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The complex shear modulus of dough over a wide frequency range

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ABSTRACT

It is shown from small strain shear rheometry and low intensity ultrasonic shear wave measurements that power law behaviour describes the frequency dependence of the complex shear modulus of dough made from a strong North American breadmaking wheat flour. This is the first characterization of the linear viscoelastic behaviour over such a wide frequency range (more than eight decades). Standard rheometry was used to determine shear moduli at low frequencies while an inclined incidence wave reflection technique was used to measure the complex shear modulus in the 10^5 Hz region. The ultrasonic data demonstrate that previous descriptions of the constitutive properties of this rheologically complex material do not incorporate a sufficiently broad range of relaxation times to comprehensively model the rheology of dough at all frequencies. Modeling the dough as a power-law gel material permitted its linear viscoelastic response to be described well over the full frequency range.

1. Introduction

Wheat flour doughs are interesting, rheologically complex materials [1–3], and to date it has not been possible to agree upon a constitutive model that comprehensively predicts their mechanical behaviour [4]. A comprehensive understanding of the constitutive properties of dough is essential because typical dough processing operations cover such a wide range of rates of stress application [5], extending from the very slow deformation induced by outgassing of carbon dioxide into bubbles within the dough during proving [6] to the high strain rates imposed during dough mixing [7], during sheeting of the dough [8] and during extrusion operations [9]. Therefore, in investigations of the rheology of dough, it is vital that an extensive range of testing rates is covered [10], otherwise extrapolation from a restricted set of rates to predict mechanical behaviour where rates of stress application are higher or lower may well lead to inaccurate results [11].

An important aspect of the mechanical behaviour of any soft solid is its response to shear solicitations in the linear viscoelastic regime [12,13]. Although there have been numerous evaluations of the shear modulus of dough, most analyses have been performed over a limited frequency range, typically less than five decades [2,14]. Two studies, one in Europe [15] and one in North America [16], have reported shear modulus values at higher rates of testing using ultrasonic techniques. However, two very different results were obtained, with the reported shear modulus being orders of magnitude higher in the North American study [16] where an all-purpose flour (presumably of North American origin) was used and values similar in magnitude to the longitudinal and bulk moduli of dough were found. It has been remarked that the “strength” of the flour may markedly influence the choice of constitutive model deemed appropriate for characterizing the properties of dough [12]. Since breadmaking flours in North America are typically made from grists of stronger wheat varieties, one would expect the resulting doughs to exhibit greater shear stiffness. However, it seems highly implausible that differences in the source of wheat flour can give rise to a 1000-fold difference in the shear modulus of dough at ultrasonic frequencies.

Therefore, the objective of this paper was to perform both small strain shear rheometry and low intensity ultrasonic shear wave measurements on doughs made from the same strong North American breadmaking wheat flour. In this way, the complex shear modulus of dough in the linear viscoelastic regime could be unambiguously characterized over a very wide frequency range.

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2. Materials and methods

A mechanical development process [17] was used to prepare all doughs from a strong breadmaking flour by mixing 100 g of Canadian Western Hard White Spring wheat flour (13.8% protein; 14% moisture) with 2.40 g of salt and 60 ml of distilled water. The dough was allowed to rest in a sealed plastic container for at least 20 min prior to specimen preparation for shear testing, either by ultrasound or by rheometry. The density of the dough (ρ) was independently determined to be $1198 \pm 7\,\mathrm{kg\,m^{-3}}$ from measurements on sub-samples using Archimedes principle [17].

To prepare specimens for rheometry, the whole dough piece was made into a sheet by passing through a dough-sheeting device with successive reductions in gap to obtain a final dough thickness of 3–4 mm. The sheeted dough was allowed to rest for 10 min and a specimen was excised using a circular steel cutter. The specimen was then carefully mounted on the rheometer.

An AR 2000 rheometer was used with 40 mm diameter parallel plates at 20 °C, essentially as described by Phan-Thien and co-workers [2,12,18]. The upper plate was lowered at $50\,\mu\mathrm{m\,s^{-1}}$ to compress the dough to a normal force of 2 N. The exposed dough surface at the specimen perimeter was coated with mineral oil. Doughs were allowed to rest for 45 min in the rheometer prior to testing, a time that sufficed to relax all but the most slowly relaxing normal stresses [2,12]. Frequency sweeps were performed (0.01–100 Hz) at a constant shear stress of 1.0 Pa (determined from preliminary analyses to be well within the linear viscoelastic region).

Due to the very strong attenuation of shear waves in dough, the complex shear modulus at ultrasonic frequencies was obtained using an inclined incidence wave reflection technique [13] that has been used to measure the shear modulus of other highly attenuative materials (e.g., high molecular weight polydimethylsiloxanes). In our experimental set-up (Fig. 1) a shear transducer emitted an ultrasonic pulse centered at 400 kHz that propagated into an acrylic block. The shear wave that was partly reflected at the acrylic–dough interface propagated again through the acrylic, was reflected at the acrylic–air interface and followed the same path back to the transducer, which was detected as a pulse whose complex fast Fourier transform (FFT) $A^*_{dough}(f)$ was determined. Measuring the reflected pulse when there was no dough at the interface provided a reference FFT, denoted $A^*_{ref}(f)$. The ratio $x^* = A^*_{dough}/A^*_{ref}$ is related to the acoustic impedances Z^* and Z_0^* and to the angles θ_i and θ_r by

$$x^* = \left(\frac{Z_0^*\cos(\theta_i) - Z^*\cos(\theta_r)}{Z_0^*\cos(\theta_i) + Z^*\cos(\theta_r)}\right)^2, \tag{1}$$

where $Z^* = \rho\upsilon^*$, ρ, and $\upsilon^* = \left[1/\upsilon_s + i\alpha/\omega\right]^{-1}$ are the (shear) impedance, density and complex velocity of the dough, with υ_s and α being its sound speed and attenuation, $Z_0^* \approx Z_0 = \rho_0\upsilon_0$ (1.62 MRay) is the impedance of acrylic (since the losses in acrylic are negligible at these frequencies, Z_0 can be taken to be real), θ_i, the angle of incidence, is 75°, and θ_r is the angle of refraction $[\sin(\theta_r) = \upsilon_s \sin(\theta_i)/\upsilon_0]$.

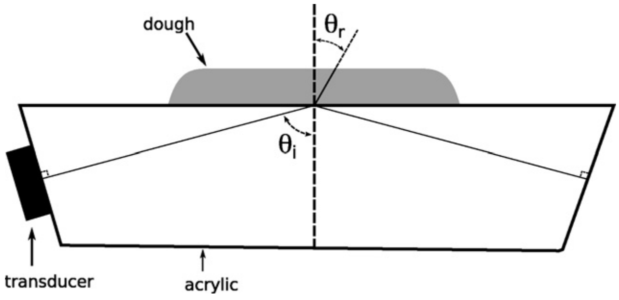

Fig. 1. Experimental set-up for the inclined incidence wave reflection method used to measure shear wave velocity of dough.

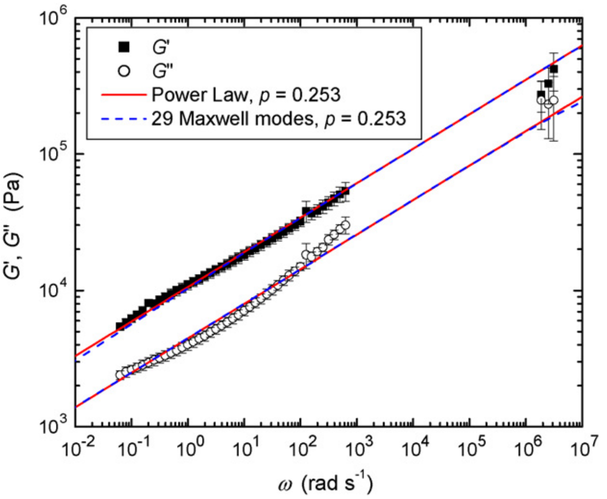

Fig. 2. Real (■) and imaginary (○) parts of complex shear modulus of dough as a function of angular frequency determined by rheometry and ultrasound. Broken blue lines are model descriptions for a discrete Maxwell relaxation spectrum (see text), while solid red lines are power-law relations ($G'(\omega) = 10600\omega^{0.253}$; $G''(\omega) = 4460\omega^{0.253}$ corresponding to $S = 9380\,\mathrm{Pa\,s^{0.253}}$).

From Eq. (1) we can extract Z^*:

$$Z^* = Z_0\frac{\cos(\theta_i)}{\cos(\theta_r)}\left(\frac{1-\sqrt{x^*}}{1+\sqrt{x^*}}\right) \tag{2}$$

Although in Eq. (2) θ_r depends on Z^*, if the shear velocity in the dough resting on the acrylic block is small compared with the shear velocity in acrylic, $\cos(\theta_r) \approx 1$. The complex quantity Z^* obtained with Eq. (2) allows the real and imaginary parts of the shear modulus to be determined:

$$G' = \Re\left(\frac{Z^{*2}}{\rho}\right) \quad \text{and} \quad G'' = \Im\left(\frac{Z^{*2}}{\rho}\right) \tag{3}$$

To prepare specimens for ultrasonic testing, sub-samples of the dough piece were excised with a very sharp pathology blade just prior to measurement (so that the fresh surface ensured good contact between the dough and acrylic block). An important issue in accuracy of these measurements is temperature stability [13]. Sub-samples were equilibrated for 5 min on the block because we observed initial signal variation, probably because of temperature changes, that essentially disappeared over this time. In addition, if the temperature changed between acquisition of the reference and the dough signals, the phase shift induced by a change in shear wave velocity in the acrylic block brought about by the temperature change would interfere with determination of the small phase shift arising from reflection at the dough-acrylic interface. To reduce the effect, we measured the signal with the piece of dough on the acrylic first (i.e., A^*_{dough}), then we removed the dough and measured the reference signal (A^*_{ref}). Results at three frequencies were reported, chosen from the working bandwidth of the transducer: 300, 400 and 500 kHz. Up to 10 different sub-samples were investigated from different mixed doughs.

3. Results and discussion

The real and imaginary parts of the complex shear modulus of doughs made from a strong North American spring wheat flour over eight decades of frequency are shown in Fig. 2. It can be seen that it is possible to extrapolate rheometry data to the ultrasonic range reasonably accurately with power laws. The best-fit lines (not shown on the figure) for the whole range of the data are described by

$$G' = 10900\omega^{0.234\pm0.004} \tag{4}$$

$$G'' = 4300\omega^{0.271\pm0.005} \tag{5}$$

Extrapolation of these fits to the MHz region indicates that G' has a value of the order of hundreds of kPa, a value close to those reported by Létang et al. [15] for MHz measurements of doughs made from a much weaker European wheat flour with only 10% protein. It therefore seems that the shear velocity for all wheat flour doughs prepared with optimal water absorption is of the order of 30 $\mathrm{m\,s^{-1}}$, and certainly not as high as that reported by Lee et al. [16]. Also, since the shear velocity in the dough is approximately 30 $\mathrm{m\,s^{-1}}$, it is indeed very small compared to the shear wave velocity of 1370 $\mathrm{m\,s^{-1}}$ in acrylic, so the approximation that $\cos(\theta_r) = 1$ in Eq. (2) was good.

A common means of modeling the rheological behaviour of complex viscoelastic materials such as dough is to use a number of discrete Maxwell relaxation modes [4,12,19,20] so that:

$$G'(\omega) = \sum_{i=1}^{N} \frac{G_i \tau_i^2 \omega^2}{1+\tau_i^2\omega^2} \tag{6}$$

$$G''(\omega) = \sum_{i=1}^{N} \frac{G_i \tau_i \omega}{1+\tau_i^2\omega^2} \tag{7}$$

As a result of a recent study [20] using dough made from Canadian flour, it was suggested that ten relaxation modes sufficed to adequately model the rheology of this strong wheat flour dough over the complete range of timescales covered by the various experimental protocols. Also recently [4,21], an analysis of a comprehensive set of rheological data from dough made from a medium strength Australian wheat flour indicated that 16 modes were required to accurately model all the time and frequency domain test data. [Australian wheat flour would still be considered “strong” by world wheat strength standards.] We applied both these constitutive models for strong doughs in an attempt to predict our small strain dynamic moduli data over the extended frequency range, but neither provided a satisfactory fit to the experimental data. We therefore used Tanner’s protocol [21] to compute the discrete spectrum for dough tested over 8 decades of frequency to demonstrate that a very broad set of relaxation times is essential if the complex shear modulus of dough is to be adequately modeled by a discrete Maxwell model [22].

Tanner et al.’s protocol is based on the method developed by Baumgärtel and Winter to represent power law behaviour of G' and G'' using a spectrum of discrete Maxwell relaxation modes. This algorithm determines the magnitudes, for a given value of the exponent p, of the different relaxation time contributions, G_i, for relaxation times equally spaced at half-decade intervals. Here, the exponent p is defined in step strain relaxation experiments by $G(t) = St^{-p}$, where S is the magnitude of the stress relaxation, or alternatively, the gel strength [3,23]. As determined from the Fourier transform of $G(t)$, both $G'(\omega)$ and $G''(\omega)$ have the same value of the frequency exponent in this approach. The exponents in Eqs. (4) and (5) are sufficiently close that it is reasonable to represent the data with the average value of the exponent, $p = 0.253$. The fit to the experimental data, shown as the dashed blue lines in Fig. 2, is satisfactory, especially considering that no adjustable parameters were employed (apart from the value of p). However, the fit did require 29 relaxation times, stretching from 10 ns, 31.6 ns, 100 ns, . . ., up to 1,000,000 s. It is clear that extending the rates of testing on dough using ultrasonic techniques necessitates a much larger spectrum of relaxation times than has been reported to date in order that the linear viscoelastic shear modulus is accurately described.

Alternatively, the rheology of dough can be described more succinctly by directly modeling it as a power law material [4,23,24]. Such a model may also have a microstructural legitimacy because a power law response is expected on the basis of the complex molecular structure of the gluten network [3]. In this model, the complex shear moduli are defined as [24]:

$$G'(\omega) = S\Gamma(1-p)\cos\left(\frac{p\pi}{2}\right)\omega^p \tag{8}$$

$$G''(\omega) = S\Gamma(1-p)\sin\left(\frac{p\pi}{2}\right)\omega^p \tag{9}$$

From a best least-squares fit of these equations to our data, the value of S was found to be 9380 $\mathrm{Pa\,s^{0.253}}$, with the fits, shown as the solid red lines in Fig. 2, being the same for angular frequencies between 10^{-1} and 10^6 $\mathrm{rad\,s^{-1}}$ as the Tanner et al model with a 29-mode discrete Maxwell spectrum. Describing the shear modulus of dough using only the two parameters required for Eqs. (8) and (9) has evident economies of description compared to the discrete Maxwell spectrum model of Eqs. (6) and (7).

An extensive oscillatory shear rheometry evaluation of the power law behaviour of doughs made from flours of different dough strength and formulated with various water contents indicated that $0.15 < p < 0.3$ and $10^3 < S/\mathrm{Pa\,s}^p < 10^4$ [23]. In contrast to gluten gels, where additional Rouse modes were required to adequately model high frequency rheometry data [3], these additional modes do not appear to be essential for capturing the high frequency rheometry response of the various doughs [23]. Indeed, with the extension of the shear modulus measurements to ultrasonic frequencies, we find that it is not possible to improve the fit to the data at *both* the upper rheometry frequencies and the ultrasonic frequencies simultaneously by the addition of short time Rouse relaxations. Thus, the ultrasonic data support the idea that dough does exhibit a power law response over a very wide frequency range.

4. Conclusions

Measurements of the shear modulus of strong wheat flour dough are extended to eight decades of frequency using ultrasound. The frequency dependence is well described by a power law fit, with the dough characterized as a soft viscoelastic solid over this wide frequency regime ($G' > G''$, $|G| \sim 400$ kPa at 2×10^6 $\mathrm{rad\,s^{-1}}$). Extension of shear modulus data up to the MHz regime shows no change in the overall functional form of the frequency dependence of the complex modulus compared with that of dough at low frequencies. This implies that the shear modulus of dough must be modeled with a broad range of relaxation times and that a progression of short timescale relaxations contributes significantly to the rheology of dough, behaviour that can be aptly described by a power-law relaxation model.

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List of symbols

A^*	complex fast Fourier transform of ultrasonic pulse
G^*, G', G''	complex, real and imaginary shear moduli
$G(t)$	stress relaxation modulus
G_i	relaxation shear moduli of Maxwell discrete spectrum
N	number of modes in Maxwell discrete spectrum

p	power law exponent parameter
S	power law strength parameter
t	time
x^*	ratio of the fast Fourier transforms of reflected ultrasonic pulses through acrylic with an acrylic/dough interface to that for reference pulses with an acrylic/air interface
v^*	complex shear wave velocity
υ_0	shear wave phase velocity in acrylic
υ_s	shear wave phase velocity in dough
Z^*	acoustic shear impedance of dough
Z_0^*	acoustic shear impedance of acrylic
α	attenuation coefficient of shear waves in dough
θ_i	angle of incidence (Fig. 1)
θ_r	angle of refraction (Fig. 1)
Γ	gamma function
ρ	density of dough
ρ_0	density of acrylic
τ_i	relaxation times of Maxwell discrete spectrum
ω	angular frequency

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