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Description of internal microstructure of agglomerated cereal powders using X-ray microtomography to study of process–structure relationships

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ABSTRACT

Agglomerated food powders are made from native particles which are assembled to form grains. The present investigation seeks to contribute to a better understanding of the three-dimensional internal morphology of food agglomerates (*i.e.* different couscous grains made from durum wheat semolina under different agglomeration process conditions) by means of microscopic methods (size, compactness) and X-ray micro-computed tomography (XMT), to bring understanding of the process–structure relationships. The three-dimensional internal morphology of food agglomerates was evaluated by means of X-ray micro-computed tomography (XMT), using a tabletop system with a resolution of several micrometers. XMT was considered to evaluate the internal porosity (or void fraction) as well as the size and spatial distribution of pores inside the grains. Closed porosity values ranged between 0.005 and 0.011. Diameter of internal pores was found to range between 13 and 155 μm which contribute to control mainly their end-properties. The experimental results were discussed to develop a better knowledge of the *process–structure relationships* for food agglomerates based on wheat powders, produced by wet agglomeration process. We demonstrated that the shearing conditions during the initial mixing stage significantly contribute to the internal structure characteristics of agglomerates. The cooking treatment is found to largely improve the mechanical strength of the agglomerates.

1. Introduction

Food powders are very present in everyday life (*e.g.* salt, pepper, spices, sugar, flour, and coffee) and also in the service of the industrial production of food as they are easy to preserve, transport, store, and process. The natural origin of the food powders brings complexity in their behavior [1]. The agglomeration is a unit operation during which native particles are gathered to form larger assemblies, called agglomerates. Agglomeration is implemented to improve the powders' functionalities such as, flow properties, dust generation, mixing capacity, aspect, dispersion, solubility, and controlled release [2–5]. Different agglomeration technologies can be used according to the aimed functional properties. Although significant scientific works have been conducted over the last 15 years, knowledge about the agglomeration of food powders still remains partial [5–8].

For food applications, the management of the agglomeration process greatly controls the structure characteristics of the agglomerates and their end-use properties [5–8]. The scientific works describing the structure of food agglomerates have been classically conducted by investigating macroscopic scale (*e.g.* size, shape, and porosity) and the molecular scale (*e.g.* chemical changes, melting yield, and denaturation levels). The description at microscopic scale has been only considered by superficial extern evaluation, through direct observations of grain surfaces using optical or scanning electron microscopy.

The description at the microscopic scale of the internal structure of the food agglomerates still represents a real scientific stake, which should allow a better understanding of process–structure or structure–property relationships. The qualification and quantification of internal granule microstructure are crucial for setting up processing maps and for describing agglomeration patterns and mechanisms [9,10].

The internal structure of agglomerates results from the spatial arrangement of the primary particles and binder components, their external organization, and internal porosity. A significant part of the agglomeration mechanisms found their origin in physical phenomena related to the establishment of capillary forces between native particles. For food powders, the physico-chemical reactivity of native particles under hydro-thermal stresses adds significant contribution of the viscous forces. The partial melting mechanisms of the native particles contribute to strengthen the internal structure of the agglomerates [8].

The internal structure of agglomerates has still rarely been investigated in literature [10–14]. Internal structure of agglomerates is related to porosity or void fraction, as well as the size and spatial distribution of pores. The presence of pores controls such basic properties as wet and dry strength, rehydration, dissolution, and disintegration in water.

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Classical methods to characterize the pore structure of materials include mercury porosimetry and gas adsorption techniques.

X-ray micro-computed tomography (known also as X-ray μCT or XMT) is a combination of X-ray imaging techniques and tomographical algorithms. Its principle is based on contrast in the X-ray images being generated by differences in X-ray attenuation arising principally from differences in density within the specimen. The resulting image is a projection of a volume on a plane (2D image). To get the 3D structure, a large number of radiographs are acquired while rotating the specimen between 0 and 180°. A mathematical inversion algorithm is then used to reconstruct the specimen 3D morphology [15]. Over the last decade, many manufacturers developed tabletop X-ray systems with a resolution of several micrometers, based on X-ray tubes with micro-focused sources, thus making it feasible to apply this technique for characterization of a wide range of materials in laboratory settings. XMT is a relevant technique to investigate both porosity and pore size distribution within an object. The microstructure of an entire granule or a tablet can be visualized down to details comparable to the resolution of the instrument, in a non-destructive way [16]. As a result, individual pores are visualized and the true internal morphology of granules is revealed. The pore size distribution could be determined without the assumption of a particular model of pore geometry. XMT does not yet possess the resolution necessary to resolve the structure of nanoparticle aggregates, but it is very well applicable to granular and porous materials that consist of larger primary particles in order to derive detailed micro-structural information [17]. Most of the morphological descriptors derived from the XMT data are not accessible in any other way.

XMT methods reveal the internal local morphology of granules to quantify their total porosity, porosity distribution and structures of pores [9,18–23]. The porosity results from mercury porosimeter are still considered to be more representative as it measures porosity of bulk granules, while XMT measures a single granule [23]. XMT is also able to describe the connectivity and the contact network built between the primary particles inside the agglomerates [17,22,24,25] as microscopic techniques do, but with the valuable advantage of being non-invasive.

Despite its adequacy, the use of XMT to investigate the internal microstructure of agglomerates produced by wet agglomeration has still been limited to few studies [18,19,24] and more particularly for food applications. The present investigation seeks two objectives: (*i*) to contribute to a better understanding of the three-dimensional internal morphology of food agglomerates (*i.e.* different couscous grains made from durum wheat semolina by different process conditions) by means of XMT; (*ii*) to bring understanding of the process–structure or structure–property relationships. XMT was considered to evaluate the internal porosity (or void fraction) as well as the size and spatial distribution of pores inside couscous grains. Grains have been characterized using a tabletop X-ray system with a resolution of several micrometers. The performances of the system are discussed in regard to the structural characteristic of the agglomerates and the number of samples. The three-dimensional internal morphology of agglomerates is discussed in regard to the process conditions used to generate them and to their end-properties.

2. Materials and methods

2.1. Raw materials

Four different couscous grains were selected to investigate the process–structure and structure–property relationships. The two industrial grain samples were produced under high shear mixing conditions during agglomeration and were steam cooked before the final drying stage. They differ by the rolling process which has been conducted in a rotating drum roller (grains named $G_{Ind\text{-}1}$) or over vibrating sifter (grains named $G_{Ind\text{-}2}$). The two industrial couscous grains were bought in a local store. Two experimental couscous grains were produced in our laboratory under low shear mixing conditions during agglomeration. They were both made by rolling over a vibrating sifter. They differ by the steam cooking stage, which was conducted (grains named $G_{Lab\text{-}1}$) or not (grains named $G_{Lab\text{-}2}$) during the process before the final drying stage.

Durum wheat semolina of industrial quality (France) was used as raw material to produce couscous grains. Semolina was first characterized using standardized methods. The water content of semolina (16.4 (±0.5) g water/100 g dry matter) was determined according to the approved method 44-15A [26], by weighing after oven drying (RB 360, WC Heraeus GmbH, Hanau, Germany) at 105 °C for 24 h. The total nitrogen content (TN) of semolina was determined by the Kjeldahl method, and the crude protein content (12.4 g protein/100 g dry matter) was calculated according to TN × 5.7 based on the AFNOR method (V 03-050). Median value of particle diameter of semolina (d_{50} = 265 (±8) μm) was measured by using laser granulometry (Coulter TMLS 230, Malvern, England) at room temperature. The semolina true density (1.478 (±0.005) $g \cdot cm^{-3}$) was measured by helium pycnometry. Semolina was stored in hermetic containers at 4 °C until experiments were carried out.

2.2. Couscous grains production

The grains named $G_{Lab\text{-}1}$ and $G_{Lab\text{-}2}$ were prepared in laboratory batch conditions according to the standard couscous processing (agglomeration, rolling, steam cooking, and drying stages) by using pilot scale equipments.

First, the agglomeration stage was conducted using a vertical low shear mixer SPI10 Labo V03 (VMI, France), equipped with a 10 liter bowl. A sample of 1.5 kg of semolina was introduced in the bowl and first mixed for 2 min at constant mixer arm (80 rpm) and mixer bowl (9 rpm) speeds, before water addition. Water was added directly over the semolina under mixing. Water addition was done at constant flow rate (18.6 $g \cdot min^{-1}$) by using a flat spray nozzle (TPU650017, Spraying System Emani, France) connected to the water supply network. Water addition was conducted to reach a water content of 35–40 g water/100 g dry matter. After the water addition step, the mixture was stirred for 2 min to homogenize the entire wet mass. The agglomerated mass was then spread over a column of 2 metallic sieves of decreasing mesh (2 and 0.9 mm). The rolling stage was conducted for 10 min at ambient temperature. The agglomerates were collected over the 0.9 mm sieve. For the steam cooking stage, the collected agglomerates were spread as a thick (3 mm) layer over a stainless steel screen and steam cooked for 15 min at 100 °C and atmospheric pressure, by using a 20 liter steam cooker (Afrem, Lyon, France). The cooked agglomerates were stabilized by drying over stainless steel screens (25 × 25 cm) using a pilot scale dryer (Afrem, Lyon, France), for 90 min at 50 °C and 50% relative humidity, and for 60 min at 70 °C and 80% relative humidity. The final couscous grains were then collected and stored in hermetic plastic cups until characterization.

2.3. Physicochemical characterization of agglomerates

2.3.1. Size distribution

The sieving stage applied at the beginning of the couscous processing has for objective to tighten the size distribution (between 0.9 and 2 mm) of the moist grains. To check that the final stages (*i.e.* cooking and drying stages) during process do not generate smaller (by erosion or rupture) or larger (by sticking) grains, size distribution of the final couscous grains was checked by a sieving method. A 300 g sample of final couscous was mildly sieved for 2 min over a column of 2 metallic sieves of decreasing mesh (2 and 0.9 mm). The content of each sieve was then weighted to check the size distribution of grains. Measurements were conducted in triplicate.