Soft food microrheology

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Abstract

Soft food materials, such as gels, pastes, emulsions and colloidal suspensions, are highly heterogeneous structures under constant dynamic interactions. For this reason, these materials frequently exhibit complicated rheological responses, many of which defy characterization by conventional techniques without disrupting their internal food microstructure. In this paper, we highlight recent advances in microrheological methods to probe rheological responses of delicate samples at the microscale. They offer a powerful new approach to characterize soft food materials in a direct and unobstrusive way. Microrheological methods promise to reveal a better fundamental understanding of the underlying structures and dynamics of soft food materials, inform novel constitutive models, and help develop new technological applications.

Keywords: food, rheology, microrheology

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1. INTRODUCTION

Food rheology is the study of how food materials deform in response to mechanical stresses. Food materials —and especially soft food materials— are typically composed of heterogeneous microstructures and metastable arrangements that lead to complicated responses when subjected to stress (Fischer and Windhab, 2011). Moreover, experimental techniques able to quantify the rheology of food materials at small length scales can provide basic insight into their underlying structure and dynamics, and ultimately into their response during processing, storage and consumption.

Microrheological techniques have been developed to complement and extend conventional bulk rheology by measuring local rheological properties of a material at the micron scale. In these techniques, the rheological information is extracted from the motion of microscopic probe particles embedded within the material. The embedded probe particles are driven by random thermal fluctuations in passive microrheology (**Squires and Mason, 2010), or by an applied external force in active microrheology (Wilson and Poon, 2011). Both active and passive microrheology are increasingly being used to characterize challenging natural and synthetic materials, but these methods have not yet been fully exploited in food rheology. Here, we briefly discuss recent progress in microrheology, and highlight benefits of these technique for studies of soft materials that play a critical role in food technology such as gels, colloids and biopolymers.

2. ACTIVE AND PASSIVE MICRORHEOLOGY

Microrheology describes a wide collection of techniques used to quantify phenomena involved in the storage and dissipation of mechanical energy in a microscopic region of a material. In the last two decades there have been important developments in microrheological techniques, which can be broadly classified as passive or active depending on the forces underlying the measurements.

Passive microrheology quantifies the mechanical properties of a sample by relating the diffusive fluctuations of an embedded probe particle to the shear modulus of the material (Fig. 1). This technique is ideally suited for soft food materials because the weak thermal energy ($\sim \kappa_B T$) required to fluctuate the particles provides a delicate probe of the internal food microstructure. And because there is no need to apply an external force, it only requires relatively simple instrumentation to trace the motion of the particles such as direct visualization (Vadas et al., 1973; Filipe et al., 2010), laser deflection tracking (Mason and Weitz, 1995; Du et al., 2012), diffusing wave spectroscopy (Mason et al., 1997; Oelschlaeger et al., 2013) or dynamic light scattering (Mason et al., 1996).

The formalism to extract rheological information from thermal fluctuations employs the generalized Stokes-Einstein relation. In the simplest case of a purely viscous material, the mean-square displacement of the fluctuating particles $\langle \Delta r^2 \rangle = 6Dt$ depends linearly on time through the diffusion coefficient D . For a probe particle of (hydraulic) radius a , the diffusion coefficient is given by the Stokes-Einstein equation $D = \kappa_B T / (6\pi\eta a)$, which relates the desired sample viscosity η to the measured mean-square displacement

(**Squires and Mason, 2010).

In a seminal work for modern microrheology, Mason and Weitz (1995) demonstrated that this approach could be generalized to characterize soft materials that exhibit viscoelastic behavior using the fluctuation-dissipation theorem (Landau and Lifshitz, 2000; Evans et al., 2016). By employing the fluctuation-dissipation theorem, Mason and Weitz (1995) generalized the Stokes-Einstein equation to relate the measured mean-squared displacement $\langle \Delta r^2 \rangle$ of the particles (Figure 1a) directly to the sample frequency-dependent viscosity $\eta^*(\omega)$, and therefore to the sample complex shear modulus $G^*(\omega) = i\omega\eta^*(\omega) = G'(\omega) + iG''(\omega)$ via integral transforms (Figure 1b). For a detailed discussion of the application of the generalized Stokes-Einstein relation to passive microrheology and conditions that can invalidate this generalization, see **Squires and Mason (2010); DePuit and Squires (2012a,b).

Active microrheology, instead, extract the mechanical properties of the material from the motion of embedded microscopic particles driven by an external force (Wilson and Poon, 2011). Therefore, besides instruments to trace the motion of the particles, active microrheology requires additional equipment to actively drive them within the material. Generally, the probe particles are pulled by a constant force using magnetic tweezers (Habdas et al., 2004) or they are driven at constant speed using optical tweezers (Furst, 2003; Sriram et al., 2010; Meyer et al., 2006). However, unlike passive techniques, active microrheology enable the characterization of the non-linear rheology of materials by using the applied force to perturb their microstructure far away from equilibrium (Zia and Brady, 2013; Gomez-Solano and Bechinger, 2014; *Gomez-Solano and Bechinger, 2015).

Currently, a frontier research area in microrheology involves combining the benefits of both active and passive approaches. In active microrheology, probe particles are driven by external forces, whereas in passive microrheology the particles are driven by Brownian fluctuations. Combined methods involve the motion of an embedded probe that is actively pulled by internal but non-Brownian stresses (e.g. by capillarity). Recently, rigorous theories elucidating the interplay between endogenous capillary forces and the hydrodynamics of embedded microscopic pores and indentations have been developed (McGraw et al., 2011; Rahmani et al., 2014; *Backholm et al., 2014; Lu and Corvalan, 2015; *Lu et al., 2015), and applied to relate the motion of the probe "micropores" (Fig. 2a) to the Newtonian (Fig. 2b, black line), shear-thinning (Fig. 2b, blue line), and viscoelastic rheology of the material (Roth and Dutcher, 2006; McGraw et al., 2011).

3. APPLICATION TO FOODS

Microrheology is particularly attractive for the study of soft materials such as gels, colloids, and biopolymers that play an ubiquitous role in food science and food technology. Because of the small size of the embedded probe particles, it can measure local rheological properties that are necessarily lumped in macroscopic measurements (Schultz and Furst, 2012). And because of the low inertia of the particles, it can extract this information at a higher range of frequencies than allowed by conventional rheology (Willenbacher and Oelschlaeger, 2007).

Consequently, an important research area open to food microrheology concerns heterogeneous materials where the rheology changes from point to

point. Either by successively tracking single probe particles or by simultaneously tracking multiple probe particles at various places in the sample, microrheology can provide insight into the spatial distribution of the internal microstructure for heterogeneous food systems — from dilute suspensions to arrested systems such as gels and colloidal glasses.

Early works in this area include suspensions of wheat gliadin (Xu et al., 2002), a protein responsible for dough viscoelasticity, and suspensions of oat beta-glucan, a bioactive component credited with health benefits (Xu et al., 2007). More recently, studies on suspensions of barley beta-glucan corroborated that microrheology can probe micro-heterogeneities in food samples that go unnoticed by macroscopic rheology (Moschakis et al., 2012).

Since the pioneering work by Mason and Weitz (1995) on hard-sphere colloidal suspensions, the microrheology of colloidal systems has been extensively studied in both the dilute limit and the arrested limit close to the glass transition. In the dilute limit, colloidal suspensions have been studied using microrheological models based on the two-body solution of the Smoluchowski equation as an approximation to the many-body interactions between colloidal particles (Zia and Brady, 2010, 2013). In this limit, microrheology enabled accurate characterization of both the loss modulus G'' , which generally dominates the dynamics, and the growth of the storage modulus G' with the volume fraction (**Puertas and Voigtmann, 2014).

Increasing the volume fraction of the colloidal particles may lead to dynamical arrest, and the suspensions may undergo a glass transition (**Puertas and Voigtmann, 2014). Close to their glass transition, dense colloidal suspensions have been studied using active and passive microrheological models

based on the Mode-Coupling theory because many-body hydrodynamic interactions generally dominate the mechanical response (Gazuz et al., 2009; Gazuz and Fuchs, 2013; Voigtmann and Fuchs, 2013). In these conditions, colloidal suspensions frequently exhibit rheological behavior similar to other soft glassy materials of interest in food technology because of their shared metastable internal disorder. Measurements largely agree on a predominantly elastic behavior with a storage modulus $G'(\omega)$ scaling as a very weak power law of frequency ω , and on the presence of a yield stress above which the material readily flows as a Herschel-Bulkley fluid (Anderson et al., 2013). For a review of recent work on the microrheology of colloidal systems, see **Puertas and Voigtmann (2014).

Microrheology enable not only simultaneous rheological measurements at different locations in a sample but also continuous measurements over time (Larsen and Furst, 2008). This makes this technique highly valuable for the study of food materials with evolving dynamic microstructures. Of particular importance for food technology is the use of microrheology to monitor the dynamics of aging (Rich et al., 2011; Gomez-Solano et al., 2012; Zia et al., 2014; Peng and McKenna, 2016) and the dynamics of gel formation (Narita and Indei, 2016).

Early research in food gels include the gelation dynamics of pectin (Vincent et al., 2007), fibrillar proteins (Corrigan and Donald, 2009), and milk gels (Cucheval et al., 2009). And more recently, the gelation dynamics of beta-lactoglobulin (Balakrishnan et al., 2010), dairy proteins (Moschakis et al., 2010), and beta-glucan solutions (Moschakis et al., 2012). For a recent review of the microrheology of food gels, see Moschakis (2013).

Recent advances have enabled the rapid characterization of fast-forming gels, and further improved the ability of microrheology to characterize gelation reactions without disturbing the dynamics and assembly of the incipient network structure. Research by Gomez-Solano et al. (2013) demonstrated the utility of microrheology to follow fast process associated to the rapid propagation of the gelation front into the sol phase during sol-gel transition, and Forde and collaborators (Shayegan and Forde, 2013; Shayegan et al., 2014) used microrheology to elucidate delicate mechanisms of self-assembly by following temporal changes in the shear modulus of gels of collagen fibrils since the very early phases of fibrillar growth .

4. CONCLUSION

In this paper, we presented a few examples that provide a glimpse of what is now possible using microrheology, and the unique benefits this technique can offer to food scientists beyond those provided by conventional bulk rheology.

Potential applications of microrheology in food technology are by no means limited to these few examples. In addition to the clear advantage of microrheology to characterize minute amounts of sample (e.g. cells, proteins) (Wirtz, 2009; Josephson et al., 2016), other potential applications include the use of microrheology to interrogate the mechanical properties of bacterial biofilms (Rogers et al., 2008; Galy et al., 2012), and the viscoelastic properties of interfaces and emulsions stabilizers (Mendoza et al., 2014; Huang et al., 2015). Microrheology has also been applied to characterize dynamical processes that modulate the release of the active ingredients in materials used

for controlled release applications (Frith et al., 2015). Moreover, leading research areas may soon enable automated microrheology instrumentation for high-throughput screening of food materials (*Shindel and Furst, 2015), as well as simpler and less expensive ways to characterize non-linear food rheology by extending microrheology to different driving modes using micropores and indentations as embedded micro and nanorheological probes (Rahmani et al., 2014; Lu and Corvalan, 2016).

Clearly, food microrheology is still in its infancy and there is much interdisciplinary work to be done to further familiarize food scientists with this technique. But the possibilities for deciphering the mechanical response of food materials at an unprecedented level of detail, and using this information to develop better theoretical constitutive models (Brader, 2010) and new technological applications, are enormous.

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Figures

Figure 1: **Passive microrheology.** Passive microrheology uses the generalized Stokes-Einstein relation (GSER) and the fluctuation-dissipation theorem to relate the Brownian mean-square displacement $\langle \Delta r^2 \rangle$ of a fluctuating probe particle embedded in a soft material as a function of time τ (a) to the storage modulus $G'(\omega)$ (red) and loss modulus $G''(\omega)$ (blue) of the material (b).

Figure 2: **Combined microrheology.** Example of a combined technique that relates the capillary velocity $v\mu/\sigma$ of a embedded contracting micropore used as a microrheological probe (a) to the shear-thinning rheological behavior of a soft material (blue line) in which the shear stress decreases ($n < 1$) with increasing shear rate (b). Here, Δt is the elapsed time, n the flow index of the material, r the radius of the micropore, and H the thickness of the fluid film.