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Droplet breakup and coalescence in a twin-screw extrusion processing of starch based matrix

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ABSTRACT

Formation of oil droplets during twin screw extrusion processing of maize starch was investigated by analyzing the droplet breakup and coalescence mechanisms separately. For this purpose, the flow was characterized by computational fluid dynamics (CFD) using material data derived from online rheological measurements. The simulated results on local flow conditions were coupled to experimental data on the dispersed phase morphology, which was analyzed by confocal laser scanning microscopy (CLSM). This was used to elucidate the influence of process characteristics relevant for droplet breakup and coalescence. The results showed that increasing screw speed does not necessarily result in smaller droplet sizes. This could be related to the contradictory effects: increasing screw speed improves droplet breakup but also increases the rate of coalescence. Smaller droplet sizes were obtained at higher blend viscosities, which could be achieved either by increasing the feed rate or by using screw configuration that applies less mechanical stress. The results suggest that an increase in blend viscosity reduced the rate of coalescence. Moreover, increasing oil content led to an increased rate of coalescence, and therefore to remarkably bigger droplets. Selection of process parameters (e.g. screw configuration, feed rate, screw speed) based on the findings of this study allowed enhancing the dispersive mixing efficiency of triglyceride droplets during extrusion processing of maize starch.

1. Introduction

Starch biopolymers are one of the most common raw materials used in food industry. The starch and its derivatives have several attributes which make them favorable for encapsulation applications. They are cost effective, vary in molecular size, and have desirable physicochemical properties such as solubility, melting and phase change (Forssell, 2004; Lakkis, 2007). In particular, extrusion technology is extensively used in the production of starch based delivery systems, since it possesses several advantages, such as its energy efficiency; the lack of process effluents; its mixing capability; and its versatility with respect to ingredient selection and the shapes and textures of products that can be produced (Lakkis, 2007). Incorporation of functional hydrophobic components (e.g. lipophilic bioactives, flavours, antimicrobials, antioxidants or drugs) into extruded starch based products has gained wide popularity by food industry as well as by chemical, pharmaceutical, and medical industries (Mano et al., 2003; Shogren et al., 1993; Yilmaz et al., 2001). In most cases, it is favorable to dissolve the functional components in lipid based delivery systems and disperse these solutions into small droplets to improve their stability, bioavailability and palatability (Horn, 1989; Horn and Rieger, 2001; Ribeiro et al., 2006). It is crucial to control the microstructure obtained during processing (i.e. size and shape of the dispersed phase), since the functional properties of hydrophobic components strongly depend on these structural characteristics.

Incorporation of health promoting lipophilic bioactives like carotenoids, fatty acids or phytosterols into an amorphous extruded starch matrix for food applications mainly involves the plasticization of starch and dispersive mixing of triglycerides in this blend. Starch plasticization requires shear and/or heat generated by the rotation of screws and heated barrels to transform the crystalline structure of starch into an amorphous phase. The lipid-based delivery system comprising bioactives is most favorably incorporated and dispersed into the plasticized starch at the end of the extruder to reduce a possible degradation of the bioactives during extrusion processing (Emin et al., 2012a). This is followed by shaping/expanding and cooling to solidify the structure and form glass, thus, restricting loss due to molecular diffusion (Lakkis, 2007; Yilmaz et al., 2001).

To the best of our knowledge, there is only one study (i.e. Yilmaz et al., 2001) concerning droplet formation in a plasticized

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starch matrix in extrusion processing. Yilmaz et al. (2001) studied the influence of processing parameters such as screw speed, screw geometry and emulsifier content on the morphology of the dispersed phase. Their study presents a good overview on the relation between the process parameters and the final droplet morphology, however, does not investigate underlying mechanisms in detail. The formation of oil droplets includes both droplet breakup and coalescence, which may occur simultaneously or stepwise. Therefore, to improve the fundamental understanding of the droplet formation in extrusion processing, clear distinction between droplet breakup and coalescence mechanisms, and identification of the important process characteristics influencing these mechanisms are crucial. The complex nature of both the process and the raw material presents several challenges due to the complex geometry of a twin screw extruder and non-Newtonian rheological behavior of the plasticized starch, respectively.

The aim of this study is to understand the dynamics of morphology development during extrusion processing by analyzing droplet breakup and coalescence separately. Recently introduced computational methodology (Emin and Schuchmann, 2013) was used to characterize local flow conditions influencing droplet breakup, whereas a new experimental method was introduced to analyze coalescence during extrusion processing. The findings were used to get insight into the formation of oil droplets in plasticized starch during twin screw extrusion processing, which involves both mechanisms.

2. Fundamentals

An initial step in dispersive mixing is the deformation of a droplet in a flow field, resulting in an increase in the interfacial area between the two phases followed by a decrease in local radii perpendicular to the flow direction (Taylor, 1934). In case of plasticized starch as continuous phase, the viscosity is high which leads to small Reynolds number ($Re \sim 10^{-3}$ at the tip of the screws). Therefore, inertia can be neglected with respect to viscous shear stresses generated by the rotation of the screws. Moreover, with respect to the interfacial effects, the buoyancy effects are negligible. Thus, the deformation of a droplet in a plasticized starch matrix is mainly governed by the ratio of deforming shear stress τ and the shape conserving interfacial stress σ/R (with σ the interfacial tension and R the local radius). This ratio is characterized by the capillary number Ca (Taylor, 1932, 1934):

$$Ca = \frac{\tau R}{\sigma} = \frac{\eta_b \dot{\gamma} R}{\sigma} \tag{1}$$

where η_b is the blend viscosity, and γ is the shear rate defined as:

$$\dot{\gamma} = \sqrt{2d : d} \tag{2}$$

with the rate of deformation tensor d. The capillary number indicates that the larger the local shear stress $\tau (= \eta_b \dot{\gamma})$ relative to the interfacial stress ($=\sigma/R$) is, the more the droplet will deform until it breaks up. The specific value, above which the droplet breaks up in an external flow field of given shear stress, is called critical capillary number (Ca_{crit}). Taylor (1934) showed that the critical capillary number depends on the viscosity ratio p:

$$p = \frac{\eta_d}{\eta_c} \tag{3}$$

where η_d and η_c are the dispersed phase and continuous phase viscosity, respectively. The dependency of the critical capillary number on the viscosity ratio was established and validated for Newtonian systems (Bentley and Leal, 1986; Debruijn, 1991; Grace, 1982). The authors found that droplet breakup in a simple shear flow occurs most favorably at an intermediate range of $0.1 < p < 1$. At lower p values as found for the systems investigated here, Ca_{crit} steadily increases with decreasing p values. No breakup is possible at $p > 4$. In contrast, no limit for droplet breakup was found in elongational flow which was reported to be more effective than simple shear flow (Grace, 1982).

The breakup time, a number and size distribution of the droplet fragments after breakup depend on system parameters and differ qualitatively for various breakup mechanisms. Thus, characterization of the decisive breakup mechanism is crucial for the reliable prediction of dispersed phase morphology. In the complex geometry of an extruder, the droplet breakup is expected to be driven by high shear regions that are generated at the tip of the screw (Cheng and Manas-Zloczower, 1997; Emin and Schuchmann, 2013; Janssen, 1993; Sundararaj and Macosko, 1995). For low viscosity ratios ($p < 1$), the flow of the droplet through these high shear regions (at which $Ca/Ca_{crit} \gg 1$) results in affine deformation, followed by disintegration into very small droplets (Grace, 1982; Janssen, 1993). The number of the fragments increases with capillary ratio for $Ca/Ca_{crit} \gg 1$, which results in smaller droplet sizes (Grace, 1982; Stone, 1994). Similarly, the total breakup time is a function of capillary ratio (Ca/Ca_{crit}), and decreases with increasing Ca/Ca_{crit} (Grace, 1982; Janssen, 1993). Therefore, the capillary ratio (Ca/Ca_{crit}) can be considered to be a kinematic variable characterizing the efficiency of dispersive mixing (Emin and Schuchmann, 2013).

Recent studies (i.e. Sibillo et al., 2004; Verhulst et al., 2009; Yue et al., 2005) further showed that the elasticity of the matrix fluid has a significant contribution on the deformation of dispersed droplets, most especially if the applied flow field is transient as in extruders. Plasticized starch is a non-Newtonian fluid. To determine the critical capillary number for the triglyceride droplets suspended in plasticized starch matrix, recently we performed experiments in simple shear flow (Emin et al., 2012b, 2012c). The results showed that critical capillary number is significantly lower compared to Newtonian fluids at the range of viscosity ratio (i.e. 10^{-7}–10^{-5}) investigated:

$$Ca_{crit} = 6 \times 10^{-5} \cdot (p)^{-0.86} \tag{4}$$

From Eqs. (1) and (4), capillary ratio for the triglyceride droplets in plasticized starch in simple shear flow can be calculated by (Emin and Schuchmann, 2013):

$$Ca/Ca_{crit} = \eta_b^{0.14} \cdot \eta_d^{0.86} \cdot \dot{\gamma} \cdot R \cdot 10^5/6\sigma \tag{5}$$

Flow induced coalescence is the second fundamental phenomenon that should be understood and properly described to evaluate the final dispersed phase morphology. Although several researchers performed both theoretical (e.g. Chesters, 1991; Janssen, 1993; Loewenberg and Hinch, 1997; Rother and Davis, 2001) and experimental (e.g. Lyu et al., 2000; Minale et al., 1998; Ramic et al., 2000; Sundararaj and Macosko, 1995; Vinckier et al., 1998) work to characterize coalescence, the modeling of the complete picture remains still very challenging due to complex multivariate nature of the coalescence phenomena (Leal, 2004). Coalescence only occurs if at least two droplets collide and the film between those two droplets drains during their contact time. Therefore, the probability of coalescence P_{coal} is a function of probability of collision P_{coll} and probability of drainage P_{drain} (Chesters, 1991; Janssen, 1993):

$$P_{coal} = P_{coll} \cdot P_{drain} \tag{6}$$

The flow field, volume fraction of the dispersed phase, viscosity, shear rate and shear stress are some of the important parameters regarding flow induced coalescence (Chesters, 1991; Leal, 2004).