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Novel approaches to oil structuring via the addition of high-pressure homogenized agri-food residues and water forming capillary bridges

Waleed Mustafa [a], Gianpiero Pataro [a], Giovanna Ferrari [a, b], Francesco Donsì [a, *]

[a] Department of Industrial Engineering, University of Salerno, Via Giovanni Paolo II, 84084, Fisciano, SA, Italy
[b] ProdAl scarl, c/o University of Salerno, Via Ponte don Melillo, 84084, Fisciano, SA, Italy

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A B S T R A C T

The suspension of micronized agri-food residues, such as tomato peels and spent coffee grounds, at 25% vol in peanut oil, results in the formation of a sample-spanning network (capillary suspension) upon the addition of a secondary immiscible fluid, such as water (at 0.17–0.57 vol with respect to the oil), to preferentially wet the particle surface, thus forming capillary bridges.

The strength of the capillary bridges, measured through the rheological characterization of the structured oil suspensions, depends on (a) the surface properties of the particles (in both cases prevalently hydrophilic, with the three-phase contact angles < 90°), (b) the fraction of added water, and (c) the mean particle size of the residues. In fact, the suspensions prepared with high-pressure homogenized particles (70 MPa, 3 passes) exhibit an apparent yield stress more than one order of magnitude higher than those prepared with coarser, high-shear mixed particles (>100 Pa vs. < 10 Pa). Finally, also the addition of a surfactant to the water phase dramatically affects the formation of the capillary bridges, reducing the interfacial tension at the oil/water interface. These results suggest a potential alternative route to vegetable oil structuring, to develop innovative foods and food ingredients based on low-calorie, health-beneficial agri-food residues, which not only induce the formation of a oleogel structure, but which also replace a fraction of the lipids.

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* Corresponding author.
E-mail address: fdonsi@unisa.it (F. Donsì).

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1. Introduction

Oils, composed of mixtures of triacylglyceride molecules with a high level of unsaturation, exhibit a liquid-like behavior at room temperature (Co and Marangoni, 2012), and therefore are unsuitable for certain applications, such as spreads, desserts and baked goods, where solid fats are preferred. Oil structuring may contribute to providing enhanced technological properties, using healthier lipid sources.

However, the conventional routes to oil structuring are generally based on blending the oil with >20 % wt solid fats, such as *trans*-fats and palm oil (Patel, 2015), generally regarded by consumers as unhealthy, at temperatures above the solid fat melting temperature, followed by a controlled crystallization process.

Alternative structuring routes are based on oleogelation, through the addition of gelators (low molecular weight organic compounds, polymeric compounds and inorganic compounds) (Patel, 2015), in which, the rheological properties are controlled by the gelator chemical type (lipid and non-lipid based oleogelators) (Dassanayake et al., 2009), as well as by the combination of different gelators (monocomponent and mixed system gels) (Co and Marangoni, 2012).

Recently, a novel structuring technique has been proposed, which is based on the addition of small amounts of a secondary immiscible fluid (for example water), to a particle suspension in a continuous phase (for example oil), in order to form a network (Koos and Willenbacher, 2011). Modification of the rheological properties of the suspension depends on the ability of the secondary fluid to create a sample-spanning particle network, supported by capillary forces (Koos and Willenbacher, 2011). Upon the addition of a secondary fluid wetting the suspended particles, the suspension will transform from a liquid-like to a gel-like state, with a noticeable increment in the apparent yield stress (Koos et al., 2011), hence forming a capillary suspension.

The strength of the capillary bridges is strongly influenced by the particle volume fraction, the volume of the capillary bridges, the density of the particles, the particle radius, the coordination number, which is the number of bridges per particle, the interfacial tension between the two phases, the mutual wettability of the ternary systems formed by (a) the continuous phase, (b) the secondary fluid and (c) the suspended particles (Patel et al., 2013).

The shape of the capillary bridges depends, instead, on the wetting ability of the particles by the secondary fluids, and hence can be expressed through the three-phase contact angle θ (Koos et al., 2012). When the secondary fluid wets the particles better than the bulk fluid, θ is $< 90^\circ$. For small bridge volumes, $\theta < 90^\circ$, causing a concave bridge shape, is classified as a capillary suspension in the pendular state. For $\theta > 90^\circ$, the bridge shape become convex, causing a positive Laplace pressure and particles repulsion, at a short distance, with the formation of a capillary suspension in the capillary state (Megias-Alguacil and Gauckler, 2010). Therefore, the transition of capillary suspensions between the pendular and capillary states is expected to occur at θ values of approximately 90° (Koos, 2014).

The capillary suspensions exhibit a superior stability against particle sedimentation and phase separation, even after long periods of storage (Goögelein et al., 2010), and therefore represent an attractive technique for the formulation of novel food products.

Due to their ease of preparation, food capillary suspensions enable the introduction of healthy ingredients in product formulation, as well as the elimination of other unhealthy components, which are conventionally used for structuring or texturization purposes.

Moreover, when used in oil structuring, capillary suspensions enable also the reduction of the caloric content, because part of the fats is replaced by a secondary fluid (at zero calories, such as water) and solid particles, such as fibrous materials.

Until now, only a few research papers have addressed the use of capillary suspensions for food applications, investigating, for example, the rheological behavior of sucrose crystals, starch and cocoa particles dispersed in oil or water continuous phases (Hoffmann et al., 2014; Killian and Coupland, 2012). In the case of sucrose crystals dispersed in an oil phase, the addition of water led to the formation of a self-supporting sucrose skeleton, through the adhesion of sugar crystals via the water capillary forces (Killian and Coupland, 2012). Similarly, in the case of starch and cocoa suspensions, the addition of small fractions of water in oil caused an increase in the apparent yield stress of the suspensions by several orders of magnitude, as a result of the capillary bridges establishing between the particles (Hoffmann et al., 2014).

This study focuses on the use of micronized agri-food residues, such as tomato peels and spent coffee grounds, in structuring a continuous peanut oil phase, via the formation of capillary suspensions. This application, which is still at a pioneering stage, faces one of the major challenge of the food industry, which is the replacement of saturated fats with vegetable oils, by proposing an alternative route to oil structuring, based on the use of naturally-derived ingredients as well as of green, purely physical processes, to develop rheological properties similar to those of solid fats. In addition, the use of agri-food residues offers multiple advantages: besides their availability at low costs, such residues are still rich in bioactive compounds, which can contribute to improving the health benefits of the structured oil, and their total reuse in food product manufacture can contribute to improving the sustainability of different sectors of the food industry. However, the use of agri-food residues also poses significant challenges related to the form, in which they are available. Therefore, this study also aims to elucidate the potential contribution of high pressure homogenization as a wet milling technique (Donsì et al., 2013, 2010b; 2009b) to control the particle size distribution of the residues (Donsì et al., 2009a), and affect the rheological behavior of the capillary suspensions (Ferrari et al., 2017), especially considering that previous studies have shown the crucial role of mean particles size on the strength of the capillary bridges (Bossler and Koos, 2016).