Early Detection of Agglomeration in Fluidized Bed Polymerisation Reactors

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Polyethylene (PE) and polypropylene (PP) make up for a large part of the total worldwide polymer production. Originally, both polymers were often produced in solution processes. However, fluidized bed reactors for polyethylene and polypropylene production gain increasing industrial interest because of the relatively low production costs, the ability to produce a wide range of polymer grades, and the environmental advantage of the absence of solvent. Nevertheless, fluidized bed production of polyolefins does have a serious drawback: agglomeration of particles is a major problem in this process (*e.g.*, Burdett *et al.*, 2001). The initial formation of particle clusters will deteriorate the otherwise uniform heat distribution in the bed. In the resulting hot pockets further agglomeration is promoted due to increased reaction rate and particle stickiness. Without proper control this process leads to excessive growth of particles in the agglomerate. This ultimately results in complete defluidization, forcing undesired plant shutdown and subsequent expensive cleaning of the reactor. To control agglomeration, operators commonly rely on average pressure drop or temperature measurements. Unfortunately, these techniques are insensitive to the actual agglomeration process: the detection is often too late.

Agglomeration is not only a problem in the production of PE and PP, but also in several other fluidized bed processes, such as combustion and gasification of waste and biomass (*e.g.*, Öhman *et al.*, 2000, Werther *et al.*, 2000) and coating of particles in the pharmaceutical industry (*e.g.*, Jono *et al.*, 2000). In the past years, we have developed DyMonT (Dynamics Monitoring Toolkit) for monitoring and controlling fluidized bed hydrodynamics. DyMonT detects agglomeration in an early stage and allows operator actions to control the agglomeration process and prevent defluidization. DyMonT is based on pressure fluctuation measurements (sample frequency in the order of 100 Hz), because pressure is a characteristic property for fluidized bed hydrodynamics. Moreover, pressure is one of the few properties that are easy to measure under operational conditions in an industrial fluidized bed (Werther, 1999). The principle of the method is to first measure of reference pressure signal (during a period of some minutes) that reflects the desired fluidization behaviour. Subsequently, the pressure signal is measured continuously and divided in blocks of fixed time periods, for example 5 minutes. The reference and every signal block to be evaluated are transformed into an 'attractor', so we obtain a reference attractor and several evaluation attractors. An attractor is a multi-dimensional distribution of delay vectors containing

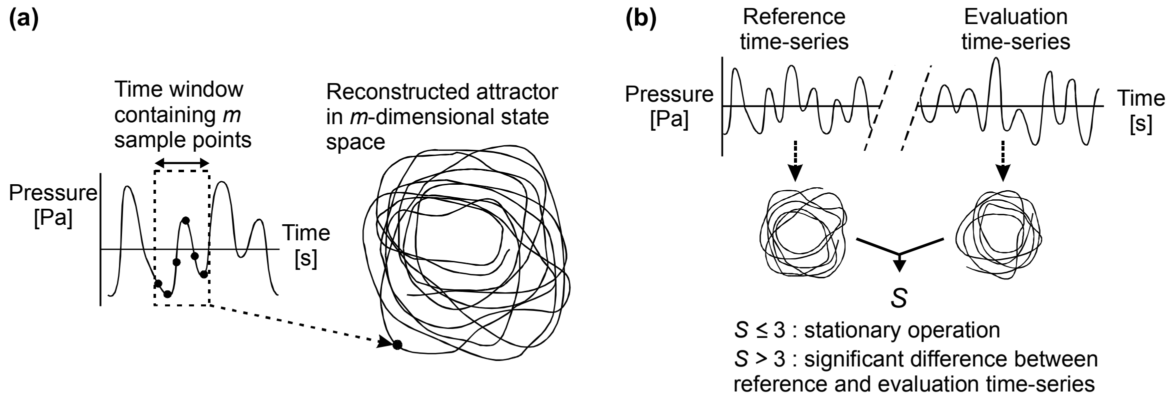

Figure 1. Schematic representation of **(a)** the reconstruction of an attractor from a pressure fluctuation signal, and **(b)** the comparison of attractors by the monitoring method.

successive pressure values (see Figure 1a). The attractor represents consecutive states of the dynamic system: it can be seen as a 'fingerprint' of the fluidized bed hydrodynamics as reflected by the pressure fluctuations in the bed. The reference attractor and each evaluation attractor are compared by calculating a statistic *S* using the Diks *et al.* (1996) test (see Figure 1b). For a more detailed description of the procedure, the reader is referred to Van Ommen *et al.* (2000). *S* represents the dimensionless distance between the two attractors. In this way all attractor properties are taken into account. For attractors generated by the same dynamics or mechanism, *S* has an expectation value of zero and a standard deviation of unity. When *S* is larger than three, we know with more than 95% confidence that the two attractors differ significantly, which means that the hydrodynamic behavior of the fluidized bed has changed.

An extensive experimental programme has demonstrated the selectivity and effectiveness of DyMonT to detect agglomeration in various types of systems (van Ommen *et al.*, 2001, Korbee *et al.*, 2003). Figure 2 gives an example of straw gasification in a lab-scale fluidized bed. In the first 20 minutes the fluidized bed hydrodynamics are stationary: the *S*-value stays

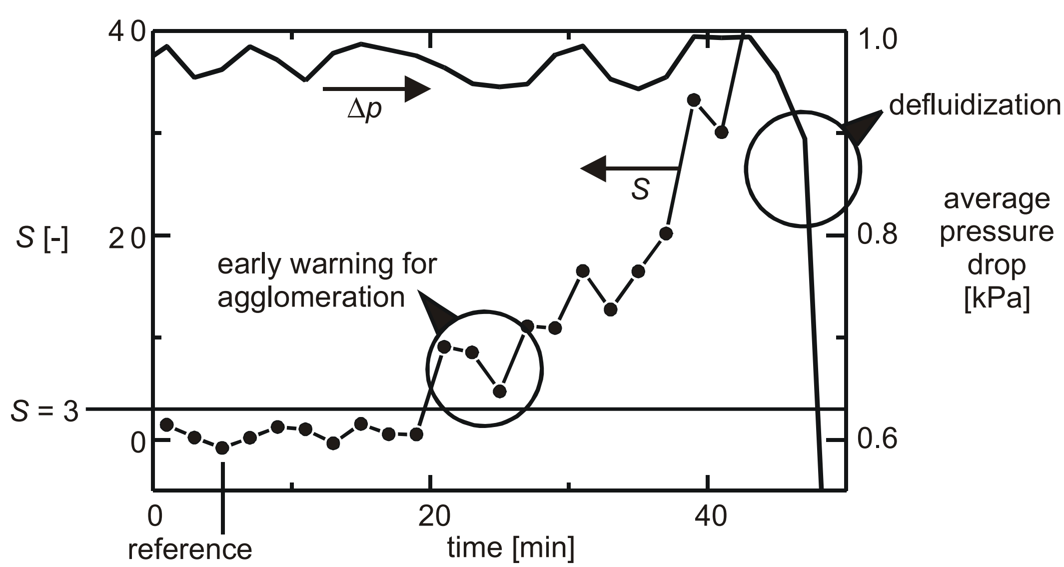

Figure 2. The application of the monitoring method during the gasification of straw. After the start-up, the *S*-value stays below 3 for some time, indicating that the hydrodynamics is similar to the reference situation. After 20 minutes, the *S*-value starts exceeding 3 indicating the onset of agglomeration. The pressure drop only shows defluidization when it is too late.

below 3. A part of this first phase is taken as reference. Then, the *S*-value already indicates in an early stage that the hydrodynamics of the bed changes, whereas the average pressure drop only indicates defluidization when it is too late. The early warning gives the opportunity to take counteractions to avoid serious problems such as defluidization. We have already demonstrated by small-scale tests that the method has the potential to return to a state of proper fluidization after the early detection of agglomeration (Korbee *et al.*, 2003). A prototype of the detection and control system is being tested in an 80 MWth wood-fired fluidized-bed combustion plant in the Netherlands. The goal is to develop solutions for typical industrial issues such as scale-effects, robustness of the system, equipment specifications, operation and maintenance, and process control (Nijenhuis *et al.*, 2004).

Currently, we are starting to test DyMonT for application in PE reactors. In these large reactors, it will be needed to measure pressure fluctuations at several positions. Besides analysing every signal, it is also possible to combine multiple signals into one *S*-value (van Ommen *et al.*, 2000). We will illustrate this by experiments with a 0.80 m diameter fluidized bed fluidized at 0.70 m/s and operated at ambient pressure and temperature. Pressure signals are measured at 0.14, 0.24, 0.34, and 0.44 m above the distributor. Initially, the bed consists of only fine sand (median particle size 532 μm; minimum fluidization velocity 0.14 m/s). This is taken as the reference situation. Gradually, increasing amounts of coarse sand (median particle diameter 1.28 mm) are added, while the bed mass is kept constant at 700 kg (giving a fluidized bed height of about 1.0 m). Figure 3 shows that both the single-signal *S*-values and the multiple-signal *S*-values increase with increasing fraction of coarse sand. However, the multiple-signal *S*-value – based on all four pressure signals simultaneously – is

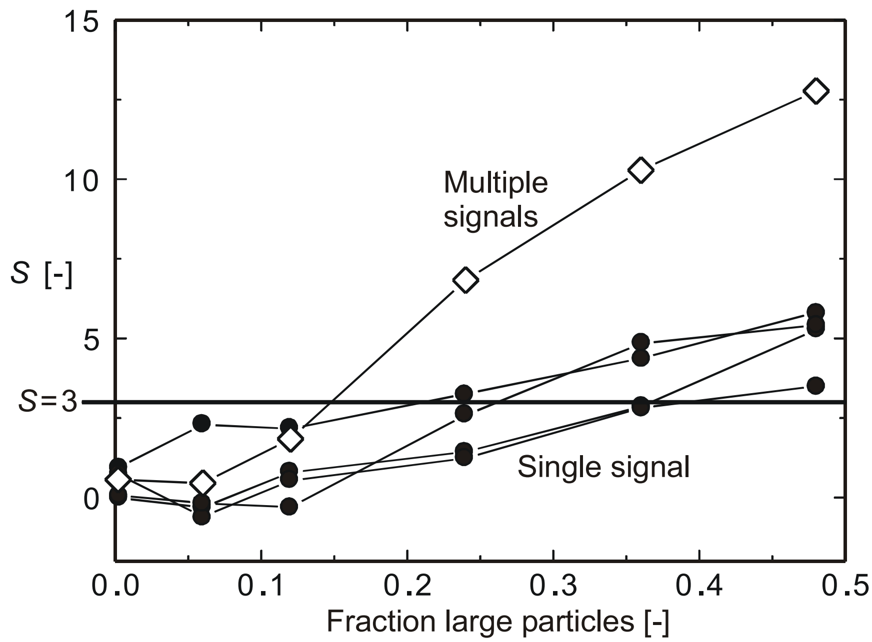

Figure 3. The *S*-value as a function of the fraction large particles in the bed. The single signal *S*-values are calculated for pressure signals from four different measurement positions; the multiple signal *S*-values for all four signals simultaneously. All symbols give the average *S*-value for five time-series. The lines are only meant to guide the eye.

much more sensitive to the coarse particle fraction and exceeds the *S*=3 limit (indicating a significant change in the hydrodynamics) already at a lower fraction of coarse particles. In practice, it is worthwhile to apply both the single signal analysis (more information about the location) and the multiple signal analysis (a higher sensitivity). Full-scale tests in industrial polymerisation reactors are scheduled for 2005. However, the use of DyMonT is not limited to early agglomeration detection in fluidized beds. It can also be used for detecting undesired misbehaviour in other multiphase systems, such as bubble columns (Villa *et al.*, 2003) and slurry reactors (Stienstra *et al.*, 2004).

References

Burdett, I.A., Eisinger, R.S., Cai, P., Lee, K.H., 'Gas-phase fluidization technology for production of polyolefins', in: Kwauk, M., Li, J., Yang, W.-C. (Eds.), *Proceedings of the Tenth Engineering Foundation Conference on Fluidization*, United Engineering Foundation, New York, USA (2001) 39-52.

Diks, C., van Zwet, W.R., Takens, F., DeGoede, J., 'Detecting differences between delay vector distributions', *Phys. Rev. E* **53** (1996) 2169-2176.

Jono, K., Ichikawa, H., Miyamoto, M., Fukumori, Y., A review of particulate design for pharmaceutical powders and their production by spouted bed coating, *Powder Technol.* **113** (2000) 269-277

Korbee, R., van Ommen, J.R., Lensselink, J., Nijenhuis, J., Kiel, J.H.A., van den Bleek, C.M., 'Early Agglomeration Recognition System (EARS)', in: *Proceedings of the 17th International Conference on Fluidized Bed Combustion*, ASME, New York, USA (2003) paper 151.

Nijenhuis, J., Korbee, R., Lensselink, J., Nijenhuis, J., Kiel, van Ommen, J.R., 'A method for agglomeration detection and control in industrial fluidized beds', presented at the 16th International Congress of Chemical and Process Engineering (CHISA 2004), 22-26 August 2004, Prague, Czech Republic (2004).

Öhman, M., Nordin, A., Skrifvars, B.-J., Backman, R., Hupa, M., 'Bed agglomeration characteristics during fluidized bed combustion of biomass fuels', *Energy Fuels* **14** (2000) 169-178.

Stienstra, G.J., Nijenhuis, J., Kroezen, T., van den Bleek, C.M., van Ommen, J.R., 'Monitoring slurry-loop reactors for early detection of hydrodynamic instabilities', accepted for publiation in *Chem. Eng. Proc.* (2004).

van Ommen, J.R., Coppens, M.-O., van den Bleek, C.M., Schouten, J.C., 'Early warning of agglomeration in fluidized beds by attractor comparison', *AIChE Journal* **46** (2000) 2183-2197.

van Ommen, J.R., Schouten, J.C., Coppens, M.-O., Lin, W., Dam-Johansen, K., van den Bleek, C.M., 'Timely detection of agglomeration in biomass fired fluidized beds', in: Geiling, D.W. (Ed.), *Proceedings of the 16th International Conference on Fluidized Bed Combustion*, ASME, New York, USA (2001) paper 131.

Villa, J., van Ommen, J.R., van den Bleek, C.M., 'Early detection of foam formation in bubble columns by attractor comparison', *AIChE Journal* **49** (2003) 2442-2444.

Werther, J., 'Measurement techniques in fluidized beds', *Powder Technol.* **102** (1999) 15-36.

Werther, J., Saenger, M., Hartge, E.-U., Ogada, T., Siagi, Z., 'Combustion of agricultural residues', *Progr. Energy Combust. Sci.* **26** (2000) 1-27.