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Synchrotron X-Ray microtomography reveals interior microstructure of multicomponent food materials such as chocolate

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ABSTRACT

The current contribution discusses the structure analysis of a solid multicomponent food product (which is in this case dark chocolate) using microtomography. The material consists of a continuous solid lipid phase, in which particles are suspended. A detailed analysis of the microstructure is needed to understand migration processes, which are e.g. responsible for major problems in the confectionery industry such as chocolate blooming. In this study it was possible to clearly distinguish the particles from the continuous phase. Particle arrangement and structural imperfections within the sample were made visible by using synchrotron radiation. The observed imperfections, which arise during the manufacturing process, might act as migration pathways, since they propagate throughout the entire sample. The captured microtomographic images proof the presence of cracks and voids within a common industrial made chocolate. Future research has to show if migration is happen along the identified microstructural defects.

1. Introduction

One of the major problems in confectionery industry is chocolate blooming. It is manifested by the formation of a whitish haze on the surface due to large fat crystals (referred to as fat blooming) or sugar crystals (indicated as sugar blooming) which scatter the incident light (Lonchampt and Hartel, 2004; Rousseau and Smith, 2008; Rousseau and Sonwai, 2008; Altimiras et al., 2007). This makes the chocolate unappealing and leads to consumer complaints and subsequent large sales losses (Afoakwa et al., 2009; Aguilera et al., 2004). Sugar bloom is mainly associated with moisture migration and recrystallization of dissolved sugar on the chocolate surface. The formation of fat bloom can be related to migration of liquid lipids through the chocolate with subsequent recrystallization on the surface (Altimiras et al., 2007; Aguilera et al., 2004; Ghosh et al., 2002; Hartel, 1999). James and Smith (2009) imaged the surface of well tempered chocolate and it seems like the blade shaped fat crystals, which cause fat bloom, are extruded from the surface of chocolate. Bloom was initiated by temperature elevation to 30 °C where chocolate is in a soft, but not molten state. The crystals might develop from low melting crystals, which melt at 30 °C and migrate to the surface and recrystallized uncontrolled (Beckett, 2008). Diffusion as well as convective flow are widely discussed as the migration mechanism (Lonchampt and Hartel, 2004; Hartel, 1999). Many research studies aiming to understand blooming of chocolate have been conducted but the exact mechanisms are still unknown (Aguilera et al., 2004). A better understanding of the microstructure of chocolate is essential to comprehend the transport processes in chocolate and to control fat bloom. Transport phenomena in food materials are closely related to microstructure and according to Aguilera (2005) they are mainly impacted by structure elements smaller than 100 μm.

By investigating possible crevices and pores in chocolate, convective flow as a possible mechanism for lipid migration might be excluded or even identified as the main transport mechanism. Apart from crevices and pores, the particle arrangement within chocolate is of high interest for explaining potential transport pathways. Thus, further information about the microstructure of chocolate without sample destruction is needed to validate the

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proposed transport mechanisms and to develop a corresponding migration model. The aim of this study is to analyze the microstructure of chocolate with a focus on potential transport pathways such as possible crevices and pores in chocolate without sample destruction and to evaluate their origin.

Chocolate is a suspension of approximately 70 wt % particles (cocoa particles, sucrose and in case of milk chocolate milk powder), which are dispersed in a continuous cocoa butter fat phase, which is partly solid and partly liquid at room temperature (Aguilera et al., 2004; Beckett, 2008). Rousseau and Sonwai (2008) compared chocolate to concrete, comparing the fat phase to cement holding the dispersed particles together. Cocoa butter, the main fat phase, is a fat mixture and exists in both the liquid and solid state at room temperature because the different fat molecules crystallize in a temperature range and develop a non-uniform crystal structure. The solid percentage is given as the solid fat content. Cocoa butter can develop six different polymorphic crystal modifications with varying stability, melting temperature, packing densities and mouthfeel (Lonchampt and Hartel, 2004). The desired crystal configuration, which ensures the right melting properties, a good snap and stability, is realized by tempering the liquid chocolate mass, which is a pre-crystallization process during chocolate manufacturing.

Chocolate surface imperfections, such as pores and protrusions (mountain like topological elements) on the surface, have been observed in various studies (Rousseau and Smith, 2008; Rousseau and Sonwai, 2008; Smith and Dahlman, 2005; Dahlenborg et al., 2012, 2011; Rousseau, 2006). It was hypothesized but not proven in these studies that these surface imperfections are voids and cracks passing through the entire chocolate sample. Hartel (1999) claimed that small cracks and crevices present in chocolate might act as routes for lipid migration. He concluded that the recrystallization of fat at the surface seems to initiate at cracks and crevices and that these irregularities are starting points of fat blooming. Studies with atomic force microscopic studies showed defects with a diameter of a few micrometers at the chocolate surface (Rousseau, 2006), which according to Smith and Dahlman (2005) might be starting points of void cavities that extend deep into the chocolate body. Furthermore, Smith & Dahlman (2005) suggested that the cracks arose from the casting process similar to metallurgical processes. However, analysis of the interior microstructure without sample destruction could not be realized with atomic force microscopy due to limited penetration depth of around 10 μm (Rousseau, 2006). Dahlenborg et al. (2012) investigated the microstructure of white chocolate up to a depth of 10 μm from the sample surface with Raman microscopy. Protrusions and pores with a length of at least 10 μm and a diameter of approximately 10 μm have been identified. Dahlenborg et al. (2011) suggested as well that these pores may be important in developing fat blooming of chocolate. Loisel et al. (1997) indirectly showed the presence of some porosity in the order of 1–2% in well tempered chocolate samples using mercury porosimetry. They found that the porosity increases with an increasing amount of particles. But due to pressure applied during measurement and the soft nature of chocolate there might have been a collapse of the structure. Rousseau and Smith (2008) observed cracks and voids with diameters of a few microns to 250 μm in cross sections of chocolate samples prepared by cutting a chocolate sample to visualize the interior structure with environmental scanning electron microscopy. They found cracks, which travel about 100 μm inside the chocolate body. But as the sample was cut to visualize the chocolate interior crack formation due to sample destruction cannot be excluded. The presence of cracks and pores inside chocolate has not been proven without sample destruction yet, which might alter the sample microstructure due to sample preparation.

X-Ray tomography is an imaging technique to investigate the three dimensional internal structure of samples without destruction nor the need for tracer components (Williams and Jia, 2003). X-Ray tomography has already been used to investigate a wide range of particulate systems because of the ability of imaging complex microstructures of soft solids to get information such as packing patterns and the structure of pore systems (Williams and Jia, 2003; Farhang et al., 2014). It has been applied as well for non-invasive analysis of the internal microstructure of cellular food products with high amounts of voids such as aerated chocolate, mousse, marshmallow, bakery products and meringues (Lim and Barigou, 2004; Cafarelli et al., 2014; Trater et al., 2005; Licciardello et al., 2012; Frisullo et al., 2010; Haedelt et al., 2005, 2007). Frisullo et al. (2010) obtained structural data such as size, shape and distribution of pores and sugar granules in Italian aerated chocolates with X-Ray tomography. A simplified overview of the experimental procedure is given in Fig. 2. In regular absorption tomography the sample is placed on a rotation stage in front of a detector. The detector consists of a scintillator screen, which converts X-Rays into visible light, microscope optics and a camera, which subsequently captures a magnified image of the scintillator screen. The sample is then rotated by 180° or 360° in small angular steps (in our case around 0.2°). At each angular step a radiographic image of the sample is taken. Reconstruction of this assembly of 2D images yields a 3D volumetric model of the sample. This 3D model can be viewed and analyzed as desired.

2. Material and methods

2.1. Sample preparation

Chocolate mass was received from a leading manufacturer and thus represents a standard industrial made dark chocolate. The chocolate mass is directly taken from the production process after refining and conching. It is composed of approximately 48 wt % sucrose, 32 wt % lipid phase (30.5 wt % cocoa butter with addition of 1.5 wt % milk fat and 0.2 wt % lecithin) and 20 wt % of cocoa solids. The average particle size of the chocolate is $x_{50,3} \approx 10$ μm determined with laser diffraction. The chocolate mass was precrystallized by traditional hand tempering on a marble top to achieve a stable chocolate sample in the desired crystal polymorph. This lead to chocolate with the characteristic gloss and snap, which contracted in the cooling process, so that sample removal was facilitated. The molding process is depicted in Fig. 1. The maximum diameter of the sample is restricted by the field of view, which is dependent on the X-Ray tomography set-up (see Subsection 2.2), thus the maximum diameter for the chosen set-up is $D = 1.8\ \text{mm} \times 2 = 3.6\ \text{mm}$. Cylindrical molds with a diameter of $D = 3$ mm out of polypropylene were used. A direct filling from the molten mass into these cylindrical molds leads to incomplete filling of the molds. Therefore, the liquid chocolate mass was filled into standard chocolate molds first and the cylindrical molds were then immediately inserted into the bed of liquid chocolate mass to ensure complete filling and wetting of the cylindrical molds. To release air entrapped during chocolate preparation, the filled molds were put on a vibratory plate for some seconds until no visible air bubbles were arising anymore. The filled mold was immediately afterwards stored at 10 °C for 2 h to allow for cooling and solidification. As chocolate mass significantly contracts during solidification, samples could easily be demolded after solidification by gently pushing the cylindrical molds from the chocolate bed (Fig. 1). The chocolate samples were subsequently stored in an air-conditioned room for 1 day at 20 °C prior to measurement.