

Stress Relaxation and Oscillatory Tests to Distinguish Between Doughs Prepared from Wheat Flours of Different Varietal Origin

Mohsen Safari-Ardi and Nhan Phan-Thien^{1,2}

ABSTRACT

Cereal Chem. 75(1):80–84

The relaxation properties of flour-water-salt doughs prepared from four different flour types (weak, medium, strong, and extra strong) at different water absorption levels from 58 to 66% with protein contents of 10.0, 10.9, 13.2, and 11.8%, respectively, were studied by imposing varying strain amplitudes of 0.1–29%. Oscillatory tests in the linear viscoelastic region of the 66% absorption strong and weak dough cannot distinguish between the two types of dough. The inability to differentiate between dough types also applied to oscillatory tests on 58% absorption weak and 66% absorption strong doughs. However, the relaxation modulus of dough (extending over time) behaved quite distinctively at high strains, where dough samples experience large deformations. At strain amplitudes of $\leq 0.1\%$ (i.e., in the linear viscoelastic region), different dough types behaved similarly. Likewise, the relaxation modulus completely relaxed at sufficiently long times. The magnitude of the modulus at intermediate-

and high-strain amplitudes were in the order: extra strong > strong > medium > weak, indicating a higher level of elasticity in the extra strong dough samples despite its lower protein content. The relaxation times spectrum of the weak flour, extracted from the relaxation modulus data, reveals a broad relaxation process. The stress relaxation data are very reproducible at high-strain amplitudes ($\approx 1\text{--}15\%$ for up to 3×10^3 sec). This work demonstrated, for the first time, the consistency in oscillatory and relaxation measurements for dough. It also clearly showed that linear viscoelastic data, although important in the characterization of time scales in dough, are largely irrelevant in differentiating between dough types. Furthermore, without proper care, a false steady-state behavior can be obtained with standard viscometric measurements due to slippage at the dough-plate interface.

Dynamic properties (Hibberd and Wallace 1966, Hibberd 1970, Hibberd and Parker 1975,) and viscometric data (Janssen 1992, Lindborg 1995) of dough have been measured in several studies. Creep and relaxation tests (Smith and Tschoegl 1970, Bagley and Christianson 1987) and large deformations in lubricated squeeze film tests (Bagley and Christianson 1986) have also been performed. The main aim has been to provide a complete characterization of dough and to find a reliable rheological test that can differentiate different dough types. In the industry, dough performance is usually measured by empirical instruments (farinograph, extensigraph, etc.) from which dimensionless data, such as resistance and extensibility, are obtained. However, due to the complex nature of the dough and the flow fields generated in such devices, it has been difficult to obtain reliable and reproducible data. More importantly, the data obtained by these devices often do not correspond to the commercial bakery performance of doughs. In addition, the interpretation of the data also poses a problem due to the inhomogeneous nature of the flow fields. Reviews of different aspects of these measurements can be found in Faridi and Faubion (1986, 1990) and Pomeranz (1971).

In previous studies (Phan-Thien et al, *in press*; Safari-Ardi et al 1996,1997), we developed a testing procedure that allows us to obtain consistent and reproducible data within the statistical variations of the sample. In oscillatory shear flows, we found a very low linear viscoelastic strain limit of $O(10^{-3})$, which is in agreement with other studies (Hibberd 1970, Hibberd and Parker 1975, Hibberd and Wallace 1966, Janssen 1992, Lindborg 1995). In shear flow, we found that both shear and normal stresses increase to peak values and then decrease rapidly with time. The values of the stress peaks are strain-rate dependent, but they occur at a strain of $O(10)$. In particular, we found that oscillatory measurements in the linear viscoelastic region on the same dough with four different water addition levels cannot distinguish between them due to the lack of significant changes in the dynamic viscosity and the storage modulus (Phan-Thien and Safari-Ardi, *in press*).

We found that high-amplitude oscillatory measurements can reveal significant changes in the stresses. However, there is no unambiguous way to decompose high-strain oscillatory measurement data into a storage and loss modulus, unless a particular rheological model is adopted. On the other hand, stress relaxation tests at high-strain amplitudes on the same dough with different water additions may be helpful in distinguishing between doughs with different water additions; they are easy to interpret physically. In Safari-Ardi et al (1997), we showed that high-strain stress relaxation measurements on some flour types can be very useful and effective in distinguishing subtle and intrinsic differences between doughs.

In this study, dough samples were characterized by both oscillatory measurements at low strain levels ($\approx 10^{-3}$), and stress relaxation measurements at varying levels of shear strains ranging from ≈ 0.1 to $\approx 29\%$. The aim is to find out whether different dough types can be delineated by large-strain relaxation tests.

MATERIALS AND METHODS

Sample Preparation

Four different flours, extra strong (baker's), strong, medium, and weak were provided by Weston Milling, Sydney, Australia. The flours were studied by preparing 66% water absorption doughs (flour basis), mixed to their respective optimum development times in a 10-g mixograph, with 200 mg of analytical-grade salt added to all mixtures. The farinograph water absorption of the four flours were 66, 64, 62, and 58% for strong, extra strong, medium, and weak flour, respectively. These farinograph absorptions were considered the "standard" cases of the respective doughs (Table I). Thus, strong standard refers to the strong dough mixed to optimal development at 66% water absorption. The 66% absorption (9.76 g of flour + 6.36 g of water + 0.2 g of salt) weak dough with a peak development time of 260 sec was also used in some tests.

TABLE I
Proportions (g) of Ingredients in Four Standard Flour-Water Doughs

| Dough | Peak Development Time | | | |
|--------------|-----------------------|-------|------|-------|
| | Flour | Water | Salt | (sec) |
| Extra strong | 9.76 | 6.24 | 0.2 | 242 |
| Strong | 9.64 | 6.36 | 0.2 | 250 |
| Medium | 9.88 | 6.12 | 0.2 | 270 |
| Weak | 10.13 | 5.87 | 0.2 | 215 |

¹ Quality Wheat CRC Ltd, Locked Bag 1345, North Ryde, NSW 2113 Australia.

² Corresponding author. E-mail: nhan@mech.eng.usyd.edu.au Department of Mechanical & Mechatronic Engineering, Sydney University, NSW 2006 Australia. Fax: +612 9351 7060.

The fully developed dough was then sealed in a plastic bag followed by mounting on a rheometer in the parallel plate configuration. Before starting the measurement, the dough was allowed to rest for 45 min. Loss of moisture was prevented by applying a layer of petroleum jelly to the edge of the sample. Slippage was prevented by using sandpaper (grade 100 cv). At the end of the each measurement, no discernible slip was noted on the dough sample. This was detected visually by the examination of the sandpaper surface while removing the dough from the gap. On the other hand, the maximum angular amplitude of $\pm 2 \times 10^{-2}$ rd given within the interval of 0.01–100% amplitude can hardly cause slippage at the sandpaper-dough interfaces. The tests were performed in an uncontrolled air-conditioned laboratory with a variation of $\pm 1^\circ\text{C}$ in the 24°C ambient temperature; ambient air humidity was not controlled during the measurements. All tests were performed on a rheometer (Bohlin VOR) operated in the parallel-plate configurations using either a 93.286 g·cm or a 2,000 g·cm torsion bar. Two different parallel plates with diameters of 30 and 40 mm were used; gaps ranged from 1 to 2 mm. Gaps <1 mm did not produce reliable data. The amount of dough loaded into the gap was calculated using the gap volume and a dough density of 1.1 g/cm^3 .

The samples were loaded onto the parallel plates of the rheometer and subjected to stress relaxation tests. The sample was subjected to a shear strain given by:

$$\gamma(t) = \begin{cases} 0 & , t < 0, \\ \gamma_0 t / \varepsilon & , 0 \leq t < \varepsilon, \\ \gamma_0 & , \varepsilon \leq t \end{cases} \quad (1)$$

in which γ_0 is the strain amplitude of 0.1–29%, ε is the rise time, set to 0.02 sec, and t is time. The relaxation modulus, defined as the ratio of the shear stress to the strain $\gamma(t)$, is then recorded as a function of time.

We also performed oscillatory tests, where the shear strain is given by:

$$\gamma(t) = \gamma_0 \sin \omega t \quad (2)$$

where γ_0 is the strain amplitude, and ω is the frequency of oscillations. The strain rate in this case is $\dot{\gamma}(t) = \gamma_0 \omega \cos \omega t$. When the strain amplitude is small, of the order 0.1% for doughs and $\approx 10\%$ for most polymer melts and solutions (Walters 1975), the response is linear viscoelastic, and the shear stress can be written as:

$$S(t) = \gamma_0 (G' \sin \omega t + G'' \cos \omega t) \quad (3)$$

where the component in phase with the strain ($\gamma_0 G' \sin \omega t$) represents the elastic response, and the component in phase with

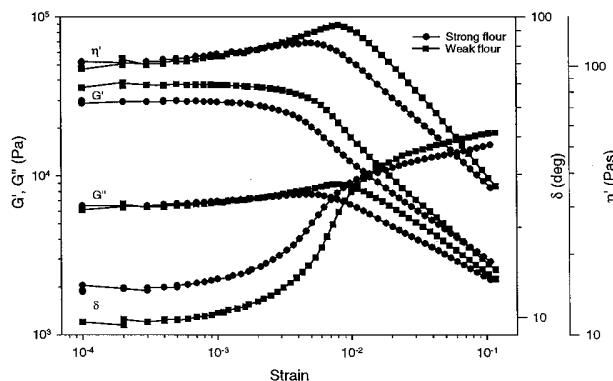


Fig. 1. Strain sweep tests of 66% absorption strong and 58% absorption weak flours. Linear behavior up to strain 0.001 is followed by nonlinear regions. G' = storage modulus; G'' = loss modulus; η' = dynamic viscosity; δ = phase lag.

the strain rate ($\gamma_0 G'' \cos \omega t$) represents the viscous response. The ratio of the elastic stress to the strain is defined as the storage modulus (G'). The ratio of the viscous stress to the strain rate is defined as the dynamic viscosity $\eta' = G''/\omega$; G'' is defined as the loss modulus. Note that the decomposition of the stress (Eq. 3) is only valid at the limit of small strain amplitude. At large strains, the stress is not linear in the strain, and it cannot be decomposed in the manner indicated: there is no validity in providing G' and G'' measurements in the nonlinear regimes. In this work, oscillatory tests were done at a strain of 0.1%, which is well in the linear viscoelastic region of doughs (Hibberd and Wallace 1966, Hibberd 1970, Hibberd and Parker 1975, Janssen 1992, Lindborg 1995).

RESULTS

The linear and nonlinear viscoelastic regions of strong (66% absorption) and weak (58% absorption) doughs, obtained through strain sweep tests at 10 Hz, are shown in Fig. 1. Both doughs exhibit linear response at a strain of $\leq 0.1\%$, and the storage modulus of weak dough is higher than that of the strong dough, contrary to the belief that strong dough with a higher protein content should have higher storage modulus. Figure 2 illustrates the shear stress as a function of time at a shear rate of 0.166 sec^{-1} , with a gap of 2 mm, plate diameter 40 mm, for the extra strong (baker's) dough with 66% absorption for all runs. Two runs were made using sandpaper on the parallel plates, and reproducibility is excellent because the two curves overlap one another for the entire measurement time. In contrast, two other runs made without using sandpaper (the dough was loaded onto the bare surfaces of the plates) reveal slippage at the dough-plate interface as they deviate substantially from the data obtained using sandpaper. One run was made on freshly mixed dough loaded into the gap and rested 45 min, and the other run was made on a dough taken from a sealed bag in which freshly mixed dough had been stored for 45 min. Thus without using sandpaper, the shear stress increases with time to a peak and then decreases until a seemingly steady-state behavior is reached. This steady-state behavior is definitely due to slippage between the dough-plate interface. Slippage must be eliminated if dough properties are to be measured, and this is effectively achieved by gluing sandpaper to the plates (Phan-Thien et al, in press; Safari-Ardi et al 1996,1997).

The storage and loss moduli, G' and G'' and the dynamic viscosity, η' , are shown in Fig. 3 as functions of the frequency taken in the linear viscoelastic region (oscillatory flows at 0.1% strain amplitude). The storage modulus of the weak standard dough is higher than that of the strong standard dough, while the storage modulus of the 66% absorption weak dough overlaps with that of the strong dough. Furthermore, the loss moduli (and hence the dynamic viscosities) of weak and strong flours are nearly identi-

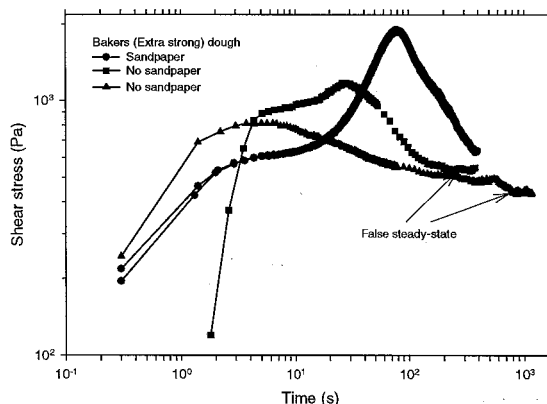


Fig. 2. Apparent shear stress vs. time for 66% absorption baker's (extra strong) dough at a shear rate of 0.166 sec^{-1} . A false steady-state region is seen, due to slippage, when no sandpaper was glued to the parallel plates.

cal. In the low frequency range where the dough is almost at rest or slightly excited, the phase lag, defined as $\delta = \tan^{-1}(G''/G')$, is of the order of 25° , i.e., fairly elastic. However, the phase lag, shown in Fig. 4, decreases with frequency, reducing to $\approx 10^\circ$ at ω 100 rd/s. This is because both moduli increase with the frequency, but G'' increases at a lower rate than G' (in fact, G'' decreases at high values of ω); the dough behaves as an elastic body at high frequencies. The phase lag versus frequency values, especially at low frequencies, are different for the strong standard and the weak standard ($\approx 10\%$), but considering the relatively low reproducibility of the results in G' and G'' ($\approx 15\%$), this is not very significant. As oscillatory measurements of the standard cases were not fruitful in distinguishing between the 66% strong and 58% weak doughs, the 66% weak dough was also tested. As seen in Figs. 3 and 4, the storage modulus and the phase lag of the 58% weak dough are higher than those of the 66% strong dough. On increasing the weak flour water absorption from 58 to 66%, the storage modulus is lowered and overlaps with that of the strong flour. We conclude that, even with the same water absorption basis, dynamic tests cannot differentiate between strong and weak doughs.

The time dependence of the relaxation modulus in the linear viscoelastic region (with a strain amplitude of $\approx 0.1\%$ and a rise time of 0.02 sec) is shown in Fig. 5. The responses of the extra strong and weak doughs almost overlap for the entire measurement time. The same applies to the other two doughs (strong and medium). However, it is widely known that these doughs have different baking properties which, therefore, cannot be detected by stress relaxation tests in the linear viscoelastic region of the doughs. We thus turn to high-amplitude relaxation tests to see whether they can differentiate different types of doughs.

A strain amplitude of $\approx 11\%$ was enough to impart somewhat

high deformation which resulted in a better resolution of the responses. An even better resolution is shown in Fig. 6, where a strain amplitude of $\approx 22\%$ was applied. To check on the reproducibility of the data, the mean (data points) and the standard deviation (error bars) of the relaxation modulus of three runs on the strong dough at 11% strain amplitude are plotted in Fig. 7. The reproducibility is excellent throughout the time range, the standard deviation is $\approx 1\text{--}15\%$ across a time range of 0.1 to 3×10^3 sec; this reproducibility applies to all other doughs tested at high strains.

The corresponding relaxation spectrum of the strong flour at a strain level of 0.1% is shown in Fig. 8, using the regularization method detailed in Honerkamp and Weese (1993) and Weese (1993). Although the relaxation spectrum is displayed for time between 10^{-3} and 10^4 sec, the input data to the inversion process are only from 2×10^{-2} to 1.584×10^3 sec, and therefore the relaxation times spectrum can only be considered reliable in this latter time interval (Weese 1993). The error bars in the figure represent ± 1 standard deviation due to simulated noise in the real data, showing clearly the range of reliability of the spectrum. The relaxation spectrum is typical of a broad molecular weight polymer, with a prominent peak at ≈ 200 sec, suggesting a highly elastic material and a power law terminal zone (Ferry 1953).

To further explore the usefulness of high-amplitude stress relaxation tests, four other flour types, unknown to us during the time of rheological testing, labeled A–D delivered by the same supplier, were tested. The type, trade name, extensigraph water absorption, and extensibility of the four supplied flours are given in Table II.

The subtle differences between these flour types were brought out very clearly with high-amplitude relaxation tests (strain of $\approx 29\%$). The result is shown in Fig. 9, with dough A behaving

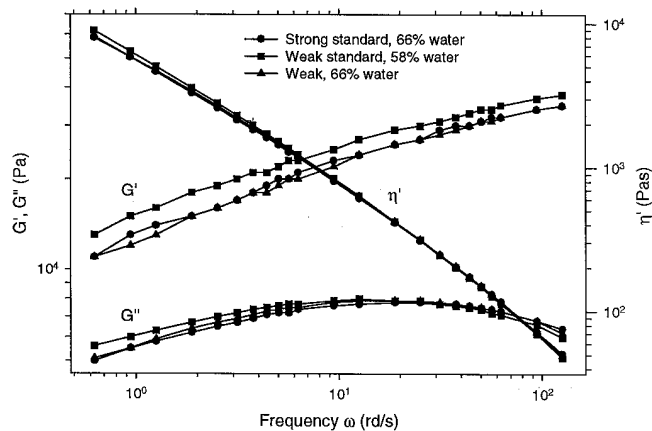


Fig. 3. Storage modulus (G') and dynamic viscosity (η') vs. frequency of doughs in the linear region.

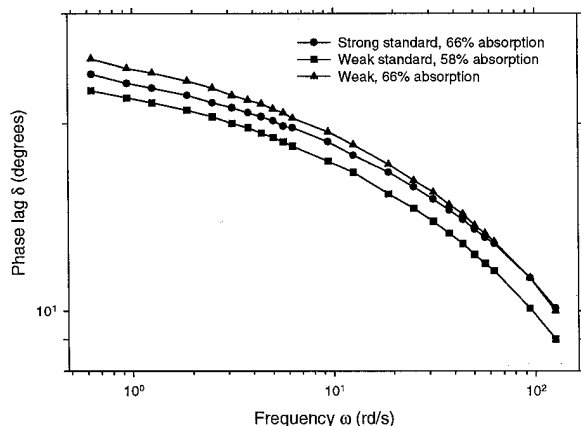


Fig. 4. Phase lag vs. frequency in the linear viscoelastic region.

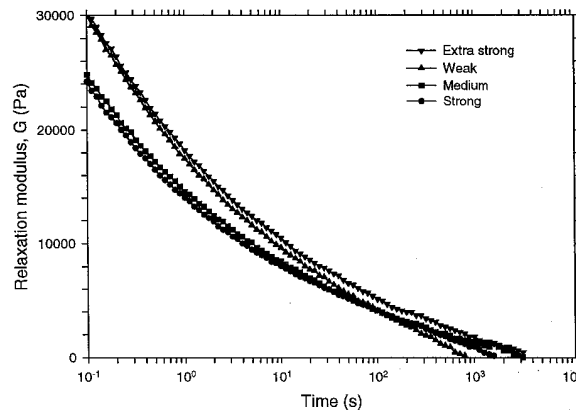


Fig. 5. Relaxation modulus vs. time for the 66% absorption doughs in the linear viscoelastic region (0.1% strain amplitude).

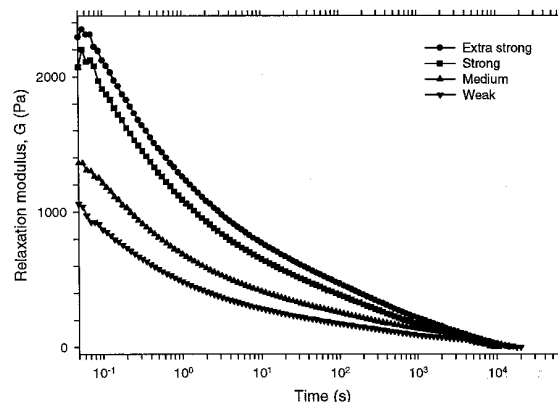


Fig. 6. Relaxation modulus vs. time of 66% absorption doughs at strain amplitude 22%.

most elastically and dough D behaving least elastically. The method, therefore, works well in characterizing and distinguishing different flour types. Test baking results of doughs A–D and the correlations between the elasticity (relaxation modulus) data and the baking performance will be reported in full.

DISCUSSION

We showed that it is quite possible to differentiate between different flour types by subjecting the doughs to high-amplitude relaxation tests. Of course, one would normally start in the linear viscoelastic region to see whether it can result in a differentiation between different types. In addition, the linear viscoelastic results can also be used to generate relaxation spectrum, which characterizes different time scales in dough responses. For the doughs tested (and we suspect for most commercially relevant flour types), oscillatory and stress relaxation measurements in the linear viscoelastic region were not fruitful in delineating different flours. High-shear strain relaxation tests, which impart relatively high deformation to the samples, may prove very useful in this context. We tested this on 66% absorption doughs after experiencing negative results from the oscillatory tests in the linear viscoelastic region of the standard cases. It was thought that different water absorption levels may have clouded the issues, therefore the water absorption level was kept at 66%. However, it was clear that linear viscoelastic properties, aside from supplying information on the time scales of the response of dough, cannot distinguish different doughs at the same level of water absorption.

Another relatively high-strain deformation that may bring out the subtle differences between different flour types is the constant shear rate flow between two parallel plates (Phan-Thien et al 1997) where the strain increases linearly with the time of deformation. There could be doughs that have identical farinograph data, not necessarily identical mixograph peak development times, that end up with different baking performances, which can be characterized by stress relaxation and viscometric tests (e.g., strong and extra strong types). It is well-known in polymer rheology that some different polymer solutions have identical viscometric properties (viscosity and normal stress differences), but their elonga-

tional properties can be quite different (polyethylene, for example). Thus, though we expect, in most cases, a high-amplitude relaxation test will be adequate to differentiate dough types, there may be cases where an elongational test may be necessary. In our laboratory, we tested two cake flours, declared identical by two milling companies based on extensigraph data, that had different baking performance (one of them yielded thicker biscuit). The mixograph peak development times were about the same for these two flours, and the low- and high-strain stress relaxation (rise time 0.02 sec) and viscometric tests (apparent shear stress vs. time plots) could not detect any differences at all. The dilemma was solved by testing the doughs in extension. The dough was suspended between a fixed and a moving grip and was pulled apart exponentially in a universal testing machine modified for the purpose to maintain a constant strain rate in the dough sample. The conclusion was that extensional properties of the doughs were quite different, although they showed identical stress relaxation and viscometric properties. It is noteworthy to mention that by applying a rise time of 5 sec, we could roughly differentiate between the samples. However, further tests are planned to examine the applicability of high rise time stress relaxation tests.

CONCLUSIONS

We showed in this study that stress relaxation tests at high shear strains can differentiate between different flour types, whereas oscillatory and stress relaxation measurements in the linear viscoelastic region of the doughs may not be able to reveal the differences between the doughs.

Oscillatory tests in the linear region are not adequate to differentiate between flour types for most cases, due in part to the poor reproducibility of the data and to a similar elastic response at low strain levels of the doughs.

TABLE II
Properties of Four Commercial Flours

| Flour | Trade Name | Extensigraph | |
|------------|------------|----------------|--------------------|
| | | Absorption (%) | Extensibility (cm) |
| A: Strong | Promax | 64.0 | 22.3 |
| B: Baker's | Extra | 64.5 | 22.8 |
| C: Medium | Eureka | 60.4 | 19.8 |
| D: Weak | Everest | 56.1 | 16.9 |

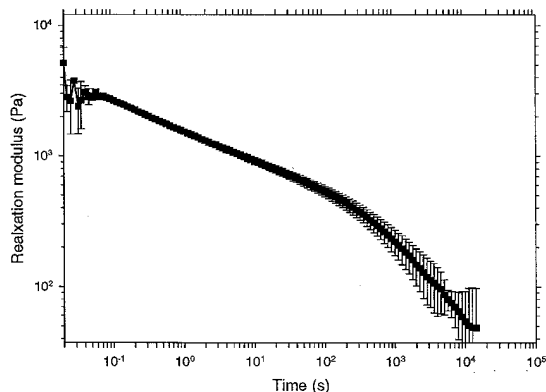


Fig. 7. Relaxation modulus mean and standard deviation vs. time for three measurements of 66% absorption strong flour dough at a strain of 11%.

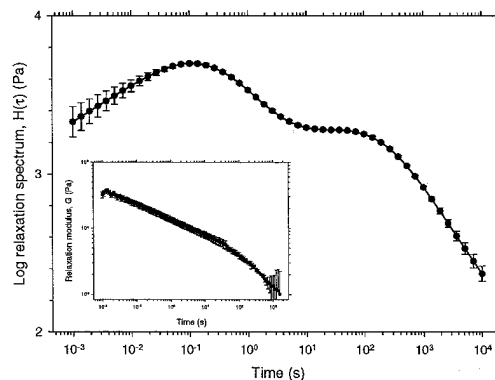


Fig. 8. Relaxation spectrum vs. relaxation times of the 66% absorption strong dough at a strain amplitude of 0.1%. Inset shows relaxation modulus and standard deviations of three measurements.

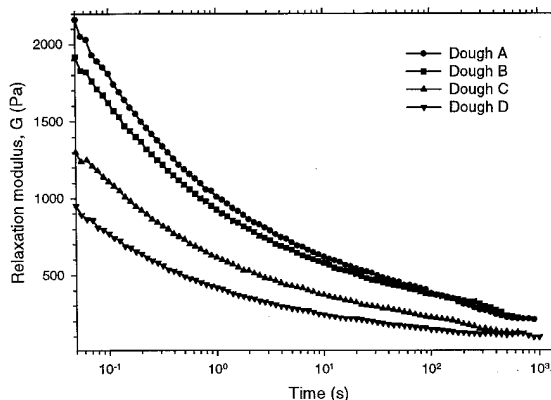


Fig. 9. Relaxation modulus vs. time of four wheat flour doughs (A–D) at 66% absorption level and strain amplitude of 29%.

Similarly, stress relaxation tests in the linear viscoelastic region of the doughs may not be able to differentiate between flour types.

On increasing the applied shear strain in a relaxation test, from low to high values, distinctive behavior between different doughs appears, suggesting that the doughs should be subjected to fairly high deformations. A strain level of the order of 20% should be sufficient in most cases.

Reproducibility of the stress relaxation data are excellent at high strains, provided that some of the systematic errors are eliminated.

The method shown in this study, so far, seems to work well as it was applied on four unknown flours A–D. We conclude that stress relaxation and (and viscometric) tests can be used by the baking industry on a routine basis.

To use the methods on a routine basis, two measurement guidelines were developed: 1) after mixing the dough, it is loaded onto the measuring geometry and rested 45 min before testing, and 2) freshly mixed dough, stored in a sealed bag for 45 min, is loaded and rested 15 min before testing. The latter method reduces the measurement time considerably for several samples that may have come from different batches.

ACKNOWLEDGMENTS

This work is supported by the Wheat Quality CRC Ltd., under project 5.3. We thank George Weston and Defiance Mills for supplying flour samples.

LITERATURE CITED

Bagley, E. B., and Christianson, D. D. 1986. Response of chemically leavened doughs to uniaxial compression. Pages 27-36 in: *Fundamentals of Dough Rheology*. H. Faridi and J. Faubion, eds. Am. Assoc. Cereal Chem.: St Paul, MN.

Bagley, E. B., and Christianson, D. D. 1987. Stress relaxation of chemically leavened dough—Data reduction using the BKZ elastic fluid theory. *J. Rheol.* 31:405-413.

Faridi, H., and Faubion, J. M. 1986. *Fundamentals of Dough Rheology*. Am. Assoc. Cereal Chem.: St Paul, MN.

Faridi, H., and Faubion, J. M. 1990. *Dough Rheology and Baked Product Texture*. Van Nostrand Reinhold: New York.

Ferry, J. D. 1980. *Viscoelastic Properties of Polymers*, 3rd ed. Wiley: New York.

Hibberd, G. E. 1970. Dynamic viscoelastic behaviour of wheat flour doughs. III: The influence of starch granules. *Rheol. Acta* 9:501-505.

Hibberd, G. E., and Parker, N. S. 1975. Measurement of the fundamental rheological properties of wheat flour doughs. *Cereal Chem.* 52:1r-23r.

Hibberd, G. E., and Parker, N. S. 1975. Dynamic viscoelastic behaviour of wheat flour doughs. IV: Non-linear behaviour. *Rheol. Acta* 14:151-157.

Hibberd, G. E., and Wallace, W. J. 1966. Dynamic viscoelastic properties of wheat flour doughs. I: Linear aspects. *Rheol. Acta* 5:193-198.

Honerkamp, J., and Weese, J. 1993. A nonlinear regularization method for the calculation of relaxation spectra. *Rheol. Acta* 32:65-73.

Janssen, A. 1992. *A Study of Factors Determining Bread Making Performance*. PhD thesis. Wageningen Agricultural University: The Netherlands.

Lindborg, K. M. 1995. *Rheological Studies of Wheat Flour Doughs: The Development of Structure During Mixing*. PhD thesis. University of Lund: Sweden.

Phan-Thien, N., Safari-Ardi, M. and Morales-Patino, A. 1997. Oscillatory shear flow of a flour-water dough: A constitutive model. *Rheol. Acta* 36:38-48.

Phan-Thien, N., and Safari-Ardi, M. *In press*. Linear viscoelasticity of flour-water doughs at different water concentrations, *J. Non-Newt. Fluid Mech.*

Pomeranz, Y. 1971. *Wheat Chemistry and Technology*. Am. Assoc. Cereal Chem.: St. Paul, MN.

Safari-Ardi, M., Phan-Thien, N., and Oliver, J. 1997. Stress relaxation measurements of wheat flour doughs at varying shear strains. In: 2nd Pacific Rim Conference, Melbourne, Victoria, Australia, July 27-31.

Safari-Ardi, M., Phan-Thien, N., and Morales-Patino, A. 1996. In: 46th Australian Cereal Chemistry Conference & Gluten Workshop, NSW, Australia, September 1-6.

Smith, T. L., and Tschoegl, N. W. 1970. Rheological properties of wheat flour doughs. *Rheol. Acta* 9:339-344.

Walters, K. 1975. *Rheometry*. Chapman and Hall: London.

Weese, J. 1993. A regularization method for nonlinear ill-posed problem, *Comp. Phys. Comm.* 77:429-440.

[Received April 24, 1997. Accepted September 19, 1997.]