

Dynamic rheological properties of wheat flour dough and proteins

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Dynamic rheological testing has become a powerful and preferred approach for examining the structure and the fundamental properties of wheat flour doughs and proteins because of its characteristic and sensitive response to the structure variation of wheat flour doughs and proteins. In the present article, the dynamic rheological properties of wheat proteins and flour doughs were reviewed. Influences of various additives on the rheological properties of gluten proteins and flour doughs are illustrated and the component interactions are emphasized. Moreover, theoretical analyses concerning the relationship between rheological behavior and structure are summarized.

Introduction

Among the cereal flours, only wheat flour can form a three-dimensional viscoelastic dough when mixed with water. Characterization of rheological properties of dough is effective in predicting the processing behavior and in controlling the quality of food products. Farinograph, mixograph, and extensograph are the most common empirical instruments used for characterizing dough rheology. Tests based on these instruments are useful for providing practical information for the baking industries while they are not sufficient for interpreting the fundamental behavior of dough processing and baking quality. Rheological

testing, especially in the linear viscoelastic region, has been used to follow the structure and properties of doughs and to study the functions of dough ingredients (Janssen, van Vliet, & Vereijken, 1996a; Miller & Hosene, 1999). This testing simultaneously measures the viscous and elastic characters of dough expressed in storage and loss moduli, G' and G'' , and loss tangent $\tan \delta$. It is generally found that doughs made from good quality flour have $\tan \delta$ values lower than doughs made from poor quality flour. The magnitude of modulus at intermediate and high strains is in the order of extra strong > strong > medium > weak (Safari-Ardi & Phan-Thien, 1998). Nevertheless, dynamic rheological tests on flour dough fail to predict the baking potential of wheat cultivars (Autio, Flander, Kinnunen, & Heinonen, 2001).

Gluten is rich in gliadins and glutenins but also contains 3.5–6.8% lipids, 0.5–0.9% minerals, and 7.0–16.0% carbohydrates. Dynamic rheological parameters of glutes are able to indicate the wheat quality. Glutes from poor quality wheat are rheologically characterized as less elastic and more viscous than those from good quality wheat (Khatkar, Bell, & Schofield, 1995). Glutes from good breadmaking wheat are crosslinked in a higher degree so that the frequency dependence of G' is smaller than that of glutes from poor breadmaking wheat (Janssen, van Vliet, & Vereijken, 1996b). G' and G'' of glutes show significant positive correlations with loaf volume (Khatkar, Fido, Tatham, & Schofield, 2002). Especially, G' of gluten doughs can be directly related to the breadmaking performance, explaining 73% of variation in loaf volume (Khatkar & Schofield, 2002a) (Fig. 1). The $\tan \delta$ values of glutes are ranked as weak glutes > strong glutes > extra strong glutes while the G' and G'' values show the reverse tendency. The weak glutes especially undergo a substantial structural change from solid-like to liquid-like behaviors with increasing frequency while the strong glutes maintain their elastic characters to a great extent (Khatkar, 2004a).

In this review, we focus our attention on dynamic rheological characteristics of wheat proteins and flour doughs. The structures and rheological behaviors of wheat proteins including gluten and gliadin- and glutenin-rich fractions are outlined. Moreover, influences of water, starch, proteins and the other constituents or additives on the rheological behaviors of flour dough are discussed. Theoretical analyses on the dynamic rheological behaviors of gluten and flour doughs are also presented.

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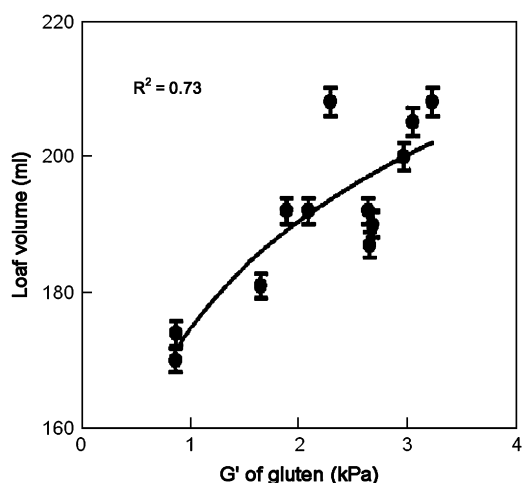


Fig. 1. Relationship between G' of gluten (stress 25 Pa, frequency 1 Hz) and loaf volume for flours reconstituted using glutes of different wheat cultivars and a constant source of starch and water-solubles (Khatkar & Schofield, 2002a).

Structures and rheological behavior of proteins

Molecular structure of proteins

Gliadins and glutenins, accounting for 80–90% of the total wheat flour proteins, are the two primary classes of storage proteins, being necessary for producing an appropriate balance of viscous and elastic properties in gluten and flour doughs. It is widely accepted that gliadins confer viscous properties whilst glutenins impart strength and elasticity (Shewry, Tatham, Forde, Kreis, & Mifflin, 1986). Gliadins with a molecular weight of 30–80 kDa are monomeric proteins and can be further classified into four groups of α , β , γ and ω . Glutenins are polymeric proteins with a molecular weight of up to 11 million and are composed of low molecular weight glutenin subunits (LMW-GS) and high molecular weight glutenin subunits (HMW-GS) of 12–60 kDa and 60–120 kDa, respectively.

Hydration gives rise to the formation of an apparent continuous water phase between gluten particles. Hydrated proteins form β -sheet structures that contribute to the network connectivity. The interactions between subunits in glutenins and aggregates involve intermolecular β -sheets located in their repetitive domains or in the region of chain entanglements. Entanglements, hydrogen bonding, hydrophobic interactions and disulfide linkages are important for hydrated gluten (Khatkar, 2004b).

Gluten

The viscoelasticities of glutes are related to glutenins, gliadin/glutenin ratio and HMW-GS/LMW-GS ratio (Popineau, Cornec, Lefebvre, & Marchylo, 1994). HMW-GS and subunit composition influence the viscoelasticity by modifying the size distribution and the protein aggregation through crosslinking. The glutenin aggregation leads to a significant rise in elastic plateau modulus G_N^0 of the network (Cornec, Popineau, & Lefebvre, 1994; Popineau *et al.*, 1994).

Gliadins might act as plasticizers in gluten so that increasing the gliadin/glutenin ratio causes a decrease in elasticity (Popineau *et al.*, 1994). It has recently been found that addition of α -, β -, γ - and ω_2 -gliadins containing cysteine residues to gluten causes unexpected increases G' and G'' (Khatkar *et al.*, 2002) (Fig. 2).

The viscoelasticity of gluten network depends on intermolecular interactions. Under mechanical actions, gradual destruction of intermolecular interactions causes the non-linear rheological behaviors above a critical strain value. The critical strain of gluten doughs at 5–10% is much larger than that of flour doughs at $\sim 0.2\%$ (Wang & Kokini, 1995a).

Thermal treatment at 20–40 °C does not induce irreversible changes in mechanical properties. At higher temperatures, however, rheological behavior changes irreversibly due to sulphhydryl/disulfide (SH/SS) exchange. The baking functionality decreases during heating with marked decreases in solubility and extractability. For the gluten fraction rich in HMW-GS, G' and G'' show a slight frequency dependence and heat treatment does not influence the rheological behavior significantly. The gluten fraction rich in LMW-GS behaves viscously before heating while heat treatment might completely change it into an elastic material (Stathopoulos, Tsiami, Dobraszczuk, & Schofield, 2006).

Gliadins and glutenins

Gliadin from the good quality flour show higher G' and lower $\tan \delta$ values than those from the poor quality flour (Toufeili *et al.*, 1999). Hydrated gliadins form a weak and highly viscous dough (Khatkar *et al.*, 1995) that shows a networking reaction at 70–115 °C (Madeka & Kokini, 1994) and does not behave as a simple viscoelastic liquid.

Glutenins from wheats with different breadmaking quality behave quite differently. Glutenins from wheats with good breadmaking quality have an elastic character greater

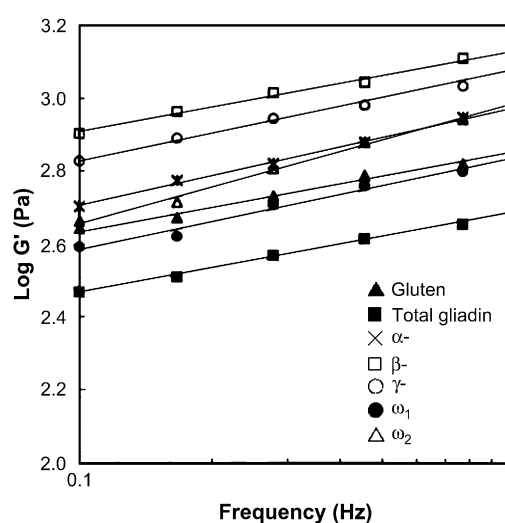


Fig. 2. Effects of addition of different gliadin subgroups on the frequency dependence of G' for cv. Hereward gluten at stress amplitude 25 Pa (Khatkar *et al.*, 2002).

than those from wheats with poor breadmaking quality (Khatkar *et al.*, 1995). Glutenin viscoelasticity is determined primarily by the interactions of large concatenations formed by HMW-GS and LMW-GS (Cornec *et al.*, 1994). HMW-GS is particularly important in influencing the breadmaking quality. The HMW-GS fractions from extra strong wheat have a higher G' value and a lower $\tan \delta$ value as compared to strong and weak wheats (Jood, Schofield, Tsiami, & Bollecker, 2000). The proportion of HMW-GS in relation to LMW-GS is also important in balancing the viscous and elastic properties. Changes in the size distribution of polymeric protein could account for all differences in dough strength.

Rheological functions of HMW-GS fractions are typical in the plateau region with long relaxation times so that they promote the network formation (Tsiami, Bot, Agterof, & Groot, 1997). The rheological spectra of LMW-GS usually represent the characteristic in the transition zone. LMW-GS fractions give rise to a plasticizing effect as indicated by their narrowing of the width of the plateau region of the HMW-GS/LMW-GS mixtures.

The 1.5% sodium dodecyl sulphate (SDS) unextractable subfraction, called glutenin macropolymer (GMP) of 80–120 kDa, is highly related to quality parameters such as loaf volume and dough properties. GMP consists of both HMW-GS and LMW-GS that assembly through disulphide bonds to form an elastic gel of particles of 5–50 μm diameter (Don, Lichtendonk, Plijter, & Hamer, 2003a). G_N^0 of the GMP gel is positively correlated to the particle size in the gel (Don, Lichtendonk, Plijter, & Hamer, 2003b; Don, Lookhart, Naeem, MacRitchie, & Hamer, 2005).

Influence of other constituents and additives

Pentosans could limit the aggregation of glutenins by covalently linking to glutenins or by competing for water. Water-soluble pentosans (WSP) are able to form a highly viscous gel under oxidative conditions. During the wet separation process, addition of WSP has a negative effect on the gluten yield but makes the resultant GMP gel be more elastic (Wang, van Vliet, & Hamer, 2004). Addition of WSP in gluten dough reinforces the network and accelerates the dissipative processes caused by structural rearrangements (Santos, Monteiro, & da Silva, 2005). Addition of water unextractable arabinoxylans to doughs increases the specific volume of GMP particles and the protein concentration in the gel (Wang *et al.*, 2003; Wang, van Vliet, & Hamer, 2005). The GMP gel becomes more elastic and stiff.

Transglutaminase catalyzes the formation of large insoluble polymers, which makes G' and G'' of gluten gels to increase 10–50 times and 2–4 times, respectively (Larre, Deshayes, Lefebvre, & Popineau, 1998). The treated gluten remains as a transient network, but its viscoelastic response is shifted toward short times (Larre *et al.*, 2000) and the dynamic moduli become less frequency dependent (Larre *et al.*, 1998). Enzyme treatment can also influence the interaction between pentosan and gluten fractions (Wang *et al.*,

2003, 2005). Xylanase, pentosanase, and laccase can weaken the GMP gel while glucoseoxidase treatment increases the content of pentosans associated with GMP thus improving the elasticity.

Lipids play an important role in the affinity of proteins to water presumably due to interaction with amino acid residuals. Defatting of gluten improves water uptake and increases crosslinking degree between proteins (Papantoniou, Hammond, Scriven, Gordon, & Schofield, 2004). Addition of surfactants increases G' of gluten dough due to increased hydrophobic interactions (Toufeili & Kokini, 2004).

The structures of non-prolamins and their contributions to rheological behaviors are not yet clear and the experimental observations might be in contradiction. A kind of low molecular weight protein rich in cysteine can decrease the rate of GMP repolymerization during resting of dough. Addition of large amounts of low molecular weight proteins increases the viscous characteristics (Weegels *et al.*, 1995). Extracting non-prolamin proteins does not alter the viscoelastic properties of gluten markedly (Hargreaves, Popineau, Marion, Lefebvre, & Le Meste, 1995). When 1.3% water-solubles are removed, gluten dough becomes more elastic and less viscous than the control gluten (Dreese & Hosney, 1990).

Rheological behavior of flour dough

Influence of water

Dough is a macroscopically homogeneous mixture of starch, protein, fat, salt, yeast, and other components. At optimum mixing, the dough is fully hydrated and has the highest elasticity. Water plays an important role in determining the viscoelastic properties of dough. Both G' and G'' decrease as water content increases. The dynamic viscoelastic behavior of flour doughs can be understood by taking into account the dual role of water that behaves as an inert filler reducing the dynamic properties proportionally and as a lubricant enhancing the relaxation (Masi, Cavella, & Sepe, 1998).

Influence of starch

Starch, making up ~80% of wheat flour on dry basis, is able to form a continuous network of particles together with the macromolecular network of hydrated gluten. These two independent networks and their interaction give rise to the rheological properties of doughs. Though the interaction plays an important role, the relative contributions of the two sources are difficult to resolve. The component interactions depend on stress level. The starch–starch interactions dominate over protein–protein interactions at low stresses while the protein–protein interactions play a dominant role at large deformations (Khatkar & Schofield, 2002b). The nonlinear rheological behavior of starch is largely responsible for the behavior of dough (Watanabe, Larsson, & Eliasson, 2002).

In starch/gluten blend with constant water content, G' increases rapidly with increasing protein content. The reconstituted doughs behave qualitatively like flour doughs with comparable compositions. When starch granules are apparently homogeneously dispersed in the gluten network,

increasing starch content gives rise to an increase in G' value (Watanabe *et al.*, 2002) thus enhancing the elasticity (Edwards, Dexter, & Scanlon, 2002). Flour doughs cannot be viewed simply as a concentrated suspension of starch granules in hydrated gluten matrix. Mixing starches from different wheat cultivars into dough with constant gluten content leads to large rheological differences, indicating an active role of starch (Petrofsky & Hosene, 1995).

Influence of proteins

The protein content of flours shows an inverse relationship with G' and G'' up to $\sim 14\%$ protein (Khatkar, 2005). Gluten contributes to the viscoelastic properties of dough to varying degrees depending on its source differing with both gliadin/glutenin ratio and LMW-GS (Edwards, Dexter, & Scanlon, 2001; Edwards, Mulvaney, Scanlon, & Dexter, 2003). Gliadin enhances viscous flow of dough. An addition of 2% gliadin results in increased dough extensibility and $\tan \delta$ as compared to gluten and glutenin additions. Glutenin addition, on the other hand, results in a more elastic dough in comparison with gluten and gliadin additions (Edwards *et al.*, 2001) (Fig. 3). Addition of glutenins at constant protein basis contributes to the dough strength with marked differences among donor cultivars (Edwards *et al.*, 2003). Increasing the glutenin/gliadin ratio improves maximum shear viscosity and dough strength (Uthayakumaran, Newberry, Keentok, Stoddard, & Bekes, 2000).

Both LMW-GS and HMW-GS contribute to overall dough strength but LMW-GS enrichment improves the elasticity by introducing greater number of physical crosslinks (Edwards *et al.*, 2001). The source of LMW-GS influences the viscoelastic characteristics of doughs while source of HMW-GS does not show such an effect (Edwards *et al.*, 2003).

Influence of other additives

Addition of carbohydrates such as arabinoxylans, β -glucans (Izydorczyk, Hussain, & MacGregor, 2001),

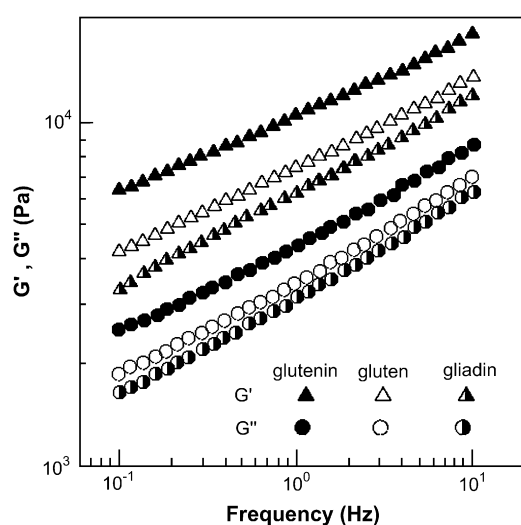


Fig. 3. Dynamic responses of durum dough enriched with 2% gluten, gliadin and glutenin, respectively (Edwards *et al.*, 2001).

carrageenan, alginate (Howell, Bristow, Copeland, & Friedli, 1998) and guar gum (Yu & Ngadi, 2006) improve the functional properties of wheat bread through associative interactions with gluten proteins that significantly increases G' of doughs at the same water content.

Defatting improves protein interaction thus increases G' and G'' significantly (Georgopoulos, Larsson, & Eliasson, 2006). Addition of nonpolar lipids to the defatted flour at their natural level might partially restore the rheological behavior while higher levels of addition have no further effect. On the other hand, addition of polar lipids has a more pronounced beneficial effect (Papantoniou *et al.*, 2004). Addition of water-solubles dramatically shortens the optimum mixing time of the reconstituted flour and decreases G' of the resultant dough (Miller & Hosene, 1999).

Added solid fat extends the strain limit where the network structure of dough begins to break down. Fat can reduce the number of effective crosslinks in gluten (Fu, Mulvaney, & Cohen, 1997; Watanabe *et al.*, 2002). However, addition of nonpolar lipid oil facilitates the aggregation of gluten and gives rise to a more elastic behavior of doughs (Watanabe, Yokomizo, & Eliasson, 2003).

Reducing agents such as glutathione or L-cysteine reduce the size of large proteins by promoting the formation of free SH groups and therefore have a great influence on the dough rheology (Lambert & Kokini, 2001; Yeh & Shiau, 1999). Increasing L-cysteine concentration in the range of 0–0.75% gradually reduces the effective degree of crosslinking in gluten (Lambert & Kokini, 2001). Addition of L-cysteine also has a dramatic effect on the relaxation spectrum and significantly decreases the relaxation modulus (Lee & Mulvaney, 2003).

Oxidation by oxidants or oxidases makes the doughs to be less sticky and more strong due to the oxidative gelation of the water-soluble pentosans or oxidation of SH groups into SS bonds (Yeh & Shiau, 1999). Peroxidase can introduce a secondary arabinoxylan network through the gluten network while glucose oxidase produces permanent crosslinks that strengthens the gluten network (Dunnewind, van Vliet, & Orsel, 2002).

Supplementation of polysaccharide hydrolyzing enzymes (Dogan, 2002) or protease (Pedersen, Kaack, Bergsøe, & Adler-Nissen, 2005) results in softening and weakening of doughs. Transglutaminase treatment can counteract the hydrolyzing effect of bug proteases (Caballero, Bonet, Rosell, & Gomez, 2005).

The SH/SS interchange reaction is pH-dependent so that edible acids or alkalis could modify the rheological and the processing properties. Doughs containing acids are initially firmer and more viscous but show less stability during mixing (Wehrle, Grau, & Arendt, 1997). Increasing pH induces a decrease in $\tan \delta$ accompanied with increases in G' and apparent viscosity (Shiau & Yeh, 2001).

Yeast fermentation makes doughs more elastic accompanied by an increase in unextractable polymeric proteins (Liao, Miller, & Hosene, 1998). The gas bubble formation

within the dough interrupts the increasingly crosslinked protein network, resulting in rheological weakness (Newberry, Phan-Thien, Larroque, Tanner, & Larsen, 2002). On the other hand, addition of sourdough leads to a less elastic and firm dough because sourdough fermentation results in solubilization and depolymerization of GMP (Angioloni, Romani, Pinnavaia, & Rosa, 2006). The rheological alterations during fermentation are mainly due to amount of carbon dioxide produced (Wehrle & Arendt, 1998).

Theoretical analyses

Wheat flour doughs and proteins are characterized by complex interactions at varying scales between the components. Several theoretical models have been examined or constructed in order to elucidate the structures and the nonlinear rheological properties. As depicted from the transient network model, the molecular weight of network strands ranges from 40 kg/mol to 150 kg/mol in the gluten/glycerol mixture, which is about 1–3 times of that of glutenin subunit (Redl, Morel, Bonicel, Vergnes, & Guilbert, 1999). Mixing at 40–80 °C increases the amount of SDS-insoluble protein with time dramatically, which is accompanied by a continuous increase of G_N^0 (Redl, Guilbert, & Morel, 2003). The equilibrium shear modulus and swelling of gluten network can be well correlated to the percentage of aggregated proteins (Domenek, Brendel, Morel, & Guilbert, 2004). The gluten network could not be modeled with the Flory–Rehner rubber elasticity of an entangled network. The very strong dependence of modulus on gluten content indicates a network of mesoscale particles with a fractal dimension of ~ 2.93 . Physical interactions at mesoscale of 0.1–100 μm affect the dough properties (Lefebvre, Popineau, Deshayes, & Lavenant, 2000). Failure of TTS in gluten/glycerol mixture indicates a chemical reaction in gluten that is not thermorheologically simple (Redl, Morel, Bonicel, Guilbert, & Vergnes, 1999).

Frequency or shear rate dependences of steady viscosity of gluten and flour doughs might be molded by the Bird–Carreau model assuming that the temporary network junctions are destroyed during deformation (Wang & Kokini, 1995b).

During all stages of food making, doughs undergo large deformations primarily dominated by the gluten fraction (Uthayakumaran, Newberry, Phan-Tien, & Tanner, 2002). The nonlinear stress–strain relationships in both uniaxial and biaxial extensions of hydrated gluten show higher strain hardening than flour dough (Sliwinski, van der Hoef, Kolster, & van Vliet, 2004). Under constant strain rate, the nonsteady-state flow exhibiting strain softening and partial failure reveals a hyper elastic contribution from the elastic gluten network and a viscoelastic contribution from the suspension of starch globules and other long chain components that do not participate in the network (Phan-Thien, Safari-Ardi, & Morales-Patino, 1997).

Stress relaxation of flour doughs reveals at least two classes of entanglements (Fu *et al.*, 1997; Safari-Ardi & Phan-Thien, 1998). In gluten fractions, gliadin and soluble glutenin show one relaxation process (Lee & Mulvaney, 2003; Li, Dobraszczyk, & Schofield, 2003). HMW-GS possesses two relaxation processes and the slow one is related to the network structure. The linear viscoelastic response of gluten or flour doughs (Fig. 4a and b) over a very broad time scale can be obtained by combining data from dynamic and retardation tests (Lefebvre, 2006; Lefebvre *et al.*, 2003). Analyses based on theoretical models reveal that the rheological behaviors of gluten are related to the transient network mainly constructed by HMW-GS while starch and the other constituents additionally contribute to the nonlinear rheological behaviors of dough.

Conclusions

It can be concluded from the available literature that the dynamic rheological technique of frequency sweep under

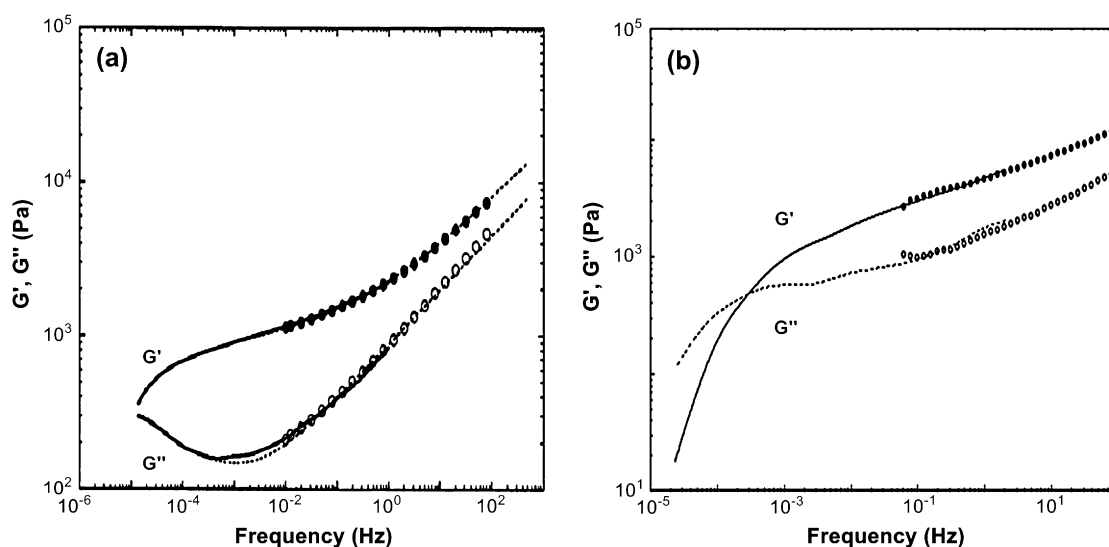


Fig. 4. Linear viscoelastic spectrum of gluten (a) and flour doughs (b) combining dynamic measurements (symbols) and retardation results (Lefebvre, 2006; Lefebvre, Pruska-Kedzior, Kedzior, Lavenant, 2003).

small deformations is highly promising for elucidating the structure of wheat proteins and the processibility of wheat flour dough. These studies demonstrate that the component interactions are fairly important for determining the rheological behaviors of gluten and flour doughs. The quality of food products based on wheat flour can be improved by enrichment of gluten, especially the glutenin- or the GMP-rich fractions. Food additives such as carbohydrates, nonpolar lipid oil, oxidants or oxidases, and transglutaminase can make the dough more elastic and strong by facilitating the aggregation of gluten proteins or the associative interactions between proteins and pentosans. On the other hand, reducing agents such as glutathione or L-cysteine significantly reduce the size of large proteins, which results in softening and weakening of doughs. Therefore, controlling the components of the gluten fraction and the structure of the gluten network is essential for improving the processibility of wheat flour dough and the quality of food products. The contributions of various additives can be readily realized from dynamic rheological parameters and their dependences on frequency, strain or stress.

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