Stress Concentration Analysis in A Heterogeneous Dough Gas Cell Wall at The Microscale: the effect of gluten/starch interaction and rheological moduli ratio

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Abstract

The unidirectional extension of a bread dough gas cell wall (GCW) composed of a single starch granule surrounded by gluten was considered in order to analyse where stress concentrated the most. The impact of gluten/starch interactions (cohesion or non-cohesion) and rheological moduli ratio on stress concentration was numerically assessed in 2D and 3D using the linear and nonlinear Structural Materials Modules available in COMSOL Multiphysics. A first **linear** viscoelastic **1-element generalized Maxwell** (1-EGM) model which is theoretically suited for infinitesimal strains (<<0.1) and small strain rates was performed. A second **1-EGM hyper-elastic** approach was performed to account for **finite strain** (large strain > 1). The two approaches were compared. The average of the Von Mises stress throughout the GCW was found to be equal using both approaches up to 0.08 strain. The Gluten/starch interactions were also modelled using the pair **Thin Elastic layer** (TEL) boundary condition to account for the gluten/starch interface without modelling a dedicated domain.

Keywords: baking, rupture, viscoelasticity, hyperelasticity, modelling.

I. Introduction

Bread dough is often described as a dispersion of gas cells in a continuous hydrated gluten/starch matrix [1]. The hydrated gluten/starch matrix located between two adjacent bubbles is called a gas cell wall (GCW) (Figure 1). The crumb structure (specific volume and porosity) of bread greatly depends on when GCWs rupture during baking [2-4]. Crumb collapse with large cells is obtained for early gas cell wall ruptures (20-50°C). Shrinkage due to the lowering in pressure at the cooling stage (just after baking) occurs when a few or even none gas cell walls rupture. Rupture of GCWs at intermediate temperatures lead to a well-distributed cell crumb structure (55-65°C).

Attempts to numerically model dough rupture are usually not performed at the GCW scale and often considered dough as continuous [3, 5, 6]. At the end of proofing and during baking, part of the thinnest GCWs is reduced to about the size of a starch granule [7-9]. When such size is reached gluten and starch granules must be considered as interacting phases in order to account for heterogeneities and appropriately describe stress concentration and points where cell walls are most likely to rupture [3, 7, 10]. The viscoelasticity of bread dough is strongly related to that of starch and gluten [11-14]. Gluten and starch are two chemically incompatible materials and should not interact very much between each other [15]. However, the existence of some interactions at the gluten/starch interface has been reported and indirectly evidenced by some authors [12, 16-18]. To our best of knowledge, the most relevant paper that accounted for gluten/starch interactions at the microscale (bonding or de-bonding) only dealt with their impact on the overall mechanical behaviour of dough but did not focused on stress concentration analysis at the starch/granule interface [18].

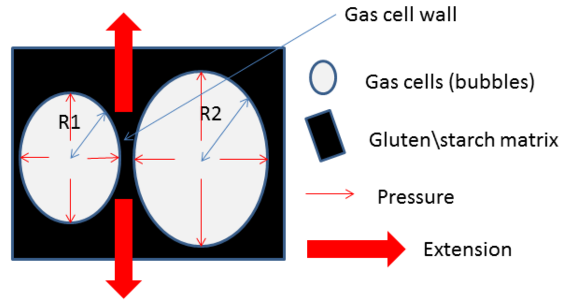

Figure 1: Gas Cell Wall (GCW) between two gas cells.

In this paper, a first analysis of the impact of starch/gluten interactions on stress concentration in GCW was proposed. An analysis of the impact of the gluten/starch elastic modulus ratio on the point where stress concentrated was proposed. Considering gluten more rigid than starch and vice versa allowed us to highlight the possible effect of starch gelatinization and gluten coagulation on the spot where stress possibly concentrated.

Those two analyses were carried out in mechanical conditions close to those actually encountered in bread dough during baking.

The strain rate involved during baking is quite low (of the order of 0.003 s^{-1}). The strain can however reach great values (of the order of 3.2, see Table. 1) and finite strain should be considered too. The strain in GCW between two growing gas cells for each step of bread processing is presented in Table. 1.

Table. 1. Strain range within gas cell wall at different stages of breadmaking.

Baking stage	Start of mixing	End of mixing	End of fermentation	End of baking
Gas cell diameter (μm)	15	50	120	500
Strain	0	2.3	1.4	3.2

To account for all the ranges of strain rate and strain encountered during baking both linear viscoelasticity and visco-hyperelasticity were considered and compared.

II. Materials and Methods

II.1 Governing Equations

Linear viscoelasticity

The equation of the conservation of momentum for quasi static problem was considered (eq. 1).

$$\nabla . \boldsymbol{S} = 0 \tag{1}$$

The total stress tensor $\boldsymbol{S}$ was split up into a purely elastic part and a viscoelastic part (eq. 2).

$$\boldsymbol{S} = \boldsymbol{\sigma} + \boldsymbol{\tau} \tag{2}$$

Where $\boldsymbol{\sigma}$ is tensor which describes the time-independent elastic behaviour and $\boldsymbol{\tau}$ the viscoelastic stress tensor. The elastic stress tensor $\boldsymbol{\sigma}$ writes (eq. 3).

$$\boldsymbol{\sigma} = \boldsymbol{C} : \boldsymbol{\varepsilon} \tag{3}$$

Where $\boldsymbol{C}$ is the fourth-order tensor of rigidity and $\boldsymbol{\varepsilon}$ is the linear strain tensor (eq. 4).

$$\boldsymbol{\varepsilon} = \boldsymbol{\nabla u} + (\boldsymbol{\nabla u})^{\boldsymbol{T}} \tag{4}$$

Where $\boldsymbol{u}$ is the displacement.

The viscoelastic stress tensor $\boldsymbol{\tau}$ derives from the constitutive equation for 1-element generalized Maxwell model (eq. 5).

$$\boldsymbol{\tau} + \lambda_r \dot{\boldsymbol{\tau}} = \lambda_r G \dot{\boldsymbol{\gamma}} \tag{5}$$

Where λ_r is the time of relaxation (gluten or starch), $\boldsymbol{\gamma}$ is the infinitesimal shear strain tensor and G the shear elastic modulus (gluten or starch).

Visco-hyperelasticity

The equation of conservation for hyperelasticity is (eq. 6).

$$\nabla . (\boldsymbol{FS})^T = 0 \tag{6}$$

Where **S**, the second Piola-Kirchofft stress tensor derives from the strain energy density function (W) (eq. 8) and **F**, the deformation gradient, is equal to $\boldsymbol{I} + \nabla \boldsymbol{u}$ where **I** is the identity tensor and $\boldsymbol{u}$ is the displacement.

$$\mathbf{S} = 2\frac{\partial W}{\partial \boldsymbol{C_c}} = \boldsymbol{S}_{vol}^{\infty} + \boldsymbol{S}_{iso}^{\infty} + \boldsymbol{Q} \tag{7}$$

Where $\boldsymbol{C_c}$ is the right green Cauchy tensor and equal to $\boldsymbol{F^T F}$, and **Q** stand for the auxiliary stress tensor from thermodynamic consideration.

$$\boldsymbol{S}_{el}^{\infty} = \boldsymbol{S}_{vol}^{\infty} + \boldsymbol{S}_{iso}^{\infty} = \det(\boldsymbol{F})^{-1} \boldsymbol{F}(\boldsymbol{C} : \boldsymbol{\varepsilon}) \boldsymbol{F^T} \tag{8}$$

$\boldsymbol{S}_{el}^{\infty}$, the elastic stress tensor was split into a volumetric and isochoric part, $\boldsymbol{S}_{vol}^{\infty}$ and $\boldsymbol{S}_{iso}^{\infty}$ [19]. $\boldsymbol{C}$ is the tensor of rigidity.

The non-linear strain $\boldsymbol{\varepsilon}$ is (eq. 9)

$$\boldsymbol{\varepsilon} = (\boldsymbol{\nabla u})^{\boldsymbol{T}} + \boldsymbol{\nabla u} + (\boldsymbol{\nabla u})^{\boldsymbol{T}} . \boldsymbol{\nabla u} \tag{9}$$

The stress evolution in the 1-element viscoelastic generalized Maxwell model is given by solving eq. 10.

$$Q + \lambda_r \dot{Q} = \lambda_r \beta \dot{\boldsymbol{S}}_{iso}^{\infty} \tag{10}$$

The dimensionless coefficients $\beta > 0$ denoted the strain energy factor.

In this study, the Neo-Hookean model was used for the strain energy density function (eq. 11).

$$W = \frac{1}{2}\mu(\boldsymbol{I_1} - 3) \tag{11}$$

where μ is the Lamé coefficient (shear modulus) and $\boldsymbol{I_1} = tr(C)$, the first invariant of the left Cauchy–Green tensor.

II.2 Model Geometry, Boundary Conditions

Geometry

We considered that some of the most stretched gas cell wall was almost reduced to the **large and lenticular starch granule** (A-type) at the end of fermentation [12]. A **gluten strip** of 100 μm in length, and 20 μm wide was considered [12]**.** In 2D, the lenticular starch granule was considered to behave like an infinite “ellipsoidal cylinder” within an infinite strip of gluten (Figure 2). The half- large diameter and half-small diameter were a = 10 µm and b = 5 µm (Figure 2).

Sides, upper and lower boundaries

The baking and the fermentation typically features strain rates ranging from 10^{-4} to $10^{-3}\, s^{-1}$ and strain above 100% (Eliasson and Larsson [15]) We considered that the strain rate was $\dot{\varepsilon} = 3.10^{-3} s^{-1}$. This strain rate was obtained using a displacement of $\delta = 15\, \mu m$ on both the upper and lower boundaries over a calculation time of t=100 s. The boundaries on the left and right-hand side were let free to move (Figure 1). Under uniaxial elongation test, the following boundary conditions were used at upper and lower sides:

$$u(x, y) = 0$$
$$u\left(x, -\frac{L}{2}\right) = -u^d;\ v\left(x, \frac{L}{2}\right) = u^d \qquad (16)$$

δ stands for strain (equal to 30, 100 or 250 %) and u^d is the applied displacement at each edge calculated in such a way to keep strain and strain rate constant:

$$u^d = \frac{\delta}{2} \cdot \frac{t}{t_{sim}} L \qquad (17)$$

Where L is gas cell wall length (Figure 2), t the instantaneous time and t_{sim} the calculation time given by:

$$t_{sim} = \frac{\delta}{\dot{\varepsilon}} \qquad (18)$$

$\dot{\varepsilon}$ is the strain rate equal to $3.10^{-3}\ s^{-1}$.

The gluten/starch interface: Thin Elastic layer

Gluten/starch interaction at the gluten/starch interface was modelled using a **Thin Elastic Layer** boundary condition. The TEL models an elastic material of a given thickness (in our case 1 µm) at gluten/starch interface. The advantage of this condition is that, the thickness of the layer can be easily defined without geometrical representation. The "Thin Elastic Layer" condition used in COMSOL Multiphysics is equivalent to the "cohesive element model" used in Abaqus by Mohammed, Tarleton [18] for modelling the cohesion and non-cohesion at the interfaces. Depending on the stiffness value of the TEL, the interaction between gluten and starch is considered as cohesive or non-cohesive. The TEL decouples the displacements between two sides of the boundary. The two boundaries are then connected by elastic. k_n and k_t are the spring constant per unit area in normal and tangential directions.

$$k_n = \frac{E_{int}(1-\nu_{int})}{e(1+\nu_{int})(1-2\nu_{int})} \qquad (12)$$

$$k_t = \frac{G_{int}}{e} \qquad (13)$$

Where E_{int} is the Young's modulus, ν_{int} the Poisson's ratio, e the thickness of the interface ($1\ \mu m$) and G_{int} is the shear modulus. The properties used for the thin elastic layer are presented in table 2.

In 3D, one must see

Figure. 3 as the plane of symmetry that contained the smallest dimension of the lenticular starch granule.

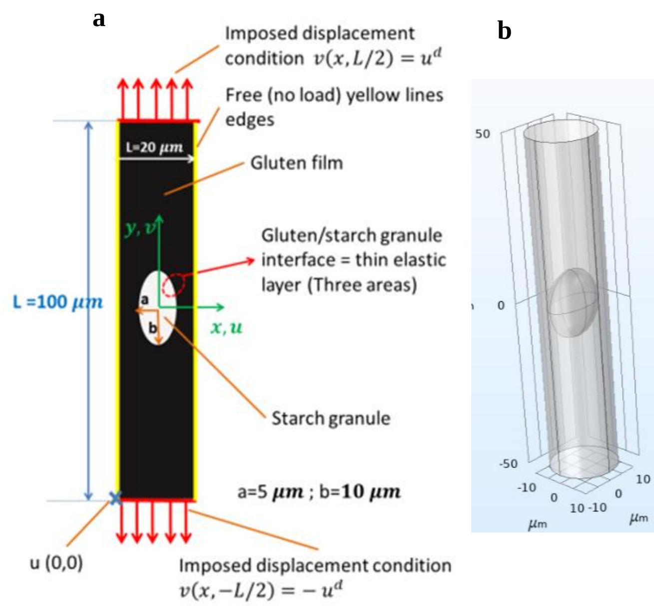

Figure 2 : 2D (.a) and 3D (.b) Geometry; summary of boundary conditions.

II.3 Material Properties

Gluten, starch and interface properties are presented in Table 1. As gluten and starch were both supposed not to exhibit a lot of time-independent elasticity the time-independent Young's modulus was set at 10 Pa. This time-independent elasticity is not well known from the literature that does not discuss very much the stress that never relaxes in the experimental works. Both the mechanical properties of gluten and starch were controlled by the elasticity allowed to relax. The same time of relaxation was used for gluten and starch. The ratio of starch to gluten Young's modulus was defined as $r = E_s/E_g$, where E_s is the starch granule Young's modulus and E_g is the Young's modulus of the gluten. The cases where r = 0.1 and 10 were considered in order to assess the effect of starch granules gelatinization and gluten denaturation phenomenon occurring during the baking (section III.3). The gluten and starch granules rheological modulus were chosen following the reported values in the literature [6, 18, 20, 21] (Table. 2). In this study, these parameters are assumed to be constant during all the extension. Table. 2. Gluten, starch granule and interface properties

Models	**Parameters**	**Unit**	**Gluten**	**Starch**	**Interface**	
					cohesion	non-cohesion
Time-independent elasticity	Young's modus	Pa	10			
Linear visco-elasticity	Young's modus (E)	kPa	10 or 100			
Visco-hyperelascticity	Strain energy factor (β)		1,000 or 100,000			
	Poisson's modulus (ν)		0.4999			
Thin elastic interface	Young's modulus (E_{int})	Pa			10^6	10^{-6}
	Poisson's modulus (ν_{int})				0.4999	0.49999999

III. Simulation Results and discussions

III.1 2D *vs* 3D

For the comparison, elliptical (2D) and lenticular (3D) shapes of starch granule were considered (Figure 2). The average of the Von Mises stress on the whole geometry was used. As showed in

Figure. 3, non-significant difference was found between 2D and 3D. These results are quite well substantiated by the work of Mohammed, Tarleton [18] who also compared simulation in 2D and 3D. This means than one could rather use 2D instead of 3D to reduce computation costs without impairing too much the results.

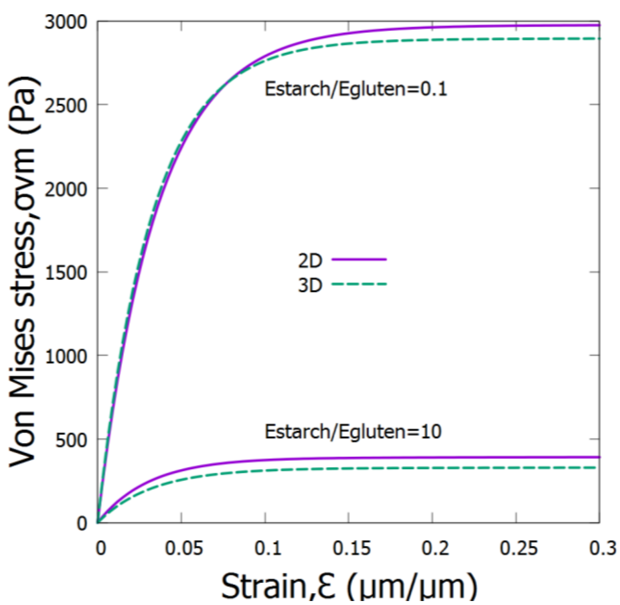

Figure. 3. Averaged Von Mises stress; **cohesive** bond at the gluten/starch interface for **r < 1** and **r > 1** ($\dot{\varepsilon} = 3.10^{-3}s^{-1}$).

III.2 Linear viscoelasticity *vs* visco-hyperelasticity

The comparison between the two approaches was carried out up 0.3 strain in order to assess the strain threshold from which the averaged Von Mises diverged. Both approaches were similar up to about 0.08 strain. Then only the visco-hyperelasticity approach was able to reproduce the expected strain softening of the bread dough at low strain rate (0.003 s^{-1}). These statements are verified regardless of interactions between the gluten and the starch granule and Young's moduli ratio.

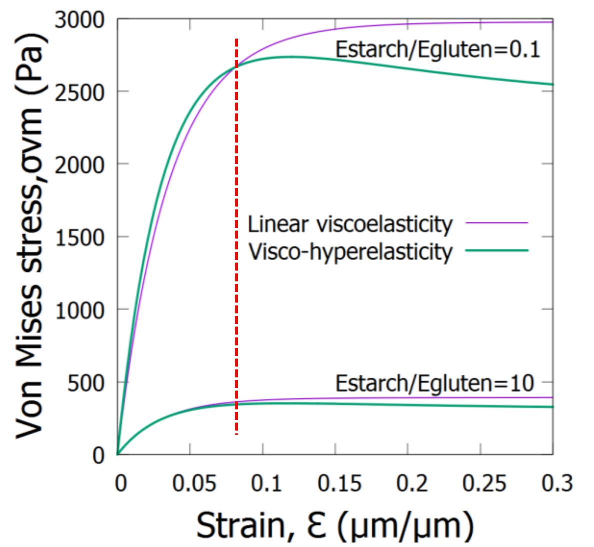

Figure 4 : Mean of the Von Mises stress, comparison between linear viscoelasticity and visco-hyperelasticity in the case of cohesive bond at the gluten/starch interface ($\dot{\varepsilon} = 3.10^{-3}$ s^{-1})

III.3 Effect of gluten/starch rheological moduli ratio and interactions

Effect of gluten/starch Young's moduli ratio

Figure 5 presents the mean Von Mises stress against strain. A great effect of gluten/starch moduli ratio on the mean Von Mises stress was found. High mean of the Von Mises stress values were found in the case where the gluten was more rigid than starch. It means that GCW extensibility is enhanced by the starch softening. At the gelatinization temperature ranging (60-65°C for wheat flour dough) starch granule viscosity softens. The softening of the starch at these temperatures probably relieves the gluten film and enhances gas cell wall extensibility. Gas cell wall under these conditions is supposed to hold on in extension much longer and rupture might occur at higher temperatures.

If starch that is more rigid than the starch granule (E_{starch}/E_{gluten}=10), gas cell wall will be less stretchy and therefore more exposed to rupture.

Effect of the nature of interactions at gluten/starch interface

The mean of the Von Mises stress shows non-significant effect of the nature of interaction between gluten and starch on gas cell wall behaviour (Figure 5). This result is in contradiction with Mohammed, Tarleton [18] results who showed the existence and effect of an interface on the behaviour of the dough film. We justify the difference of our result by the low starch volume proportion in the used geometry (7% for starch and 93% for gluten).

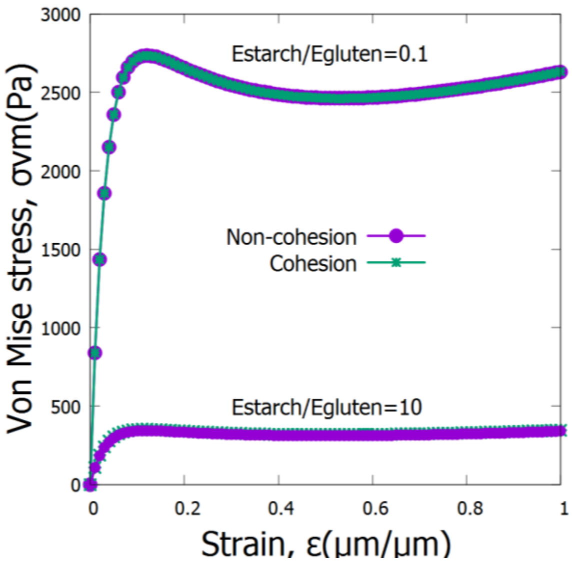

Figure 5 : Mean of the Von Mises stress in the case of **cohesive** and **non-cohesive** bond at the gluten/starch interface for **r<1** and **r>1** ($\dot{\varepsilon}$ = $3.10^{-3}s^{-1}$)

III.4 Stress concentration analysis: critical rupture spot within the GCW

r < 1 (the gluten is more rigid than the starch granule)

When the gluten is more rigid than the starch granule, the Von Mises stress fields were globally quite similar for both situations of cohesive and non-cohesive bonds at the interface. The difference appears close to the starch granule (zooms in Figure 6). Starch granules are more stretched (flattened) in the case of a cohesive bond compared to the non-cohesive case (Figure 6). The stress was transmitted to the starch granule which reinforced the gluten. In this case the rupture is quite equally likely to happen within either the gluten or starch.

On the opposite, the non-cohesive bond caused a stress concentration within the gluten located close to the stiff starch granule. The starch granule does not reinforce the gluten in this case. The gluten flows around the starch granule. In this case, the rupture is so more likely to happen within the gluten at the spot where the stress was the greatest i.e. very close to the rim of the starch granule (Figure 6.b).

r >1 (the starch granule is more rigid than gluten)

Non-significant difference was noticed in the Von Mises stress fields for both cohesive and non-cohesive bond at the starch/gluten interface for (Figure 7). The effect of gluten/starch interactions was cancelled by the fact that the starch granule was less rigid than the gluten. The extension of the GCW led to a thinning of the GCW at the location of the starch granule (Zoom Figure 7). This is the spot where the GCW is the most likely to rupture whether the bond is cohesive or non-cohesive.

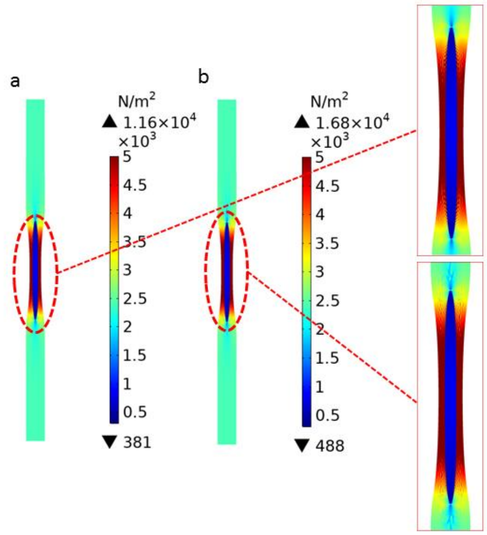

Figure 6 : The Von Mises stress fields in the case of **a.** cohesive and b. non-cohesive bond at the gluten/starch interface for **r<1** ($\dot{\varepsilon}$ = $3.10^{-3}s^{-1}$ and $\varepsilon = 1$).

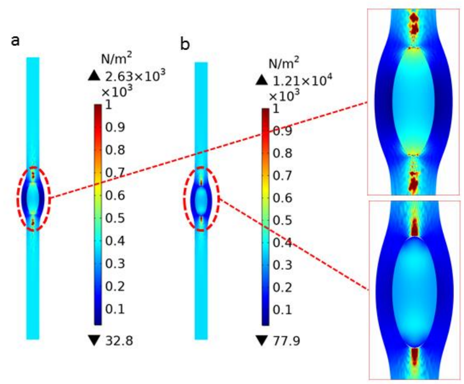

Figure 7 : The Von Mises stress fields in the case of a. cohesion and b. non-cohesion bond at the gluten/starch interface for **r>1** ($\dot{\varepsilon}$ = $3.10^{-3}s^{-1}$ and $\varepsilon = 1(\mu m/\mu m)$).

IV. Conclusion

The ability of both linear viscoelasticity and visco-hyperelasticity to reproduce the mechanical response of a gas cell wall (GCW) in bread dough was evaluated. Advantages and disadvantages of both approaches were discussed. It was found that visco-hyperelasticity involving finite strain was more suitable to mimic the extension of a GCW at strain and strain rate close to those encountered during baking.

Locations of Von Mises stress concentration were identified and discussed. Experimental observations are though required in order to identify where the rupture is really initiated and bring light to these numerical results. This is the purpose of on-going study.

This study needs also to be extended by adding the variations in material properties due to the increase in temperature and water content variations encountered during baking. The study of the behaviour of multiple starch granules is also required in order to respect the real proportions of starch and gluten.

V. Acknowledgements

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VI. References

1. Gan, Z., et al., *The microstructure and gas retention of bread dough.* Journal of Cereal Science, 1990. **12**(1): p. 15-24.
2. Hayman, D.A., R.C. Hoseney, and J.M. Faubion, *Effect of Pressure (Crust Formation) on Bread Crumb Grain Development.* Cereal Chemistry, 1998. **75**(5): p. 581-584.
3. Hayman, D.A., et al., *Factors controlling gas cell failure in bread dough.* Cereal Chemistry, 1998. **75**(5): p. 585-589.
4. Grenier, D., T. Lucas, and D. Le Ray, *Measurement of local pressure during proving of bread dough sticks: Contribution of surface tension and dough viscosity to gas pressure in bubbles.* Journal of cereal science, 2010. **52**(3): p. 373-377.
5. Singh, A.P. and M. Bhattacharya, *Development of dynamic modulus and cell opening of dough during baking.* Journal of Texture Studies, 2005. **36**(1): p. 44-67.
6. Dunnewind, B., et al., *The kieffer dough and gluten extensibility rig - an experimental evaluation.* Journal of Texture Studies, 2003. **34**(5-6): p. 537-560.
7. Sandstedt, R., *The microscopic structure of bread and dough.* Cereal Chem., 1954. **31**: p. 43-49.
8. Bloksma, A.H., *Rheology of wheat flour doughs.* Journal of Texture Studies, 1972. **3**(1): p. 3-17.
9. Dobraszczyk, B.J., *The rheological basis of dough stickiness.* Journal of Texture Studies, 1997. **28**(2): p. 139-162.
10. Baker, J.C. and M.D. Mize, *Effect of temperature on dough properties - II.* Cereal Chemistry, 1939. **16**: p. 682-695.
11. Bloksma, A., *Dough structure, dough rheology and baking quality.* Cereals Food World, 2, 35, 237-244, 1990.
12. Van Vliet, T., et al., *Strain hardening of dough as a requirement for gas retention.* Journal of Texture Studies, 1992. **23**(4): p. 439-460.
13. Sliwinski, E.L., P. Kolster, and T. van Vliet, *Large-deformation Properties of Wheat Flour and Gluten Dough in Uni- and Biaxial Deformation*, in *The Gluten Proteins*. 2004, The Royal Society of Chemistry. p. 211-214.
14. Schofield, R.K. and G.W.S. Blair, *The relationship between viscosity, elasticity and plastic strength of a soft material as illustrated by some mechanical properties of flour dough.-III.* Proc. R. Soc. Lond. A, 1933. **141**(843): p. 72-85.
15. Eliasson, A.-C. and K. Larsson, *Cereals in breadmaking: a molecular colloidal approach*. 1993: Marcel Dekker.
16. Petrofsky, K. and R. Hoseney, *Rheological properties of dough made with starch and gluten from several cereal sources.* Cereal Chemistry, 1995. **72**(1): p. 53-57.
17. He, H. and R. Hoseney, *Factors controlling gas retention in non-heated doughs.* Cereal Chem, 1992. **69**(1): p. 1-6.
18. Mohammed, M.A.P., et al., *Mechanical characterization and micromechanical modeling of bread dough.* Journal of Rheology, 2013. **57**(1): p. 249-272.
19. Holzapfel, G.A., T.C. Gasser, and R.W. Ogden, *A new constitutive framework for arterial wall mechanics and a comparative study of material models.* Journal of elasticity and the physical science of solids, 2000. **61**(1-3): p. 1-48.
20. Tschoegl, N.W., J.A. Rinde, and T.L. Smith, *Rheological properties of wheat flour doughs I.—Method for determining the large deformation and rupture properties in simple tension.* Journal of the Science of Food and Agriculture, 1970. **21**(2): p. 65-70.
21. Ng, T.S., G.H. McKinley, and M. Padmanabhan, *Linear to non-linear rheology of wheat flour dough.* Applied Rheology, 2006. **16**(5): p. 265-274.