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Early Detection of Agglomeration in Conical Spouted Beds using Recurrence Plots

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ABSTRACT

Agglomeration of particles in a conical spouted bed was investigated using recurrence plot (RP) and recurrence quantification analysis (RQA) of the pressure fluctuations (PFs) and acoustic emission (AE) signals. Experiments were carried out in a 45° conical spouted bed with sugar particles ($d_p = 720 \mu\text{m}$; $\rho_p = 1580 \text{ kg/m}^3$). Water was sprayed incrementally into the bed to produce agglomerates during the operation. Several recurrence quantification parameters were calculated during agglomeration process and the most suitable ones were chosen for early prediction of the agglomeration in the bed. The results show that recurrence rate, determinism and laminarity of PFs and AE signals increase during agglomeration process which indicate that bed behaves more periodic and deterministic in nature. Additional examination of the RQA parameters show that AE signals are substantially more sensitive to the hydrodynamic changes occurring in the bed as compared to those of PFs and therefore can detect changes earlier with more accuracy.

Keywords: Conical spouted bed; Agglomeration; Pressure fluctuation; Acoustic emission; Recurrence plot

1. INTRODUCTION

Spouted beds are gas-solid contactors in which particles are suspended through the flow of a gas from a small orifice at the bottom of the bed. Although there are several different design configurations, a conventional spouted bed (also known as conical-cylindrical spouted bed) is a bed with a conical base to eliminate dead spaces at the bottom followed by a cylindrical section. The bed material fills up the entire conical section and extends to the cylindrical section. Conventionally, spouted beds have been designed for handling large particles (i.e., Geldart group D particles) due to their well-received advantages as compared with fluidized beds. In a conical spouted bed, on the other hand, only the conical section is filled up with the bed material. One of the main advantages of conical spouted beds is their short gas residence times with narrow distribution.¹ In these beds, liquids can easily be sprayed into the bed through the nozzle placed at top or bottom.¹ Thus, conical spouted beds have been successfully applied for numerous industrial processes such as gasification, combustion, granulation, coating and drying of suspensions, solutions, and pastry materials.¹⁻³

Coating and granulation processes usually involve the presence of a liquid in the bed. The liquid increases the particle cohesiveness and promotes de-spouting, leading to the bed malfunctioning. Also, in high temperature applications of spouted beds, such as gasification and combustion, some compounds may melt, or partly melt, and the sticky particles can stick to each other and form larger agglomerates.⁴⁻⁶ Therefore, hydrodynamics of a spouted bed can be changed over time due to instabilities imposed by particle size changes and cohesiveness. The instabilities here refer to changes in spout shape such as, intense oscillation in fountain height, vigorous swings of the spout from side to side, choking and consequent slugging of the spout. The degree of these instabilities depends on the bed configuration and operating conditions, such as inlet gas

velocity, temperature, pressure, liquid content, as well as physical and chemical properties of the gas, particles and liquid.^{7, 8} Therefore, monitoring of spouted beds parameters is important since the performance of these beds strongly depends on their hydrodynamics.⁹ Detecting bed instability is not only useful for preventing undesirable situations (e.g., agglomeration and despouting), but also is crucial to control the product quality, both in batch and continuous operations. However, there is a lack of information on early detection of the possible changes in the spouting behavior when liquids are injected into spouted beds. This knowledge can be very useful in monitoring the hydrodynamic status of spouted bed coaters, granulators and dryers to avoid the bed collapse and control the product quality.

Many techniques have been proposed in the literature for the hydrodynamic characterization of spouted beds. These techniques are based on different measurement methods such as, pressure fluctuations (PFs) measurements,¹⁰⁻¹³ radioactive particle tracking (RPT),¹⁴ fiber optic (FO)¹⁵⁻¹⁷ and acoustic emissions (AE) measurements.¹⁸ Since PFs are easily measurable, many researchers have used this method for hydrodynamic characterization of spouted beds.¹⁰⁻¹³ Moreover, PFs include the effect of various phenomena in the spouted bed, including individual and bulk movement of particles and formation and movement of agglomerates.¹⁹ Mostoufi et al.¹⁹ identified different hydrodynamic phenomena (including movement of bulk of solids, particle transport in spout, clustering and motion of agglomerates through the bed) in a conical spouted bed by analyzing PFs in the frequency domain. Sari et al.²⁰ characterized the gas-solid flow in a conical spouted bed of high density zirconia particles. They indicated a dominant frequency of 12 Hz in the stable spouting region which drops to 9 Hz in the jet spouting regime. The dominant frequency was attributed to the frequency of the particle motion passing thorough the spout.

Measurement of acoustic emissions, as a non-intrusive, low cost, reliable measurement technique, has shown a potential to improve process understanding and to provide a basis for on-line monitoring and control of different processes.²¹ Analysis of AE signals is applicable to a wide range of process conditions. This technique has also been widely used to characterize the hydrodynamics of fluidized beds.²²⁻²⁴ Despite existence of numerous published papers on application of monitoring the AE for detecting the hydrodynamics of fluidized beds, to the authors' knowledge, there is only one published study on spouted beds based on the AE signal. Oliveira et al.¹⁸ evaluated the feasibility of characterizing the hydrodynamic of a spouted bed based on acoustic emissions. The 1/3-octave band spectra of AE signals was calculated in their study and the results indicated that, regardless of the operating conditions studied, these spectra were practically indistinguishable at frequencies below ~200 Hz. This undue insensitivity may be attributed to the signal analysis method. At higher frequencies, the 1/3-octave spectra showed some variations with the spouting conditions, but appeared to be dominated by the flow through the inlet orifice.

Various nonlinear analysis methods have been used for analyzing the dynamic changes in the spouted beds.¹⁰⁻¹³ Although these nonlinear analysis methods can give a better understanding of the system state, they are accompanied with some drawbacks and limitations such as, long term data sampling, time consuming numerical calculations and uncertainty in the determination of embedding parameters. Nonlinear analysis by recently introduced recurrence plot (RP) technique eliminates the need for long term data sampling and storage. Unlike other methods of nonlinear analysis, numerical calculation of RP method is not time consuming and also, does not require complex algorithms for determination of embedding parameters.²⁵ Although nonlinear analysis by RP is now being used in the characterization of fluidized bed hydrodynamics,²⁶⁻²⁸ application

of this method to spouted beds is rare.²⁹ In spite of the fact that this method seems to be a promising tool for flow regime recognition in spouted bed, no previous work can be found on the analysis of PFs and AE signals based on the RP method to monitor hydrodynamic changes and detect the de-spouting in conical spouted beds.

This work focuses on developing a novel methodology for monitoring of the hydrodynamics of spouted beds, especially early detection of agglomeration, based on PFs and AE measurements in a bottom liquid sprayed gas-solid spouted bed. For this purpose, recurrence plots were obtained for both PFs and AE signals and parameters of recurrence quantification analysis (RQA) were compared at different time of water injection into a bed of sugar particles.

2. THEORY

2.1. Recurrence Plots

Recurrence plot, introduced by Eckmann et al.,²⁵ visualizes recurrences in the dynamics of a dynamical system. The RP represents the times at which states of a phase space of the system is repeated. The RP is a two-dimensional squared matrix, R , which is mathematically expressed as:

$$R_{i,j} = \theta(\varepsilon - \|\vec{x}_i - \vec{x}_j\|) \quad i, j = 1, 2, 3, \dots, N \quad (1)$$

where N is the number of state space points, $\vec{x}_i, \vec{x}_j \in R^m$ are i -th and j -th points of the m -dimensional state space trajectory, ε is a threshold distance, $\|\cdot\|$ is the norm and $\theta(\cdot)$ is the Heaviside function. In fact, the matrix R compares the states of a system at times i and j . If the states are similar (the norm is less than ε), this would be marked by a one in the matrix, i.e., $R_{i,j}=1$, and a black spot would appear on the plot at coordinate (i, j) . If, on the other hand, $\vec{x}_i$ and $\vec{x}_j$ are rather different (the norm is greater than ε), the corresponding entry in the matrix would be

$R_{i,j}=0$ and a white spot would appear on the plot. In other words, this matrix can tell when similar states of the underlying system occur. Figure 1 represents the reconstruction of RP graphically.

2.2. Recurrence Quantification Analysis

Several methods have been proposed for measuring the complexity of structures in a RP.³⁰⁻³³ These methods quantify small-scale structures of RPs and are known as recurrence quantification analysis. The RQA involves estimation of recurrence point density as well as diagonal and vertical lines in a RP. Patterns within a RP (and subsequently the RQA parameters) are related to different dynamics of the system. Hence, valuable information about the dynamics can be extracted from the RP patterns and RQA analysis. Among various RQA parameters, recurrence rate (RR), determinism (DET), laminarity (LAM) and entropy (ENT) were used in this study. In order to reduce the sensitivity of the method to small changes in the superficial gas velocity, the time series were first normalized as following:³⁴

$$x_{i,n} = \frac{x_i - \bar{x}}{\sigma_x} \quad (2)$$

Recurrence rate, or per cent recurrence, is the simplest measure in the RQA which represents the density of recurrence points in the RP and is defined as:

$$RR = \frac{1}{N^2} \sum_{i,j=1}^N R_{i,j} \quad (3)$$

where the summation of the right hand side is the total number of repeated points and N^2 is the total number of points in the recurrence matrix.

Determinism is the ratio of recurrence points that form diagonal lines (of at least length l_{min}) to all recurrence points and is defined as:

$$DET = \frac{\sum_{l=l_{min}}^N lP(l)}{\sum_{l=1}^N lP(l)} \quad (4)$$

where $P(l)$ is the number of lines with the length of l , the summation is the number of black dots forming the diagonal lines and l_{min} is the minimal length of diagonal lines. Determinism measures the predictability (rule-obeying) of the system and is low for a stochastic system and high for a periodic system.

Similar to the definition of determinism, the ratio of recurrence points forming vertical lines with length v to all recurrence points can be computed and is called laminarity:

$$LAM = \frac{\sum_{v=v_{min}}^N vP(v)}{\sum_{v=1}^N vP(v)} \quad (5)$$

where $P(v)$ shows the number of lines with the length v , the summation represents the number of dots forming the vertical lines and v_{min} is the minimal length of vertical lines. The laminarity shows the probability of occurrence of states which do not change or change very slowly (laminar states). Thus, laminarity corresponds to the amount of laminar states in the system.

Entropy refers here to the Shannon information entropy of probability distribution of diagonal line length. The probability distribution of diagonal line length was defined as follows:

$$p(l) = \frac{P(l)}{N_l} \quad (6)$$

where N_l is the number of diagonal lines. Entropy is defined as:

$$ENT = - \sum_{l=l_{min}}^N p(l) \ln p(l) \quad (7)$$

Entropy reflects the complexity of RP structures. For example, entropy of an uncorrelated noise is rather small, indicating low complexity of structures in its RP. In contrast, large entropy is typical of a chaotic behavior which reflects high complexity of the system.

3. EXPERIMENTS

3.1. Experimental set-ups

A schematic of the conical spouted bed ($\gamma = 45^\circ$) with bottom spray is shown in Figure 2. The bed was made of polyoxymethylene (also known as Delrin) and consisted of a 150 mm ID cylinder with a conical section at the base, connected to a 6 mm ID gas inlet tube (D_o). The cone bottom diameter of the bed, D_i , was 25 mm and the height of the conical section, H_c , was 151 mm. A changeable wire mesh (300 μm) was placed on the inlet orifice to prevent particles from falling into the inlet tube. Compressed air at ambient temperature was used as the spouting gas. The air was supplied from a screw type compressor at 8 bar with a maximum flow rate of 0.05 m^3/s . A pressure regulator and a 30 L air tank were placed between the supply line and the spouted bed gas inlet to eliminate possible fluctuations of the air flow rate. Water was sprayed into the bed with a nozzle positioned at the center of the air inlet orifice. The spouting gas velocity at the inlet orifice was high enough to act as the atomizing gas for the liquid spray. It is worth mentioning that the inlet gas velocity is defined based on the inlet diameter, D_o . The gas velocity corresponding to the point at which stable external spouting collapses with decreasing gas flow rate was defined as the minimum spouting velocity, U_{ms} .¹

Experiments were also carried out in a half column conical spouted bed (having exactly the same dimensions as the full column) to visualize the flow pattern during water injection and drying periods. The half conical spouted bed was constructed by slicing a full column and attaching a flat Plexiglas sheet to the front open surface for visual observation and photography. Images were acquired with a camera (UI-2210SE, IDS Company) equipped with a 16 mm PENTAX lens having a resolution of 640×480 pixels with the frame rate up to 100 frame/s (fps). The images were captured by uEYE software and analyzed using the MATLAB Image Processing Toolbox.

Pressure fluctuations were measured by differential pressure transducers (PX163-120D5V, OMEGA Engineering) connected to the inlet line of the bed, base of conical section and bed internal wall at 9 cm above the base of conical section. Analysis of results revealed that PFs measured at the inlet line and at a height of 9 cm above the inlet were not able to detect the imposed instabilities and agglomeration in the bed early enough. Therefore, only PFs measured at the base of conical section, as shown in Figure 2, is reported here. The other line of the pressure transducer was left open to atmosphere. The pressure transducers have a response time of 1 msec. A fine mesh net was used at the tip of the pressure line to avoid its blockage. The pressure signals were digitized by a 16 bit data acquisition board (National Instruments, USB-6351). The sampling frequency was set as 500 Hz following recommendation of Johansson et al.³⁵ and van der Stappen et al.³⁶ who suggested sampling frequency between 100 and 600 Hz which is 5-100 times the average cycle frequency.

The AE signals were measured by an omnidirectional back electret condenser microphone (Panasonic, WM-61 A) which had a frequency response of 20-20000 Hz (sensitivity: -35 ± 4 dB, signal to noise ratio: more than 62 dB). The outlet signal from the microphone was recorded by a

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3 USB interface sound analyzer (ART-SA16, ARTIMAN Instruments) with a sampling frequency
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5 of 44 kHz. The AE sensor was glued by silicon grease externally to the outer surface of the bed
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7 at 12 cm above the base of the conical section. Oliveira et al.¹⁸ recorded AE signals of spouted
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9 bed at four positions: near the inlet zone, adjacent to the medium core zone, between the core
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11 zone and fountain zone and in the fountain. Their results showed that AE signals obtained at
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13 different measurement positions were very similar, expect for those corresponding to the inlet
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15 region, where the difference was probably caused by spouting gas entering at high velocity
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17 through the inlet orifice. Therefore, the selection of the location of the acoustic sensor for
18
19 collecting AE signals was not decisive, provided that the static bed height position remained
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21 constant for all experiments.
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29 **3.2. Agglomeration Experiments**

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31 Sugar particles, 720 μm in diameter and a density of 1580 kg/m^3 , were used in agglomeration
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33 experiments and water was used as the binding agent. These materials were used in this work
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35 since they can easily form agglomerates. In each experiment, 400 gr sugar particles was used
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37 which resulted in a static bed height of 12 cm. The minimum stable spouting gas velocity (U_{ms})
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39 of dry particles was measured as 40.3 m/s. Agglomeration experiments were carried out at inlet
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41 gas velocities of 10%, 15%, 20%, 25% and 30% higher than U_{ms} .
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47 To eliminate possible effects of the initial packing status on the measurements, the bed was
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49 spouted for 10 min prior to spraying of water in each experiment. Water was then injected every
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51 2 minutes into the bed to form agglomerates. The water was sprayed at a flow rate of 3 mL/min
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53 for 10 s in every injection. In other words, 0.5 mL of water was added to the bed in every
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55 injection interval. Water injection was continued until de-spouting of the bed was observed. AE
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and PFs signals were recorded continuously during each experiment. The signal measurements were started 2 minutes prior to the first water injection. Visual observation of the bed hydrodynamics was performed in order to characterize the bed behavior during experiments. Preliminary experiments were conducted to assess the spraying and drying times. Experiments were repeated three times and it was found that all values and trends reported in the following sections are reproducible.

4. RESULTS AND DISCUSSION

4.1. Agglomeration Tests

In all of the agglomeration tests, with the periodic injection of water, a gradual decrease of the fountain height was observed. A point was then reached where a sudden collapse of the spout was monitored leading to the complete cessation of spouting. The time it took for the complete cessation of spouting from the beginning of water injection is defined as de-spouting time in this study. The effect of spouting air velocity on de-spouting times is summarized in Table 1. After each test, the whole bed material was sieved to see the particle size ranges of the agglomerates. Results of sieve analyses are also included in Table 1. A photo of the particles collected at the end of the agglomeration test performed at $U/U_{ms} = 1.2$ is illustrated in Figure 3.

As can be seen in Table 1, the de-spouting times increased by increasing the inlet gas velocity. In dry conical spouted bed, the drag force is the dominant force among other forces exerted on particles in the spout region, due to high gas velocity. When particles are wetted, de-spouting occurs as the drag force is balanced by the liquid bridge force³⁷. Since the drag force is proportional to U^2 , a greater drag is exerted on particles at higher gas velocities which can counterbalance a greater liquid bridge force. Therefore, longer de-spouting times are observed at

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3 higher inlet gas velocities. Moreover, as indicated in the Table 1, the fraction of larger particles
4 increases at higher gas velocities. Fine droplets of water form a liquid layer at the particle
5 surface. When a wet particle collides with another, a liquid bridge forms between the two
6 particles. As spraying of the liquid stops, the water evaporates and a solid bridge forms due to
7 solidification of the binder³⁷. Repetition of these steps (wetting, collision, drying) leads to the
8 growth of size of bed particles due to agglomeration. At higher gas velocities, greater drag force
9 is exerted on particles which can handle greater liquid bridge force. Therefore, more water can
10 be sprayed into the bed due to longer time of operation which results in higher mass fraction of
11 larger particles.
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26 Agglomeration resulting from continuous spraying of water into the bed causes noticeable
27 changes in the hydrodynamics. Figure 4 demonstrates the change in bed behavior during water
28 spraying and drying periods in half bed at $U/U_{ms} = 1.2$. Black ink was added to the water for
29 better observation of the flow. The total amount of water injected to the system from the start of
30 the experiments is also shown on each photo. The flow structures are characterized as stable
31 spouting (Figures 4a and b), unstable spouting (Figures 4c, d, e and f) and bed collapse (Figures
32 4g, h and i). As shown in Figure 4a, dry particles form a stable spout in the bed with small
33 oscillations in the fountain height. Particles travel rapidly up the spout until they are thrown out
34 forming a fountain above the bed surface. The gas velocity decreases in the fountain region, then
35 particles disengage from the gas and move downward in the annulus. The downward-moving
36 particles re-enter the spout, mainly at the bottom, and this cycle of particle movement repeats
37 again. Agglomeration caused by the liquid injection affects spout, fountain and annulus regions
38 in many different ways. As can be seen in Figures 4d and e, after spraying of a small amount of
39 water (1 mL in total), the fountain height oscillates more vigorously which can be attributed to
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the presence of agglomerates in the spout. Water also affects the voidage of the annular region. The annular region becomes more consolidated because the liquid occupies the void volume between the particles and makes it difficult for the spouting gas to diffuse into the annular region. Compared to the dry bed, more gas passes through the spout region decreasing the solids hold-up in the spout leading to lower solids circulation, which can also be observed visually. By spraying more water, liquid bridge forms between wet particles in contact. After the water evaporates, this bridge consolidates into a solid bridge, causing formation of particle agglomerates. Number of wet particles increases with the injection of more water into the bed. Therefore, the adhesion tendency of particles intensifies due to increase in the number of bridges between particles. By further spraying of water, the height of fountain decreases and turns out to be oblique as shown in Figures 4 e and f. Eventually, the gas flow rate ceases to be sufficient for keeping a stable spouting regime leading to bed collapse (Figures 4g, h and i).

4.2. Sensitivity Analysis of Input Parameters of RQA

Prior to recurrence plot analyses, it is essential to conduct a sensitivity analysis on the input parameters of RQA (m , τ , ε , l_{min} , v_{min} , N). It should be noted that the sensitivity analysis of input parameters were done using signals measured at inlet velocity of $1.2U_{ms}$ in the dry full bed and optimum input parameters obtained were then used in the analyses of all other signals. Firstly, the length of time series (N) for the RQA should be determined. Llop et al.³⁸ investigated the effect of time series length on several RQA variables. They concluded that, for pressure fluctuations, values of recurrence rate, determinism and laminarity do not change significantly when more than 8192 data points are used. Furthermore, Wang et al.²⁹ and Babaei et al.²⁶ used 1025 and 3500 data points with sampling frequencies of 600 and 400 Hz, respectively, for

calculating RQA parameters of pressure fluctuations. Effect of number of data points was also investigated in this study and it was found that the RQA variables of interest (RR, DET, LAM and ENT) for PFs and AE signals do not change when at least 4000 (8 sec.) and 4400 (0.1 sec.) data points are used, respectively. This shows that the RP method can provide useful information about the hydrodynamics of spouted bed with a small number of data.

Embedding dimension (m) is the second recurrence parameter to be calculated and can be estimated by the false nearest neighbors (FNN) algorithm proposed by Kennel et al.³⁹ In this algorithm, m is increased until the number of neighbors of points in the embedding space become unchanged. For each point, x_i , the Euclidean distance to its nearest neighbor, x_j , is calculated in both m and $m+1$ dimensional spaces. The point is treated as a false neighbor when the distance between points (i, j) increase by increasing the embedding dimension. Therefore, the number of FNN is calculated for several embedding dimensions until the fraction of these false neighbors reaches zero (or at least becomes very small). At this proper value of m , information of the system is at maximum and there is no need to explore higher embedding dimensions. The fraction of FNN for PFs and AE signals are shown in Figure 5. This figure indicates that the proper embedding dimensions for pressure fluctuation and acoustic emission are 7.

The attractor reconstruction is a function of time delay (τ). The mutual information function between time series of $x(t)$ and $x(t + \tau)$ was used to determine proper time delay.^{30,40} The mutual information function is defined as:⁴⁰

$$MI(x(t), x(t + \tau)) = \sum_{x(t+\tau)} \sum_{x(t)} p[x(t), x(t + \tau)] \log_2 \left\{ \frac{p[x(t), x(t + \tau)]}{p[x(t)]p[x(t + \tau)]} \right\} \quad (8)$$

where $p[x(t), x(t + \tau)]$ is the joint probability distribution function of $x(t)$ and $x(t + \tau)$, and $p[x(t)]$ and $p[x(t + \tau)]$ are marginal probability distribution functions of $x(t)$ and $x(t + \tau)$. The common way of selecting a proper time delay includes finding the first minimum in the mutual information function of the time series.⁴⁰ The mutual information function is zero if $x(t)$ and $x(t + \tau)$ are independent. Figure 6 shows the mutual information with time delay for PFs and AE signals. As illustrated in this figure the proper time delay for all time series is 1. Grassberger et al.⁴¹ demonstrated that the time delay is a non-critical parameter and most systems are robust and stable against changes in this parameter. This was confirmed in this work for PFs and AE signals of the spouted bed by computing RQA variables at different values of time delay and finding that these variables are not sensitive to the time delay. This is advantageous in characterization of spouted bed hydrodynamics by RQA since it is not necessary to find an optimum time delay value.

Another important input parameter is the radius threshold (ε), which determines the number of points that will appear in the RP. In this study, the proper value of ε was selected following three guidelines proposed by Webber and Zbilut³³: (i) the radius threshold should fall within the linear scaling region of recurrence rate vs. ε in a full logarithmic plot; (ii) recurrence rate must be as low as possible (e.g., 0.1% to 10%); and (iii) the radius threshold should not coincide with the first minimum of DET vs. ε . Figure 7a and b show the variations of RR and DET with the radius threshold for the dry spouted bed at inlet gas velocity of $1.2U_{ms}$, respectively. It can be seen in these figures that the values of 1 and 2 for AE and PFs signals, respectively, satisfy all the above mentioned guidelines. In the present work, the values of ε were set such that RPs of all signals show the same RR. This keeps the RR constant, providing the same number of black points in each RP.

A typical value for the minimal length of diagonal and vertical lines (l_{min} and v_{min}) is 2.^{26, 42, 43} Babaei et al.⁴³ analyzed determinism of the Lorenz system and found that the proper l_{min} is 2 and used this value for investigating the hydrodynamics of fluidized beds. Therefore, the minimal length of vertical and diagonal lines was considered to be 2 in this work.

4.3. RP Structures

Figures 8a-d show the normalized PFs signals and AE signals as well as the corresponding RPs of these signals measured in dry stable spouting bed at $U/U_{ms} = 1.2$. It is apparent from Figure 8b that dots in the PFs are scattered and cannot form a regular pattern such as long diagonal lines, like what is expected from the PFs-RP of fluidized beds.²⁶⁻²⁸ This demonstrates that PFs in the fluidized beds are more regular and periodic than those in the spouted beds. Marwan et al.³⁰ pointed out that RP of stochastic, periodic and chaotic systems have different typological structures: (i) stochastic system: characterized by uniform distribution of black points (ii) periodic system: if the plot contains several long diagonal lines and structures of the local zones are repeated to complete all RP; (iii) chaotic system: when non-regular short and long diagonal lines are present besides single isolated points. As can be seen in Figure 8b, the homogeneous single spots (which are the distinct feature of stochastic system) can be rarely found in RP of the spouted bed, indicating non-stochastic dynamics of spouted beds. The RPs of spouted bed also include short diagonal lines, some vertical and horizontal lines. Therefore, it can be concluded that the gas-solid flow dynamics in a conical spouted bed can be characterized as a combination of periodic and stochastic systems.

The difference between PFs-RPs of spouted and fluidized bed is related to the difference in the origin of PFs. Many researchers^{26-28, 44, 45} have used PFs for studying the hydrodynamics of the

fluidized beds and concluded that PFs represent interaction of three different phenomena in a fluidized bed: (i) high amplitude PFs of low frequency correspond to macro structures (eruptions and movement of large bubbles), (ii) medium amplitude PFs of higher frequency represent dynamics of agglomerates in the dense phase as well as small bubbles and (iii) low amplitude PFs of very high frequency arise from interactions among single particles and fluid and also include noise. On the other hand, the PFs of spouted bed at stable spouting are of low amplitude (hence small standard deviation) which indicates that a stable spouted bed is free of bubbles and slugs². In fact, PFs of spouted bed originate mostly from interparticle and gas-particle interactions. Since the hydrodynamic behavior of bubbles and agglomerates is more periodic and predictable than single particles, the PFs of fluidized beds are more periodic, thus, they are predictable for longer times in comparison with spouted beds. In other words, the PFs of a spouted bed are more chaotic than those of a fluidized bed. This is reflected in Figure 8b in which diagonal lines do not exist. Freitas et al.⁴⁶ found that the largest Lyapunov exponent of pressure fluctuations in stable spouting reaches maximum and the chaotic behavior is dominant in stable spouting, which is consistent with the results obtained in this work.

As can be seen in the RP of AE signal in Figure 8d, the contribution of diagonal lines is relatively more than that of the RP of PFs signals (Figure 8b). This indicates that the AE signals measured at the surface of the spouted bed is more deterministic than PFs signals measured at the base of the conical section. Here, the AE signals in a spouted bed mainly originate from particle-particle and particle-wall collisions at the surface of annulus and the fountain region while PFs signals mainly reflect the collective motions of single particles at the base of the spout. The gas velocity decreases in the fountain region and particles disengage from the gas and move downward in the annulus. Particle velocity in the fountain is much lower than in the base of the

spout. The hydrodynamic changes in the base of the spout are faster and more chaotic than at the bed surface. Therefore, RP of AE signals contains more diagonal lines compared to that of PFs.

4.4. Recurrence Quantification Analysis

As previously mentioned, overall patterns in a RP and its typology can provide useful insight on a dynamical system. However, detecting changes of finer patterns need quantitative analysis. In this study, the graphical display of the RP is quantified by RQA in terms of recurrence rate (RR), determinism (DET), laminarity (LAM) and entropy (ENT) for both PFs and AE signals. For this purpose, variations of RR, DET, LAM and ENT with time for three inlet gas velocities are shown in Figures 9-12 and discussed below. It should be mentioned that for calculations of RQA parameters, the original signal was partitioned into smaller windows or epochs (4000 points for PFs and 4400 points for AE signal). Adjacent windows offset was set to 4 seconds which means that each window was shifted 4 seconds over the main signal and then the RQA parameters were calculated for the new window. The number of segments does not affect the RQA parameters trends. Increasing number of segments only increases the number of windows and accordingly the number of data points in the RQA plots with time. However, to be able to capture small variations in the RQA parameters with small distances and at the same time to be computationally efficient, 4 seconds was selected through a sensitivity analysis in this study. Moreover, instability of spouting behavior was detected by visual observation throughout this work. The approximate start time of instabilities in the bed are presented by small arrows in Figures 9-12. In order to find a criterion for agglomeration detection based on RQA parameters, 95% confidence interval was calculated for each RQA parameter based on the data up to the start of instabilities and is shown by horizontal lines in Figures 9-12.

4.4.1. Recurrence Rate

Figures 9 a-c illustrate variations of RR of PFs and AE signals during the agglomeration process at the inlet gas velocities of $1.1U_{ms}$, $1.2U_{ms}$ and $1.3U_{ms}$, respectively. These figures show that the RR of PFs and AE signals increase during the agglomeration process, regardless of the gas velocity. As mentioned previously, the RR simply counts the black dots in the RP, excluding points on the main diagonal line (since $R_{i,j} = 1$ for any i) and measures the relative density of recurrence points in the RP. The trend of RR for PFs and AE signals illustrates that the relative density of black dots increases with increase in the water content of the bed. As discussed above, the adhesion tendency of particles intensifies by spraying water into the bed, causing formation of liquid bridges and particle agglomeration. Decrease in movement of particles is the main reason for observing PFs and AE signals with higher recurrence points. Number of black dots in the RP increases when amplitude of the time series becomes low. In contrary, if there are fluctuations with a relatively high amplitude, some points of the time series have a distance longer than the radius threshold, thus, are not considered as recurrence points.

A 95% confidence interval is shown in Figures 9a-c for RR parameter (horizontal dashed lines) and 3rd degree polynomial trend lines are drawn through each data set. As shown in these figures, the RR of AE signal passes its 95% confidence limit sooner than PFs. Therefore, it can be concluded that although RR of both PFs and AE signals are responsive to hydrodynamic changes in the spouted bed, RR of AE signals is substantially more sensitive to these changes. In other words, early detection of slight hydrodynamic changes of the spouted bed may be delayed or even missed by PFs, but detected sooner and/or more accurately by RR of the AE signals.

4.4.2. Determinism

Figures 10a-c show variations of determinism for PFs and AE signals during agglomeration at various inlet gas velocities. The trend of determinism is similar to RR and it increases with increasing water content of the bed. As mentioned above, determinism quantifies the fraction of black points which form lines parallel to the main diagonal line and quantifies the existence of deterministic structures. As water is sprayed into the bed, contribution of the bulk movement of particles and agglomerates increases. On the other hand, increase in the determinism indicates that PFs and AE signals approach a periodic situation and the hydrodynamic status of the bed becomes more periodic and more deterministic. Therefore, it can be concluded that the hydrodynamic behavior of bulk movement of particles and agglomerates is more predictable and deterministic than the movement of single particles. In other words, RP of time series in the presence of larger particles, clusters and agglomerates includes more deterministic structures.

Figures 10a-c also demonstrates that DET trend lines of AE signals cross the 95% confidence lines before PFs at all gas velocities. This fact reveals that although both AE and PFs techniques are responsive to hydrodynamic changes in spouted beds, such as instabilities in the fountain, stagnant zones in annulus and agglomeration, the AE measurement is substantially more sensitive to these changes. The ability to recognize differences in collisions of particles with only small changes in hydrodynamic behavior of particles reflect the capability of AE measurement technique and its potential for monitoring the spouted bed hydrodynamics. Comparison of Figures 9a-c with Figures 10a-c reveals that trend lines of RR and DET of PFs and AE signals cross the corresponding confidence lines at the same times. This means that sensitivity of RR and DET to agglomeration is the same.

4.4.3. Laminarity

Figures 11a-c demonstrate the variations of laminarity of PF and AE signals during agglomeration at various inlet gas velocities. It can be seen in these figures that the trend of laminarity of PFs and AE signals against the operation time is similar to that of determinism. Laminarity of PFs and AE time series increases with injection of water into the bed. As mentioned earlier, laminarity is the ratio of recurrence points that form vertical lines to all recurrence points and vertical lines reflect the probability of occurrence of specific states that do not change or change slowly. Agglomerate fraction increases when water is sprayed into the bed and interactions of agglomerate-agglomerate and gas-agglomerate are slower than those of single particles. Therefore, hydrodynamics of the bed becomes slower when agglomerates form and grow, thus, the laminarity of PFs and AE signals, which is related to the amount of laminar structures (whose state change slowly) in the bed, increases. On the other hand, contribution of the finer structures in the associated signals, like motion of single particles and small agglomerates decreases during the agglomeration process. These small structures show a more turbulent behavior and their laminarity is low.

Figures 9, 10 and 11 reveal that the three RQA parameters, RR, DET and LAM, increase during the agglomeration process. However, an increase in the recurrence rate and determinism is more pronounced and, they can detect agglomeration before collapse of the bed while laminarity does change significantly during the agglomeration process. Moreover, comparison of Figures 10 and 11 reveals that determinism and laminarity of AE signals are greater than those of PFs at all inlet gas velocities. This means that AE signals of a spouted bed measured at the bed surface are more deterministic and laminar than the corresponding PFs measured at the base of conical section of the bed which confirms the results obtained by visual observation of Figures 8 b and d. The PFs

measured at the base of conical section mainly reflect the particle motions at the base of the spout while AE signals represent movement and mixing of particles in the fountain region. As mentioned earlier, the gas velocity decreases in the fountain region, particles disengage from the gas and move downward in the annulus. Particle velocity in the fountain is much lower than in the base of spout. Rapid movement of particles in front of the pressure probe produce PFs with a more chaotic behavior than AE signals measured at the bed surface. Therefore, both determinism and laminarity of AE signal are greater than those of PFs signals.

4.4.4. Entropy

Evolution of RP entropies during water injection is demonstrated in Figures 12a-c for both PFs and AE signals at various inlet gas velocities. These figures show that the entropy of RP figures remains almost constant with time, both before and after water injection. This may seem to be in contradiction with the interpretation of entropy. In fact, entropy measures recurrence complexity of the RP figures. Entropy of a periodic system is low (small diversity in diagonal line lengths) while it is high in a chaotic system (large diversity in diagonal line lengths). It was shown by determinism and laminarity that the bed status approaches a periodic state with spraying of water while it is expected that entropy to decrease in this case. Llop et al.³⁸ and Wang et al.²⁹ also reported this contradiction in fluidized beds and computed greater values of entropy for the slugging regime than in the bubbling regime, although slugging is more periodic than bubbling. To solve this apparent contradiction, Llop et al.³⁸ investigated the effect of frequency of the signal on the entropy of RP and concluded that the number of truncated diagonal lines is different at different frequencies. This causes computation of a false large entropy although the system complexity is reduced. However, this contradiction between definition of Shannon

information entropy of probability distribution of diagonal line length and disorganization of the system reveals the need for another definition for entropy of RP. Therefore, it can be concluded that RR and DET are useful in diagnosing complex spouted bed hydrodynamics during agglomeration process while the entropy still requires standardization and verification.

5. CONCLUSIONS

In this study, the feasibility of detecting agglomeration in conical spouted beds via pressure fluctuations and acoustic emissions was investigated. The hydrodynamics of a conical spouted bed during agglomeration process was inspected through recurrence quantification analysis of both PFs and AE signals. For this purpose, RR, DET, LAM and ENT of the signals were extracted from corresponding recurrence plots and following conclusions were drawn:

- RR parameter of both PFs and AE signals increased during water spraying into the bed which shows that amplitude of signals decreased during time reflecting local de-spouting conditions. Increase in the RR of both PFs and AE signals can be attributed to the formation of liquid bridges leading to particle agglomeration.
- DET of PFs and AE signals increased during agglomeration process which indicates that bed behavior becomes more periodic and predictable. Therefore, it can be concluded that the hydrodynamic behavior of the bulk movement of particles and agglomerates is more predictable and deterministic than the movement of single particles.
- The laminarity of PFs and AE signals, which is related to the amount of laminar structures in the bed increased with water spraying. The decrease in particle velocity resulted in PFs and AE signals with higher laminar structures in the RP.

- The entropy of the signals was found to be independent of time indicating that ENT parameter is not suitable to evaluate the bed behavior during agglomeration of particles.
- Examining RQA parameters showed that the change in RR and DET parameters were more pronounced than other parameters investigated and, therefore, can be used as early indicators for agglomeration detection in conical spouted beds.
- Although both PFs and AE signals are responsive to dynamical changes in the spouted bed, AE signals were found to be substantially more sensitive and therefore, can predict the agglomeration sooner and/or more accurately.

It can be concluded that the acoustic emission measurements can detect differences in particle collisions, highlighting the sensitivity of such measurements for monitoring the spouted beds. Therefore, acoustic emissions can be used to design a non-intrusive, on-line and real time monitoring system for gas-solid flow in conical spouted beds.

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■ NOMENCLATURE

DET	determinism
D_c	column diameter, m

1		
2		
3		
4	D_i	cone bottom diameter, m
5		
6	D_o	gas inlet diameter, m
7		
8	d_p	particle diameter, m
9		
10		
11	D_{ij}	elements of distance matrix
12		
13	ENT	entropy
14		
15	f_s	sampling frequency
16		
17		
18	H_b	static bed height
19		
20	H_c	height of conical section
21		
22	l	diagonal line parameter
23		
24		
25	L	length of epoch
26		
27	LAM	laminarity
28		
29		
30	m	embedding dimension
31		
32	M	number of state space vectors
33		
34	MI	mutual information function
35		
36		
37	N	number of time series points
38		
39	N_l	number of diagonal lines
40		
41	p	probability distribution
42		
43		
44	$P(l)$	number of diagonal lines of length l
45		
46	$P(v)$	number of vertical lines of length v
47		
48	Q	volumetric flow rate, m ³ /s
49		
50		
51	R_{ij}	recurrence plot matrix
52		
53	RR	recurrence rate
54		
55		
56	U	inlet gas velocity, m/s
57		
58		
59		
60		

U_{mf} minimums fluidization velocity, m/s

U_{ms} minimums spouting velocity, m/s

$\vec{x}_i$ i-th point of state space trajectory

Greek letters

Θ Heaviside function

γ cone angle

ε radius threshold

σ standard deviation

τ time delay vector

υ vertical line parameter

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Table 1. Observed de-spouting times and mass fractions of particles collected at the end of the agglomeration test performed in full bed.

Velocity (U/U_{ms})		1.10	1.15	1.20	1.25	1.30
De-spouting time (min.)		2.8	4.7	6.5	30.5	83.2
Mass fractions (%)	$d_p > 1.4$ mm	4	4.5	5.1	7.2	8.9
	$1\text{ mm} < d_p < 1.4$ mm	39	42.5	46	50	55
	$0.71\text{ mm} < d_p < 1$ mm	53	50	46.4	40.7	34.6
	$d_p < 0.71$ mm	4	3	2.5	2.1	1.5

Figure Captions

Figure 1. Construction of a recurrence plot

Figure 2. Schematic of the conical spouted bed

Figure 3. A photo of the particles collected at the end of the agglomeration test (bed collapse) performed at $U/U_{ms} = 1.2$

Figure 4. Camera images of half bed during water (with black ink) injection, (a) dry bed, (b) start of water injection, (c), (d), (e) and (f) unstable spouting and (g), (h) and (i) bed collapse (despouting) at $U/U_{ms} = 1.2$.

Figure 5. Variations of fraction of number of false neighbours with embedding dimension for the dry bed operating at $U/U_{ms} = 1.2$.

Figure 6. Variation of mutual information with time delay for the dry bed operating at $U/U_{ms} = 1.2$.

Figure 7. Variation of (a) recurrence rate and (b) determinism of PFs and AE signals for the dry bed operating at $U/U_{ms} = 1.2$

Figure 8. (a) Normalized PFs signals, (b) RP of PFs with $m = 7$ and $\varepsilon = 2.5$, (c) AE signal, (d) RP of AE signal with $m = 7$ and $\varepsilon = 1.5$ (measured in dry bed operating at $U/U_{ms} = 1.2$).

Figure 9. Recurrence rate vs time for PFs and AE signals at (a) $U/U_{ms} = 1.1$, (b) $U/U_{ms} = 1.2$, (c) $U/U_{ms} = 1.3$ (Horizontal dashed lines are 95% confidence intervals).

Figure 10. Determinism vs time for PFs and AE signals at (a) $U/U_{ms} = 1.1$, (b) $U/U_{ms} = 1.2$, (c) $U/U_{ms} = 1.3$ (Horizontal dashed lines are 95% confidence intervals).

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Figure 11. Laminarity vs time for PFs and AE signals at (a) $U/U_{ms} = 1.1$, (b) $U/U_{ms} = 1.2$, (c) $U/U_{ms} = 1.3$ (Horizontal dashed lines are 95% confidence intervals).

Figure 12. Entropy vs time for PFs and AE signals at (a) $U/U_{ms} = 1.1$, (b) $U/U_{ms} = 1.2$, (c) $U/U_{ms} = 1.3$.

Figure 1. Construction of a recurrence plot.

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Figure 7. Variation of (a) recurrence rate and (b) determinism of PFs and AE signals for the dry bed operating at $U/U_{ms} = 1.2$

Figure 8. (a) Normalized PFs signals, (b) RP of PFs with $m = 7$ and $\varepsilon = 2.5$, (c) AE signal, (d) RP of AE signal with $m = 7$ and $\varepsilon = 1.5$ (measured in dry bed operating at $U/U_{ms} = 1.2$).

Figure 9. Recurrence rate vs time for PFs and AE signals at (a) $U/U_{ms} = 1.1$, (b) $U/U_{ms} = 1.2$, (c) $U/U_{ms} = 1.3$ (Horizontal dashed lines are 95% confidence intervals).

Figure 10. Determinism vs time for PFs and AE signals at (a) $U/U_{ms} = 1.1$, (b) $U/U_{ms} = 1.2$, (c) $U/U_{ms} = 1.3$ (Horizontal dashed lines are 95% confidence intervals).

Figure 11. Laminarity vs time for PFs and AE signals at (a) $U/U_{ms} = 1.1$, (b) $U/U_{ms} = 1.2$, (c) $U/U_{ms} = 1.3$ (Horizontal dashed lines are 95% confidence intervals).

Figure 12. Entropy vs time for PFs and AE signals at (a) $U/U_{ms} = 1.1$, (b) $U/U_{ms} = 1.2$, (c) $U/U_{ms} = 1.3$.

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