Development of 3D morphological descriptors for agglomerates with complex structures

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Abstract

Most of the food powders like maltodextrin are often produced in agglomerate form in spray fluidized beds in order to improve their consumer properties for daily uses. During the production of agglomerates moist amorphous particles are amalgamated together due to the strong adhesion forces. The produced agglomerates mostly have thus complex pore structures with irregular shapes. The description of the internal structure and morphology of the food agglomerates at the microscopic scale has still rarely been investigated in literature. The main reasons for this are the difficulty to measure the internal morphology of the agglomerates with fragile structures. In this work, a nondestructive X-ray microtomography technique is used as an appropriate experimental method to overcome this lack of data by a thorough characterization of the three-dimensional internal structure of agglomerates. X-ray images are not free of artifacts especially for maltodextrin agglomerates with low density and highly porous structure. Therefore, to decrease high-frequency noise and scattered X-ray beams, suitable scanning parameters are defined. A sequence of image processing steps is applied to the reconstructed X-ray images (voxel size 2.2 μm) of agglomerates in order to obtain a 3D view and extract necessary data for morphological characterizations. The internal porosity and the pore size distribution of agglomerates are obtained and evaluated. Diameter of internal pores is found to be in range between 20 and 90 μm. The open pores of agglomerate comprised of relatively large cavities and channels are also determined directly from the X-ray images. The sphericity is also calculated from the voxel value in 3D X-ray images for different agglomerates.

Keywords: Maltodextrin agglomerate, 3D morphological description, Internal microstructure, X-ray micro computed tomography

Introduction

In various industries fine powders are often produced in agglomerated form in order to improve their flowability, instant properties or simply to improve the optical appearance of the products [1]. Most food, pharmaceutical and chemical powders, which are agglomerated, are amorphous and water soluble like maltodextrin. Spray fluidized bed is one of the important apparatus, widely used in food industry for agglomeration process. The properties of an agglomerate are influenced by the microstructure which is created during processing [2]. The internal structure of agglomerates results from the spatial arrangement of the primary particles and pores inside the agglomerates. Despite of some research on maltodextrin agglomeration, the internal microstructure and morphology of this kind of agglomerate with complex structure has rarely been investigated especially in three dimensions.

A common problem in morphological analysis of this type of material is that three-dimensional information of its microstructure is required. Monitoring microstructure of this material using X-ray microtomography (XMT) allows us to begin to bridge this gap since a three-dimensional image of the specimen can be reconstructed from non-destructive, serial sections and can be processed to show and measure three-dimensional features [3]. There are many factors influencing the precision and accuracy of 3D information obtained from X-ray images. Image quality depends on real density variation within the object, and the energy of the X-rays has to be chosen so that the differences in resulting linear attenuation coefficients between the contrasting detail and surrounding material increase the contrast more than it increases the image noise [4]. In this study the suitable scanning parameters are defined and the artifact during scanning is decreased. By applying a sequence of image processing steps the morphological descriptors are derived from the XMT. The resulted images are used to quantify the internal local morphology of agglomerates and their internal porosity, porosity distribution, structures of pores, open porosity and sphericity.

Spray fluidized bed agglomeration

In spray fluidized bed, primary particles are fluidized and a binder solution is sprayed onto the fluidized particles, creating liquid bridges which form agglomerates. As soon as a desired size of agglomerates is achieved, spraying is stopped and further evaporation of liquid may lead to the solidification of binders at contact points between primary particles. Amorphous water-soluble substances like maltodextrin absorb significant quantities of water if they are exposed to an increasing relative humidity of the surrounding air [5]. Water migrates into the particles and solid material dissolves in the binder droplets spreading on the particle surface. Following this colliding particles adhere to each other at areas which are wetted or plasticized by the impacting liquid droplets. Therefore, maltodextrin agglomerates have complex and irregular structures. In this study, the agglomeration procedure was performed in a lab-scale batch fluidized bed granulator with a transparent, cylindrical fluidization chamber with152 mm inner diameter and 450 mm height (Glatt GmbH, Germany) (Fig. 1). Pure water was sprayed as a binder (plasticizing agent) with two-fluid nozzle which was provided by Düsen-Schlick GmbH (Untersiemau, Germany, model 940). The nozzle was placed on top of the chamber at the height of 150 mm from distributer plate with relative air pressure of 0.5 bar. For each experiment, 50 gr of powder was fluidized using a constant fluidization air flow rate of 70 kg/h, heated by an electrical heater up to 50 °C before it enters into the chamber.

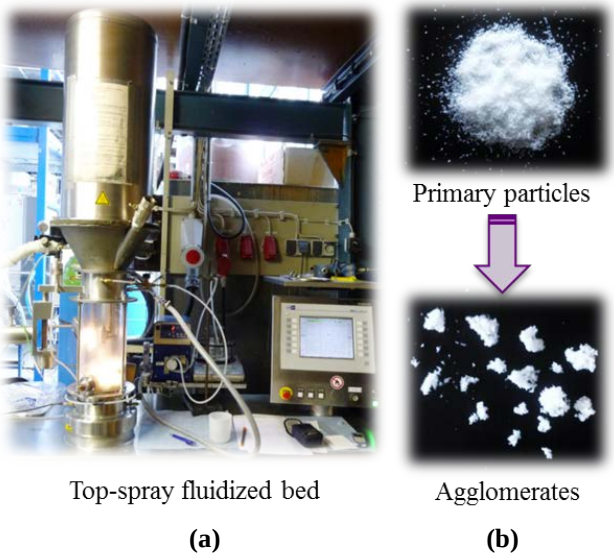

Figure 1: (a) The experimental lab-scale set-up for spray fluidized bed and (b) maltodextrin powders and agglomerates.

X-ray computed tomography scanning

The internal microstructure of the agglomerates was investigated using X-ray micro tomography device, manufactured by ProCon X-ray GmbH, Germany, denoted by CT Procon alpha 2000. Each agglomerate was scanned individually within the entire range of 0-360° with a rotation step of 0.3°. The agglomerate was placed on the sample holder and rotated to obtain radiographic projection from these angles. X-rays passing through agglomerate are absorbed according to a linear attenuation coefficient that has some spatial variation depending on the average atomic number, density and thickness of the agglomerate. Sufficient data obtained from the projection images was recorded by a 2D panel detector of 2048×2048 pixels to reconstruct slices of the three-dimensional agglomerates (Fig. 2). In order to obtain images with sufficient contrast and resolution by which the structural constituents can be easily separated, the X-ray source has been set at 50 Kv and 110 μA. Three images were taken per angular position, with an exposure time of 1500 ms. With these setting although captured images have good intensity and contrast, the images suffer from some artifacts like a shadow around agglomerate. This shadow is probably caused by scattering of X-ray beam during the scanning while the maltodextrin agglomerate has low density with high porosity. In order to eliminate this shadow, different experiments were done which are summarized in Table 1. This is observed that by using plastic filter in front of X-ray tube the shadow around the object can be removed.

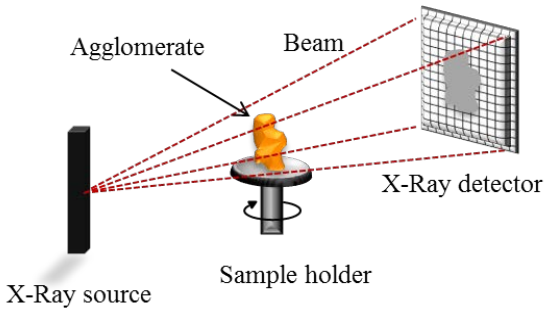

Figure 2: Principle of μ-computed tomography.

Table 1: Setting parameters used for X-ray scanning of an agglomerate.

	Main measurment	**Try 1**	**Try 2**	**Try 3**	**Try 4**	**Try 5**
Exposure time (ms)	1500	1500	1500	2000	1500	1500
Number of image averaging	3	1	1	1	1	1
Skip	1	0	0	0	0	0
Voltage (kv)	50	50	45	35	50	50
Current (uA)	140	140	110	100	140	140
Energy (W)	5	5	3.4	2.5	5	5
Number of projection	1200	1200	1200	1200	1200	1200
Approx. Voxel size (μm)	2.2	2.2	2.2	2.2	2.2	2.2
Measurement time (h)	1:58	1:00	1:00	1:00	1:00	1:00
Remark	--	Decreasing the number of images taking from each angel	Decreasing the energy of X-ray beam	Decreasing the energy of X-ray beam	Use aluminum foil in front of X-ray tube	**Use PMMA plastic in front of X-ray tube**
Result	Shadow around the particle, the image contrast is good	Shadow around the particle	Shadow around the particle, the image contrast decrease	Shadow around the particle, the image contrast decrease	Shadow around the particle	**No shadow around the particle**

Image processing

In order to obtain data required for morphological analysis, different steps of image processing need to be performed over the X- ray images. For image processing and quantitative analysis, the MAVI software of the Fraunhofer Institute for Technical and Industrial Mathematics in Kaiserslautern, Germany, was used. The common image processing steps are as follows: extraction of the region of interest, segmentation (binarization), removal of measurement noise by filtering, removal of sample holder effect by labeling, object filtering, and finally visualize volume image and internal micro structure of agglomerates (Fig. 3). Then, the qualitative and

quantitative analyses were performed on these binary images. For Further morphological analysis, more image processing operations and additional steps are necessary.

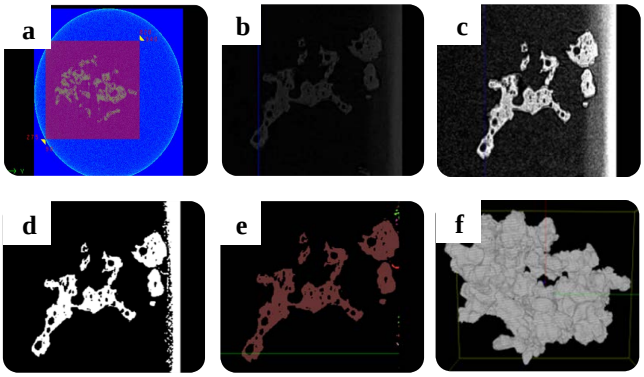

Figure 3: The procedure of image processing: (a) the cross sectional view of original image, (b) extract the region of interest, (c) binarizing the image, (d) removeing noise by filtration, (e) removing sample holder effect by labelling and object filter, (f) volume rendering.

Morphological descriptors

Porosity of closed pores (Internal porosity)

Closed porosity is defined as the ratio of the inner pore volumes to the agglomerate volume including internal pores (V_{agg}). This porosity is mostly attributed to the structures of primary particles. The solid volume can be obtained directly from binary images, but further image processing should be done in order to obtain the V_{agg}. The procedure is shown in Fig. 4 and shows a two-dimensional slice of the volume image. 3D views of the agglomerate before and after filling its internal pores are illustrated in Fig. 5.

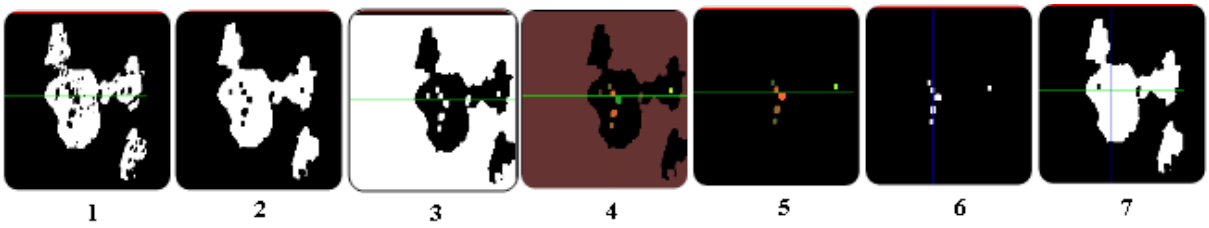

Figure 4: An image processing sequence for filling internal pores and for calculating volume of agglomerate including internal pores (V_{agg}): (1) Binray image, (2) Closing-cube (fill small pores), (3) Complement (invert the images), (4) Labeling (identify the objects), (5) Object filter (filter out the air), (6) Binarization, (7) fill all pores (combine steps 6 and 2).

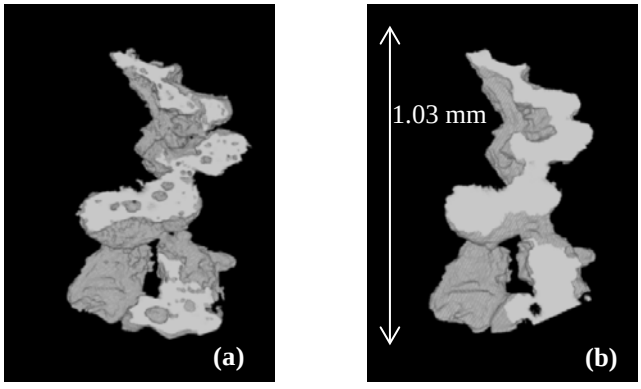

Figure 5: Cross-sectional 3D volume view of an agglomerate: (a) before and (b) after filling its internal pores.

In total, the internal porosity of agglomerates which is not connected to surface was found to be around 18.8%. Similar values (0.121–0.206) were reported for the internal porosity of cereal powder agglomerates in [6]. No significant differences in closed porosity values were found among the individually investigated maltodextrin agglomerates. This is because the internal porosity of agglomerates mostly belongs to the structure of the primary particles. The internal porosity of primary particles before agglomeration was also investigated and is around 25%. This result shows that the porosity of primary particles after agglomeration is decreased by 5%, because some of the internal pores are blocked and filled during the spray fluidized bed agglomeration process. The size distribution of internal pores is evaluated by the spherical granulometry function. In mathematical morphology, granulometry is an approach to compute the size distribution of grains in binary images, using a series of morphological opening operations. The granulometric analysis is applied to 3D µ-CT images of internal pores and it yields a volume weighted generalized pore size distribution. The 3D view of the spatial distribution of internal pores (Fig. 6b) illustrates the true internal morphology of the agglomerate. Marked with blue color are smaller pores which were defined by the closing mathematical morphology operators, while larger pores which were distinguished with the complementing and labeling method are marked orange. The density size distribution of internal pores displays pore sizes ranging between 16 and 90 µm (Fig. 7).

Moreover, if we compare the internal porosity of agglomerate with images possessing shadow artifact, it is noticed that the internal porosity for the same agglomerate increases by 5%. Therefore, with shadow artifact, the internal porosity investigation doesn't yield exact value and it is overestimated. In the images with shadow, there is free space between this layer of shadow and real object and all this space is accounted for the calculation of internal porosity.

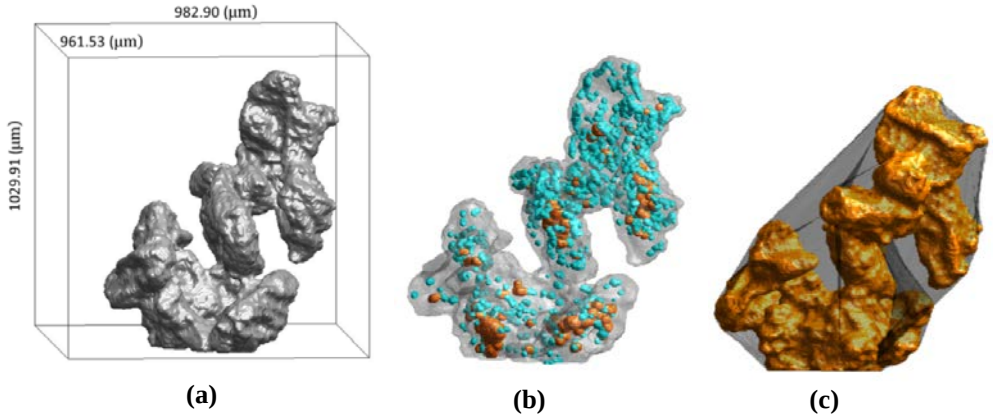

Figure 6: (a) The overall 3D view of maltodextrin agglomerate from X-ray images, (b) the spatial distribution of internal pores, (c) the total volume of agglomerate defined by convex hull method to calculate the porosity of open pores.

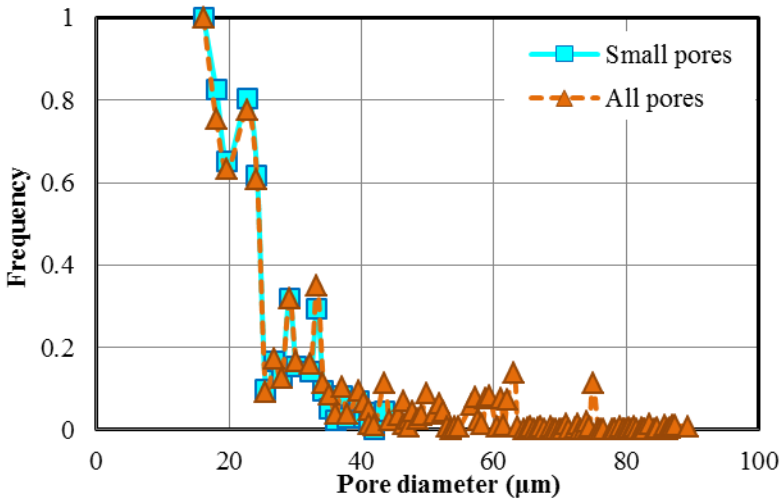

Figure 7: Normalized frequency distribution of internal pores represented by applying closing morphology and labeling method for distinguishing small and large pores.

Porosity of open pores

An open pore is a cavity or channel that communicates with the surface of the particle and has noticeable influence on strength and permeability of agglomerate. The porosity of such pore is defined:

$$\emptyset = 1 - \frac{V_{agg}}{V_{Total}} \quad (1)$$

The total volume of each agglomerate can be calculated by different ways but for a complex structure with irregular shape such as maltodextrin agglomerate the convex hull method is more suitable [7]. By this method, all channels and cavities will be considered in calculation of total volume (Fig. 6c). Convex hull volume can be directly obtained from XMT data.

The value of the open pore porosity for maltodextrin agglomerate which is calculated by the convex hull method is around 80%. This value is higher than the reported value (about 63%) for glass bead agglomerates [8]. This behaviour is attributed to properties of maltodextrin as amorphous water-soluble substance. During agglomeration, the viscosity of the residual water increases due to the dissolved amorphous substance. Therefore, sticky and plasticized surfaces of maltodextrin particles lead to the creation of more irregularly shaped agglomerates with open structures in the spray fluidized bed. In calculation of open pore porosity, there is not that much differences between the value of the image with and without shadow artifact. Therefore, appearance of the thin layer of shadow around the particle has minor effect on porosity of open pores.

Sphericity

In three dimensional shape analyses, sphericity describes how closely the particle resembles a sphere. It is defined as the surface area of a sphere, with the same volume of agglomerate (V_{agg}) divided by the surface area (S) of the 3D object (Eq. 2) [6,9].

$$Sphericity = 6\sqrt{\pi}\,\frac{V_{agg}}{\sqrt{S^3}} \quad (2)$$

The precision and accuracy of volume and surface area are important for sphericity analysis and it can be obtain from X-ray images. X-ray μ-CT data for sphericity, which are based on micro-scale resolution 3D images, are shown in Fig. 8. The mean sphericity value of agglomerates from X-ray images is 0.19.

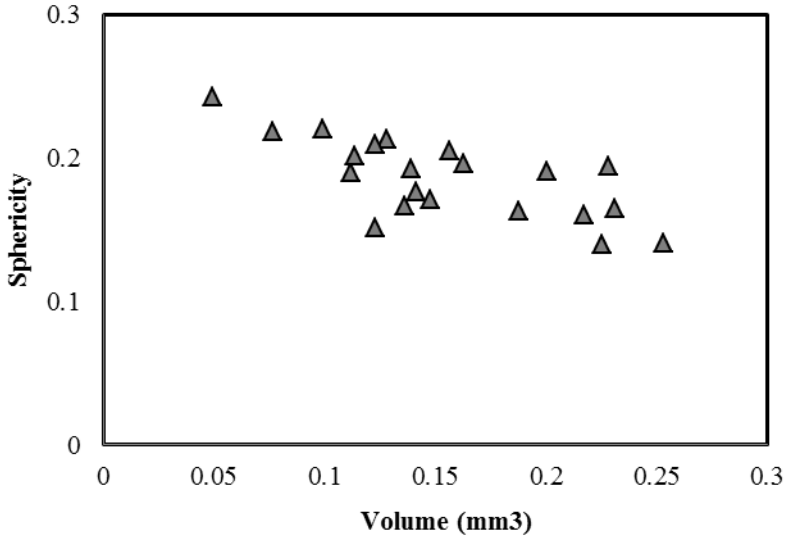

Figure 8: Sphericity of different agglomerates calculated from 3D X-ray images.

Conclusion

In this study, maltodextrin agglomerates were produced in a spray fluidized bed with spraying pure water. The internal microstructure of agglomerate was investigated by means of X-ray μ-computed tomography technique. The suitable scanning parameters were found for maltodextrin agglomerate which has low density with high porosity. The shadow artifact was removed by using PMMA plastic as filter during scanning. The

internal porosity, open porosity and sphericity were investigated for series of agglomerates. It was observed that removal of the shadow has noticeable effect in characterizing the internal structure of agglomerate like internal porosity. Moreover, the high value of open porosity and law value of sphericity proved that maltodextrin agglomerates produced in a spray fluidized bed exhibit irregular and open structure. In general, the present work demonstrates the ability of the X-ray μ-CT method to describe the 3D morphology and the internal microstructure of food agglomerates. The most serious limitation concerns the time and effort necessary for measurement and, especially, for image processing, which limits the number of agglomerates that can be analysed and assessed.

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