



Influence of droplet clustering on the rheological properties of emulsion-filled gels



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ABSTRACT

The aim of this work was to determine the effect and magnitude of fat droplet clustering on the rheological properties of emulsion-filled gels. To this end, we investigated experimentally the effect of fat hardness and emulsion droplet clustering in gelatin gels. Fat droplet clustering was induced or avoided by changing the temperature at which the emulsion was mixed with the gelatin. Rheological properties of the filled gels were investigated by uniaxial compression. The obtained experimental data show that next to changes in droplet hardness and matrix hardness, also droplet clustering contributes to the gel stiffness; even in the absence of discrete bonds between adjacent droplets. The experimental data was compared with the Kerner model, the Kerner model corrected by the effective volume fraction approach and the Kerner model considering an inhomogeneous distribution of the droplets. The Kerner model gives a good prediction for the gels filled with liquid droplets without clustering. The correction for an inhomogeneous distribution of the droplets best described the influence of emulsion droplet clustering in emulsion-filled gels, showing a quantitative evaluation on the role of droplet clustering in the rheological behaviour of emulsion-filled gels. This may provide guidelines for developing a strategy to control certain product properties.

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1. Introduction

Consumption of fat rich in saturated and trans fatty acids has been associated with the increment of illnesses such as obesity, type II diabetes and cardiovascular disease (Hu et al., 1997; Kris-Etherton et al., 2001; Ravnskov, 2010; Willett, 2012). Although the health implications of fats containing these saturated and trans fatty acids is debated (Ravnskov, 2010, 2012), there is a huge consumer and political pressure to reduce these type of fats (Rogers, 2009), and an important consumer-driven trend is observed for foods containing healthier fats by using different combinations of fatty acids. Examples of food products that are high in saturated fats are dairy products (e.g. sour cream, set yoghurt and cheese) and processed meat, and can be described as gels filled with emulsions

of semi-solid fat. An improvement of the health image of these products can be achieved by replacing the highly saturated semi-solid fat by a more unsaturated liquid oil. However, successful replacement of semi-solid fats by more liquid oils, while retaining the original texture, is not easily achieved by simply substituting the solid fat, since solid fats play an important role in the rheological and structural properties of these food products.

To understand the role of the emulsion droplets and their effect on the properties of emulsion-filled gels, these gels have been subject of extensive study by food scientists. It is well established that the rheological properties of emulsion-filled gels depend on the physico-chemical properties of the gel matrix and the emulsion droplets, the volume fraction of the emulsion droplets, the size and spatial distribution of emulsion droplets, and the strength of the interaction between the emulsion droplets and the gel matrix (van Vliet, 1988). Depending on the nature of this interaction, the emulsion droplets are classified as either active or inactive fillers (Ring & Stainsby, 1982). Active fillers are mechanically connected to the gel network and, depending on the ratio between their stiffness and that of the matrix, they either increase or decrease gel stiffness;

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thus, the gel stiffness will increase if the filler particles have a higher stiffness than the matrix, but will decrease if the filler particles are less stiff than the matrix. Inactive fillers have little chemical or physical affinity for the molecules forming the gel network and always decrease gel stiffness. These two main types of droplet–matrix interactions have, alternatively, been named “bound” or “unbound”, respectively (Sala, van Aken, Stuart, & van de Velde, 2007).

As mentioned above, the extend of the reinforcement of active droplets is influenced by its stiffness (i.e.: deformability). The droplet deformability is linked to the physical state of the fat and therefore to fat crystallinity or hardness. In the case of liquid oil droplets, the droplet deformability is dependent on the size and surface properties of the droplets, determining the Laplace pressure within the droplets (van Vliet, 1988). Several studies have shown that the stiffness of emulsion-filled gels decreases when solid fat is replaced by liquid oil due to a larger deformability (low stiffness) of the dispersed droplets (Houzé, Cases, Colas, & Cayot, 2005; Mor-Rosenberg, Shoemaker, & Rosenberg, 2004; Mor, Shoemaker, & Rosenberg, 1999; Xiong & Kinsella, 1991). Recently, we have also demonstrated experimentally that an increase in hardness of the fat droplets (by increasing the solid fat content) increases the stiffness of the overall gel, in line with the expectation that stiffer fillers increase the stiffness of the emulsion-filled gel (Oliver, Scholten, & van Aken, 2015). However, the magnitude of this reinforcement often significantly exceeds the predictions of existing theoretical models. As suggested by several authors (Chen & Dickinson, 1998; Kim, Renkema, & van Vliet, 2001; Lewis & Nielsen, 1970; Oliver et al., 2015; Sala et al., 2007), this enhancement is not only an effect of an increase in droplet stiffness, but may also be caused by an inhomogeneous distribution of the fat droplets, often as a result of droplet aggregation or clustering, thereby increasing the effective volume fraction of the droplets in the gel matrix. However, a qualitative analysis of the separate effect of droplet stiffness and an inhomogeneous distribution of the fat droplets (e.g: clustering or aggregation) on the rheological properties of emulsion-filled gel is still lacking.

The aim of the work described in this paper was to determine the effect and magnitude of fat droplet clustering on the rheological properties of emulsion-filled gels. To this end, we investigated experimentally the effect of fat hardness and emulsion droplet clustering in gelatin gels by controlling the degree of droplet clustering. The relative stiffness of the fat droplets was varied by selection of the fat type (sunflower oil, milk fat and beef fat) and the stiffness of the gel matrix was controlled by changing the gelling agent concentration. Moreover, droplet clustering (aggregation) was altered by the gelation kinetics, mixing the emulsion with the gelling agent at different temperatures (Sala et al., 2007). This resulted in two sets of samples: gelatin gels with non-clustered emulsion droplets and gelatin gels with clustered emulsion droplets. This allowed studying the effect of droplet stiffness and droplet clustering independently from each other with a constant composition of the gel matrix. The experimental results were compared to existing theories describing the mechanical behavior of composite gels and the validity of these theories will be discussed.

2. Theory

To understand the general approach used in this paper, we will provide a brief summary of the theoretical models used to evaluate our data (for a more detailed explanation see: van Aken, Oliver, & Scholten, 2015; Yang, Rogers, Berry, & Foegeding, 2011).

Several theoretical models have been developed to link the rheological behaviour of composite gels (emulsion-filled gels) to

the properties of the filler particles (emulsion droplets). The model established by Kerner (1956) for a composite gel and the model developed by Palierne (1990) for concentrated suspension, are two well-known models being applied in many studies. The Kerner model includes the compressibility of the materials, but neglects the interfacial properties of the filler particles, while the Palierne model includes the interfacial properties, but neglects the compressibility of the materials. In our case, the Palierne model should be considered in the case that emulsion droplets are completely liquid (since this model considers the interfacial properties), and the Kerner model is more appropriate in the case of emulsified droplets high in solid fat content. In the latter case, the effect of the interfacial tension becomes relatively less important than the fat crystal network inside the droplets (Yang et al., 2011). However, in order to be able to compare the results between systems with liquid oil and solid fat, we have used the Kerner model considering that all parts of the system are incompressible. For liquid droplets, we assume that a certain incompressibility is obtained from the Laplace pressure exerted by the large curvature of the emulsified droplets.

The Kerner model relates the shear modulus of a composite gelled material to the rigidity of homogeneously distributed filler particles as (Kerner, 1956):

$$\frac{G'_{gel}}{G'_{matrix}} = \frac{15(1 - \nu_m)(M - 1)\phi_f}{(8 - 10\nu_m)M + 7 - 5\nu_m - (8 - 10\nu_m)(M - 1)\phi_f} + 1 \quad (1)$$

where $M = \frac{G'_{filler}}{G'_{matrix}}$ and G'_{gel} , G'_{filler} and G'_{matrix} are the shear moduli of the overall gel, the filler particles and matrix, respectively, ϕ_f is the particle volume fraction and ν_m is the Poisson ratio of the matrix, which is assumed to be 0.5 in the case the gel behaves as an incompressible material in the linear viscoelastic region (van Vliet, 1988).

The Kerner model (Eq. (1)) does not consider the crowding effect of filler particles (particle interactions) at high volume fractions. This effect can be understood by the principle that the crowded/aggregated fillers behave as particles with a higher effective volume fraction than the single fillers, as water or gelled matrix material is entrapped between the fillers in the aggregate. To model this effect of crowding at high volume fractions, Lewis and Nielsen extended the Kerner model (Eq. (1)) by substituting the actual volume fraction of fillers (ϕ_f) with the effective volume fraction of fillers as ($\psi\phi_f$) (Lewis & Nielsen, 1970):

$$\frac{G'_{gel}}{G'_{matrix}} = \frac{15(1 - \nu_m)(M - 1)\psi\phi_f}{(8 - 10\nu_m)M + 7 - 5\nu_m - (8 - 10\nu_m)(M - 1)\psi\phi_f} + 1 \quad (2)$$

The function $\psi\phi_f$ takes into account the crowding effect of fillers, and in its simplest form can be given as (Lewis & Nielsen, 1970):

$$\psi\phi_f = \left[1 + \left(\frac{1 - \phi_{max}}{\phi_{max}^2} \right) \phi_f \right] \phi_f \quad (3)$$

where ϕ_{max} is the maximum volume fraction of the filler particles. The value of ϕ_{max} varies with the packing geometry of the filler particles. Theoretically, it is either 0.64 for a random close packing or 0.74 for a hexagonal close packing of uniform spheres (Scott & Kilgour, 1969). Originally, these models were proposed using the shear modulus of composite systems, but alternatively, for incompressible matrices ($\nu_m = 0.5$), also the Young's modulus (stiffness) of the gels can be used (Oliver et al., 2015).

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