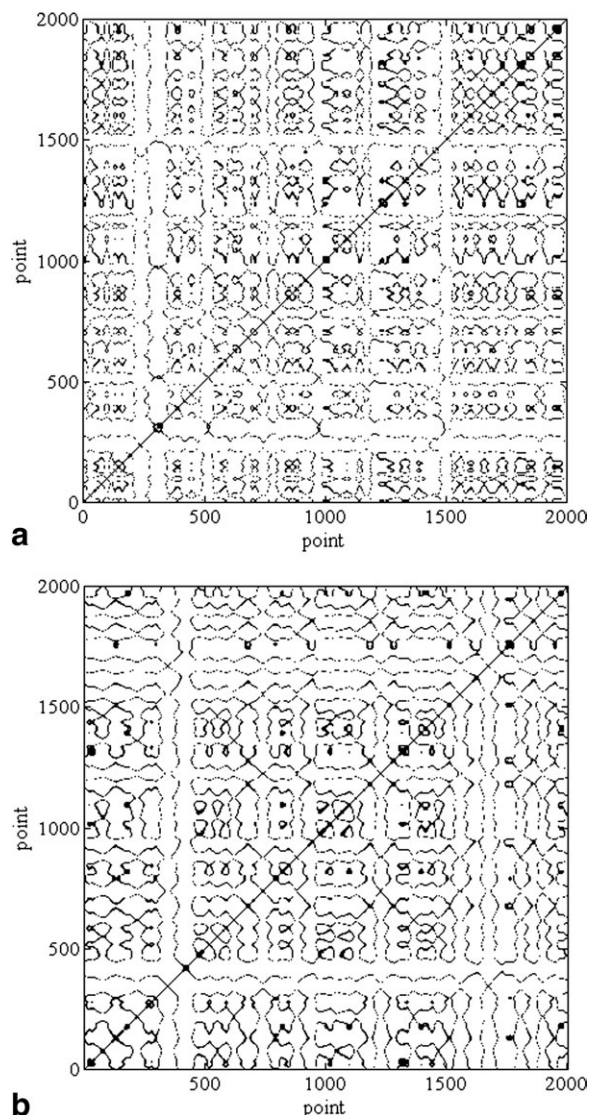
Figure 1. Recurrence plots of the (a) reference state, and (b) the state at which 50% of particles of the reference state were replaced with particles of size 490 μm.

Normalized DM. Radius threshold = 0.17.

The RPs of the reference state and an unideal state have been shown in the Figures 1a, b. As can be seen in these figures, there are many recurrence points with clear regular patterns, indicating the cyclic behavior. The Recurrence points forming diagonal and horizontal lines indicate that special structures exist in the time series of pressure fluctuations related to the dynamics of the bed. Different phenomena taking place in the bed, e.g., bubble generation, eruption, coalescence, and splitting, form these structures. Indeed, the RP represents hydrodynamics of the bed graphically and any change in the fluidized bed hydrodynamics affects the corresponding RP. Because the differences between two RPs can not be easily detected visually, RQA methodology is applied. To quantify the RP, first of all the input parameters should be determined.

Determination of input parameters

Length of epochs (L) and radius threshold are needed parameters that their values should be carefully determined before evaluating the value of Det. Each epoch contains a portion of the signal to be analyzed. The epoch length should be selected in such a way that it maintains main dynamical features of the whole time series. A good choice for L is the value by which each epoch contains the main phenomena occurring in the bed. In this case, the hydrodynamic characteristic of the bed is reflected within each epoch. Therefore, the epoch length should be enough larger than the sampling frequency to the main frequency ratio. Because the main frequency of pressure fluctuations of fluidized bed corresponds to 1.5 Hz,[16] epoch length is considered to be enough larger than 266 points with sampling frequency of 400 Hz. An epoch length of 2000 points is selected in this work. This length contains 2.5 s of the fluid beds hydrodynamics. Because Det gives an extent of repetitive patterns existed in the systems dynamic, to prevent loss of information, the epoch shift is selected as half of the epoch length (1000 points). Therefore, in a time series of length $2000 \times n$ there are $2n$-1 epochs of length 2000.

Statistically, any random variable (in this work Det), even at highly controlled condition, does not have a fixed value. The probability distribution provides the probability that the value of the random variable is within any specific range. There are many types of probability distribution; however, the normal probability distribution is the most common type of distributions. Therefore, it is investigated if Det of pressure fluctuations signal has a normal distribution. Figure 2 shows the distribution of Det calculated for various epochs of pressure fluctuations signal obtained from a certain operating condition. This figure shows that, is Det data distribution (horizontal axis) deviating from normal distribution (vertical axis)? Using this curve, if the data does come from a normal distribution, the plot will appear linear and symmetrical. As this figure shows the points are distributed symmetrically about the mean and they are approximately linear. This means that the assumption of normal distribution for Det is reasonable. However, mean and standard deviation of the distribution may change with the radius threshold. Because a low value of the standard deviation leads to a narrow confidence interval and consequently enhances the sensitivity of the method to changes in operation parameters, a good selection for the radius threshold is a value that gives the lowest standard deviation.
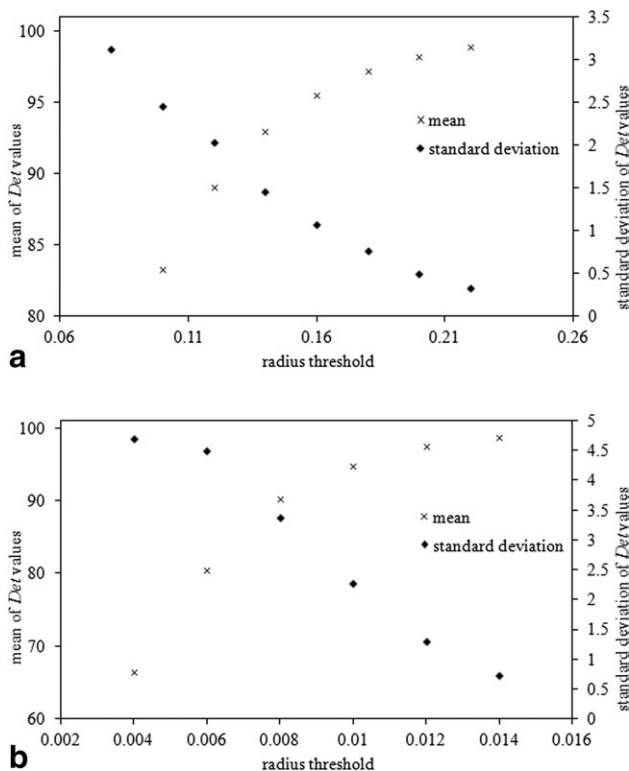
Figures 3a, b shows the mean and the standard deviation of the Det distribution of the reference time series at
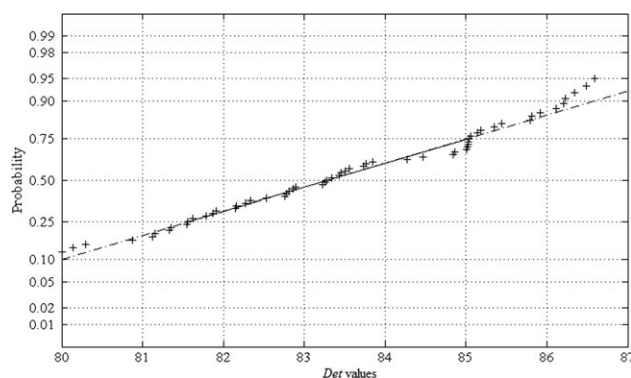
Figure 2. Probability distribution of *Det* values of various epochs for pressure fluctuations time series obtained from the reference state.

The line represents normal distribution.

Figure 3. Mean and standard deviation of the normal probability distribution of *Det* as a function of radius threshold.
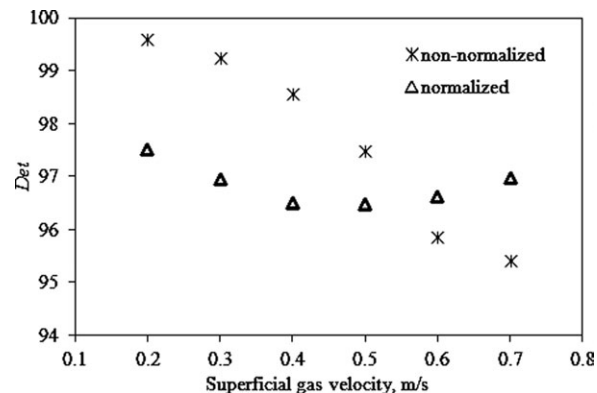
(a) normalized DM and (b) non-normalized DM.

Figure 4. *Det* values at different superficial gas velocities for the bed containing sand type 1, radius threshold = 0.17.

different values of radius threshold for the normalized and non-normalized DMs, respectively. As it can be seen in both Figures 3a, b, the mean increases and standard deviation decreases with radius threshold. In addition, at very high values of radius threshold *Det* is saturated (it approaches 100%) and consequently standard deviation becomes zero. According to these figures the value of radius threshold is selected as $\varepsilon = 0.17$ for normalized DM and $\varepsilon = 0.011$ for non-normalized DM. These values give lowest standard deviation while the value of *Det* has not been saturated yet.

For normally distributed random variables, 95% of the values will be within ± 2.06 standard deviations from the mean. Therefore, for the reference state, according to Figures 3a, b, the 95% confidence interval of *Det* of normalized and non-normalized time series are obtained as 94.75–98.19 and 93.23–99.87, respectively. If the value of *Det* is not within this range, the null hypothesis is rejected and it is concluded that with 95% confidence the fluidization hydrodynamics has been changed.

Sensitivity to superficial gas velocity variations

Figure 4 shows *Det* at different superficial gas velocities for non-normalized and normalized DM, respectively. As this figure shows, *Det* obtained from non-normalized DM decreases with superficial gas velocity, indicating its sensitivity to superficial gas velocity variations whereas *Det* changes very slightly when DM is normalized, indicating that *Det* is insensitive to minor variations in superficial gas velocity when DM is normalized. In other words, the effect of minor variations in the superficial gas velocity is eliminated through DM normalizing. This is a very considerable

property for *Det* because minor superficial gas velocity variations can not be avoided in industrial applications.

Sensitivity to small changes in particle size

To investigate the sensitivity of *Det* to the particles size, two sand types with mean particles sizes of 150 and 490 μm were examined. These sand types have been shown in Table 1. Figure 5 shows the value of *Det* for the signals obtained from the bed operating at the superficial gas velocity of 0.5 m/s, $L/D = 1$ and different fractions of sand type 2 in sand type 1. At this figure, DM has been normalized. Although *Det* was not sensitive to superficial gas velocity; however, as Figure 5 shows, is sensitive to small changes in particles sizes. As different fractions of sand type 2 (particles of size 490 μm) are added to finer particles of size 150 μm (sand types 1) the value of *Det* increases. In other words, the *Det* value increases as the mean particles size increases. The 5, 12, and 50% fractions show this trend. As larger particles are added to the bed, the minimum and turbulent fluidization velocities increase and the bubbles begin to shrink in the number and upward movement velocity and growth in the size. As the bed passes from a stormy situation to a calm situation gradually, its behavior becomes more regular. This regularity causes the *Det* value to increase. The ACF reduction with the addition of larger particles also implicitly confirms the regularity. ACF is the number of cycles per signal total time. The number of cycles is the number of times that the time series crosses its average value divided by two. Figure 6 shows ACF of the signals obtained from the bed operating at different particles size. A minimum in ACF of the pressure fluctuations signal indicates a minimum deviation from periodicity of the bed[16,17] and, thus, higher *Det*. As it can be seen the ACF decreases as the average particles size of the bed increases. The ACF reduction means the higher cycle time of the signal. The high cycle time of the signal is due to larger bubbles.

Table 1. Composition of the Mixtures Used in the Experiments to Testify Sensitivity of the Method to Particle Size
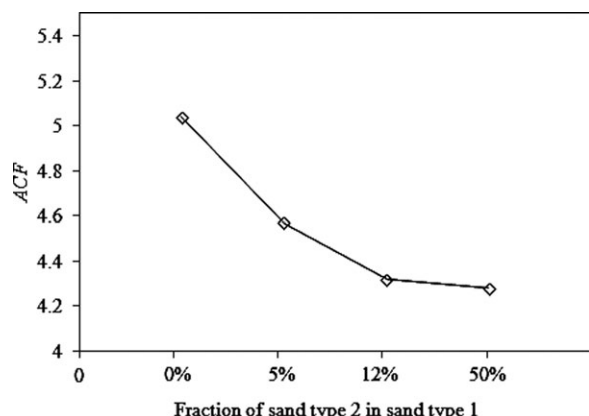
	Average Particle Size (μm)	Density (kg/m^3)
Sand type 1 (reference state)	150	2640
Sand type 2	490	2640
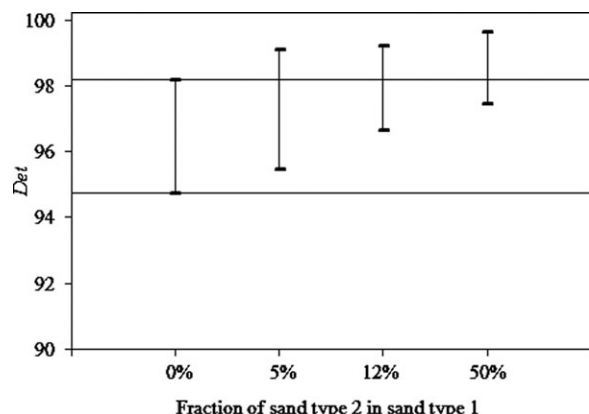
Figure 5. *Det* values for sand types 1 blended with various fractions of sand type 2 at superficial gas velocity of 0.5 m/s and *L/D* = 1, normalized DM.

An important matter in regards to the results is that the largest change in *Det* value occurs when fractions of sand type 2 is added to mono-range particles of size 150 μm. The difference between the bed containing 5 and 12% fractions of larger particles is lower than the difference between the bed containing mono-range particles of size 150 μm and 5% fraction. This is an ordinary and normal event because the largest change in the particles distribution, in percent, occurs at the onset of addition of larger particles to the mono-range bed. This feature is independent of the studying method. However, the method is useful to detect small changes in particle size (5% of sand type 2 in sand type 1). In other words, in the applications wherein some particles within the bed may grow gradually (for example, agglomeration) this method will be effective.

To enhance the sensitivity of the method to change in particles size, the length of the epoch is increased. However, it causes increase in the number of calculations exponentially. Using central limit theorem, the sensitivity can be increased without epoch length increment. This theorem indicates that the average of k normally distributed independent random variables with mean μ and standard deviation σ has normal distribution with same mean and standard deviation $\sigma/(k)^{1/2}$. If central limit theorem is used, the standard deviation of the distribution decreases and the acceptance interval of the null hypothesis become smaller and thereby the method becomes more sensitive to the changes in particles size. To apply this theorem, a time series of length $n \times L$ is needed to calculate *Det*. For example, a time series of length 18,000 points, $n =$ 9, (containing 17 epochs of length 2000 with epoch shift 1000) acceptance interval for the reference state is 96.06–96.87. Figures 7 and 8 show *Det* for the different sand types and different superficial gas velocities, respectively. These figures indicate that using central limit theorem, the sensitivity of *Det* to changes in particles sizes increase whereas it is insensitive to minor superficial gas velocity variations. It can be concluded that the larger lengths of the time series makes the method more sensitive to variations in particles size whereas it is insensitive to minor superficial gas velocity variations.

As it was discussed in detail, the presented method is sensitive to small change in particles sizes whereas it is not sensitive to minor changes in superficial gas velocity. This can be advantageous for the industrial cases at which small variations in superficial gas velocity are unavoidable. In addition, this method eliminates need for time series embedding and long-term data sampling, both necessary in nonlinear based monitoring methods.

Numerous methods have been proposed in the literature for detection of change in particle size in fluidized beds (see "Introduction" section). Among the methods so far proposed for detection of change in particle size, attractor comparison[6] (*S* statistic) used by van Ommen et al., are seen having the highest accuracy and acceptability in the literature. Therefore, to show novelty of this method, it is compared with the attractor comparison method. The comparison is made on the basis of input parameter settings, mathematical relationships, time complexity, and sensitivity to small changes in particle size. These aspects have been shown in the Table 2.

The main advantage of this method to the attractor comparison method (*S* statistic) is straightforward parameter settings. The calculation of *Det* needs to set just two input parameters, radius threshold and epoch length, whereas calculation of *S* statistic requires three input parameters, band width, segment length, embedding dimension (time window), to be set. The radius threshold and epoch length correspond to band width and segment length, respectively. While band width and segment length in *S* statistics method are difficult to
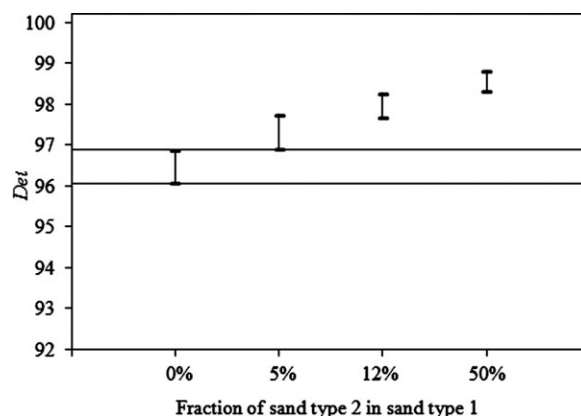

Figure 6. ACF for the pressure fluctuations signal obtained from the bed operating at superficial gas velocity of 0.5 m/s and sand types 1 blended with various fractions of sand type 2.

Figure 7. *Det* values when central limit theorem is applied, for sand types 1 blended with various fractions of sand type 2 at superficial gas velocity of 0.5 m/s and *L/D* = 1, normalized DM. time series of length 18,000 points.
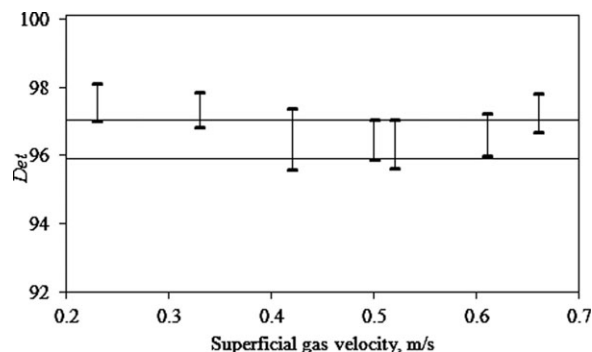
Figure 8. *Det* values when central limit theorem is applied, at different superficial gas velocities for the bed containing sand type 1, normalized DM. time series of length 18,000 points.

calculate and need optimized values, radius threshold and epoch length are selected easily. As it was mentioned, the radius threshold is selected in the way that *Det* is near to 100 (but not exactly equal to 100). Such a selection decreases the standard deviation of *Det* of the reference signal, and consequently it leads to narrower confidence interval.

The main role of epoch length is to reduce calculation time through time series shortening. Therefore, this parameter does not considerably influence the results. In other words, the time series is divided to subseries having length *L*. The strategy for the selection of *L* is in the way that each subseries contain main hydrodynamic phenomena occurring in the bed. In this case, each subseries reflect almost all the hydrodynamic characteristics of the fluidized bed. Since the main frequency of the hydrodynamic phenomena occurring in the bed is about 1.5 Hz, the epoch length should be enough larger than 266 points (the epoch length should be enough larger than sampling frequency to main frequency

ratio). As it was discussed, the epoch length is selected as 2000 points.

Figures 9a, b reconstruct the Figure 7 with the different values of epoch length. As it can be seen, this parameter has not great impact on the results.
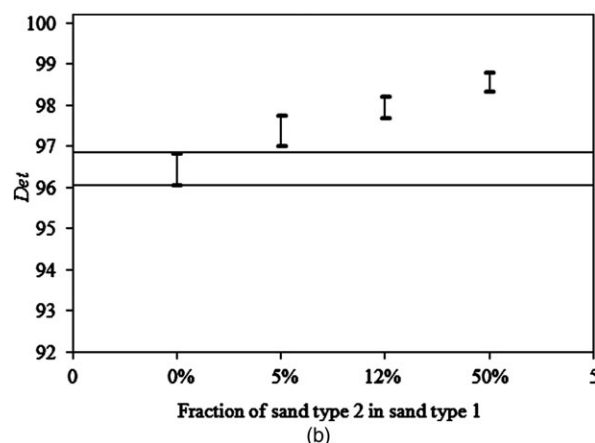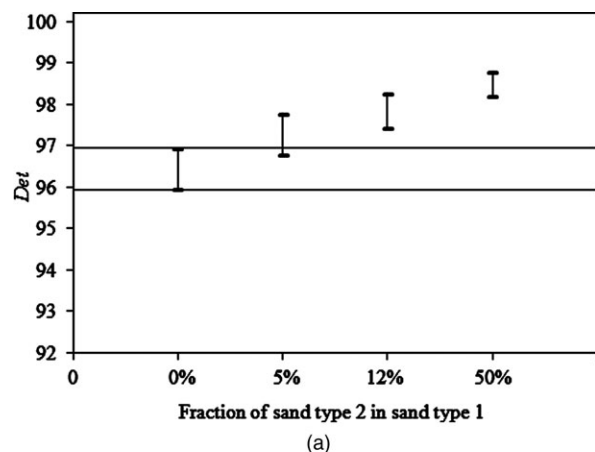
The need to set embedding dimension (time window) as input parameters of *S* statistic has been eliminated in this method. Finally, it is concluded here that input parameter setting in this method is faster and simpler than attractor comparison method.

The mathematical relationships and equations of the both methods have been shown in the Table 2. These equations clearly prove the claim that calculation of *Det* is mathematically simpler than the calculation of *S* statistic. The large number of summations in the attractor comparison method increases the calculation time of *S* statistic. In addition, the minimum length of time series needed in the attractor comparison method (65,535 points)[13] is much more than the length required in this method (18,000 points).

The comparison of two methods in terms of the speed can be carried on through measurement of the running time of each program. However, as the computer programs of the two methods may have not been written in the optimum mode, the comparison is done using complexity computation. The time complexity analysis avoids wrong results associated with running time comparison; therefore, the time complexity of the *S* statistic and *Det* is estimated to compare them in terms of the speed. In this method, *Det* is calculated through a 2-D matrix (DM matrix). The algorithm used to calculate *Det* contains two nested loops. Thus, time complexity of *Det* algorithm is a polynomial of order 2. As it can be seen in the Table 2, the asymptotic time complexity of H_{pq} is $O(n^2)$. Since there are nested loops in the calculation of Q and V_c from H, the order of the time complexity of the algorithm of *S* statistic rises much higher than 2. In addition, the lower length of the input time series for the

Table 2. Comparison of the Present Method (*Det*) with Attractor Comparison Method (*S*)

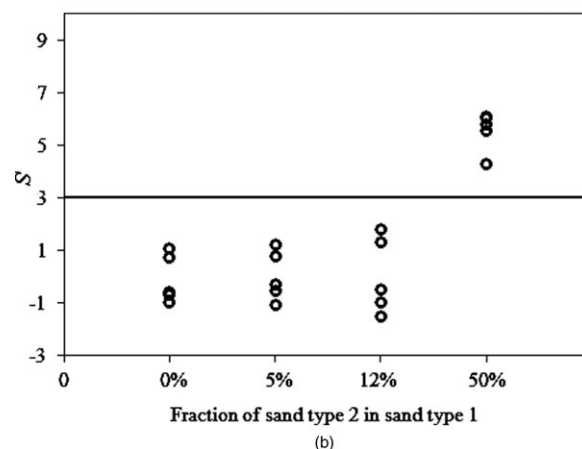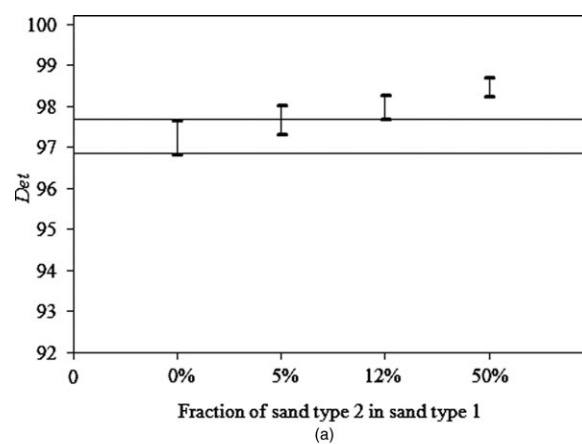|	*S* Statistics	*Det*
Parameter settings	Three input parameters; time window (embedding dimension), band width, segment length	Just two input parameters; radius threshold and epoch length
Mathematical relationships	$S = \dfrac{\widehat{Q}}{\sqrt{V_C(\widehat{Q})}}$ $V_C(\widehat{Q}) = \dfrac{4(N-1)(N-2)}{N_1(N_1-1)N_2(N_2-1)N(N-3)} \sum\sum_{1 \le p < q \le N} \psi_{pq}^2$ $\widehat{Q} = \dfrac{2}{N_1(N_1-1)} \sum\sum_{1 \le p < q \le N_1} H_{pq} + \dfrac{2}{N_2(N_2-1)} \sum\sum_{N_1+1 \le p < q \le N} H_{pq} - \dfrac{2}{N_1 N_2} \sum_{p=1}^{N_1} \sum_{q=N_1+1}^{N} H_{pq}$ $\psi_{pq} = H_{pq} - g_p - g_q$ $H_{pq} = \mathrm{H}_{pq} - \dfrac{2}{N(N-1)} \sum\sum_{1 \le p' < q' \le N_1} \mathrm{H}_{p'q'}$ $g_p = \dfrac{1}{N-2} \sum_{\substack{q \\ p \ne q}} H_{pq}$ $\mathrm{H}_{pq} = \dfrac{1}{L^2} \sum_{i=1}^{L}\sum_{j=1}^{L} h(Z_{(p-1)L+i}, Z_{(q-1)L+j})$ $h(Z_i, Z_j) = e^{-|Z_i - Z_j|^2/(4d^2)}$	$\mathrm{Det} = \dfrac{\sum_{l=2}^{N} lP(l)}{\sum_{l=1}^{N} lP(l)}$ $R_{i,j} = \Theta(\varepsilon - \|x_i - x_j\|),$ $i, j = 1, 2, 3, \ldots, N$
Complexity of calculations	Order is higher than 2	A polynomial of order 2
Length of the signal (time series)	Minimum 65535	18,000 is sufficient in this work
Sensitivity to small changes in particle size	Less sensitive	More sensitive

Figure 9. *Det* values for sand types 1 blended with various fractions of sand type 2, at superficial gas velocity of 0.5 m/s and *L/D* = 1, normalized DM, with the application of central limit theorem, at different values of epoch length, (a) *L* = 2500, (b) *L* = 1500.

Figure 10. Comparison of the sensitivity of *Det* with *S* at high superficial gas velocity, with the application of central limit theorem for *Det* (a) *Det*, (b) *S*.

calculation of *Det*, contributes to lower calculation time. The lower length of the time series (18,000 for *Det* at the present paper vs. 65,535 for *S* statistic) also eliminates the problems associated with long-term data sampling.

To emphasize how surprisingly this method is sensitive to small changes in particle size in compare with early warning *S* indicator, the comparison is made at high superficial gas velocities where the sensitivity of the *S* statistic to changes in particle size distribution is low.[6] Sensitivity of both methods to small changes in particle size at high superficial gas velocities are shown in the Figures 10a, b. These figures show that the attractor comparison method (*S* statistic) is not able to detect small changes in particle size at the high superficial gas velocities (velocities about *U*t). Despite the attractor comparison method, *Det* shows good sensitivity to small change in particle size at high superficial gas velocities. While the sensitivity of the both methods (*Det* and *S* statistic) to changes in particle size distribution is reduced at higher gas velocities, the sensitivity of the *Det* to changes in particle size distribution is more than *S* statistic at high superficial gas velocities.

Both *Det* and *S* statistic are selectively sensitive to change in particle size. In other words, while these indicators can detect change in particle size they are not sensitive to minor variations in gas velocity. This reveals that the particle size

influences the fluidization hydrodynamics in a different ways in comparison to the gas velocity. The both particle size and gas velocity influence the hydrodynamics of the fluidized bed. The particle size influences the hydrodynamics through the variation of minimum fluidization velocity (U_{mf}) and turbulent fluidization velocity (U_t) while the gas velocity causes the bubbles to change in the size and the number. Minimum fluidization and turbulent velocity are the function of particle size (also gas and particle properties). Indeed, *Det* and *S*-statistic detect the variations of U_{mf} and U_t. This is the reason that why these indicators are not sensitive to minor gas velocity variations. Table 3 shows the U_{mf} and U_t for the mixtures used in the experiments. As it can be seen in Table 3, the addition of coarse particles to the bed causes the U_{mf} and U_t to increase. In addition, at low gas velocities, bubble hold up is proportional to the difference between superficial velocity and U_{mf} rather than high gas velocities. At low gas velocities as U_{mf} varies (because of the change in particle size), the difference between U and U_{mf} and respectively

Table 3. The U_{mf} and U_t of the Mixtures Used in the Experiments

	U_{mf} (m/s)	U_t (m/s)
100% sand type 1	0.024	0.80
5% sand type 1 + 95% sand type 2	0.031	0.84
12% sand type 1 + 88% sand type 2	0.042	0.91
50% sand type 1 + 50% sand type 2	0.126	1.10

bubble hold up are varied and the indicators detect the change in particle size. At higher gas velocities, the bubble hold up is weakly linked with the difference between U and U_{mf}. Therefore at the higher gas velocities the indicators become less sensitive.

Conclusions

A method was investigated for small changes in fluidized bed hydrodynamic monitoring using pressure fluctuations signal. It was shown that Det of the signal has normal probability distribution. A 95% confidence interval was determined for Det of the reference state. It was supposed that if Det of the signal obtained from the bed operating at different particle sizes does not lie within the confidence interval, the null hypothesis, fluidization hydrodynamics has remained unchanged, is rejected. Sensitivity of the method to small change in fluidization parameters, particle sizes, and superficial gas velocity were examined. The method illustrated high sensitivity to small changes in particles size especially when central limit theorem was used. The sensitivity of the method is adjustable through manipulating time series length. A higher length of time series leads to higher sensitivity. Insensitivity of Det to minor changes in superficial gas velocity, lower length of needed time series, adjustable sensitivity, easy numerical calculations and no need to time series embedding can be regarded as advantages of the presented method. While both Det and S statistic indicators are selectively sensitive to change in particle size, the sensitivity of the Det to changes in particle size distribution is more than S statistic at high superficial gas velocities.

Notation

Roman letters

$a_{i,j}$ = elements of distance matrix
ACF = average cycle frequency
Det = determinism
d_p = average diameter of particles, μm
l = line parameter
L = length of epoch
L/D = height to diameter of fluidized bed
n = number of epochs
N = number of time series points
O = the notation of time complexity
$P(l)$ = number of diagonal lines of length l
$R_{i,j}$ = recurrence plot matrix
R^d = d-dimensional space
S = S-statistics (attractor comparison method)
U = superficial gas velocity, m/s
U_{mf} = minimum fluidization velocity
U_t = superficial gas velocity of flow regime transition from bubbling to turbulent
x_i = i-th point of space state trajectory

Greek letters

Θ = Heaviside function
ε = radius threshold
σ = standard deviation

Abbreviations

DM = distance matrix
RP = recurrence plot
RQA = recurrence quantification analysis

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