Journal of Food Engineering 391 (2025) 112450

Contents lists available at ScienceDirect

Journal of Food Engineering

journal homepage: www.elsevier.com/locate/jfoodeng

Non-linear rheology of melted cheddar cheese

Jake Song

Department of Mechanical Engineering, Stanford University, CA, 94305, United States

ARTICLE INFO

Keywords:
Food rheology
Melted cheese
Non-linear viscoelasticity
Fractional models
Mutation

ABSTRACT

The rheology of melted cheese is a fundamental parameter in the preparation of cheese for consumer foods, but remains poorly understood. We show that the linear and non-linear viscoelasticity of melted cheddar cheese can be captured by a fractional linear viscoelastic model and a strain-softening damping function, respectively. However, we show that a time-strain separable constitutive equation of the K-BKZ type using these two components fails to capture the dramatic strain stiffening observed in the steady shear response of melted cheddar cheese. We show that this stiffening effect arises due to the rapid thermally-induced phase separation, dehydration, and solidification of melted cheddar cheese, and that incorporating an appropriate mutation function in the K-BKZ equation to account for this effect results in the complete description of the non-linear shear rheology of melted cheddar cheese. We thus elucidate the origins of the solid-like behavior of melted cheese commonly seen under non-linear deformations, and provide broad insight into the modeling of the non-linear rheology of soft matter systems that exhibit temporal mutations in mechanical properties.

1. Introduction

Viscoelasticity is an essential property of melted cheese, and must be precisely tuned for the use of cheese as a food ingredient in a wide variety of contexts. For instance, the use of melted cheese as a topping ingredient in foods such as pizzas and burgers requires a delicate control of elastic (solid-like) and viscous (liquid-like) properties to prevent the cheese from becoming too tough or runny. In addition, the viscoelastic properties of melted cheese must be intimately understood in order to facilitate the reverse engineering of dairy-free alternative cheeses that mimic these properties using plant-based ingredients. However, despite decades of research (Atik and Huppertz, 2023; Lucey et al., 2003), a deep understanding of the rheology of melted cheese has remained limited for several reasons. First, melted cheese – like many other food systems – exhibits complex linear viscoelastic properties which often require modeling approaches beyond the classical Maxwell or Kelvin-Voigt models of viscoelasticity (Faber, Jaishankar and McKinley, 2017a,2017b; Kuo et al., 2000; Song et al., 2023; Wagner et al., 2017). Second, the rheology of melted cheese requires consideration of viscoelasticity in the non-linear regime, which is particularly important for their stretchability while melted. Third, the rheology of melted cheese is fundamentally a transient phenomenon as it occurs at elevated temperatures (40 °C and above (Lucey et al., 2003)), where casein undergoes protein-protein aggregation, fat molecules and water exude from the gel matrix, and the cheese undergoes irreversible hardening (Atik and Huppertz, 2023; Kim et al., 2011; Lucey et al., 2003). Accounting for the interplay of these three factors is challenging, but an essential prerequisite for understanding the non-linear rheology of melted cheese.

Several useful heuristic measures have been developed to rapidly characterize these non-linear rheological properties of melted cheddar cheese in industrial settings, broadly encompassed in terms such as "meltability" and "stretchability". These include the Schreiber test and the Arnott test, which measure the diameter spread and the height decrease of melted cheese at elevated temperatures respectively (Muthukumarappan et al., 1999; Ustunol et al., 1994; Wang and Sun, 2002), and the Fork test, which measures the vertical pull and thus the stretchability of melted cheese using a fork-shaped probe (Fife et al., 2002). More quantitative measures have also been introduced to characterize meltability of cheese, including the use of squeeze-flow rheometers to measure melted cheese rheology under constant stress (Campanella et al., 1987; Kuo et al., 2000; Wang et al., 1998), a rapid visco-analyzer to measure the viscosity of melted cheese (Prow and Metzger, 2005), and a conventional rheometer to measure the complex shear modulus G^* of melted cheese (Ustunol et al., 1994). While these measures capture different aspects of the non-linear rheology of melted cheese and can indeed be useful for fingerprinting purposes, they do not provide deeper insights into the underlying physics governing the various aspects of the rheology of melted cheese.

In parallel to these developments, studies have also examined non-

E-mail address: jakesong@stanford.edu.

https://doi.org/10.1016/j.jfoodeng.2024.112450
Received 1 August 2024; Received in revised form 16 December 2024; Accepted 17 December 2024
Available online 18 December 2024
0260-8774/

linear rheological properties of melted cheese under controlled deformations, for instance under extension (Muliawan and Hatzikiriakos, 2007), and under large-amplitude oscillatory shear (LAOS) deformations (Wang et al., 2001) – a technique which is becoming increasingly common for measuring the non-linear rheology of solid cheeses as well (Dahl et al., 2024; Melito et al., 2013; Piñeiro-Lago et al., 2023). However, how these rheological properties interplay with (or arise as a result of) other fundamental properties such as linear viscoelasticity and thermal properties to give rise to "meltability" remain unexplored.

Therefore, the objective of this work is to provide a detailed characterization of the linear viscoelasticity, the non-linear viscoelasticity, and the thermo-mechanical properties of melted cheddar cheese, and critically examine how these distinct properties are manifested in the complex non-linear rheological response of melted cheddar cheese – thus providing insight into key engineering variables that determine the processability of melted cheese. We show that melted cheddar cheese exhibits a power-law linear viscoelastic response, a strain-softening non-linear viscoelastic response, and rapid thermal solidification after melting, and that all three of these properties are manifested in the ultimate non-linear shear rheological response. We show that the Kaye-Bernstein Kearsley and Zapas (K-BKZ) integral constitutive equation – a time-strain separable equation commonly used to characterize the non-linear response of polymer melts and foods (Jaishankar and McKinley, 2014; Keshavarz et al., 2017; Larson, 1985) – which is modified to include contributions from thermally-induced sample mutations can provide a complete description of the non-linear shear rheology of melted cheddar cheese. Overall, we provide physical insight into key engineering variables that determine the processability of melted cheese (especially those underlying heuristic measures such as "meltability" and "stretchability"), and provide broad insight into the modeling of the non-linear rheology of soft matter systems that exhibit temporal mutations in mechanical properties.

2. Materials and methods

2.1. Sample preparation

All tests were performed on a commercially available cheddar cheese (Sharp Cheddar aged 6 months by Wegmans, purchased in 2021) using a stress-controlled ThermoFisher HAAKE MARS 60 rheometer. All samples were placed on a Peltier system which is heated to $T = 40$ °C, and loaded under a 35 mm stainless steel parallel plate geometry which is slowly brought down to the sample at a rate of 1 mm/min down to a 1.2 mm gap. All samples were trimmed and then sealed with sunflower oil to minimize evaporation of water prior to rheological testing. Samples were equilibrated at target temperatures for 2 min before subsequently starting the experiments listed below.

2.2. Temperature- and time-dependent property measurements

To determine the behavior of cheese as a function of temperature and time, we performed temperature and time sweep experiments. For temperature sweep experiments, we rapidly cooled the cheese to $T = 20$ °C, equilibrated at this temperature for 2 min, and then characterized the storage modulus G' and loss modulus G'' with an oscillation frequency of $f = 1$ Hz through a temperature ramp from $T = 20$ °C to 90 °C, and then back down to 20^oC, at a rate of 1 °C/min. For measuring the temporal evolution of G' and G'' of cheddar cheese at the melting temperature, we imposed an oscillatory stress of $\sigma_0 \geq 10$ Pa at $T = 55$ °C over 5000 s.

2.3. Linear viscoelastic property measurements

To measure linear viscoelastic properties, we performed oscillatory shear and creep compliance measurements. For oscillatory shear, we imposed an oscillatory stress of $\sigma_0 = 10$ Pa on the melted cheddar cheese at $T = 55$ °C over a range of frequency f of 0.03 Hz $\leq f \leq$ 20 Hz. For creep compliance measurements, we imposed a step stress of $\sigma_{step} = 10$ Pa $T = 55$ °C.

2.4. Non-linear viscoelastic property measurements

To measure non-linear viscoelastic properties, we performed step strain and steady shear experiments. For step strain experiments, we imposed incremental amounts of step strain γ_0 at $T = 55$ °C, measuring the relaxation response at each γ_0 for 100 s. For stress amplitude experiments, we imposed increasing oscillatory stress σ_0 on melted cheddar cheese at a frequency of 1 Hz and temperature of $T = 55$ °C. For steady shear experiments, we imposed a constant strain rate $\dot{\gamma}_0$ on the melted cheddar cheese at $T = 55$ °C over 100,000 s.

2.5. Model fitting

All fitting of our data to the various models in the paper were done by minimizing the weighted residual sum of squares which is rescaled to the magnitude of the data point:

$$RSS_{wi} = \sum_{i=1}^{n} \left(\frac{y_i - f(x_i)}{y_i} \right)^2 \quad (1)$$

which computes the sum of the difference between data y and model $f(x)$, which is then scaled by the magnitude of the data y.

3. Results

3.1. Linear thermoviscoelasticity of cheddar cheese

We first studied the thermoviscoelastic properties of cheddar cheese through a temperature ramp experiment (Fig. 1A). We find that cheddar cheese begins to undergo melting (indicated by the crossover in G' and G'') at ~ 55 °C, and that irreversible stiffening begins to occur at higher temperatures, as evidenced by the re-entrant increase in G' at $T \sim 75$ °C and the higher G' and G'' measured in the cooling curve. We expect this melting behavior to be heating rate dependent as the phase separation of proteins which occurs at elevated temperatures – consisting of protein denaturation, protein-protein aggregation driven by hydrophobic interactions, water and fat expulsion, and consequent irreversible hardening of cheese (Atik and Huppertz, 2023; Kim et al., 2011; Lucey et al., 2003) – is both time and temperature dependent. Indeed, when monitoring G' and G'' as a function of time at $T = 55$ °C, we find that G' and G''

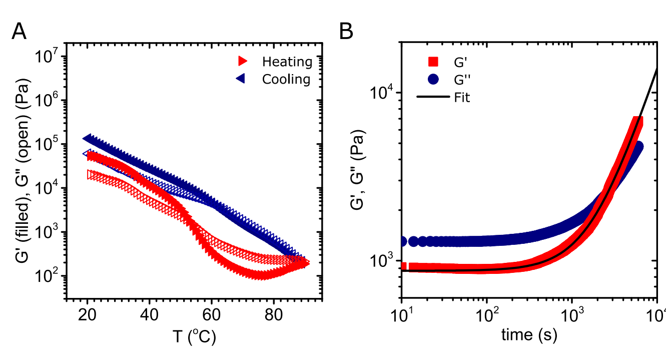

Fig. 1. Temperature- and time-dependent linear viscoelasticity of melted cheddar cheese. A) Storage modulus G' and loss modulus G'' of cheddar cheese measured through a temperature ramp experiment from $T = 20$ °C to 90 °C, and then back down to 20 °C, at a ramp rate of 1 °C/min. The G' and G'' in the cooling curve are higher than those of the melting curve, indicative of thermally-induced solidification in melted cheddar cheese. B) Time-dependent measurement of G' and G'' at $T = 55$ °C, confirming the thermally-induced solidification. G' is fitted to Eqn. (2).

both exhibit a steep increase (Fig. 1B). The rapid increase in G' can be empirically captured through a power-law mutation function of the form:

$$G' - G'(0) = kt^b \quad (2)$$

yielding parameters of $k = 0.009$ and $b = 1.54$. Here, the term mutation is used broadly to describe the active structural change in the material, such as chemical gelation, phase transitions, and phase separations, which consequently affect the rheology of the material (Mours and Winter, 1994). As these stiffening behaviors are driven by protein aggregation and dehydration of the cheese, we expect the fitting exponents to be sensitive to factors that affect the dehydration process. For instance, while the oil coating of the sample during rheological characterizations attempts to minimize this effect, water pervaporation through an oil layer can still occur at these elevated temperatures, and contribute to this mutation process. In practical food application environments where cheese is melted in open air, we expect the dehydration process to occur more rapidly.

3.2. Linear viscoelasticity of melted cheddar cheese

We next studied the linear viscoelastic properties of cheddar cheese at the melt temperature of $T = 55\ ^\circ$C. Frequency sweep experiments show that melted cheddar cheese acts as a viscoelastic fluid, with a non-trivial dependence of G' and G'' on frequency that deviate from classical viscoelastic models such as the Maxwell model (Fig. 2A). To more quantitatively understand the linear viscoelasticity of melted cheddar cheese, we performed creep measurements under constant stress conditions (Fig. 2B). We find that the linear viscoelasticity of melted cheddar cheese can be described by a single power-law. To model this mechanical response of melted cheddar cheese, we utilize a spring-pot – representing a mechanical element which has a constitutive equation that interpolates between a spring $\left(\sigma = G\left(d^0\gamma/dt\right)\right)$ and a dashpot $\left(\sigma = \eta\left(d^1\gamma/dt\right)\right)$ via the relation $\sigma = \mathbb{V}(d^\alpha\gamma/dt)$ (Jaishankar and McKinley, 2013; Song et al., 2023). This interpolated behavior of the spring-pot is compactly described by two parameters – the quasi-property $\mathbb{V}$, and the power-law exponent α. Solving the constitutive equation of the spring-pot for the case of a step stress yields the creep compliance (Jaishankar and McKinley, 2013):

$$J(t) = \frac{\gamma(t)}{\sigma_{step}} = \left(\frac{1}{\mathbb{V}}\right)\frac{t^\alpha}{\Gamma(1+\alpha)} \quad (3)$$

Fits of the compliance measurement of melted cheddar cheese to Eqn. (3) yields parameter values of $\mathbb{V} = 351.5$ Pa s$^\alpha$ and $\alpha = 0.73$. We thus find that melted cheddar cheese exhibits a single power-law relaxation kernel, which is similar to other food systems such as agarose (Pommella et al., 2020), casein (Keshavarz et al., 2017), and hard cheeses, (Faber, Jaishankar and McKinley, 2017a, 2017b), and also observed in a larger variety of soft materials more generally (Song et al., 2023). This power-law rheological response can be attributed to a broad range of underlying physical phenomena (Song et al., 2023), for instance an underlying fractal microstructure of network-forming proteins (Bantawa et al., 2023; Muthukumar, 1985), or colloidal hydrodynamic interactions (Varga and Swan, 2018). Simultaneous rheo-imaging (Hsiao et al., 2012; Massaro et al., 2020) of the casein protein in melted cheese under step stress would likely yield important insights into the underlying physical process.

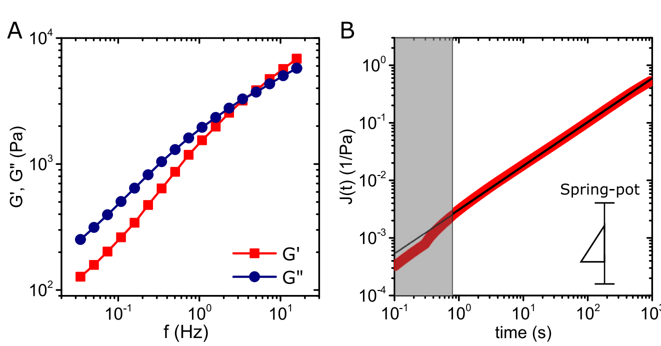

Fig. 2. Linear viscoelasticity of melted cheddar cheese. A) Frequency-dependence of G' and G'' in melted cheddar cheese at $T = 55\ ^\circ$C. B) Creep compliance of melted cheddar cheese at $T = 55\ ^\circ$C. Solid line indicates fit to the constitutive relation of a spring-pot, a fractional viscoelastic element (Eqn. (3)). Data taken below ~0.8 s (greyed area) are measured prior to the equilibration of step stress σ_{step} and are beyond the resolution of the instrument.

3.3. Non-linear viscoelasticity of melted cheddar cheese

We next performed a series of step strain tests to determine the evolution of the relaxation modulus as a function of increasing step strain (Fig. 3A). At small strain amplitudes, the relaxation response is fitted to the spring-pot constitutive relation (Jaishankar and McKinley, 2013):

$$G(t) = \frac{\sigma(t)}{\gamma_0} = \mathbb{V}\frac{t^{-\alpha}}{\Gamma(1-\alpha)} \quad (4)$$

We find that the parameters obtained from the creep compliance measurements in Fig. 2B yields an exact fit to the $G(t)$ response at small strains, i.e. $\gamma_0 < 1$. We find that the relaxation modulus exhibits the same power-law with time, but at a lower magnitude, indicative of a softening

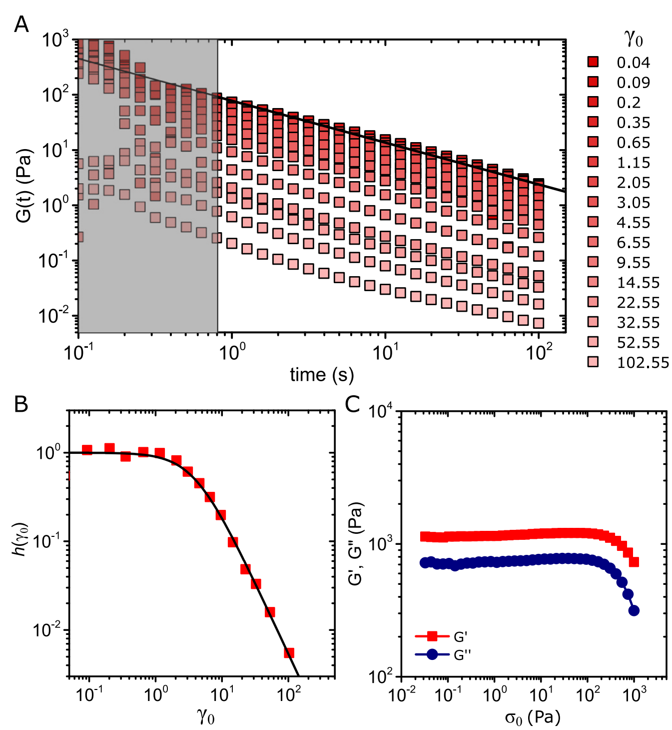

Fig. 3. Strain-dependent mechanics of melted cheddar cheese. A) Relaxation modulus $G(t)$ of melted cheddar cheese measured with increasing step strain γ_0 at $T = 55\ ^\circ$C. Solid line indicates fit to the constitutive relation of a spring-pot (Eqn. (4)). Data taken below ~0.8 s (greyed area) are measured prior to the equilibration of γ_0 and are beyond the resolution of the instrument. B) Strain-dependent damping function of melted cheddar cheese, obtained by normalizing the γ_0-dependent $G(t)$ between 1 and 100 s to the mean $G(t)$ value at $\gamma_0 < 1$ between 1 and 100 s, i.e. $h(\gamma_0) = \langle G(\gamma_0, t)/\langle G(\gamma_0 < 1, t)\rangle\rangle_{1<t<100}$. The data are fitted to a damping function (Eqn. (6)). C) Oscillatory stress amplitude dependence of G' and G'' in melted cheddar cheese, measured at $T = 55\ ^\circ$C.

effect due to accumulated internal damage in the material (Rolón-Garrido and Wagner, 2009). This softening effect can be quantified by a damping function $h(\gamma_0)$, which we define by normalizing the γ_0-dependent $G(t)$ between 1 and 100 s to the mean $G(t)$ value at $\gamma_0 < 1$ between 1 and 100 s, i.e.:

$$h(\gamma_0) = \frac{G(\gamma_0, t)}{G(0, t)} = \left\langle \frac{G(\gamma_0, t)}{\langle G(\gamma_0 < 1, t)\rangle} \right\rangle_{1<t<100} \tag{5}$$

The damping function for melted cheddar cheese is monotonically decreasing (Fig. 3B), and can be fitted to a simple damage model commonly used in polymeric systems of the form (Jaishankar and McKinley, 2014; Rolón-Garrido and Wagner, 2009; Soskey and Winter 1984):

$$h(\gamma_0) = \frac{1}{1 + c\gamma_0^d} \tag{6}$$

yielding parameters of $c = 0.105$ and $d = 1.6$. These parameters are comparable to those observed in polymer melts. (Soskey and Winter 1984) To independently verify that the strain softening response in Fig. 3B is not due to artifacts associated with taking step strain experiments at incremental strain, we also performed a large-amplitude oscillatory shear (LAOS) experiment, which similarly shows a monotonic softening in G' and G'' at large stress amplitudes (Fig. 3C).

Overall, we find that the non-linear viscoelasticity of melted cheddar cheese can be described by a strain-softening damping function. That melted cheese exhibits a purely strain softening response is somewhat surprising, but the same finding was observed in a study on Mozzarella cheese (Hatzikiriakos and Vlassopoulos, 1996), in which a purely strain softening response was observed under extensional rheology. These results suggest that additional mechanisms may be important in the solid-like response of melted cheese under non-linear deformations.

3.4. Integral constitutive equation for the non-linear viscoelasticity of melted cheddar cheese

We now attempt to combine the (time-dependent) linear viscoelasticity and (strain-dependent) damping function of melted cheddar cheese to fully quantify the non-linear rheology of melted cheddar cheese. Prior works on polymers (Larson, 1985) and food systems (Jaishankar and McKinley, 2014; Keshavarz et al., 2017) have shown that non-linear rheological responses can be described by using a K-BKZ time-strain separable integral equation of the form (Keshavarz et al., 2017):

$$\sigma(t) = \int_{-\infty}^{t} G(t - t')h(\gamma)\dot{\gamma}(t')dt' \tag{7}$$

where $G(t - t')$ is the relaxation modulus at an arbitrary time interval of $t - t'$, $h(\gamma)$ is the damping function, and $\dot{\gamma}(t')$ is the strain rate. Thus, with the knowledge of the linear viscoelasticity (Eqn. (4)), and the damping function (Eqn. (6)), one can solve for the K-BKZ integral to predict the non-linear shear stress behavior of a material at a defined shear strain or strain rate.

We first test this hypothesis by performing steady shear rate experiments on the melted cheddar cheese at $\dot{\gamma}_0 = 0.001$ 1/s at a temperature of $T = 55$ °C (Fig. 4A). We find that the steady shear response can be characterized by an initial power-law regime, followed by a steep increase in stress at ~1000 s. We attempt to model this response with the K-BKZ relation in Eqn. (7), substituting our existing information on linear viscoelasticity (LVE) (Eqn. (4)) and non-linear damping function (Eqn. (6)), to give:

$$\sigma(t) = \int_{-\infty}^{t} \frac{\mathbb{V}t^{-\alpha}}{\Gamma(1-\alpha)} \frac{1}{1 + c(\dot{\gamma}_0 t')^d} \dot{\gamma}_0 dt' \tag{8}$$

Using previously obtained parameters of $\mathbb{V} = 351.5$ Pa s$^\alpha$, $\alpha = 0.73$, $c = 0.105$, $d = 1.6$ and performing numerical integrations of Eqn. (8), we find that the LVE portion of the integral equation allows the accurate prediction of the initial power-law increase in shear stress, but the addition of the damping function results in a strain-softening prediction which deviates substantially from the stiffening response observed in the experimental data (Fig. 4A). It is evident that an additional contribution must be considered to fully model the non-linear rheology of cheddar cheese.

We hypothesized that the stiffening response observed in Fig. 4A might arise from the solidification of cheddar cheese arising due to thermal aggregation. To test this hypothesis, we included the mutation function in Eqn. (2) which is obtained from the time-dependence of G' measured in Fig. 1B, yielding a modified K-BKZ type integral constitutive equation of the form:

$$\sigma(t) = \int_{-\infty}^{t} \frac{\mathbb{V}t^{-\alpha}}{\Gamma(1-\alpha)} \frac{1}{1 + c(\dot{\gamma}_0 t')^d} (1 + kt'^b) \dot{\gamma}_0 dt' \tag{9}$$

Though the above relation is phenomenological in nature, using previously obtained parameters of $k = 0.009$ and $b = 1.54$, we find that

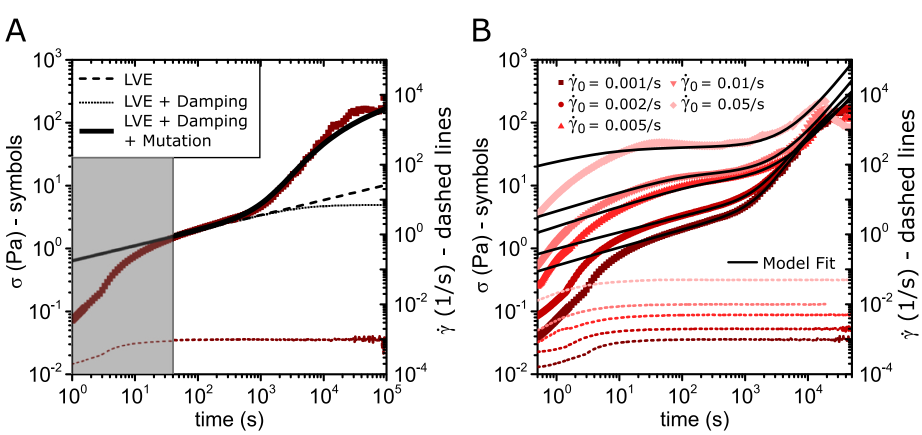

Fig. 4. Steady-shear-rate response of melted cheddar cheese at $T = 55$ °C. A) Typical response of melted cheddar cheese at an imposed strain rate of $\dot{\gamma}_0 = 0.001$ $1/s$ (solid symbols). The rheometer typically takes longer to equilibrate to the strain rate (dashed line, right axis). Data taken before this equilibration are beyond the resolution of the instrument (greyed area). The black lines are *parameter-free fits* of the integral constitutive equations using results from linear viscoelasticity (LVE), LVE and the damping function, and LVE, the damping function, and the mutation function (Eqn. (9)). B) Steady-shear response of melted cheese at different strain rates. Data which are measured after the equilibration of the strain rate are fitted to the integral constitutive equation in Eqn. (9), now with parameters k, b, c and d treated as fitting parameters.

the experimentally measured steady shear response is in good agreement with a *parameter-free fit* of Eqn. (9) obtained via numerical integration, by considering the linear viscoelastic response, the damping function, and the mutation arising from thermal aggregation (Fig. 4A). These results show that the stiffening effect arising at long times may be attributed to the thermally-induced solidification of the melted cheddar cheese (Fig. 1B). This is furthermore apparent in steady shear measurements performed at different shear rates – we find that stress increases at roughly the same time for all shear rates considered ($\dot{\gamma}_0 = 0.001$ 1/s to $\dot{\gamma}_0 = 0.05$ 1/s), indicating that the stiffening effect is a function of time, rather than strain (Fig. 4B). While the damping function was not relevant for describing the total stress response of melted cheddar cheese at the slowest strain rate considered of $\dot{\gamma}_0 = 0.001$ 1/ s, the damping effect becomes more important at the fastest strain rate considered of $\dot{\gamma}_0 = 0.05$ 1/s, as seen in Fig. 4B.

Given that the purely strain-dependent mechanical response of melted cheddar cheese is monotonically strain softening (Fig. 3B), these results show that the "stretchable" and solid-like non-linear rheological responses of melted cheese observed under continuous deformations may be attributed to the solidification of cheese (Fig. 4), which is triggered by thermal input and dependent on time and temperature (Fig. 1). We note that, in practical food application contexts, the stretching of melted cheese also involves the rapid cooling of cheese in air (case hardening (Lucey et al., 2003)), as well as rapid dehydration effects, both resulting in a more rapid increase in stiffness (Fig. 1A). It may be possible to modify the protocols of this study to incorporate a thermal quenching protocol during steady shear to better mimic the behavior of melted cheese in such industrial applications.

More broadly, these results imply that mutation effects *must be explicitly considered* in modeling the non-linear rheology of soft materials. These effects are often not considered when modeling the non-linear steady shear rheology of soft materials, as these studies are done either in equilibrated systems (Jaishankar and McKinley, 2014; Keshavarz et al., 2017; Larson, 1985; Ng and McKinley, 2008) or in "soft glassy" systems in which steady shear experiments are considered to halt aging effects and rejuvenate the sample (Fielding, 2020). However, we can expect these mutation effects to be significant in materials systems that are undergoingactive structural changes, as shown here with melted cheddar cheese which undergoes a dramatic phase separation involving protein-protein aggregation and solvent (fat and water) expulsion at elevated temperatures *en route* to solidification. Based on these results, we expect these phenomena to apply to a wide range of food systems that exhibit thermally-induced mutations, such as albumin (protein denaturation) and amylopectin (starch gelatinization), as well as other soft matter systems exhibiting thermally-induced mutations.

3.5. Time-strain inseparability in melted cheddar cheese

Considering the slight deviation between the parameter-free K-BKZ fits to the data in Fig. 4A, we next fitted the rate-dependent stress responses in Fig. 4B with Eqn. (9), using four fitting parameters $k, b, c,$ and d. The first two parameters describe the onset of mutation effects arising from thermally-induced solidification, and the second two parameters describe the onset of damping arising from non-linear strains. Overall we find that our modified integral equation provides an excellent description of the non-linear rheology of melted cheddar cheese. The fitted parameters are shown in Fig. 5A, and suggest two main trends: a later and steeper onset of thermally-induced solidification with increasing strain rate (indicated by a decrease in k and increase in b respectively), and an earlier and softer onset of strain-induced damping with increasing strain rate (indicated by an increase in c and decrease in d respectively). We note that the k and b values deviate from this behavior at the fastest strain rate considered of $\dot{\gamma}_0 = 0.05$ 1/s, but note that the strain rate equilibration in the instrument is quite poor at these regimes, which results in a poor fit to the data (Fig. 4B). Overall, these results show that time-temperature and strain are *inseparable,* and cannot be straightforwardly separated in describing the non-linear rheology of melted cheddar cheese. Investigating the exact interrelation between solidification and damping – and furthermore, how these relationships depend on temperature – would be an important topic for future studies to better understand the non-linear rheology of melted cheddar cheese.

Nonetheless, the inseparability of time-temperature and strain observed in Fig. 5A appear to be consistent with the underlying physics of the non-linear rheology of melted cheddar cheese. The later onset of thermally-induced solidification with increasing strain rate can be rationalized by the disruption in the buildup of the microstructure of the material from shear deformations, a phenomenon commonly seen in other gelling food systems (Nelson et al., 2022; Omari et al., 2003; Ronsin et al., 2011). In fact, this can be observed directly in our system: time-dependent oscillatory shear experiments with increasing oscillatory stress σ_0 indeed show that thermally-induced stiffening occurs at later time points (Fig. 5B). The earlier onset of damping effects with increased strain rate can also be rationalized by the fact that slower deformations allow the buildup of the microstructure of the material, which can further resist non-linear deformations and delay the onset of damping effects.

4. Discussion and conclusion

This work provides a broad exploration of the non-linear rheology of

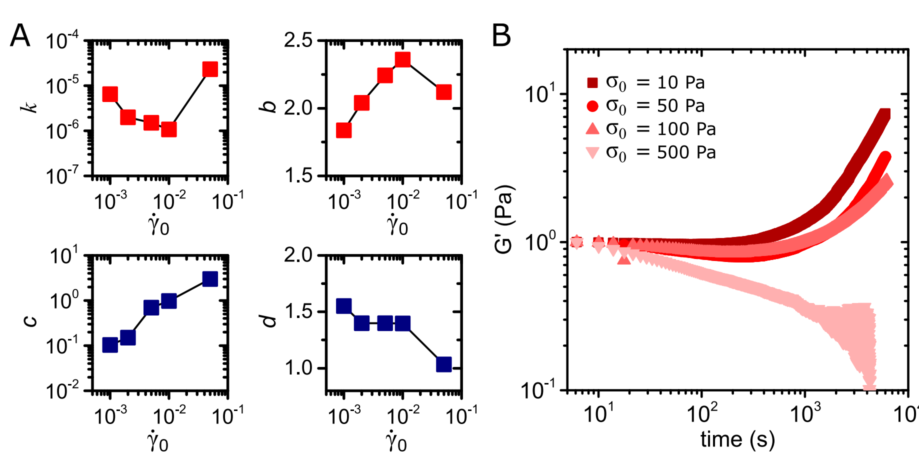

Fig. 5. Time-strain inseparability in melted cheddar cheese. A) Fitting parameters from Fig. 4B, for time- and temperature-dependent solidification (top row, red symbols) and strain-dependent damping (bottom row, blue symbols), as a function of strain rate. B) Time-dependence of G' at $T = 55$ °C with different magnitudes of oscillatory stress σ_0.

melted cheddar cheese. We find that the linear viscoelasticity of melted cheddar cheese can be described by a fractional relaxation model, and that the non-linear viscoelasticity of melted cheddar cheese can be described by a monotonically decreasing damping function of a form commonly seen in other soft materials. Importantly, however, we show that a K-BKZ type integral constitutive model that consists of the time-dependent fractional linear viscoelasticity and the strain-dependent damping function is not sufficient for capturing the steady shear response of melted cheddar cheese. We find this to be due to the thermally-induced solidification of melted cheese, which arises from hydrophobic protein aggregation, phase separation, and solvent expulsion of the cheese sample. This results in the stiffening of the steady shear response that causes substantial deviations from the K-BKZ predictions based on the measured linear and non-linear viscoelastic properties alone.

These results provide important physical insight to the so-called "stretchability" of melted cheddar cheese, as arising from a thermally-induced solidification process. We find that the complete steady shear response of melted cheddar cheese can be quantitatively described by a modified K-BKZ model which includes additional contributions from the thermally-induced solidification of melted cheddar cheese. These findings elucidate the physical origins of the solid-like behavior of melted cheese commonly seen under non-linear strain, and provide guidance on the modeling of the non-linear rheology of soft matter systems that exhibit temporal mutations in mechanical properties.

Looking ahead to the potential impact of our findings in the context of food engineering, our study provides physical insights into how different materials properties of cheese are manifested in the overall non-linear rheological properties of melted cheese. These insights can serve as a useful guide for precisely engineering the processability of melted cheese – which we have shown to be possible by tuning the time-, strain-, and temperature-dependent viscoelastic properties of cheese – rather than utilizing fingerprinting techniques to rapidly screen for "meltability" of cheese using coarse-grained metrics. Our findings can be particularly useful in the context of the preparation of cheese-based processed foods – for instance, in the use of cheese as a topping in prepared foods such as pizza and burgers – and in the reverse engineering of cheese textures with dairy-free ingredients using alternative proteins and food hydrocolloids.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Jake Song was an employee of Motif FoodWorks Inc. at the time of the study, and received financial support, administrative support, equipment access, and supplies from Motif FoodWorks Inc.

Acknowledgements

J.S. acknowledges helpful discussions with M. Boehm (Motif FoodWorks), G. H. McKinley (Massachusetts Institute of Technology), and J. Du (Stanford University).

Data availability

Data will be made available on request.

References

Atik, D.S., Huppertz, T., 2023. Melting of natural cheese: a review. Int. Dairy J., 105648
Bantawa, M., Keshavarz, B., Geri, M., Bouzid, M., Divoux, T., McKinley, G.H., Del Gado, E., 2023. The hidden hierarchical nature of soft particulate gels. Nat. Phys. 19 (8), 1178–1184.
Campanella, O., Popplewell, L., Rosenau, J., Peleg, M., 1987. Elongational viscosity measurements of melting American process cheese. J. Food Sci. 52 (5), 1249–1251.
Dahl, J.F., Gregersen, S.B., Andersen, U., Schulz, H.-J., Corredig, M., 2024. Small and large deformation rheology on pizza cheese as an example of application to study anisotropic properties of food soft materials. Food Hydrocolloids 148, 109456.
Faber, T., Jaishankar, A., McKinley, G., 2017a. Describing the firmness, springiness and rubberiness of food gels using fractional calculus. Part I: theoretical framework. Food Hydrocolloids 62, 311–324.
Faber, T., Jaishankar, A., McKinley, G., 2017b. Describing the firmness, springiness and rubberiness of food gels using fractional calculus. Part II: measurements on semi-hard cheese. Food Hydrocolloids 62, 325–339.
Fielding, S.M., 2020. Elastoviscoplastic rheology and aging in a simplified soft glassy constitutive model. J. Rheol. 64 (3), 723–738.
Fife, R.L., McMahon, D.J., Oberg, C.J., 2002. Test for measuring the stretchability of melted cheese. J. Dairy Sci. 85 (12), 3539–3545.
Hatzikiriakos, S.G., Vlassopoulos, D., 1996. Brownian dynamics simulations of shear-thickening in dilute polymer solutions. Rheol. Acta 35 (3), 274–287.
Hsiao, L.C., Newman, R.S., Glotzer, S.C., Solomon, M.J., 2012. Role of isostaticity and load-bearing microstructure in the elasticity of yielded colloidal gels. Proc. Natl. Acad. Sci. USA 109 (40), 16029–16034.
Jaishankar, A., McKinley, G.H., 2013. Power-law rheology in the bulk and at the interface: quasi-properties and fractional constitutive equations. Proceedings of the Royal Society A 469 (2149), 20120284.
Jaishankar, A., McKinley, G.H., 2014. A fractional K-BKZ constitutive formulation for describing the nonlinear rheology of multiscale complex fluids. J. Rheol. 58 (6), 1751–1788.
Keshavarz, B., Divoux, T., Manneville, S., McKinley, G.H., 2017. Nonlinear viscoelasticity and generalized failure criterion for polymer gels. ACS Macro Lett. 6 (7), 663–667.
Kim, S.Y., Lim, S., Gunasekaran, S., 2011. Protein interactions in reduced-fat and full-fat Cheddar cheeses during melting. LWT–Food Sci. Technol. 44 (2), 582–587.
Kuo, M.-I., Wang, Y.-C., Gunasekaran, S., 2000. A viscoelasticity index for cheese meltability evaluation. J. Dairy Sci. 83 (3), 412–417.
Larson, R., 1985. Constitutive relationships for polymeric materials with power-law distributions of relaxation times. Rheol. Acta 24, 327–334.
Lucey, J., Johnson, M., Horne, D., 2003. Invited review: perspectives on the basis of the rheology and texture properties of cheese. J. Dairy Sci. 86 (9), 2725–2743.
Massaro, R., Colombo, G., Van Puyvelde, P., Vermant, J., 2020. Viscoelastic cluster densification in sheared colloidal gels. Soft Matter 16 (10), 2437–2447.
Melito, H., Daubert, C., Foegeding, E., 2013. Relationships between nonlinear viscoelastic behavior and rheological, sensory and oral processing behavior of commercial cheese. J. Texture Stud. 44 (4), 253–288.
Mours, M., Winter, H.H., 1994. Time resolved rheometry. Rheol. Acta 33, 385–397.
Muliawan, E.B., Hatzikiriakos, S.G., 2007. Rheology of mozzarella cheese. Int. Dairy J. 17 (9), 1063–1072.
Muthukumar, M., 1985. Dynamics of polymeric fractals. J. Chem. Phys. 83 (6), 3161–3168.
Muthukumarappan, K., Wang, Y.-C., Gunasekaran, S., 1999. Modified Schreiber test for evaluation of Mozzarella cheese meltability. J. Dairy Sci. 82 (6), 1068–1071.
Nelson, A.Z., Wang, Y., Wang, Y., Margotta, A.S., Sammler, R.L., Izmitli, A., Katz, J.S., Curtis-Fisk, J., Li, Y., Ewoldt, R.H., 2022. Gelation under stress: impact of shear flow on the formation and mechanical properties of methylcellulose hydrogels. Soft Matter 18 (7), 1554–1565.
Ng, T.S., McKinley, G.H., 2008. Power law gels at finite strains: the nonlinear rheology of gluten gels. J. Rheol. 52 (2), 417–449.
Omari, A., Chauveteau, G., Tabary, R., 2003. Gelation of polymer solutions under shear flow. Colloids Surf. A Physicochem. Eng. Asp. 225 (1–3), 37–48.
Piñeiro-Lago, L., Ramlawi, N., Franco, I., Tovar, C.A., Campo-Deaño, L., Ewoldt, R.H., 2023. Large amplitude oscillatory shear stress (LAOStress) analysis for an acid-curd Spanish cheese: afuega'l Pitu atroncau blancu and roxu (PDO). Food Hydrocolloids 142, 108720.
Pommella, A., Cipelletti, L., Ramos, L., 2020. Role of normal stress in the creep dynamics and failure of a biopolymer gel. Phys. Rev. Lett. 125 (26), 268006.
Prow, L., Metzger, L., 2005. Melt analysis of process cheese spread or product using a rapid visco analyzer. J. Dairy Sci. 88 (4), 1277–1287.
Rolón-Garrido, V.H., Wagner, M.H., 2009. The damping function in rheology. Rheol. Acta 48, 245–284.
Ronsin, O., Caroli, C., Baumberger, T., 2011. Microstructuration stages during gelation of gelatin under shear. The European Physical Journal E 34, 1–7.
Song, J., Holten-Andersen, N., McKinley, G.H., 2023. Non-Maxwellian viscoelastic stress relaxations in soft matter. Soft Matter 19 (41), 7885–7906.
Soskey, P.R., Winter, H.H., 1984. Large step shear strain experiments with parallel-disk rotational rheometers. J. Rheol. 28 (5), 625–645.
Ustunol, Z., Kawachi, K., Steffe, J., 1994. Arnott test correlates with dynamic rheological properties for determining Cheddar cheese meltability. J. Food Sci. 59 (5), 970–971.
Varga, Z., Swan, J.W., 2018. Normal modes of weak colloidal gels. Phys. Rev. 97 (1), 012608.
Wagner, C.E., Barbati, A.C., Engmann, J., Burbidge, A.S., McKinley, G.H., 2017. Quantifying the consistency and rheology of liquid foods using fractional calculus. Food Hydrocolloids 69, 242–254.
Wang, H.H., Sun, D.W., 2002. Correlation between cheese meltability determined with a computer vision method and with Arnott and Schreiber tests. J. Food Sci. 67 (2), 745–749.
Wang, Y.-C., Gunasekaran, S., Giacomin, A., 2001. The lodge rubberlike liquid behavior for cheese in large amplitude oscillatory shear. Appl. Rheol. 11 (6), 312–319.
Wang, Y.C., Muthukumarappan, K., Ak, M., Gunasekaran, S., 1998. A device for evaluating melt/flow characteristics of cheeses. J. Texture Stud. 29 (1), 43–55.