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Application of microwave tomography to investigation the wet gas-solids flow hydrodynamic characteristics in a fluidized bed*

H.Q. Che^{1,2}, H.G. Wang^{1,2}, J.M. Ye¹, W.Q. Yang³ and Z.P. Wu³

1. Institute of Engineering Thermophysics, Chinese Academy of Sciences, Beijing 100190, China

2. University of Chinese Academy of Sciences, Beijing 100049, China

3. School of Electrical and Electronic Engineering, University of Manchester, Manchester, M13 9PL, UK

Abstract

Fluidized beds are widely used in pharmaceutical industry for particles coating and granulation. Due to the complex gas solids flow process, it is extremely desirable that a real time monitoring and control system is used in the process in order to enable a bubbling stable fluidization and reduce the chance of defluidization due to agglomeration with high moisture contents particles. The microwave tomography (MWT) technology has been developed for more than twenty years and was mainly applied in medical sectors. MWT has a wide range of frequency (1~2.5 GHz) and can be used to measure materials with high permittivity and conductivity. Therefore, it is a good choice for the monitoring of fluidized bed coating and granulation processes. In this paper, a MWT system with 16 antennas was designed and applied for the monitoring of wetting and coating processes in a lab-scale fluidized bed with top-spray and bottom-spray system. The hydrodynamics characteristics of wet gas-solids flow with different moisture content were investigated by MWT technology. This is the first time that MWT was applied in a real dynamics gas-solids fluidized bed. Experiment result shows that the MWT is available for the online monitoring of the gas solids flow with different moisture content. Different flow regimes were revealed based on the MWT images combined with pressure signals. The results indicate that the MWT provides valuable information to identify the defluidization in the fluidized bed process with high moisture contents materials. It is evident that microwave tomography has clear advantages over single point based measurement technology for process fault diagnose.

Keywords: Fluidized bed, Coating, Microwave tomography, Defluidization, Moisture content

1. Introduction

Fluidized beds are frequently applied in pharmaceutical, chemical and food industries for particles granulation, coating and drying. After the fluidized bed process, the quality of products can be significantly improved by increasing the mechanical resistance of particles, providing controlled release of active principles and protecting from external physical agents (da Silva *et al.* 2014). Normally, the fluidized bed process can be classified into three types, i.e. top spray, bottom spray and side (tangential) spray, based on the location of spray nozzle (Srivastava *et al.* 2010 and Henrich *et al.* 2015). The top spray fluidized bed is commonly used for granulation/agglomeration, during which water or water-based binder solution is sprayed into the bed and in contact with the particle surface to increase adhesion forces so that agglomerating excipients can be formed (Zhang *et al.* 2016). In the meantime, the product is dried in the same vessel. The bottom spray fluidized bed which was firstly introduced by Dale Wurster in the late 1950s (Wurster 1959) and is commonly used for pellet coating.

* Correspondence author, Tel.: 0086-10-82543140, E-mail: wanghaigang@iet.cn (H.G. Wang)

For a bottom spray fluidized bed system, there exists a concentric cylindrical tube which is called Wurster tube inside an outer conical chamber, and the flow region mainly includes two parts, i.e., the coating zone (inside the Wurster tube) and annulus zone (outside the Wurster tube). During the coating process, a pellet circulation exists between these two zones. Firstly, pellets move downward in the annulus zone and then enter the coating zone through the gap between the Wurster tube and air distributor. Secondly, the pellets move upward in the coating zone and are wetted by the coating solution. Finally, the pellets fall back to the annulus zone. The pellets are coated and dried constantly during the circulation until a coating layer is formed.

For the wet granulation processes in a fluidized bed, the moisture of bed material is one of the most relevant variables related with the stability of the fluidization regime transition. When the solution is over sprayed, the particle moisture increases rapidly, the particles may adhere to each other to form large entities. This undesired agglomerating is totally different from the desired process, which fixes the primary granules together so that larger porous secondary granules are formed. These entities can easily stick on to the hot wall and are difficult to dry, resulting in a non-uniform product at the end of the process (Wang *et al.* 2016). Another issue induced by the excessive solids moisture is defluidization inside the bed and it is defined as any condition where a well-fluidized bed loses fluidization in partial or in total (Lin *et al.* 2011). To determine the onset of defluidization is of high interest for fluidized bed process monitoring, which may reduce downtime and production loss and avoid the cleaning and start-up efforts resulting from a full plant shutdown (Shabaniyan *et al.* 2017a). Various techniques have been employed to detect the onset of defluidization, and these methods are mainly based on the processing of single-point based measurement data, including pressure, temperature and humidity (van Ommen *et al.* 2004, Parise *et al.* 2011, Serrano *et al.* 2015). However, these techniques all need sophisticated data processing methods and can only give an overall estimation of the fluidization state due to limited measurement information.

Thus, it is extremely desirable that a real time monitoring system is used in the fluidized bed coating and granulation processes to guarantee the product quality with a wide range of moisture content. Process tomography technologies, including electrical capacitance tomography (ECT), microwave tomography (MWT) and X-ray etc., have received a lot of attentions from various industrial sectors, as they can reconstruct the material distributions by non-intrusive measurements (Abdullah 2012, Semenov 2009, Zhang *et al.* 2014). Among these process tomography technologies, ECT is a commonly applied method to investigate the gas-solids flow characteristics (Chaplin and Pugsley 2005, Rimpiläinen *et al.* 2012, Ge *et al.* 2016). However, when the moisture of pellets is higher than a critical value, the application of ECT has its limitation due to the conductivity effect (Wang *et al.* 2016). In contrast, MWT is suitable for the measurement of wet solids due to its wide frequency band and can be used to measure materials with high permittivity and conductivity. Up to date, the MWT has been studied for application in the medical sector and is less developed for industrial applications including the real gas-solids flow application (Semenov 2009, Wu 2016, Töpfer *et al.* 2017). The basic principle of MWT is to determine the complex permittivity distribution of an object being imaged from the scattered electromagnetic fields measured around the periphery of the object by using electromagnetic waves from different incident angles, creating multi-views of the object (Wu *et al.* 2009). Wang *et al.* compared the measurement results of ECT and MWT for static measurements and it was found that both ECT and MWT work well with low moisture content particles (Wang *et al.* 2016). However, for high moisture content particles, MWT can give better image reconstruction results compared with ECT. This thus demonstrated that MWT can be applied to monitor the granulation, coating and drying process, in which the moisture content of solids may dramatically change.

The objective of the work in this paper is to investigate the wet gas-solids flow characteristics in a fluidized bed with top-spray and bottom-spray systems based on the MWT technology. A MWT sensor with 16 antennas is applied for the monitoring of wetting and coating processes in a lab-scale fluidized bed with top-spray and bottom-spray systems. This is the first time that MWT is applied in a real dynamics gas-solids fluidized bed. Experimental results show that the MWT is

applicable for online monitoring of the gas-solids flows with a wide range of moisture content. The gas-solids flow characteristics of different types of fluidized bed, pellet and spraying liquid are analyzed and compared. In addition, the standard deviation of normalized permittivity and the dominating frequency and the standard deviation of pressure signal are used and compared for the detection of defluidization of the bed, which gives a new tool for the fault diagnose of fluidized bed processes with high moisture content materials. Further details are given in the following sections.

2. Experimental setup and test materials

2.1. Test facility

The experimental setup for the fluidized bed system used in this paper is depicted in Fig. 1. The main frame of the fluidized bed is made up of a plenum in the bottom, a conical chamber, an expansion chamber and a filter on the top. Two pressure transducers (Setra 264) are installed at the positions of 50 mm and 300 mm above the air distribution respectively to measure the pressure drop with a sampling rate of 100 Hz. The inlet air is preheated by an air heater and the air velocity is adjusted by a roots blower. The outlet and inlet air temperatures are measured by two temperature sensors (HMP110). The liquids for both top and bottom spraying system are supplied by a peristaltic pump (LONGER BT100-1F) to a double fluid nozzle atomizer (Schlick 970). The atomizing air is provided by an air compressor of 0.1 MPa.

The bottom spray experiments are performed by a fluidized bed with Wurster tube. The fluidized bed is constructed by adding a plastics tube into the original conical chamber and moving the spray nozzle to the center of the air distributor. The air distributor also has open areas of holes lager in the central region (35%) and smaller in the annular region (4%). The Wurster tube has a length of 15 cm and a diameter of 6.5 cm, and the gap between the tube and air distributor is 20 mm.

< Fig. 1>

2.2. Test materials and experimental parameters

Sugar pellets and cellulose pellets are used in this paper and their properties are listed in Table 1. In this work, for the top-spray fluidized bed, wetting experiments are conducted and distilled water is used as the spraying liquid. For the bottom-spray fluidized bed with Wurster tube, both wetting and coating experiments are conducted and the coating solution is made up of 90% Polymethacrylate aqueous solution (Wantai Co., Ltd) and 10% coloring agents (Dyestuffs Co., Ltd). Table 2 gives the experimental parameters for different tests and in total nine tests are investigated. During the experiments, 3~5 g samples are taken from the chamber every 5~10 minutes and their moisture contents are measured by the loss on drying (LOD) method.

<Tables 1 and 2>

3. MWT sensor and dynamic calibration

3.1 MWT sensor and image reconstruction

A MWT system with 16 antennas from Sensona Ltd, UK is used in this work. The antennas are arranged with equal angular spacing along the circumference of a circle of 207 mm in diameter. The measurement region of the MWT sensor is in the range of 12 cm and 22 cm above the air distributor in the conical chamber, and the imaging cross-section is divided by 41x41 pixels. The top-spray and bottom-spray fluidized beds with MWT sensor are shown in Fig. 2. The operating frequency of the microwave system ranges from 1 GHz to 2.5 GHz and the sampling rate is 31 frames per second.

< Fig. 2>

Compared with other process tomography, likes electrical capacitance tomography (ECT) and electrical resistance tomography (ERT), image reconstruction of MWT is more complicated due to the scattering effect (Wu *et al.* 2009). Normally, the image reconstruction for the MWT is based on the scattered electromagnetic fields by using electromagnetic waves at different incident angle and frequency (Wu *et al.* 2009, Wang *et al.* 2016). Until now, a good number of iteration algorithms have been developed for MWT image reconstruction. In this work, due to the strong nonlinear problem, an improved Newton iterative approach based on the Levenberg-Marquardt (LM) method is used to solve the nonlinear and ill-posed inverse problem (Wang *et al.* 2016, Wu and Wang 2017). The solution is first determined by solving a Newton equation, which is a linearized approximation of the MWT inverse problem. The relationship between the microwaves scattered field E^S and the dielectric constant ϵ can be described as

$$F(\epsilon) = E^S \quad (1)$$

The dielectric contrast ϵ is generally complex. However, at microwave frequencies, for low-loss dielectric granules considered in this work ϵ is dominated by its real part. Hence, the image reconstruction is focused on the mapping of its real part, i.e. $\text{Re}[\epsilon]$.

Considering the change in the scattered electric field E^S with respect to the increment in dielectric contrast ϵ , the following relationship can be established

$$F'(\epsilon)\delta\epsilon = \delta E^S \quad (2)$$

where δE_k^S can be assigned to

$$\delta E_k^S = E_M^S - F(\epsilon_k) \quad (3)$$

With the solved value of $\delta\epsilon_k$, the new solution of ϵ at iteration step $(k+1)$ is given by

$$\epsilon_{k+1} = \epsilon_k + \delta\epsilon_k \quad (4)$$

In this work, multiple antennas, i.e. 16 antennas, are used and equation (2) can be arranged into a matrix form as

$$[D_k][\epsilon_k] = [b_k] \quad (5)$$

where $[D_k]$ is the matrix form of $F'(\epsilon_k)$, $[\epsilon_k]$ is the matrix form of $\delta\epsilon$, and $[b_k]$ is the matrix form of δE^S .

Equation (5) contains more unknowns than the measurements, it is strong ill-posed. Its solution takes in general the form

$$[\epsilon_k] = [\psi([D_k^*][D_k], \alpha)][D_k^*][b_k] \quad (6)$$

where $[D_k^*]$ is the conjugate transpose of the $[D_k]$, and α is the regularization parameter.

With Tikhonov regularization, equation (6) becomes,

$$[\epsilon_k] = ([D_k^*][D_k] + \alpha[I])^{-1}[D_k^*][b_k] \quad (7)$$

The regularization parameter α in the above equations lies between 0 and 1. Its choice is important to the iteration process and the accuracy of the solution as well as the stability of the solution. There are no regular rules for the choice of α (Wang *et al.* 2016). For noise-less data, α can be very small, e.g. $\alpha = 10^{-15}$. However, for the noise data as presented in reference (Nugroho and Wu, 2015), the regularization parameter is typically chosen to be $\alpha = 4 \times 10^{-5}$. The convergence of the above iteration can be evaluated from the value of

$$\text{Error} = \frac{\|\delta E_k^s\|^2}{\|E_M^s\|^2} \quad (8)$$

The iterative process stops when it meets the pre-set convergence criterion or the number of iterations is reached, and all of the reconstructed images show smooth feature after the iteration. The efficiency of this method has been validated in several applications (Joachimowicz *et al.* 1991, Franchois and Pichot 1997, Wu and Wang 2017). Details of the iterative algorithm as well as the evaluation of this algorithm are given in reference (Wu and Wang 2017).

3.2 Dynamic calibration

Before the measurement, the experimental system operates firstly with a set of low and high reference calibration. The measurements taken at low reference calibration correspond to placing a lower permittivity material (e.g. air) fully in the imaging area. On the other hand, the measurements taken at high reference calibration correspond to placing a higher permittivity material fully in the imaging area. The grey value of a pixel in the MWT image denotes the normalized permittivity (ϵ_N) of the object or substance in the imaging region and it is written as

$$\epsilon_N = \frac{\epsilon_M - \epsilon_L}{\epsilon_H - \epsilon_L} \quad (9)$$

where ϵ_L and ϵ_H is the relative permittivity of the low and high ϵ calibration material respectively, ϵ_M is the relative permittivity of the object or substance being imaged in the imaging area.

For a wetting and coating process, the permittivity value in the measuring area is a function of both solids concentration and moisture content, i.e. water content. The permittivity of water at 25 °C is 78 in the frequency of 1 GHz (Weast *et al.* 1989). In contrast, the permittivity of dry bulk sugar or cellulose pellets in the same frequency range is around 3. In this research, the maximum moisture content is 35% and the mixing permittivity of solids can be estimated in the range of 3 and 7.1 by the Maxwell mixing model (Chaplin and Pugsley 2005). Therefore, it is desirable to get the high reference calibration data by equation (9) during the whole wetting and coating process to account the moisture effect on the image reconstruction. Similarity the ECT calibration procedure introduced in references (Yang *et al.* 2004, Wang *et al.* 2009), high dynamics calibration for MWT is used in this research and the procedure is given as follow.

Firstly, the low reference calibration is performed by an empty bed, and then a series of high reference calibrations are performed with the moisture contents ranging between 7% and 35% with an interval of 2%. Once all the calibrations have been performed, the reference data are saved in the database for online measurement. The normalized permittivity is re-written as

$$\epsilon_N = \frac{\epsilon_M - \epsilon_L}{\epsilon_H(X) - \epsilon_L} \quad (10)$$

where $\varepsilon_H(X)$ is the high reference calibration data with reference moisture value of X .

For a subsequent wetting and coating experiments, after the moisture content of pellets is determined by the offline samples, the high reference calibration database with the closest value is used for image reconstruction based on the normalized permittivity value of (10). This dynamic calibration method is an effective approach to reconstruct images with varying solids moisture content, and it has been applied and validated in references (Wang *et al.* 2018 and 2019). Fig. 3 gives several reconstructed images of the same measurement data (from both static and dynamic processes) but with different high calibration data. The reconstructed images clearly show different solids distributions with different high references by equation (10). To obtain an accurate image, it is necessary to apply the dynamic high reference calibration based on the normalized permittivity equation (10).

< Fig. 3 >

4. Results and discussion

4.1 MWT validation with static measurement

As previously introduced, the MWT sensor has a high operating frequency ranging from 1~2.5 GHz. To validate the MWT measurement and choose a proper operating frequency for the measurement, several static tests are carried out before the dynamic tests. In static tests, three types of solids distributions, i.e. one rod, two rods, three rods are considered (see Fig. 4). The solids moisture content is 20%. To characterize the image quality, the correlation coefficient (C_{coef}) is introduced and it is defined as (Wang *et al.* 2014)

$$C_{\text{coef}} = \frac{\sum_{i=1}^P (\hat{g}_i - \bar{\hat{g}})(g_i - \bar{g})}{\sqrt{\sum_{i=1}^P (\hat{g}_i - \bar{\hat{g}})^2 \sum_{i=1}^P (g_i - \bar{g})^2}} \quad (11)$$

where P is the total pixel number in the reconstructed image, $\hat{g}$ and g are the true and reconstructed permittivity distributions, respectively.

Fig. 4 gives the MWT images measured from different solids distributions. Fig. 5 compares the C_{coef} of those images. As it can be seen, for different solids distributions, the reconstructed MWT images can give reliable information. For two rods and three rods types, the objects in the images tend to stick together with the increase in operating frequency. This feature can be seen more clearly in terms of C_{coef} , the highest C_{coef} values are generally associated with the operating frequency of 1 GHz (Fig. 5). Therefore, only a fixed frequency, i.e. 1 GHz, is used in the following investigations. More details on the validation MWT measurement are given in reference (Wang *et al.* 2016).

< Figs. 4 and 5 >

In the following, a series of top spray fluidized bed wetting and drying dynamics processes are given and analyzed first. Then another series of bottom spray fluidized bed coating and drying processes are given. To clearly indicate the change of measurement signals, low pass filter is applied to the measured data and shown as smooth line in the following curves as a solid line.

4.2 Gas solids flow characteristics in a top spray fluidized bed process

Fig. 6 gives the variation of normalized permittivity ϵ_N and the MWT images at different time for test 1. The value ϵ_N is obtained with a fixed calibration data, and it is the averaged value of one measuring frame. However, the MWT images are displayed using the dynamic calibration. As it can be seen from the figure, ϵ_N is very sensitive to the moisture content of solids when the moisture content is in the range between 4.7% and 35%. Two stages can be identified from the curve of ϵ_N , i.e., wetting and drying stages. It should be noticed that the noise level or instability of ϵ_N as shown in this figure is higher than the results obtained by electrical capacitance tomography with the same fluidization conditions (Che *et al.* 2017). There are three sources which contributed to the instability. The first one is contributed to the noise generated by the MWT system itself. This can be reduced by the application of low-pass filter to the measured data which can be seen clearly in the same figure. The second reason for this instability is due to choice of regularization parameter of α in equations (6) and (7) in the image reconstruction. An optimized regularization parameter should be given. The third source is contributed to the particle's surface electrostatic charge during fluidising the particles. The electrostatic charge will affect both the microwave and capacitance signals (Fotovvat *et al.* 2017).

In the wetting stage, the fluidization air temperature is kept at 20°C without heating and ϵ_N increases constantly due to the increase in solids moisture content. From the MWT images, in the time of 10 min, the solids moisture content is about 6.5% and the flow is bubbling flow regime. With the moisture content increased to 21% in the time of 50 min, the bubbling size increases obviously and most of the fluidization air pass through the center of the measurement region. The flow structure is similar with those measured by ECT in a same fluidized bed (Zhang *et al.* 2016, Ge *et al.* 2016). With continuous wetting, the moisture content is further increased to 35% in the time of 80 min and the MWT images show the gas solids flow transits from bubbling flow regime to intermittent flow regime. One possible reason for the transition of flow regime is due to the effect of interface force between wet particles with the increase of moisture content. Another possible reason is the swelling of the particles by absorbing water during the wetting process which can be observed from the bed surface expansion.

At the time of 82 min, the process is in the drying stage with the spray nozzle switched off and the air heater turned on. As shown in Fig. 6, the air temperature increases to 40°C in few minutes and then continuously increases up to 51°C in the rest of drying stage. From the MWT images, the gas solids flow recovers to bubbling flow regime at time of 120 min.

< Fig. 6>

The fitting lines of solids moisture content against the normalized permittivity ϵ_N is shown in Fig. 7 and the data obtained from the wetting and drying stages are illustrated separately. With the same solids moisture content, the normalized permittivity ϵ_N of the wetting stage is slightly higher than that of drying stage. This feature is possibly caused by the different operating temperatures in these two stages. Considering that the permittivity of solids is mostly determined by their moisture content, the above result agrees with the fact that the permittivity of water decreases with the increase in temperature (Weast *et al.* 1989). It is possible to apply the fitting data online to determine the solids moisture content in the process.

< Fig. 7>

Fig. 8 shows the variation in the normalized permittivity ϵ_N and the MWT images for test 2. This test is operated under the fluidization air temperature of 51°C, and the solids moisture content has a narrow range from 5.5% to 9.98%. The variation in ϵ_N due to solids moisture can be clearly seen from Fig.8. However, the flow regime of solids is not obviously changed after the water is sprayed into the bed. As it can be seen, the flow regime is kept in bubbling flow.

<Fig. 8>

Fig. 9 shows the normalized permittivity ϵ_N with MWT images for test 3, the bed material is changed to sugar pellets. This test involves two sets of wetting-drying processes, which are operated under two different fluidization air velocities, i.e. 1.73 m/s and 2.54 m/s. As it can be seen, the value ϵ_N increases during the wetting stage and decreases during the drying stage. In addition, the value ϵ_N decreases with the increase in fluidization velocity. From the offline samples, the moisture content of solids ranges narrowly in 1.04% ~ 1.77% during the test. The reason for the low moisture level of sugar pellets is that further wetting of sugar pellets may result in severe solids agglomeration or even bed defluidization, which will be discussed later. It can be observed from this test that solids agglomeration is much easier to occur when using sugar pellets as bed material. Considering that the variation in solids moisture is relatively small, the main reason for the increase in ϵ_N during the wetting process is the solids agglomeration. When solids agglomeration occurs, the solids are much harder to fluidize and it results in more solids collected in the measuring region.

<Fig. 9>

4.3 Gas solids flow characteristics in a bottom spray fluidized

Four coating and drying processes (tests 4~7) are performed by the bottom spray fluidized bed system with Wurster tube. Fig. 10 gives the variation in ϵ_N and MWT images of coating and drying test using cellulose pellets as the bed material (tests 4~5). It can be observed that the variation in ϵ_N for coating and wetting processes are similar and follow the trend of solids moisture change. However, the change of ϵ_N for the wetting test is smoother than that in the coating test, which indicates the fluidization is much stable with spraying pure water into the bed. From the MWT images, for both coating and wetting processes, the solids concentration in the measurement region increases when the solutions are sprayed into the bed, and it is hard to distinguish the boundary of solids flow between the coating and annulus regions.

<Figs. 10 and 11>

The MWT measurement results for wetting and coating test using cellulose pellets (tests 6~7) are given in Fig. 11. The variation in ϵ_N for coating and wetting test follows the similar trend in the early stage of spraying, and the solids fluctuation turns weak after 5~7 minutes of spraying. At the same time, most of the solids collect in the bottom region inside the conical chamber as shown in MWT images. In these two tests, the bed material turns fluffy due to spraying liquid and the bed height increases noticeably. The flow is actually a plug flow and nearly no bubbles exist in the annulus zone. In this flow regime, the solids move downward uniformly in the annulus zone and still circulate inside the bed periodically (Wang et al. 2010). It should be noted that the flow behaviours of the above two tests are different when the nozzle spraying is stopped. In the wetting test (test 6), the flow regime recovers to the bubbling flow while in the coating test (test 7) it is kept in the plug flow regime. This phenomenon can be confirmed both by the ϵ_N curve and MWT images. The main reason is that the pellets are agglomerated together by liquid bridge after the liquid spraying. For the wetting test, the agglomeration is largely eliminated after water is vaporized by hot air. However, for the coating test, Polymethacrylate retains after the coating solution is vaporized. As a result, pellets are still agglomerated by the dry Polymethacrylate, and the solids are kept in plug flow.

Table 3 compares the degree of agglomeration (R_{agg}) of the tests performed by the bottom spray fluidized bed. The degree of agglomeration of pellets is defined by

$$R_{agg} = \frac{m_{agg}}{m_{end}} \times 100\% \quad (12)$$

where m_{end} stand for the end point mass of the product and m_{agg} is the mass of the agglomerated pellets. m_{agg} is determined by sieving the coated pellets through a sieve with a nominal opening size 100 μm larger than the initial mean pellet size, and the mass of pellets retained by the sieve was considered as m_a . From table 3, R_{agg} of the tests using Polymethacrylate (coating test) is much higher than those using water (wetting test), this result is consistent with above discussions.

<Table 3>

4.4 Flow regime identification based on MWT measurement

The stability of the fluidization regime plays an important role in the coating and wetting process as discussed in above. It is necessary to combine different operating parameters, i.e. fluidization velocity, bed temperature, initial bed materials, spraying flow rate and atomizing air pressure, to achieve a stability flow regimes and make the fluid system stable (da Silva *et al.* 2014). Inappropriate flow regime might reduce the heat and mass transfer coefficient between particles and flow gas and damage the coating and wetting processes. Further, partial or fully defluidization is an undesired phenomenon in industrial applications involving fluidized bed (Parise *et al.* 2011). In the following part, we will discuss the defluidization analysis based on the MWT results.

The identification of defluidization in gas solids fluidized bed is normally based on pressure signals via different signals analysis mythologies, i.e. Gaussian spectral (Parise *et al.* 2011), standard deviation (van Ommen *et al.* 2004) and chaos analysis (Shabanian *et al.* 2017b). In the following part, two additional wetting tests (tests 8 and 9) using sugar pellets are performed to investigate the solids defluidization. For tests 8 and 9, water is continuously sprayed into the bed until the solids defluidization is attained.

<Figs. 12 and 13>

Fig. 12 gives the variation in ϵ_N and several photos of the bed surface for test 8, these photos show different gas-solids flow regimes including bubbling, defluidization and fixed bed state during the process. Fig. 13 gives MWT images for test 8 and the time interval between adjacent images in the same row is 1s. As it can be seen from Fig. 12, when defluidization appears, the solids concentration in the measurement region suddenly increases to a high level, and the fluctuation of solids flow turns weak. This feature can be confirmed by the MWT images in Fig. 13, with the spraying of water, the difference between adjacent image frames becomes less and less, and finally the solids distribution keeps nearly unchanged, which indicates it is in a fixed bed state. In this regard, the standard deviation (SD) of the normalized permittivity ϵ_N can be used to detect the defluidization. Therefore, the standard deviation of ϵ_N in five typical regions (a~e, see Fig. 14 and Fig. 15) in the cross-sectional MWT images are selected to analyse the defluidization process, and each region contains 4 x 4 pixels. The dominant frequency of power spectral density (PSD) and the standard deviation of pressure drop are also adopted in this work to compare the MWT results.

Fig. 14 gives the process curves of test 8, and these curves include variations in the dominant frequency of PSD, the standard deviation of ϵ_N and pressure drop. For both ϵ_N and pressure, the standard deviation is calculated with a 30s time period along the time-series and a 20 s overlap between the periods. As shown in Fig. 14, the SD curves of both ϵ_N and pressure show similar trends. The decline of SD curves starts around 2 min after the spraying of water, which indicates that the solids flow has been obviously impacted by the increasing solids moisture. With further spraying of water, the defluidization occurs when the SD curves decreases to around 20% of their original level. The value of SD decreases to zero when a fixed bed condition is reached. In practical, a threshold must be selected to differentiate fluidization and defluidization when using the SD value and the value of threshold should be adjusted with different systems (Serrano *et al.* 2015). In addition, one can observe that the SD values in different regions are not fully in the same level, which indicates

the solids flow in bed has a non-uniform distribution actually. The non-uniformity is more obvious for test 9. Finally results in the partial defluidization will be introduced in following.

<Fig. 14>

On the other hand, the turning point of dominant frequency of PSD can be observed when solids defluidization occurs, and a sharp decrease from 3 Hz to zero exists. However, for test 8, the SD decreases for nearly 3 minutes before the defluidization, which gives enough time for adjusting the operational parameters, such as reducing the solution spraying rate.

The process curves and the MWT images for test 9 are given in Fig. 15 and Fig. 16, respectively. Test 9 has a higher fluidization air velocity and contains two stages of spraying. In the first stage, water is sprayed with 20 mL/min during the time period from 5.5 to 9.7 min. The SD of ϵ_N shows different trends in different regions. The value of region “a” decreases while the values of regions “b”, “c” and “e” increase. This phenomenon indicates the appearance of stagnation zones near the region “a”, and this further results in more fluidization air passing through other regions. In the second spraying stage, the water flow rate is increased to 25 mL/min. The SD of ϵ_N of all regions decreases sharply. As a result, the SD of ϵ_N of region “a” decreases to nearly zero which indicates the solids in this region is fixed. However, the SD of ϵ_N in other regions decreases to a value above zero. By visual observation of the bed surface, many pellets stick on to a part of the wall of fluidized bed chamber and the fluidized bed is actually in partial defluidization state. This feature can be clearly seen in the MWT images (Fig. 16), the grey level is nearly unchanged in one part of the images. It should be noted that the fixed bed condition is not reached in this test.

In contrast, both of the SD of pressure signal and the dominant frequency of PSD decreased constantly when water is sprayed with 25 mL/min, and finally decreases to a value above zero. Table 4 qualitatively describes the variation of process curves and flow regimes of tests 8 and 9. It is obvious that pressure signal can hardly give reliable information in terms of partial defluidization of the fluidized bed. The advantage of process tomography monitoring is that it gives more information of the fluidized bed process than those single point measurement strategies.

<Figs. 15 and 16>

5. Conclusion

The MWT sensor has been successfully applied to monitor the dynamic processes of two types of fluidized bed, and dynamic calibration strategy is used for image reconstruction. Experiments with different types of solids and spraying liquids have been carried out. The conclusions are as follows.

In the top spray fluidized bed process, the changes in the flow regimes due to the variation in solids moisture and air velocity can be recorded and recognized by the MWT measurements. For cellulose pellets, the MWT measurement is applicable with the solids moisture even is increased to 35%, and the measured normalized permittivity value follows the same trend with the solids moisture. In addition, the flow regime of the bed changes from bubbling flow to intermittent flow when the solids moisture increased to 35%. For sugar pellets, the slightly increased moisture would result in the increase in the solids concentration. This is induced by the pellet agglomeration.

MWT monitoring of wetting and coating experiments with bottom spray fluidized bed showed that both the types of pellets and spraying solutions can affect the flow regimes in fluidized bed. Sugar pellets can be more easily affected by the change of spraying liquids compared with cellulose pellets. In addition, for sugar pellets, plug flow in annulus zone is observed

after several minutes of nozzle spraying, which indicates the appearance of pellet agglomeration.

MWT images can give more valuable information for the identification of defluidization, especially for partial defluidization. It is evident from the study in this paper that microwave tomography has clear advantages over single point based measurement technology for process fault diagnose.

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Nomenclature

C	Correlation coefficient (-)
E	Microwave scattered electric field (-)
Error	Iteration error of microwave tomography (-)
g	Grey level (-)
k	Iteration number (-)
m	Mass of material (g)
P	Pixel number in a reconstructed image (-)
PSD	Power spectral density (-)
R	Degree of agglomeration (-)
Re	Real part of the complex permittivity (-)
SD	Standard Deviation (-)
u	Fluidization air velocity (m/s)
χ	Moisture content of particle (%)

Greek symbols

ε	Permittivity (-)
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Sup scripts

$\wedge$	True distribution
-	Averaged
S	Scattered electric field

Subscripts

agg	Agglomeration
coeff	Correlation
end	End of process
H	High calibration
i	Index of pixel
k	Iteration number
L	Low calibration

M	Measurement
N	Normalized permittivity

References

- Abdullah, M.Z., 2012. Tomographic techniques for computer vision in the food and beverage industries. 64-96.
- Chaplin, G., Pugsley, T., 2005. Application of electrical capacitance tomography to the fluidized bed drying of pharmaceutical granule. *Chemical Engineering Science*. 60, 7022-7033.
- Che, H.Q., Wu, M., Ye, J.M., Yang, W.Q., Wang, H.G., 2017. Monitoring a Lab-scale Wurster Type Fluidised Bed Process by Electrical Capacitance Tomograph. *Flow Meas. Instrum.* In press
- da Silva, C.A.M., Butzge, J.J., Nitz, M., Taranto, O.P., 2014. Monitoring and control of coating and granulation processes in fluidized beds—A review. *Advanced Powder Technology*. 25, 195-210.
- Fotovat, F., Bi, X.T., Grace J.R., 2017. Electrostatic in gas-solid fluidized beds: A review. *Chemical Engineering Science*. 173, 303-334.
- Franchois, A., Pichot, C., 1997. Microwave imaging-complex permittivity reconstruction with a Levenberg-Marquardt method. *IEEE Transactions on Antennas and Propagation*. 45, 203-215.
- Ge, R., Ye, J., Wang, H., Yang, W., 2016. Investigation of gas–solids flow characteristics in a conical fluidized bed dryer by pressure fluctuation and electrical capacitance tomography. *Drying Technology*. 34, 1359-1372.
- Heinrich, S., Dosta, M., Antonyuk, S., 2015. Chapter Two - Multiscale Analysis of a Coating Process in a Wurster Fluidized Bed Apparatus, in: Guy, B.M., Jinghai, L. (Eds.), *Advances in Chemical Engineering*. Academic Press, pp. 83-135.
- Joachimowicz, N., Pichot, C., Hugonin, J.-P., 1991. Inverse scattering: An iterative numerical method for electromagnetic imaging. *IEEE Transactions on Antennas and Propagation*. 39, 1742-1753.
- Lin, C.-L., Peng, T.-H., Wang, W.-J., 2011. Effect of particle size distribution on agglomeration/defluidization during fluidized bed combustion. *Powder Technology*. 207, 290-295.
- Parise, M.R., Silva, C.A.M., Ramazini, M.J., Taranto, O.P., 2011. Identification of defluidization in fluidized bed coating using the Gaussian spectral pressure distribution. *Powder Technology*. 206, 149-153.
- Rimpiläinen, V., Heikkinen, L.M., Vauhkonen, M., 2012. Moisture distribution and hydrodynamics of wet granules during fluidized-bed drying characterized with volumetric electrical capacitance tomography. *Chemical Engineering Science* 75, 220-234.
- Semenov, S., 2009. Microwave tomography: review of the progress towards clinical applications. *Philosophical Transactions of the Royal Society of London A: Mathematical, Physical and Engineering Sciences*. 367, 3021-3042.
- Serrano, D., Sánchez-Delgado, S., Sobrino, C., Marugán-Cruz, C., 2015. Defluidization and agglomeration of a fluidized bed reactor during *Cynara cardunculus* L. gasification using sepiolite as a bed material. *Fuel Processing*

Technology. 131, 338-347.

- Shabanian, J., Sauriol, P., Chaouki, J., 2017. A simple and robust approach for early detection of defluidization. *Chemical Engineering Journal*. 313, 144-156.
- Shabanian, J., Sauriol, P., Mostoufi, N., Chaouki, J., 2017. Performance evaluation of different approaches for early detection of defluidization. *Powder Technology*. 316, 139-147.
- Srivastava, S., Mishra, G., 2010. Fluid bed technology: overview and parameters for process selection. *International Journal of Pharmaceutical Sciences and Drug Research*. 2, 236-246.
- Töpfer, F., Oberhammer, J., 2017. Microwave cancer diagnosis, Principles and Applications of Rf/microwave in Healthcare and Biosensing. Academic Press, pp. 103-149.
- van Ommen, J.R., de Korte, R.-J., van den Bleek, C.M., 2004. Rapid detection of defluidization using the standard deviation of pressure fluctuations. *Chemical Engineering and Processing: Process Intensification*. 43, 1329-1335.
- Wang, H., Li, Y., Qiu, G., Song, G., Yang, W., 2014. Measurement of gas–solids flow in loop seal and external heat exchanger in a circulating fluidized bed. *Powder Technology*. 266, 249-261.
- Wang, H., Senior, P., Mann, R., Yang, W., 2009. Online measurement and control of solids moisture in fluidised bed dryers. *Chemical Engineering Science*. 64, 2893-2902.
- Wang, H., Zhang, J., Ramli, M., Mao, M., Ye, J., Yang, W., Wu, Z., 2016. Imaging wet granules with different flow patterns by electrical capacitance tomography and microwave tomography. *Measurement Science and Technology*. 27, 114007.
- Wang, L.K., Heng, P.W.S., Liew, C.V., 2010. Classification of annular bed flow patterns and investigation on their influence on the bottom spray fluid bed coating process. *Pharmaceutical Research*. 27, 756-766.
- Weast, R.C., Astle, M.J., Beyer, W.H., 1989. CRC handbook of chemistry and physics. CRC press, Boca raton FL.
- Wu, Z.-P., 2016. Microwave Tomography as a New Modality for Biomedical and Biopharmaceutical Imaging, *Medicine and Biopharmaceutical: Proceedings of the 2015 International Conference*. World Scientific, pp. 687-692.
- Wu, Z., McCann, H., Davis, L., Hu, J., Fontes, A., Xie, C., 2009. Microwave-tomographic system for oil-and gas-multiphase-flow imaging. *Measurement Science and Technology*. 20, 104026.
- Wu, Z., Wang, H., 2017. Microwave Tomography for Industrial Process Imaging: Example Applications and Experimental Results. *IEEE Antennas and Propagation Magazine*. 59, 61-71.
- Wurster, D.E., 1959. Air-suspension technique of coating drug particles - a preliminary report. *Journal of the American Pharmaceutical Association*. 48, 451-454.
- Yang, W.Q., Chondronasios, A., Natrass, S., Nguyen, V.T., McCann, H., 2004. Adaptive calibration of a capacitance tomography system for imaging water droplet distribution. *Flow Measurement and Instrumentation*. 15, 249-258.
- Zhang, J., Mao, M., Ye, J., Wang, H., Yang, W., 2016. Investigation of wetting and drying process in a gas-solid fluidized

bed by electrical capacitance tomography and pressure measurement. Powder Technology. 301, 1148-1158.

Zhang, W., Wang, C., Yang, W., Wang, C.-H., 2014. Application of electrical capacitance tomography in particulate process measurement – A review. Advanced Powder Technology. 25, 174-188.

Tables:

Table 1 Properties of the pellets

Table 2 Experimental parameters of different tests

Table 3 The degree of agglomeration of the tests performed by bottom spray fluidized bed

Table 4 Qualitative description of the process curves and flow regimes for tests 8 and 9.

Table 1 Properties of the pellets

Material	Cellulose pellets	Sugar pellets
Mean diameter (μm)	690	800
Apparent density (kg/m^3)	896	863

Table 2 Experimental parameters of different test

Test No.	Fluidized bed type	Bed material	Solution type/Spray rate (mL/min)	Inlet air temperature ($^{\circ}\text{C}$)	Fluidization air velocity (m/s)
1	Top spray	Cellulose	Water/35	20, 51	2.54
2	Top spray	Cellulose	Water/30	51	1.73
3	Top spray	Sugar	Water/15, 25	51	2.54, 1.73
4	Bottom spray	Cellulose	Water/30	50	1.73
5	Bottom spray	Cellulose	Polymethacrylate/30	50	1.73
6	Bottom spray	Sugar	Water/20	50	1.73
7	Bottom spray	Sugar	Polymethacrylate/15	50	1.73
8	Top spray	Sugar	Water/20	50	1.73
9	Top spray	Sugar	Water/20~25	50	2.54

Table 3 The degree of agglomeration of the tests performed by bottom spray fluidized bed

No.	Solution/pellet	R_{agg}
Test 4	Water/cellulose	0%
Test 5	Water/cellulose	13.7%
Test 6	Polymethacrylate/sugar	5.1%
Test 7	Polymethacrylate/sugar	35.0%

Table 4 Qualitative description of the process curves and flow regimes for test 8 and 9.

Test No.	Time range (min)	Flow regime	SD from MWT image					Pressure drop	
			a	b	c	d	e	SD	Dominant Frequency
8	0~4	Bubbling	→	→	→	→	→	→	→
	4~8	Bubbling	↘	↘	↘	↘	↘	↘	→
	8~11.4	Defluidization	↘	↘	↘	↘	↘	→	↘
	11.4~12	Fixed bed	→	→	→	→	→	→	→
9	0~5	Bubbling	→	→	→	→	→	→	→
	6~9.4	Bubbling	↘	↗	↗	→	↗	→	→
	10~12	Bubbling	↘	↘	↘	↘	↘	↘	↘
	12~20	Partial defluidization	→	→	→	→	→	↘	↘

- “→” indicating constant, “↗” indicating increase and “↘” indicating decrease

Figures:

Fig. 1 Experiment set up

Fig. 2 MWT sensor with two types of fluidized bed

Fig. 3 Image reconstruction using the same measurement data but different calibration files

Fig. 4 MWT images in static tests

Fig. 5 Correlation coefficient of static tests

Fig. 6 Variation in the normalized permittivity and MWT images for test 1

Fig. 7 Relationship between moisture and normalized permittivity under the fluidization air velocity of 2.0 m/s

Fig. 8 Variation in the normalized permittivity and MWT images for test 2

Fig. 9 Variation in the normalized permittivity and solids moisture for test 3

Fig. 10 Variation in normalized permittivity and MWT images for tests using cellulose pellets

Fig. 11 Variation in normalized permittivity and MWT images for tests using sugar pellets

Fig. 12 Variation in normalized permittivity normalized permittivity (test 8)

Fig. 13 MWT images at different times of test 8

Fig. 14 Process curves for test 8 (total defluidization)

Fig. 15 Process curves for test 9 (partial defluidization)

Fig. 16 MWT images of at different time of test 9

Fig. 1 Experiment set up

Top spray

Bottom spray

MWT sensor

Measuring region

Fig. 2 MWT sensor with two types of fluidized bed

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Fig. 15 Process curves for test 9 (partial defluidization).

Fig. 16 MWT images of at different time of test 9