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Studying model suspensions using high resolution synchrotron X-ray microtomography

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ABSTRACT

The addition of minor quantities of secondary liquids to suspensions may lead to a transition from a fluid-like structure to paste-like structure for the system. Previous studies have shown how rheological properties such as viscosity and yield stress are affected, however, qualitative visual observation on the micro-scale during both short and long term storage has yet to be achieved or reported.

This research focuses on the movement of a secondary immiscible liquid (water or saturated sucrose solution) when added to a model food system. The model food system used in this study is a suspension of sucrose particles in a continuous oil phase to better understand the interactions between the particles and the liquid phases present. This was accomplished using dynamic X-ray computer tomography to study the behaviour of the sample. This non-destructive approach allowed the movement of the secondary liquid as well as the solid particles from the bulk suspension to be monitored through a time lapse of scans. This was achieved by observing the changes in the grey scale range of the droplet with time, which was then correlated to the uptake and movement of sucrose into the secondary liquid using an innovative method. This movement was due to the hydrophilicity and solubility of sucrose with gravity/sedimentation playing a minimal role.

1. Introduction

Several food products comprise of suspensions in which solid hydrophilic particles are suspended as a disperse phase throughout a continuous hydrophobic liquid phase. When a small proportion of a secondary immiscible liquid such as water or glycerol is added to a suspension of this kind, it can lead to behavioural changes from a fluid-like material to a paste-like material (Koos and Willenbacher, 2011; Johansson and Bergenståhl, 1992). This transformation has been ascribed to the formation of liquid bridges between the solid particles by the secondary liquid thereby modifying the rheological properties of the suspension such as its yield stress and viscosity which can increase several-fold (Koos and Willenbacher, 2011, 2012; Koos et al., 2012; Negreiros et al., 2015). The secondary liquid in this case has a greater affinity to the primary particles in comparison to the continuous phase because of their hydrophilic nature.

In food systems, the movement of moisture is generally considered from the view point of molecular diffusion. Moisture can enter from the surrounding atmosphere and/or may diffuse between different regions within the product. This movement is driven by the water activity differential between the different domains. This is of particular importance when considering the preservation of food products and the behaviour changes which may occur during storage. Past studies in the literature have focused on the assumption of diffusion being the dominant mechanism for moisture movement within such systems, and models are presented based on apparent or effective diffusivities to simplify the kinetics taking place (Yuan et al., 2009, 2012). These studies focused on the bulk mass transfer as an overall average of the sample, but did not investigate the localised movement. A more plausible explanation would be that a combination of transport mechanisms are responsible for the movement of a secondary liquid phase in a suspension including gravity, capillary and diffusion effects

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(Ghosh et al., 2004). However, at present, there is limited work within the literature that has been able to follow the localised movement of a secondary immiscible liquid within a suspension and qualitatively track the interactions and structural changes taking place.

X-ray computed microtomography (μCT) has been used previously to characterise several properties of food products in three dimensions (3D) such as the cellular structure of cereal bars (Chevallier et al., 2014; Falcone et al., 2004), analyse bread, biscuits and breadsticks (Frisullo et al., 2010a; Falcone et al., 2006), coffee beans (Pittia et al., 2011), the microstructure of apples stored at various environmental conditions (Herremans et al., 2014), the microstructure of mayonnaise (Laverse et al., 2012) and of various chocolates (Frisullo et al., 2010b; Reinke et al., 2016) to name a few.

The current work uses a state-of-the-art non-invasive method based on phase-contrast synchrotron X-ray μCT to monitor the movement of a secondary liquid within a suspension through a series of scans taken at different time intervals (dynamic or 4D μCT). Using this method it was possible to track behavioural effects over timescales of interest for food stability studies.

2. Materials and methods

2.1. Materials

Sucrose suspensions (icing sugar, ZMR Zuckermühle Rupperswil AG, Germany) and high oleic sunflower oil (Surface tension 35.5 mN/m, FTA125. Viscosity 49 mPa s, Malvern Kinexus lab+. Measured at 25 °C) were prepared according to the method described by Islam et al. (2016). The mass fractions of the solid and liquid phases were 50% wt for both; this amount was chosen based on the particle size distribution of the sucrose; ensuring adequate sunflower oil is present to completely coat all particles. The solid particles had a size distribution of 9–110 μm, with a median particle size (d_{50}) of 32 μm (measured using Malvern Mastersizer S, Malvern Instruments, UK).

Once the suspensions were prepared, they were mixed manually with a glass rod for a further 30 s and then transferred into cellulose straws (9 mm diameter, Fishers Ideas UK) which were glued (to prevent leaking of sample) to a plastic chuck specifically designed for the X-ray μCT experiments using a disposable pipette. A specifically prepared lid with a hole in the centre to fit the pipette tip was then placed on top, which ensured that the addition of the secondary liquid was to the centre of the suspension. The secondary liquid, which was either distilled water or a saturated sucrose solution, was added to the surface of the suspension using an electronic pipette (Eppendorf Xplorer, UK), locating the tip of the pipette close to the surface of the suspension, though not actually touching the surface. The distance between the surface of the suspension and the pipette tip was approximately 5 mm. Once the secondary liquid was added, another lid was placed which contained no holes, to prevent the evaporation of the secondary liquid during the experiment.

2.2. Rheological properties

The rheological properties were measured using a rotational rheometer (Kinexus Lab+, Malvern, UK). For liquid samples a cone & plate geometry was used (1/50, with a 1° angle and 50 mm diameter). Measurements were conducted using a shear rate ramp between the range of 10–1000 s^{-1}, ensuring steady state had been reached at each step before proceeding to the next set shear rate.

For the suspension sample, a serrated parallel geometry was used (diameter of upper and lower plate of 40 mm, 1 mm gap). The serrated plate geometry was used to prevent slippage of the sample, as the bulk solid can settle creating layer oil on the surface. Measurements were conducted using a shear rate ramp between the range of 0.1–100 s^{-1}, ensuring steady state had been reached at each step before proceeding to the next set shear rate. The data was fitted to a Herchel–Bulkley model to calculate the yield stress using Eq. (1):

$$\sigma = \sigma_y^H + k\dot{\gamma}^n \quad (1)$$

where σ is the shear stress (Pa), σ_y^H is the yield stress (Pa), $\dot{\gamma}$ is the shear rate (s^{-1}) with k and n are constants (Mewis and Wagner, 2012). All experiments were conducted at a temperature of 25 °C.

2.3. Scanning Electron Microscope (SEM)

Images were taken using a Scanning Electron Microscope (SEM) (JEOL, JSM-6010LA) to visually observe both primary particles, as well as, the mixture of water/sucrose droplet after removal from the sucrose and sunflower oil suspension.

Images were taken to visualise the surface topography of the sample after an initial gold coating which is needed for obtaining an image after applying a focused beam of electrons. Scans were conducted at a moderate energy level of 20 kV, which was found to be appropriate after trialling various energy levels for the samples being scanned.

2.4. Synchrotron X-ray μCT measurements

Transmission X-ray μCT is a non-destructive technique based on the mapping of the linear attenuation coefficient of X-rays crossing the studied sample (Kak and Slaney, 1988) and allowing the internal 3D microstructure of a sample to be visualised and analysed. This is achieved by capturing a series of projections, as the sample is rotated between the X-ray source and detector. These projections are then reconstructed using an algorithm which allows the 3D internal structure at the micron-scale spatial resolution to be seen.

All samples studied in this work have been imaged at the SYRMEP beamline of the Elettra-Sincrotrone Trieste laboratory (Basovizza, Italy). A schematic view of the employed experimental setup can be seen in Fig. 1.

Performing synchrotron X-ray μCT experiments at a third-generation synchrotron source, such as Elettra, highly improves the image quality compared to the use of conventional X-ray sources. In fact, a synchrotron X-ray beam features properties that significantly increase the data quality, in terms of contrast and spatial resolution, and then extend the imaging possibilities. These properties are the beam monochromaticity, its high intensity, high spatial coherence and nearly-parallel geometry. A monochromatic and highly-intense X-ray beam leads to reconstructed slices free

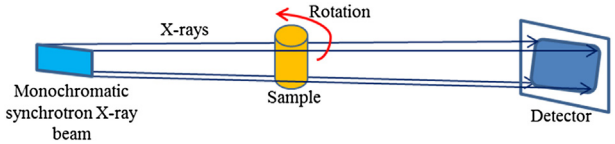

Fig. 1 – Schematic view of the experimental set up used for X-ray μCT at the SYRMEP beamline of Elettra.