



Short survey

Review: rheology and the breadmaking process

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Abstract

The applications of rheology to the main processes encountered during breadmaking (mixing, sheeting, fermentation and baking) are reviewed. The most commonly used rheological test methods and their relationships to product functionality are reviewed.

It is shown that the most commonly used method for rheological testing of doughs, shear oscillation dynamic rheology, is generally used under deformation conditions inappropriate for breadmaking and shows little relationship with end-use performance. The frequency range used in conventional shear oscillation tests is limited to the plateau region, which is insensitive to changes in the HMW glutenin polymers thought to be responsible for variations in baking quality. The appropriate deformation conditions can be accessed either by long-time creep or relaxation measurements, or by large deformation extensional measurements at low strain rates and elevated temperatures.

Molecular size and structure of the gluten polymers that make up the major structural components of wheat are related to their rheological properties via modern polymer rheology concepts. Interactions between polymer chain entanglements and branching are seen to be the key mechanisms determining the rheology of HMW polymers. Recent work confirms the observation that dynamic shear plateau modulus is essentially independent of variations in MW of glutes amongst wheat varieties of varying baking performance and also that it is not the size of the soluble glutenin polymers, but the secondary structural and rheological properties of the insoluble polymer fraction that are mainly responsible for variations in baking performance. Extensional strain hardening has been shown to be a sensitive indicator of entanglements and long-chain branching in HMW polymers, and is well related to baking performance of bread doughs. The Considere failure criterion for instability in extension of polymers defines a region below which bubble walls become unstable, and predicts that when strain hardening falls below a value of around 1, bubble walls are no longer stable and coalesce rapidly, resulting in loss of gas retention and lower volume and texture. Strain hardening in doughs has been shown to reach this value at increasingly higher temperatures for better breadmaking varieties and is directly related to bubble stability and baking performance.

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1. Introduction—what is rheology?

Rheology is the study of the flow and deformation of materials. Generally, to measure rheological behaviour, a controlled, well-defined deformation or strain is applied to a material over a given time and the resulting force response is measured (or vice versa) to give an indication of material parameters such as stiffness, modulus, viscosity, hardness, strength or toughness of the material. The general aims of rheological measurements are:

- to obtain a quantitative description of the materials' mechanical properties
- to obtain information related to the molecular structure and composition of the material
- to characterise and simulate the material's performance during processing and for quality control

Rheological principles and theory can be used as an aid in process control and design, and as a tool in the simulation and prediction of the material's response to the complex flows and deformation conditions often found in practical processing situations which can be inaccessible to normal rheological measurement.

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113 For example, it is difficult to access dough during mixing,
 114 sheeting, proving and baking without interrupting the
 115 process or disturbing the structure of the material. We
 116 therefore have to predict the range of conditions the dough
 117 experiences during a given process and then extrapolate
 118 from rheological measurements made under simple, well-
 119 defined laboratory conditions, e.g. via mathematical model-
 120 ling using analytical or computational methods (Scott and
 121 Richardson, 1997). For example, mixing and sheeting of
 122 doughs has been simulated (Love et al., 2002; Binding et al.,
 123 2003; Morgenstern et al., 2002) and bubble growth during
 124 proof of bread doughs (Shah et al., 1999) and expanded
 125 cereal extrudes has been modelled through the simulation
 126 and growth of bubbles (Fan et al., 1994).

127 Rheology can be related to product functionality: many
 128 rheological tests have been used to attempt to predict final
 129 product quality such as mixing behaviour, sheeting and
 130 baking performance. This is based on the structural
 131 engineering analysis of materials, where small-scale
 132 laboratory measurements of mechanical properties have
 133 successfully been extrapolated to the behaviour of large
 134 engineered structures such as bridges, buildings, pressure
 135 vessels etc. resulting in the idea that controlled tests on well-
 136 defined small samples of food in the laboratory can be
 137 related to the larger, more complex multi-component
 138 situations found in practical processing conditions.

141 2. Rheological testing

143 Rheological tests attempt to measure the forces required
 144 to produce given controlled deformations, such as squashing
 145 (compression), bending or pulling apart (tension), and to
 146 present them in such a way as to be independent of sample
 147 size, geometry, and mode of testing. They measure a well-
 148 defined property, such as stress, strain, stiffness or viscosity.
 149 A small test piece of the material is usually deformed in a
 150 controlled way, normally on a motor driven machine, and
 151 the force is measured as well as the distance moved or
 152 displacement of the object. The force is then usually plotted
 153 against the displacement to give a force-displacement curve.
 154 Normally for stiff materials we would divide the force and
 155 displacement by the *original* sample dimensions to obtain
 156 stress (force/original cross section area) and Cauchy, or
 157 Engineering strain (displacement /original dimension, e.g.
 158 dL/L_0 , where dL = extension, L_0 = original length),
 159 because the changes in sample dimensions are small and
 160 uniform with displacement; this allows us to remove the
 161 sample size as a variable. Many food materials are not stiff
 162 and undergo large deformations in practice, where the
 163 geometry often changes in a non-uniform and unpredictable
 164 manner giving large and non-uniform stresses and strains
 165 along the sample. For example, dough thins out non-
 166 uniformly when stretched, in common with many polymers,
 167 giving rise to large stresses and strains not correctly
 168 calculated by the conventional method of dividing by

169 original sample dimensions. In this case, it is necessary to
 170 normalise by *actual* change in dimensions during deforma-
 171 tion to obtain true stress (force/instantaneous area) or
 172 Hencky strain (displacement/instantaneous length (L), e.g.
 173 $dL/L = \ln(L/L_0)$), in which case sample dimensions should
 174 be measured locally and independently by using contact
 175 extensometers or non-contact techniques such as laser,
 176 video or photographic techniques. For materials which flow
 177 under normal measurement time scales, stress is normally
 178 divided by strain rate (strain/time of strain application) to
 179 give viscosity.

180 Rheological properties should be independent of size,
 181 shape and how they are measured; in other words, they are
 182 universal, rather like the speed of light or density of water,
 183 which do not depend on how much light or water is being
 184 measured or how it is being measured. It would be
 185 comforting to know that the stiffness of bread or viscosity
 186 of dough measured in one laboratory will be the same
 187 measured in any laboratory in the world, even if they are
 188 measured using different tests, sample sizes or shapes. An
 189 objective of rheology is to determine properties reproduc-
 190 ibly in a manner that allows comparison between different
 191 samples, test sizes and shapes, and test methods.

194 3. Rheological test methods

196 There are many test methods used to measure rheological
 197 properties. It is not possible to describe all the available
 198 testing geometries here, and the reader is referred to general
 199 reviews of rheology (Whorlow, 1992; Ferry, 1980; Barnes
 200 et al., 1989), rheological testing of foods (Sherman, 1970;
 201 Carter, 1990; Rao and Steffe, 1992; Dobraszczyk and
 202 Vincent, 1999; van Vliet, 1999) and cereal products
 203 (Bloksma and Bushuk, 1988; Faridi and Faubion, 1990;
 204 Muller, 1975; Faridi and Faubion, 1986). It is common to
 205 categorise rheological techniques according to the type of
 206 strain imposed: e.g. compression, extension, shear, torsion
 207 etc. and also the relative magnitude of the imposed
 208 deformation, e.g. small or large deformation. The main
 209 techniques used for measuring cereal properties have
 210 traditionally been divided into descriptive empirical tech-
 211 niques and fundamental measurements (Bloksma and
 212 Bushuk, 1988).

215 4. Descriptive rheological measurements

217 Within the cereals industry there has been a long history
 218 of using descriptive empirical measurements of rheological
 219 properties, with an impressive array of ingenious devices
 220 such as the Penetrometer, Texturometer, Consistometer,
 221 Amylograph, Farinograph, Mixograph, Extensigraph,
 222 Alveograph, various flow viscometers and fermentation
 223 recording devices, reviewed by Muller (1975) and Shuey
 224 (1975), (Table 1). Empirical tests are easy to perform and

Table 1
Rheological methods used for cereal products

Method	Products	Property measured
<i>Empirical methods</i>		
Mixers: farinograph, mixograph, reomixer	Dough	Mixing time/torque apparent viscosity
Extensigraph	Dough	Extensibility
Taxt2/Kieffer Rig	Dough, gluten	Extensibility
Alveograph	Dough, gluten	Biaxial extensibility
Amylograph, RVA	Pastes, suspensions	Apparent viscosity, gelatinisation temp
Consistometer	Sauces, fillings	Apparent viscosity
Flow cup	Fluids, sauces, batters	Apparent viscosity
Falling ball	Fluids	Apparent viscosity
Flow viscometers	Fluids, pastes	Apparent viscosity
Fermentometers	Dough	Height, Volume
Penetrometers	Semi-solid foods, gels	Firmness, hardness
Texturometer, TPA	Solid foods	Texture, firmness
<i>Fundamental methods</i>		
Dynamic oscillation, concentric cylinders, parallel plates	Fluids, pastes, batters, doughs	Dynamic shear moduli, dynamic viscosity
Tube viscometers: capillary, pressure, extrusion, pipe flow	Fluids, sauces, pastes, dough	Viscosity, viscosity, in-line viscosity
Transient flow: concentric cylinders, parallel plates	Semi-solid (visco-elastic) materials	Creep, relaxation, moduli and time
Extension: uniaxial, biaxial, dough inflation system, lubricated compression	Solid foods, doughs	Extensional viscosity, strain hardening

are often used in practical factory situations, providing data which is useful in evaluating performance during processing and for quality control. The instruments are often robust and capable of withstanding demanding factory environments, and do not require highly skilled or technically trained personnel. They have provided a great deal of information on the quality and performance of cereal products such as consistency, hardness, texture, viscosity etc. However, these instruments do not fulfil the requirements of a fundamental rheological test, since:

- the sample geometry is variable and not well defined
- the stress and strain states are uncontrolled, complex and non-uniform
- it is therefore impossible to define any rheological parameters such as stress, strain, strain rate, modulus or viscosity.

Therefore, these tests are purely descriptive and dependent on the type of instrument, size and geometry of the test sample and the specific conditions under which the test was performed. For example, empirical

tests have been used to characterise the behaviour of bread doughs during processing. Many of these are used as 'single point' tests, where a single parameter is often arbitrarily selected from a whole range of data acquired during the test as, for example, in selecting the peak torque from a mixing trace and then using this to correlate with performance, which neglects a large part of the recorded data (Dobraszczyk and Schofield, 2002; Wikström and Bohlin, 1996), and is appropriate only to the set of conditions under which that test was performed and is generally not applicable to any other deformation conditions. Since dough experiences a wide range of conditions of stress states and strain rates during processing and baking, and the rheological properties of dough are dependent both on time and strain, there is often a discrepancy between such single point type tests and actual performance in the plant, where conditions of strain and strain rate may be poorly defined and very different to those in the laboratory test. Whilst this may give the illusion of a 'scientific' test by being performed on a machine (frequently with a computer attached), and may give satisfactory correlations with a textural or processing parameter, it is impossible to compare results between different testing machines, e.g. between Farinograph, Mixograph or Extensigraph, or to extrapolate the results to other deformation conditions, such as during baking.

Most food materials are viscoelastic and therefore their properties depend on how quickly the test is performed (the strain rate or frequency). This is important in many aspects of dough processing: if the dough is deformed quickly, such as in mixing or sheeting, then the rheological properties of the dough will be very different if measured at the typically slower rates of deformation found in conventional testing machines. Alternatively, during processing dough will experience strains very different in magnitude and nature than those generally available in a rheological test. Many food processes operate under large deformation extensional flow, whilst most rheological tests on foods are performed in small deformation shear oscillation. Measurements under large deformations often show very different rheological response to those in small deformation, especially if the material contains high molecular weight (HMW) polymers. Tests under only one particular set of conditions of rate, temperature and strain will almost certainly not be applicable to another set of deformation conditions. What is necessary is to define the set of deformation conditions which the food sees in practice and perform tests under similar conditions.

5. Fundamental rheological tests

Problems encountered with such fundamental tests are: complex instrumentation which is expensive, time consuming, difficult to maintain in an industrial environment and

require high levels of technical skill, often inappropriate deformation conditions, difficulty in interpretation of results, and slip and edge effects during testing.

The most common types of fundamental rheological tests used in cereal testing are: (i) small deformation dynamic shear oscillation; (ii) small and large deformation shear creep and stress relaxation; (iii) large deformation extensional measurements; and (iv) flow viscometry (Table 1).

6. Dynamic oscillation measurements

Adapted from techniques developed for measuring viscoelastic properties of polymer melts and concentrated solutions (Ferry, 1980; Barnes et al., 1989), this is one of the most popular and widely used fundamental rheological techniques for measuring cereal doughs and batters. These tests measure rheological properties (such as elastic and viscous moduli) by the application of sinusoidally oscillating stress or strain with time and measuring the resulting response. They have the advantage of a well developed theoretical background, readily available instrumentation, and simultaneous measurement of elastic and viscous moduli, while the non-destructive nature of the test enables multiple measurements to be performed as temperature, strain or frequency are varied.

Disadvantages of the dynamic oscillation method are that the deformation conditions are often inappropriate to practical processing situations, because they are carried out at rates and conditions very different from those experienced by the dough during processing or baking expansion. For example, rates of expansion during proof and oven rise in bread doughs have been calculated between 5×10^{-3} and $5 \times 10^{-4} \text{ s}^{-1}$, compared with measuring rates in rheological tests several orders of magnitude greater (Bloksma, 1990).

Conventional oscillatory shear rheological tests usually operate in the linear region at small strains in the order of up to 1%, whilst strain in gas cell expansion during proof is known to be in the region of several hundred percent (Amemiya and Menjivar, 1992). Furthermore, most dynamic rheological tests are carried out in shear, whilst most large-strain deformations in dough (i.e. extrusion, sheeting, proof and baking) are extensional in nature. It is not widely recognised within the cereals science community that measurements in large deformation shear show a completely different response to extensional measurements. Polymer melt fluid dynamics show that very different rheology occurs under shear deformation than under extension due to the different physical effects large deformations can have on networks of HMW polymers (Ferry, 1980; Padmanabhan, 1995). From extensional studies on long-chain high molecular weight polymer melts it is known that entirely different rheological properties are obtained in shear than in tension, especially if the polymer chains are branched. For example, the elongational

viscosity of low-density (branched-chain) polyethylene melts increases with both strain and strain rate (strain hardening), whilst the shear viscosity decreases with strain and strain rate (shear-thinning), giving widely different values in final viscosities between elongation and shear (Fig. 1a). For dough, shear and elongational viscosities at low strains are similar, with extensional viscosity slightly higher than shear viscosity but at higher strains they diverge and the elongational viscosity rises steeply to give a value 2 orders of magnitude higher at failure (Fig. 1b). This increased strain hardening is attributed to entanglement of long-chain molecules during extensional flow, whereas in simple shear they remain coiled and can slip past each other, giving rise to observed shear thinning at higher strains (Cogswell, 1981).

Therefore, many of the rheological tests for doughs reported in the literature are inappropriate in predicting end-use quality because they do not measure the system under the appropriate deformation conditions and they are not sensitive to the molecular structures responsible for baking quality. Small deformation dynamic shear rheological tests are frequently performed and applied almost indiscrimi-

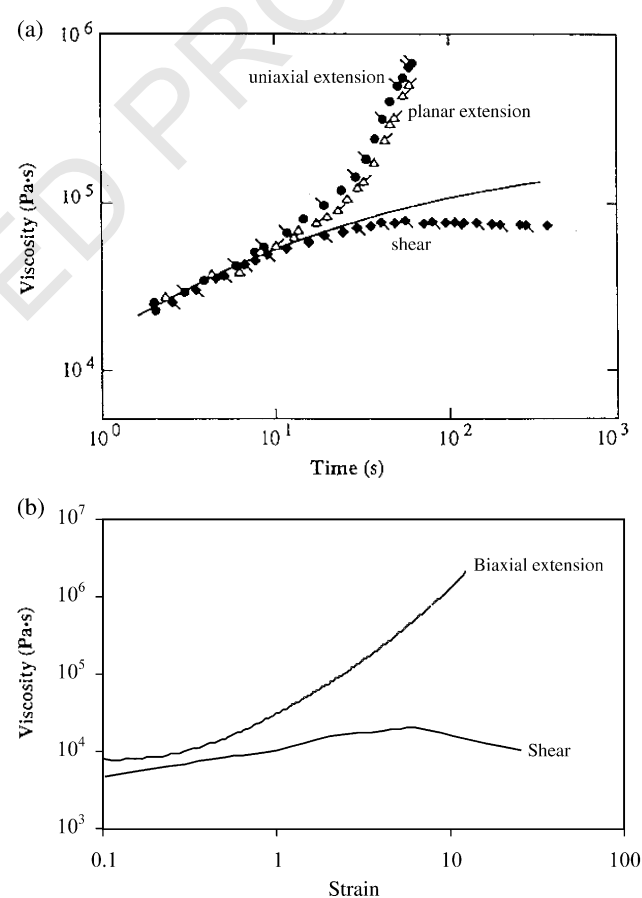


Fig. 1. (a) Shear and extensional viscosity of LDPE (low density polyethylene) at 125 °C at constant strain rate (0.05 s^{-1}). From MacLeish and Larson (1998). (b) Large deformation shear and biaxial extensional viscosities vs. Cauchy strain for dough at constant strain rate (0.1 s^{-1}).

nately in predicting end-use quality regardless of whether they are valid or not.

7. Creep and relaxation measurements

In stress relaxation measurements, deformation is held constant and the force response is measured, whilst in creep the stress is held constant and the deformation is measured. Schofield and Scott Blair (1937) first measured the creep response of dough and obtained values for elastic moduli and viscosity which are still relevant. Bloksma and Bushuk (1988) surveyed the experimental results from the literature for stress relaxation measurements for a number of doughs. None of the curves showed the exponential decay typical of a single relaxation time, which would indicate a single molecular mechanism responsible for relaxation, but correspond to a decay typical of a broad spectrum of relaxation times. This shows a broad distribution of molecular mechanisms is responsible for the relaxation process within dough, which can be related to the wide molecular weight distribution of gluten.

Many authors have shown that a slower relaxation time is associated with good baking quality (Bloksma, 1990; Launay, 1990; Wang and Sun, 2002), with relaxation time relatively independent of water, mixing time or temperature. Stress relaxation measurements on dough and gluten in shear showed that the relaxation behaviour of dough could be described by two relaxation processes (Bohlin and Carlson, 1980): a rapid relaxation over 0.1–10 s and a slower process occurring over 10–10,000 s. The rapid relaxation process has been associated with small polymer molecules which relax rapidly, and the longer relaxation time has been linked to the HMW polymers found within gluten. Measurements of large-deformation creep and shear stress relaxation properties were found to be useful in discriminating between different wheat varieties of varying quality, and were found to be closely associated with baking volume (Safari-Ardi and Phan-Thien, 1998; Wikström and Eliasson, 1998) and strength of durum wheat varieties (Edwards et al., 1999, 2001). At small strain amplitudes (0.1%), doughs with different baking quality showed no differences in relaxation behaviour, but at a range of large strains (up to 29%) their creep and relaxation behaviour was closely correlated with the baking behaviour of dough (Safari-Ardi and Phan-Thien, 1998).

Doughs exhibited a characteristic bimodal distribution of relaxation times, with the second peak clearly discriminating between cultivars with varying strength and quality, which reflects the differences in the MW distribution of glutenin polymers (Rao et al., 2001; Li et al., 2003). The second relaxation peak is related to the entanglement properties of high molecular weight insoluble glutenin polymers, and has been shown to be directly related to the insoluble fraction of the high MW glutenins (Li et al., 2003). Relaxation properties of doughs relate well to MWD and

particularly to entanglements of HMW glutenin polymers and may be used as a rapid method of discriminating variations in MWD between varieties which vary in baking quality.

8. Extensional techniques

There are many types of extensional flow measurements, including: simple uniaxial tension, fibre wind-up or spinning, converging flow, capillary extrusion, opposed jets, lubricated compression and bubble inflation (Whorlow, 1992; Macosko, 1994). There are several methods which have been used to measure the rheological properties of dough in extension: simple uniaxial extension, where dough is stretched in one direction; and biaxial extension, where the dough is stretched in two opposing directions, which can be achieved either by compression between lubricated surfaces or by bubble inflation (Dobraszczyk and Vincent, 1999; Huang and Kokini, 1993; Bagley and Christianson, 1986).

Many food materials undergo large deformations in practice during processing and eating, and many of these have a large extensional component. For example, extensional flows are important in mixing and sheeting of pastry and dough, converging and diverging flow such as in extrusion and pumping, spreading of soft solids such as butter, cheeses and pastes, and expansion of bubbles in foams such as bread dough, cakes and heat extruded snacks. Unfortunately, most tests in flow are carried out in shear under small deformations, mainly because most conventional viscometers operate in shear, because the equipment is readily available and the technique well established. However, measurements carried out in shear and using small deformations do not provide information about material behaviour under large extension. Therefore there is an obvious need to perform measurements under conditions relevant to those experienced by the material in actual practice.

9. Uniaxial extension

One of the oldest and most widely used test methods to measure materials properties is the uniaxial tensile test. A strip of material is clamped at both ends and pulled apart at a fixed rate in a suitable testing machine, and the force measured at the same time as the displacement of the object. The force is generally plotted against the displacement (extension) to give a force–extension curve. Tensile tests may produce an approximately uniform extension of a sample provided necking does not occur. Normally the force and extension are divided by the original sample dimensions to obtain stress and strain, and allows removal of the sample geometry as a variable, but for doughs undergoing large extensional deformation the actual change in dimensions must be measured or calculated. The slope of

561 the stress–strain curve then gives the elastic modulus or
562 stiffness. Many test methods attempt to measure the uniaxial
563 extensional properties of doughs, such as the Simon
564 Research Extensometer, Brabender Extensigraph, Stable
565 Micro Systems Kieffer dough and gluten extensibility rig,
566 but none of these gives rheological data in fundamental
567 units of stress and strain, because the sample geometry is not
568 defined or measured, dimensions change extensively and
569 non-uniformly during testing, and it is therefore impossible
570 to define any rheological parameters such as stress, strain,
571 strain rate, modulus or viscosity.

572 Studies on the fundamental uniaxial extensional rheolo-
573 gical properties of doughs have been carried out by many
574 workers (Schofield and Scott Blair, 1932; Tschoegl et al.,
575 1970; Tschoegl et al., 1970; Rasper, 1975; Uthayakumar
576 et al., 2000). Some of the earliest attempts to characterise
577 the fundamental rheological properties of dough were in a
578 series of uniaxial extensional measurements by Schofield
579 and Scott Blair (1932) who stretched a cylinder of dough
580 floated on a mercury bath and measured the elongation and
581 force. Plastic and elastic components of deformation were
582 resolved and viscosity and elastic modulus were calculated.
583 They showed that the rheological behaviour of dough is
584 non-linear with strain and strain rate, i.e. elastic modulus
585 and viscosity vary with both rate and strain. Tschoegl et al.,
586 1970a,b) measured the large extension properties of doughs
587 by extending a ring of dough suspended in a liquid of
588 density equivalent to that of the dough between two hooks at
589 constant deformation rates until rupture. The stress-strain
590 curves showed considerable strain hardening, and strain and
591 stress at rupture was considerably less for poor quality flours
592 than good quality flours.

593 10. Biaxial extension

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596 In biaxial extension a sample is stretched at equal rates in
597 two perpendicular directions in one plane, as in an
598 expanding spherical balloon. The most widely used methods
599 for measuring biaxial extension properties of food materials
600 are inflation techniques and compression between flat plates
601 using lubricated surfaces, which produce purely extensional
602 flow provided no friction occurs (Dobraszczyk and Vincent,
603 1999; Chatraei et al., 1981).

604
605 Inflation was first used as an empirical technique to
606 measure wheat gluten and bread dough extensibility in
607 1920s (Hankoczy, 1920; Chopin, 1921). This method was
608 later developed to assess rheological parameters (Launay
609 et al., 1977) and further developed to measure the fracture
610 and biaxial extensional rheological properties of wheat
611 doughs and glutes, and to assess the quality of wheat flour
612 doughs and glutes (Dobraszczyk and Roberts, 1994;
613 Dobraszczyk, 1997). The major advantage of this test is
614 that the deformation closely resembles practical conditions
615 experienced by the cell walls around the expanding gas cells
616 within the dough during proof and oven rise, i.e. large

617 deformation biaxial extension. Extensional rheological
618 properties can be measured at large strains up to failure
619 and low strain rates, and the gripping problems normally
620 associated with uniaxial tests can be minimised. Extensional
621 rheological properties of wheat doughs have been measured
622 using lubricated compression and bubble inflation (Huang
623 and Kokini, 1993; Bagley and Christianson, 1986; Uthaya-
624 kumar et al., 2000; Chatraei et al., 1981; Hankoczy, 1920;
625 Chopin, 1921; Launay et al., 1977; Dobraszczyk and
626 Roberts, 1994; Dobraszczyk, 1997; van Vliet et al., 1992;
627 Wikström and Bohlin, 1999a,b). Differences in extensional
628 strain hardening between varieties of different baking
629 quality were found to relate to baking quality, with good
630 breadmaking varieties showing greater strain hardening and
631 extensional viscosity (Dobraszczyk and Roberts, 1994;
632 Dobraszczyk, 1997; van Vliet et al., 1992; Wikström and
633 Bohlin, 1999a,b).

634 11. Applications of rheology to cereal products

635 11.1. Mixing

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640 Mixing is a critical operation in food processing where,
641 apart from the obvious function of mixing ingredients, the
642 structure of the food is often formed. For example, in
643 the production of batters, pastes and doughs, the nature of
644 the mixing action develops the viscoelastic properties of
645 gluten and also incorporates air, which has a major effect on
646 their rheology and texture. There is an intimate relationship
647 between mixing, aeration and rheology: the design and
648 operation of the mixer will develop texture, aeration and
649 rheology to different extents (Campbell and Shah, 1999),
650 and conversely the rheology of the food will affect the time
651 and energy input required to achieve optimal development.
652 This is seen in the great variety of mixers used in the food
653 industry and the fact that certain mixers are required to
654 produce a desired texture or rheology in a food (Campbell,
655 1995).

656 Studies on the rheology of mixing have focused on a
657 number of areas: (i) the effects of mixer design and
658 operation on the development of rheology and texture; (ii)
659 empirical measurement of rheology during mixing from
660 mixer torque or power consumption; (iii) effect of rheology
661 on mixing patterns and performance; and (iv) simulation
662 and prediction of mixing flow deformation patterns as
663 functions of mixer geometry and rheology.

664 Most of the studies on doughs have been on the
665 relationships between mixing, rheology and baking per-
666 formance, because of the rheological changes that occur in
667 the gluten viscoelastic network during mixing and their
668 importance for product quality. Despite the obvious
669 importance of mixing in the development of rheology and
670 texture in doughs, there is very little information in the
671 literature on these changes during the different stages in the
672 mixing process. Most work has either concentrated on

empirical measurement of mixer motor torque, voltage or power consumption during mixing as a qualitative indication of changing rheology, or measurement of rheological changes at some time after mixing. Problems associated with these approaches are: failure to take into account motor and drive losses, frictional and surface effects between the dough and the mixer, varying signal damping and data acquisition rates, effects of aeration on rheology, and rheological relaxation effects. Since dough is a viscoelastic material which shows rapid relaxation after deformation, which varies between different flours, such measurements are not ideal and run the risk of giving misleading information. Nevertheless, much useful information has been obtained about the effect of mixing on gluten structure (Weegels et al., 1996; Skerit et al., 1999), rheology and baking performance. Extensive work on dough mixing has shown that mixing speed and energy (work input) must be above a certain value to develop the gluten network and to produce satisfactory breadmaking (Kilborn and Tipples, 1972), and an optimum in work input or mixing time has been related to optimum breadmaking performance (Skeggs, 1985), which varies depending on mixer type, flour composition and ingredients (Mani et al., 1992). For example, mixing doughs by elongational flow in sheeting to achieve optimum development required only 10–15% of the energy normally used in conventional high speed shear mixers (Kilborn and Tipples, 1974), suggesting that much higher rates of work input can be achieved due to the enhanced strain hardening of doughs under extension. Numerous studies have shown that rheological measurements after mixing parallel changes in mixer torque and power consumption (Mani et al., 1992; Zheng et al., 2000; Anderssen et al., 1998), especially if rheological measurements are made under large, non-linear deformation conditions closer to those experienced in the mixer (Mani et al., 1992; Hwang and Gunasekaran, 2001). Recent studies have suggested that qualitative elongational rheological information during mixing can be derived directly from the torque/power consumption of a dough mixer (Gras et al., 2000).

Another approach is the experimental measurement, simulation and modelling of deformation patterns within a mixer using high speed imaging techniques and 3D numerical computational fluid dynamics (CFD). Here the objective is to predict the effect of changing mixer geometry and rheological properties on deformation during mixing to produce mixers better designed to achieve optimum dough processing. Close correspondence between actual and predicted flow patterns indicates the validity of such models, and can be used to obtain indirect information on rheological behaviour during mixing. Model 3D simulation and experimental studies have been carried out for complex rotating flows associated with dough mixing (Binding et al., 2003). Laser Doppler Anemometry was used to obtain the actual flow profiles and velocities during mixing and compared with the numerical modeling simulation to

validate the predicted flow. The problem with such approaches is that many models contain highly simplistic rheological relationships (such as a Newtonian power law shear flow for dough (Levine and Drew, 1990)) which do not take into account the viscoelastic shear and extensional properties of doughs at large deformations, and render such models inaccurate.

11.2. Sheeting

Sheeting of dough with rolls is a common operation in the manufacturing of products from cereals. There is a range of different products and raw materials that are sheeted. The purpose of sheeting is different for each product. Bread dough is sheeted in a moulder to shape a dough and to control the bubble size distribution. Repeated sheeting can be used to develop the gluten network in bread dough. Biscuit dough is sheeted both to form the dough and to develop the gluten. Corn masa is sheeted to produce a coherent dough sheet and pastry dough is sheeted and folded to produce a layered structure. There are several different sheeter arrangements, ranging from a single rolling pin to multi-rollers. Generally, the shape of the dough changes during sheeting and the rheological properties of the dough determine the stresses and strains during sheeting. Care must be taken to use rheological properties relevant to the particular application, taking the magnitude of the strain and the rate of strain into account.

Few studies have been done to find the relationship between fundamental rheological properties and sheeting. Most studies are empirical in nature and relate flour properties to end product quality. Bread dough has been studied most extensively. When bread dough is sheeted repeatedly the gluten forms a network, which increases the elasticity in the dough. This gluten network is an essential structure for bread making. Its development is usually measured by the amount of energy that is used to form the dough. Kilborn and Tipples (1974) measured the energy requirement and found that sheeting uses 10–15% of the energy required for dough mixing. It is obvious that the energy requirement is not an inherent flour or dough property as it depends on the mixer or sheeter it has been produced on. To measure rheological properties during sheeting the force or torque on the rolls can be measured. The force and torque depend on the flour strength: a stronger flour produces higher forces. But the roll diameter and roll gap also affect the force and torque (Kilborn and Preston, 1982; Raghavan et al., 1996). Rheological properties can be measured immediately after sheeting. Extensigraph resistance and extensional viscosity increase with repeated sheeting and then decrease again (Moss, 1980; Morgenstern et al., 1999). The maximum viscosity coincides with highest bread volume, showing that the repeated sheeting affects the dough in a similar way to mixing.

Puff pastry is made by enveloping a sheet of butter or fat in dough, sheeting it, folding it to increase the numbers of

785 layers and sheeting it again to reduce the thickness. This
 786 process is repeated a number of times to obtain a layered
 787 structure with thin fat/dough layers. The rheological
 788 properties of the pastry dough are quite different from a
 789 bread dough. The dough is less elastic and ruptures at
 790 smaller deformations. Dough with high extensibility and
 791 resistance (measured by Extensigraph) produces best lift
 792 (rise) during baking (Hay, 1993). When pastry dough is
 793 sheeted the stresses relax over a period of up to 1 h. This
 794 stress relaxation is related to shrinkage of baked puff pastry
 795 and therefore a resting time is usually observed between
 796 sheeting and baking (Newberry et al., 1996).

797 There is no standardised test for measuring rheological
 798 properties of a dough sheet. Many studies use the
 799 Extensigraph. However, the Extensigraph was developed
 800 for bread dough and the standard test uses rested (relaxed)
 801 dough with a water level related to bread dough. Clearly,
 802 applications with different water levels will have very
 803 different rheological properties than measured with the
 804 Extensigraph test. An adapted extensional test was proposed
 805 by Morgenstern et al. (1996) to measure extensional
 806 viscosity on a dough sheet. Measurements on bread dough
 807 demonstrated power law behaviour and strain hardening for
 808 large deformations.

809 For a fundamental understanding of the sheeting
 810 operation Levine (1985) used a mathematical model,
 811 borrowed from polymer calendaring, to predict sheet
 812 thickness, roll separation force and energy consumption of
 813 sheeting. Dough rheology was described with a power law
 814 relationship between stress and strain rate. This model has
 815 been extended to include finite width sheets, to include air
 816 bubbles, to describe flaking, and to include the effect of
 817 ingredient levels (Levine and Levine, 1997; Raghavan et al.,
 818 1995; Levine, 1998; Levine et al., 2002; Morgenstern et al.,
 819 2002). The power law, however, does not incorporate
 820 viscoelasticity, and predictions of sheeting behaviour have
 821 been only satisfactory in a qualitative sense. A recent study
 822 has incorporated elasticity in the models (Morgenstern et al.,
 823 2002; Love, 2002).

825 11.3. Fermentation and baking

826
 827 Fermentation is an important step in the breadmaking
 828 process, where the expansion of air bubbles previously
 829 incorporated during mixing provides the characteristic
 830 aerated structure of bread, which is central to its appeal
 831 (Dobraszczyk et al., 2000). Although fermentation is clearly
 832 important in breadmaking, most rheological tests are
 833 performed on doughs without yeast and at room tempera-
 834 ture. Few studies have been made on the changing
 835 rheological properties during fermentation and baking.
 836 Direct rheological measurements have been made on
 837 yeasted bread doughs (Kilborn and Preston, 1981), cake
 838 batters (Massey, 2002; Sahi, 1999), sour doughs (Wehrle
 839 and Arendt, 1998), and cracker sponge and dough (Oliver
 840 and Brock, 1997). Such measurements suffer from

841 the problem of the evolving gas volume and metabolites
 842 from fermentation confounding the rheological data. The
 843 decrease in density as a result of increasing gas volume
 844 would be expected to have the effect of decreasing modulus
 845 and viscosity, but the compressibility of air may counteract
 846 this effect, especially at higher gas volumes and low
 847 densities where the moduli of the solid and gas phases
 848 converge, such as in cake batters, where shear modulus is
 849 directly related to the air content (Massey, 2002). Fermenta-
 850 tion metabolites such as lactic and acetic acid, may also
 851 exert rheological effects through changes in pH (Wehrle
 852 et al., 1997).

853 Other approaches have been to measure the increase in
 854 height or volume of the fermenting product using devices
 855 such as the rheofermentometer or risograph, but these
 856 provide no direct information about the rheology of the
 857 material, since they do not measure force or deformation per
 858 change in unit dimensions. Changes in aeration have been
 859 predicted from modelling the increase in dough height
 860 (Shah et al., 1999), or by directly measuring internal gas
 861 pressure during fermentation (Matsumoto et al., 1975).
 862 Another approach has been to prevent fermentation by
 863 inactivating the yeast by freezing and thawing (Newberry
 864 et al., 2002), or by mixing under oxygen to rapidly saturate
 865 the yeast activity (Chamberlain and Collins, 1979).

866 Rheological studies on doughs related to baking have
 867 normally been performed in small deformation shear
 868 oscillation at room temperature. Such dynamic rheological
 869 measurements on doughs have been investigated in many
 870 studies (Hibberd and Wallace, 1966; Hibberd and Parker,
 871 1975a,b; Abdelrahman and Spies, 1986; Smith et al., 1970;
 872 Dreese et al., 1988). Viscous (G'') and elastic (G') moduli for
 873 dough are measured over a range of frequencies. Elastic
 874 properties predominate over viscous properties, and the
 875 moduli are slightly frequency dependent, which is typical of
 876 a cross-linked polymer network (Rao and Steffe, 1992).

877 No convincing relationship has been established between
 878 dynamic rheological properties and baking performance
 879 (Khatkar et al., 1995; Kokelaar et al., 1996; Hayman et al.,
 880 1998; Autio et al., 2001). Abdelrahman and Spies (1986)
 881 compared two flours of different baking quality and
 882 measured lower values of elastic (storage) modulus (G')
 883 for the higher baking quality flour. Similarly, many reports
 884 have found lower values of G' corresponded with better
 885 baking quality (Faridi and Faubion, 1986; Weipert, 1988;
 886 Weipert, 1992; Schober et al., 2002). Others, however, have
 887 found that a higher value of G' for glutes and doughs
 888 relates to better baking performance (Amemiya and
 889 Menjivar, 1992; Mani et al., 1992; Attenburrow et al.,
 890 1990; Janssen et al., 1996), see Table 2. Small deformation
 891 dynamic rheological measurements have provided useful
 892 information about the properties of isolated glutes and their
 893 sub-fractions in relation to baking quality, but similar
 894 measurements on whole dough systems have not shown any
 895 significant differences (Khatkar et al., 1995). It is thought
 896 that these conflicting results are obtained because most of

these tests are carried out at rates and deformation conditions very different from those experienced by the dough during baking expansion, and also because dynamic rheological parameters in the plateau region are generally insensitive to differences in molecular weight (MW) of polymers. However, these parameters are highly sensitive to changes in polymer concentration and diluents such as water, which are almost never kept constant in these experiments on doughs. Most dynamic rheological tests on doughs and glutes have been carried out over a relatively narrow frequency range in the plateau zone, which is known to be insensitive to changes in molecular weight (Ferry, 1980; Macosko, 1994). If, as is generally accepted, large MW glutenin polymers are responsible for the variations in breadmaking performance between different wheat varieties, then it is to be expected that measurements of the plateau modulus will not be good indicators of baking performance and are therefore not appropriate to baking.

During proof and baking the growth of gas bubbles determines the expansion of the dough and therefore the ultimate volume and texture of the baked product (He and Hoseney, 1991). The limit of expansion of these bubbles is related directly to their stability, due to coalescence and the eventual loss of gas when the bubbles fail. The rheological properties of the bubble walls will therefore be important in maintaining stability against premature failure during baking, and also in relation to gas cell stabilization and gas retention during proof, and thus to the final structure and

volume of the baked product (Dobraszczyk et al., 2000). The relevant rheological conditions around an expanding gas cell during proof and baking are biaxial extension, large strain and low strain rate. Any rheological tests which seek to relate to baking performance should therefore be performed under conditions similar to those of baking expansion, such as large strain biaxial extension and low strain rates. Methods such as bubble inflation and lubricated compression potentially offer the most appropriate method for measuring rheological properties of doughs. The major advantage of these tests is that the deformation closely resembles practical conditions experienced by the cell walls around the expanding gas cells within the dough during proof and oven rise, i.e. large deformation biaxial extension, and can be carried out at the low strain rates and elevated temperatures relevant to baking (Dobraszczyk et al., 2003).

Extensional rheology is sensitive to polymer chain branching and entanglement interactions between high MW polymers at large deformations (MacLeish and Larson, 1998; Wagner et al., 2000; Münstedt et al., 1998), the theory is simple and relatively well developed, and it generally provides good correlations with the relevant large deformation processing quality parameters (Table 2). Biaxial extensional rheological properties can be measured at large strains up to failure, and the gripping problems normally associated with uniaxial tests can be minimised. Disadvantages are that there is no single well-defined and accepted method for extensional measurement, with many different

Table 2
Correlations between rheological properties and baking performance

Rheological parameter	Baking parameter	Correlation	References
<i>(i) Small deformation shear oscillation</i>			
G' 1 Hz (dough)	Volume	0.15 ($n = 48$)	Autio et al. (2001)
G' SLOPE		0.72	
G' 10 Hz (wet gluten)	Volume	-0.85 ($n = 27$)	Schober et al., 2002
	Form ratio (W/H)	0.65	
G' 1 Hz (gluten)	Volume	N.S ($n = 20$)	Tronsmo et al. (2003)
TAN DELTA	Form ratio (H/W)	0.69	
	(H/W)	-0.71	
G' 1 Hz	Loaf height	-0.64 ($n = 8$)	Uthayakumaran et al. (2000)
TAN DELTA		N.S.	
G'	Volume	N.S. ($n = 4$)	Safari-Ardi and Phan-Thien (1998)
<i>(ii) Large deformation</i>			
Biaxial extensional	Volume	0.89 ($n = 20$)	Tronsmo et al. (2002)
Strain hardening	Form ratio	0.80	
Max. Uniaxial, extensional viscosity	Loaf height	0.81	Uthayakumaran et al. (2002)
Biaxial strain hardening	Volume	0.92–0.97 ($n = 6$)	Dobraszczyk and Roberts (1994), Dobraszczyk (1997), and Dobraszczyk et al., 2003
Biaxial strain, hardening	Volume	Good	Wikström and Bohlin (1999a,b)
Biaxial strain hardening	Volume	Good	Kokelaar et al. (1996)
Relaxation	Volume	Good	Wikström and Eliasson (1998)
Creep		0.94 ($n = 23$)	Wang and Sun (2002)
Relaxation	Quality	Good	Safari-Ardi and Phan-Thien (1998)
Shear relaxation, shear viscosity	Quality	Good	Amemiya and Menjivar (1992)

methods being used depending on the type and viscosity of the material being studied; the tests often use large amounts of material and the tests are destructive. The measurement of extensional flow is often difficult, because the deformation is large and non-uniform and it is therefore impossible to calculate strain directly from the machine displacement, requiring the direct measurement or calculation of changes in sample dimensions, often at high speeds. In biaxial extension, for example, inaccuracies in the calculation of rheological properties can be introduced due to deviations of the bubble shape from assumed ideal sphericity (Charalambides et al., 2002a,b).

The failure of gas cell walls in doughs has been shown to be directly related to the elongational strain hardening properties of the dough measured under large deformation biaxial extension (Dobraszczyk and Roberts, 1994; Dobraszczyk, 1997; Dobraszczyk et al., 2003). Strain hardening is shown as an increase in the slope of the true stress-strain curve with increasing extension, giving rise to the typical J-shaped stress-strain curve observed for highly extensible materials (Fig. 2) and has been shown to be necessary for stability in any operations which require large extensions, such as cold-drawing of polymer fibres, inflation of polymer films and blow moulding of plastic bottles, and the expansion of gas cells within expanding bread doughs (Dobraszczyk and Roberts, 1994; Kokelaar et al., 1996). Strain hardening in doughs is thought to arise mainly from entanglement coupling of the larger glutenin molecules which gives rise to the high viscosities observed at large strains (Singh and MacRitchie, 2001). Under extensional flow, entangled polymers exhibit strain hardening which is enhanced for polymers with a broad MW distribution, particularly a bimodal distribution (Watanabe, 1999) and branching (Wagner et al., 2000; Münstedt et al., 1998). It is therefore expected that the broad bimodal MW distribution and branched structure of gluten will result in enhanced strain hardening and a bimodal distribution of relaxation times (Bohlin and Carlson, 1980; Li et al., 2003).

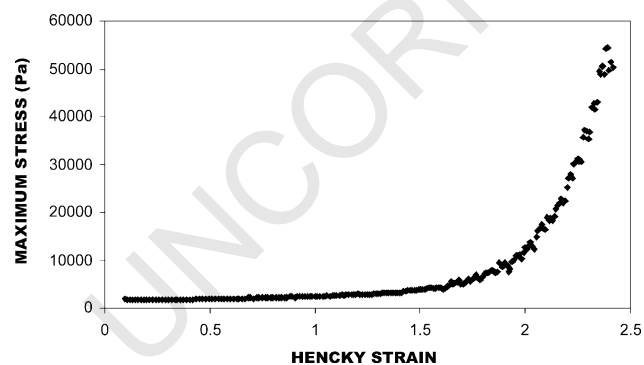
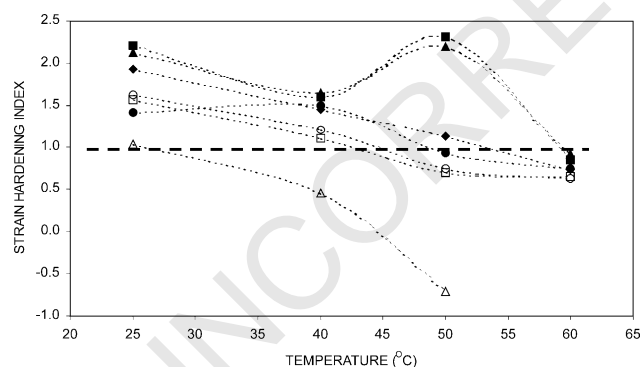


Fig. 2. Typical J-shaped stress-Hencky strain curve in biaxial extension for a dough bubble inflated at 50 °C and constant strain rate (0.1 s⁻¹). Bubble inflation using SMS Dough Inflation System, maximum stress and Hencky strain calculated for bubble wall polar region.

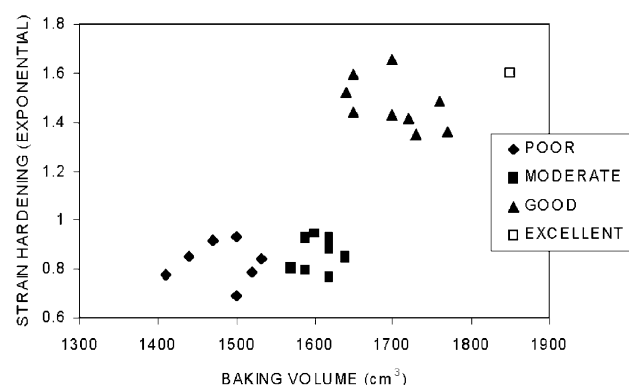
Gas cell wall failure in expanding dough bubbles can be predicted using the Considere criterion for instability in extension for polymers (Dobraszczyk and Roberts, 1994; Wikström and Bohlin, 1999). This criterion states that the stability in extension of a viscoelastic material is guaranteed provided the strain is less than that at which a maximum occurs in the force-extension plot and defines a critical strain beyond which failure is inevitable on further extension. Uniform extension of a viscoelastic membrane during inflation is guaranteed provided the strain does not reach this maximum. Beyond this critical value of strain the criterion states that the material cannot be extended homogeneously and instead undergoes a dynamic failure event (MacKinley and Hassager, 1999). During large extension of materials, plastic strain is uniform throughout the sample up to the point of maximum force. Beyond this point, force begins to decrease and it is at this point that localised and non-uniform plastic deformation begins to occur. The cross-sectional area begins to change in a non-uniform way, and a neck or localised constriction forms, which can either stabilise or propagate in an unstable manner to failure. If the cross section at any point is slightly less than elsewhere or there are any irregularities in the sample when the force is increasing in plastic flow, the stress will increase locally. Whilst the force is increasing the deformation will be stable, i.e. any local constrictions are self-arresting. In contrast, under a decreasing force the deformation is no longer stable, leading to the formation and cumulative increase of necking and eventual failure. Hence the force maximum defines a point of instability in tension, beyond which fracture is inevitable. The occurrence of strain hardening (sometimes called work hardening) in a material stabilises any regions of incipient localised thinning that could lead to unstable necking and eventual fracture during high extensions, and can allow much larger extensions before rupture than would otherwise be possible.

Recent work has shown that bread doughs exhibit strain hardening in large deformations such as bubble expansion, and that these extensional rheological properties are important in baking performance (Dobraszczyk and Roberts, 1994; Dobraszczyk, 1997; van Vliet et al., 1992; Wikström and Bohlin, 1999a; Janssen et al., 1996; Dobraszczyk et al., 2003). Strain hardening allows the expanding gas cell walls to resist failure by locally increasing resistance to extension as the bubble walls become thinner, and appears to provide the bubbles with greater stability against early coalescence and better gas retention. It is expected therefore that doughs with good strain hardening characteristics should result in a finer crumb texture (e.g. smaller gas cells, thinner cell walls and an even distribution of bubble sizes) and larger baked volume than doughs with poor strain hardening properties. It has been shown that good bread-making doughs have good strain hardening properties and inflate to larger single bubble volume before rupture, whilst poor bread-making doughs inflate to lower volumes and

1121 have much lower strain hardening (Dobraszczyk and
 1122 Roberts, 1994; Dobraszczyk, 1997). Loaf volume for a
 1123 number of commercial white flour doughs has been
 1124 related directly to the failure strain and strain hardening
 1125 properties of single dough bubbles measured at elevated
 1126 temperatures in biaxial extension (Dobraszczyk et al.,
 1127 2003). Strain hardening and failure strain of cell walls
 1128 were both seen to decrease with temperature, with cell
 1129 walls in good breadmaking doughs remaining stable and
 1130 retaining their strain hardening properties to higher
 1131 temperatures (60 °C), whilst the cell walls of poor
 1132 breadmaking doughs became unstable at lower tempera-
 1133 tures (45–50 °C) and had lower strain hardening (Fig. 3).
 1134 Strain hardening measured at 50 °C gave good correla-
 1135 tions with baking volume, with the best correlations
 1136 achieved between those rheological measurements and
 1137 baking tests which used similar mixing conditions. As
 1138 predicted by the Considere failure criterion, a strain
 1139 hardening value of around 1 defines a region below
 1140 which gas cell walls become unstable, and discriminates
 1141 well between the baking quality of a range of
 1142 commercial flour blends of varying quality (Fig. 4). It
 1143 is suggested that changes in strain hardening and bubble
 1144 stability in this temperature region are important in
 1145 relation to bubble coalescence. When strain hardening
 1146 falls below a value of 1, bubble walls are no longer
 1147 stable and coalesce rapidly, resulting in loss of gas
 1148 retention and lower volume and texture. Bubble walls
 1149 with good strain hardening properties remain stable for
 1150 longer during baking, allowing the bubbles to resist
 1151 coalescence and retain gas for much longer. Strain
 1152 hardening in poorer breadmaking varieties starts to
 1153 decrease at much lower temperatures, giving earlier
 1154 bubble coalescence and release of gas, resulting in lower
 1155 loaf volumes and poorer texture (Dobraszczyk et al.,
 1156 2003).



1171 Fig. 3. Mean bubble cell wall strain hardening values ($n = 6$) for a number
 1172 of wheat varieties inflated at constant strain rate (0.1 s^{-1}) at various
 1173 temperatures on the SMS Dough Inflation System. The dashed line at strain
 1174 hardening index = 1.0 defines the region below which expanding bubble
 1175 walls become unstable. ■ = Pillsbury, ▲ = Soissons, ● = Rialto 1999,
 1176 ◆ = Hereward, ○ = Rialto 1998, □ = Charger, △ = band (From
 1177 Dobraszczyk et al. (2003)).



1177 Fig. 4. Discrimination of CBP baking performance for a range of
 1178 commercial flour blends of varying baking quality using mean bubble
 1179 cell wall strain hardening properties obtained at 50 °C and constant strain
 1180 rate (0.1 s^{-1}). (From Dobraszczyk et al. (2003)).

12. Rheological and polymer molecular structure– function relationships in breadmaking

1194 Gluten is the major protein in wheat flour doughs,
 1195 responsible for their unique viscoelastic behaviour. It is now
 1196 widely accepted that gluten proteins are responsible for
 1197 variations in baking quality, and in particular it is the
 1198 insoluble fraction of the HMW glutenin polymer, which is
 1199 best related to differences in dough strength and baking
 1200 quality amongst different wheat varieties (MacRitchie and
 1201 Lafiandra, 1997; Weegels et al., 1996). However, the exact
 1202 molecular mechanisms responsible for this variation still
 1203 remain unclear, largely because information about the
 1204 molecular size and structure of this fraction is inaccessible
 1205 by conventional polymer size characterization techniques
 1206 such as GPC and SE-HPLC due to its insolubility. Therefore
 1207 other techniques sensitive to changes in polymer MWD and
 1208 structure are necessary.

1209 One such technique is rheology, which is increasingly
 1210 being used as a sensitive indicator of changes in the
 1211 structure of HMW polymers. It has long been recognized
 1212 that the key mechanisms determining the rheology of HMW
 1213 polymer melts and concentrated solutions arise from
 1214 physical structural interactions between polymer molecules
 1215 (Doi and Edwards, 1986), and that much of the rheological
 1216 behaviour of these polymers is independent of their
 1217 chemistry. Recent work has shown that the elongational
 1218 rheology of HMW polymer melts and concentrated
 1219 solutions is a sensitive indicator of changes in secondary
 1220 molecular structure such as small increases in the size of the
 1221 highest end of the MW distribution, or the presence of long
 1222 chain branching (LCB) (Wagner et al., 2000; Münstedt et al.,
 1223 1998). The most significant property is elongational strain
 1224 hardening (the non-linear rapid increase in viscosity with
 1225 increasing strain) which is highly sensitive to the degree of
 1226 entanglement and LCB of the polymer. For example, LCB
 1227 has been shown to explain the radically different behaviour
 1228 of branched and linear polymers. Polymer melts which show

1233 no LCB, such as High Density Polyethylene (HDPE) show
 1234 no extensional strain hardening, whilst branched polymers
 1235 such as LDPE (Low Density Polyethylene), exhibit
 1236 pronounced strain hardening (Kurzbeck et al., 1999). The
 1237 presence of chain branches is important in giving rise to
 1238 strain hardening, which is a necessary property for stability
 1239 of polymers undergoing large deformation. Since HMW
 1240 subunits of gluten have been shown to have a branched
 1241 structure (Humphris et al., 2000) with branches occurring
 1242 every 40–50 nm, it is expected that this structure will result
 1243 in strain hardening in doughs, and will contribute strongly to
 1244 its resistance to extension and bubble wall stability under
 1245 large deformation.

1246 Modern polymer physical models relate the molecular
 1247 size and structure of polymers to their rheological properties
 1248 and end-use performance. The pom-pom model, recently
 1249 proposed by MacLeish and Larson (1998) describes the
 1250 rheological behaviour of HMW branched polymer melts.
 1251 This class of polymers is envisaged as a relatively flexible
 1252 HMW backbone to which are attached a number of branches
 1253 (pom-poms) radiating out from either end of the backbone.
 1254 This model accurately predicts, for the first time, the well-
 1255 known phenomenon of strain hardening under uniaxial and
 1256 planar extensional flow and strain softening (shear thinning)
 1257 in shear seen in branched HMW polymer melts (Fig. 1a) and
 1258 in gluten (Fig. 1b). The branches entangle with the
 1259 surrounding polymers and stretching of the flexible back-
 1260 bone between entanglements produces strain hardening.
 1261 Predictions of this model have shown that the number of
 1262 branches and the distance between entanglements have the
 1263 most effect on strain hardening (Fig. 9).

1264 Beyond a critical molecular weight (MW_c), character-
 1265 istic for each polymer, zero-shear viscosity (η_0) starts to
 1266 increase rapidly with increasing MW, following a
 1267 relationship $\eta_0 = MW^{3.4}$ for linear polymer melts,
 1268 independent of polymer chemistry. Above this critical
 1269 MW, the polymers start to entangle, giving rise to the
 1270 observed rapid increase in viscosity with MW (Fig. 5).
 1271 Branching has the effect of increasing viscosity and
 1272 strain hardening still further, although the rapid increase
 1273 in viscosity is shifted to higher MW with increasing
 1274 branching, giving an effective decrease in viscosity at a
 1275 constant MW. Entanglements also give rise to the steep
 1276 increase in stiffness and viscosity with strain known as
 1277 strain hardening. Entanglements can be viewed as
 1278 physical constraints between segments of the polymer
 1279 chain, rather like knots, where the polymer chains are
 1280 stuck and are not free to move past each other (Fig. 6).
 1281 As can be seen in Fig. 5, a relatively small variation in
 1282 the highest end of the MWD or branching can give rise
 1283 to a large increase in viscosity and strain hardening, and
 1284 is likely to have a large effect on baking performance.
 1285 Molecular entanglements of HMW polymers under large
 1286 deformation are related to changes in their relaxation
 1287 spectrum and extensional strain hardening, which reflect

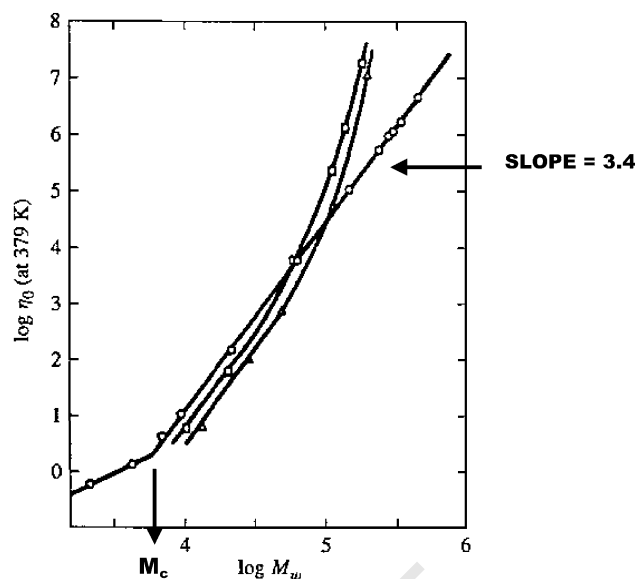


Fig. 5. Effect of MW and branching on zero-shear viscosity for polymer melts. Beyond a critical molecular weight for entanglements (M_c) zero-shear viscosity (η_0) increases rapidly for linear polymer (O) as $MW^{3.4}$. (\square) = 3 armed branched polymer (linear with one long side chain), (Δ) = 4-armed branched polymer. Source: rheological properties of multichain polybutadienes, Kraus and Gruber (1965) Journal of Polymer Science, John Wiley & Sons.

the expected qualitative differences in the underlying MWD of glutenin polymers.

Dynamic shear properties such as G' and relaxation time are sensitive to MW and entanglements. Lefebvre et al. (2000) have shown that the height of the plateau modulus increased with the proportion of the highest MW fractions extracted from heated gluten. Dynamic oscillation measurements are usually made over a relatively narrow window of frequencies, which are not relevant to the strain rates observed during fermentation and baking, and which do not reflect the intermolecular interactions of the HMW glutenin polymers known to be responsible for baking performance. The frequency range of most conventional oscillatory shear rheological tests on doughs is limited to a part of the plateau range, at which the HMW polymers are entangled and are

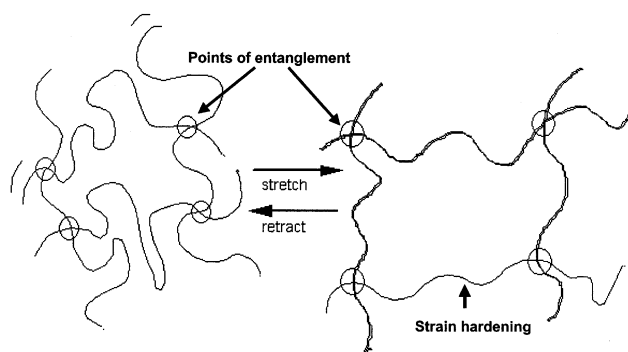


Fig. 6. Model of entanglement network in a high MW polymer during stretching.

1345 insensitive to changes in the MWD (Graessley, 1974,
 1346 1982). Therefore, it is necessary to extend these
 1347 measurements into the low frequency range or the
 1348 terminal zone, which is known to reflect the disentangle-
 1349 ment of the HMW polymers. Because of the extreme
 1350 polydispersity in MW of the gluten, it is thought
 1351 necessary to separate out fractions of varying solubility
 1352 and MW from the gluten (gliadins, soluble glutenins and
 1353 insoluble glutenins) and then to seek to use additive laws
 1354 in a recombination of their properties (Tsiami et al.,
 1355 1997). Fig. 7 shows the effect of increasing MW on the
 1356 dynamic shear modulus for a narrow MW linear
 1357 polystyrene polymer melt. As the MW increases, a
 1358 plateau in modulus begins to appear, which increases in
 1359 width as the MW increases further. The plateau
 1360 represents the effect of entanglements, which at a certain
 1361 polymer size effectively lock the polymer structure into a
 1362 temporary 3D network with a fixed modulus, the height
 1363 of which is independent of MW. At some frequency the
 1364 polymer network begins to disentangle, and the modulus
 1365 starts to decrease rapidly into the terminal zone, where
 1366 the polymer chains are free to move about and act as a
 1367 viscous liquid. The larger the polymer, the longer it stays
 1368 entangled, and therefore the wider the frequency range
 1369 over which the plateau remains. Thus it is the width of
 1370 the plateau, or the point at which it descends into the
 1371 terminal zone, which is defined by the MW of the
 1372 polymer. Unfortunately, most rheological measurements
 1373 on dough and gluten have been performed in the plateau
 1374 region, which is the region most insensitive to differences
 1375 in MW. For example, Fig. 8a shows the relationship
 1376 between the wt% fraction of the various polymer MW
 1377 size fractions calculated from the total MWD of gluten
 1378 up to values $>10^8$ (measured by flow-field flow
 1379 fractionation and light scattering (Stathopoulos, 2003)),

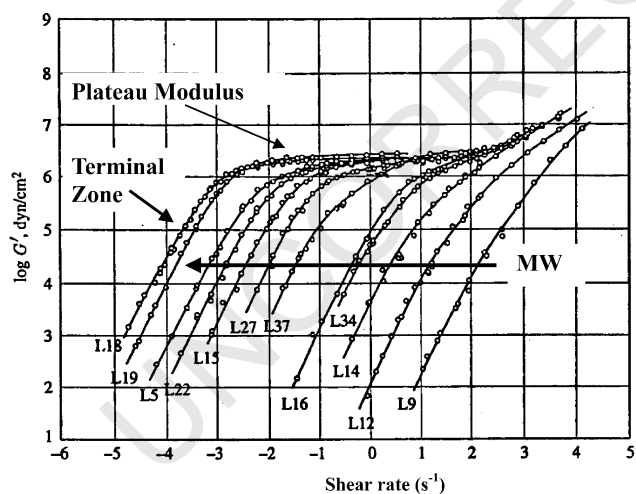


Fig. 7. Effect of increasing MW on dynamic shear modulus for a series narrow MW linear polystyrene polymer melts. (From: Onogi, Masuda, and Kitagawa (1970) *Macromolecules* 3: 109–116.)

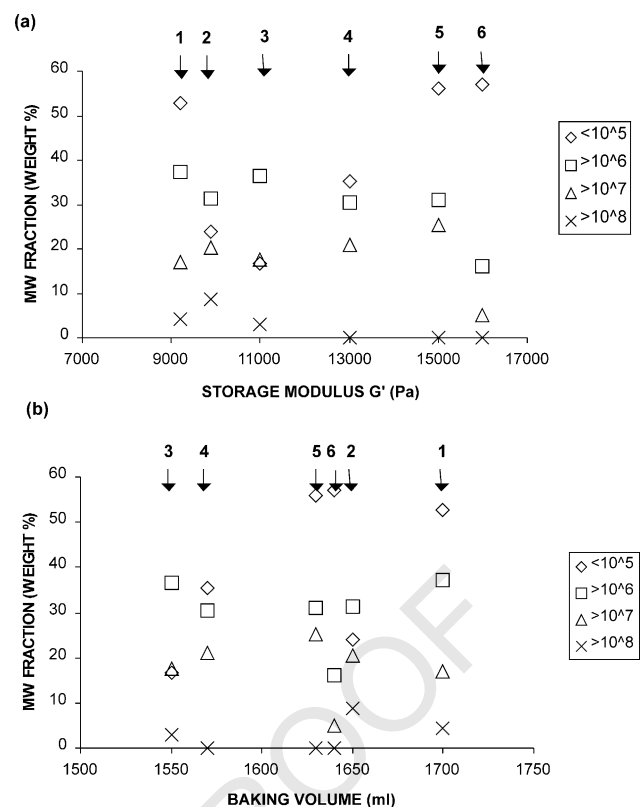


Fig. 8. (a) Relationship between molecular weight fractions and plateau storage modulus (G' at 1 Hz). Molecular weight expressed as wt% of fractions greater than certain molecular sizes calculated from the total molecular weight distributions for glutes obtained from five UK wheat varieties and one US commercial flour: 1 = Hereward 2 = Pillsbury 3 = Riband 4 = Soissons 5 = Charger 6 = Rialto. MW data obtained by light scattering from Stathopoulos (2003)). (b) Relationship between molecular weight fractions from (a) and CBP baking volume for a number of UK wheat varieties and one US commercial flour: 1 = Hereward 2 = Pillsbury 3 = Riband 4 = Soissons 5 = Charger 6 = Rialto.

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plateau storage modulus (G' at 1 Hz) of gluten and baking quality (Fig. 8b) from a number of wheat varieties of varying baking performance. These show that neither plateau modulus nor baking volume is related to MW up to a size of about 5×10^8 . This confirms the observation that plateau modulus is essentially independent of MW and also that it is not the size of the soluble glutenin polymers, but the insoluble polymer fraction that is mainly responsible for baking performance. This does not support the commonly held view that the MW of the glutenin polymers is related to their small deformation shear rheology and to baking quality, but that that is more likely to be the secondary molecular structure of the insoluble glutenin that is responsible for baking performance. Recent evidence suggests that these insoluble HMW polymers are entangled with a corresponding long relaxation time (Fig. 9) (Li et al., 2003), they are branched, and form extensive intermolecular secondary structures held together by covalent and hydrogen bonding (Belton, 1999; Popineau et al., 1994; Feeney et al., 2003).

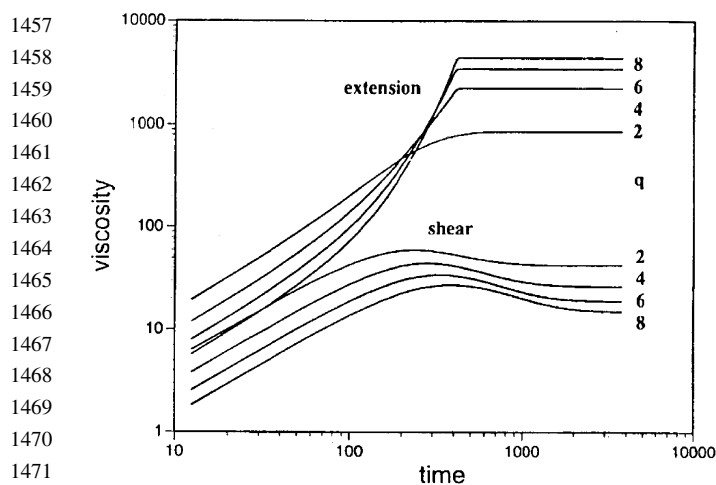


Fig. 9. Modelling the effect of polymer chain branching on shear and extensional rheology. (q = no. of branch points). From: MacLeish and Larson (1998).

13. Conclusions

It would appear that for HMW polymers such as gluten, large deformation extensional rheological properties are more sensitive to changes in MWD, polymer entanglements and branching than small deformation dynamic shear properties, based on sound polymer physics principles and experimental data. Insoluble HMW glutenins have been shown to be best related to variations in baking quality, and to the presence of long relaxation times, indicating entanglements of the HMW polymers. Strain hardening, which has been shown to be a sensitive indicator of entanglements and long-chain branching in HMW polymers, is seen in large extensional deformation of doughs and glutes, and is well related to bubble wall stability, long relaxation times and to variations in baking performance amongst different wheat varieties.

15. Uncited References

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