Title: Starch replacement in gluten free bread by cellulose and fibrillated cellulose

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Highlights:

- The incorporation of (fibrillated) cellulose strengthened gluten free doughs
- The generalised Maxwell model was applied to analyse dough rheology
- The incorporation of (fibrillated)cellulose influenced the later stage of proofing
- Volume and water competition dominates the thermal rheological behaviours
- Loaves with (fibrillated)cellulose were smaller with harder, denser but finer crumb

Keywords:

Fibrillated cellulose, cellulose, gluten free bread, dough rheology, bread quality

Abstract

This study investigated starch reduction and replacement by purified cellulose (FC0) and fibrillated cellulose (FC60) which included a comprehensive investigation on dough properties, proofing behaviours, cooking performance, and bread qualities. Replacing flour with FC0 and FC60 was found to strength the doughs without, however, altering the extension of the structural network according to the weak gel model. The relaxation time calculated by the generalised Maxwell model was found to be shorter than the deformation rate during proofing which suggests that the doughs behave like fluids during proofing. The relaxation time was less influenced by the additions of FC0 and FC60. Although the initial stage of proofing was less influenced, the later stage was significantly affected by the additions of FC0 and FC60 which increased dough rigidity and restrained the volume growth. The pasting properties were significantly influenced by the competition for water and volume between FC0/FC60 and flour. The bread qualities were characterised in terms of loaf volume and crumb properties. Loaves containing FC0 and FC60 had smaller specific volume and harder crumb. However, the addition of FC0 and FC60 is beneficial to the generation of a finer crumb structure. Fibrillation process is detrimental to maximise the addition of fibres. However, a small amount of fibrillated cellulose is beneficial to workability and crumb structure.

1. Introduction

Wheat flour is the main ingredient of bread, the staple food in many regions. However, many people around the world are intolerant or sensitive to gluten, which leads to coeliac disease and other gluten-trigged health problems (Catassi et al., 2012; Czaja-Bulsa, 2015; Sapone et al., 2012). In 2014, Aziz et al. (2014) conducted a survey in Sheffield, UK, and found that 13% of the population reported gluten sensitivity and the prevalence of diagnosed Celiac disease was 0.8%. An early study reported that the prevalence of the coeliac disease in the world is approximately 1 in every 100 people (Zandonadi, Botelho & Araujo, 2009). Celiac disease, the most widely studied gluten-triggered disease, is an autoimmune genetically determined chronic inflammatory intestinal disorder (Fasano & Catassi, 2001; Schuppan, 2000) which can be influenced by both genetic (intrinsic) and environmental (extrinsic) factors (Di Sabatino & Corazza, 2009; Schuppan, 2000; Wahab, Meijer, Goerres & Mulder, 2002). On the other hand, ‘free from’ is becoming a global trend of a healthy lifestyle choice, which further promotes the market of gluten free products. The production and improvement of gluten free bread have been widely studied and the current methodologies can be categorised as the application of alternative flour blends, enzyme treatments and sourdough applications, and other treatments. The application of alternative flour blends is the most investigated which includes investigations on formulations (Haque & Morris, 1994; Lazaridou, Duta, Papageorgiou, Belc & Biliaderis, 2007; Nishita, Roberts, Bean & Kennedy, 1976; Ronda, Pérez-Quirce, Angioloni & Collar, 2013) and structured gel and colloids (Van Riemsdijk, van der Goot, Hamer & Boom, 2011). Both dough rheological properties and bread qualities have been widely investigated. In addition, the development of the dough microstructure and during baking, which can be influential in the production of bread and other bakery goods, have also been studied (Babin et al., 2006; Baldino, Laitano, Lupi, Curcio & Gabriele, 2018; Bousquières, Michon & Bonazzi, 2017; Wagner, Quellec, Trystram & Lucas, 2008).

In addition, the proportion of overweight adults and children has increased around the world and obesity has become a global health challenge during the past three decades (Ng et al., 2014). The prevalence of obesity around the world is one of the main reasons for the increased morbidity rate of type-2 diabetes (Shaw, Sicree & Zimmet, 2010). However, the removal of gluten from the diet easily lead to a significant decrease in carbohydrate oxidation rate, increase in body fat stores, and weight gain in celiac patients (Capristo et al., 2000; Hager et al., 2012).

Therefore, food containing low calories and/or glycaemic index is recommended and the production of gluten-free bread with low-calorie content and glycaemic index will be beneficial to deal with gluten intolerance, obesity and type 2 diabetes simultaneously. The additions of dietary fibres or fibre enriched materials, most by-product or cell wall materials, have been studied to improve the nutritional profiles of gluten free bread. Demirkesen, Mert, Sumnu and Sahin (2010) produced gluten free bread with desirable qualities in terms of loaves, texture, colour and sensory perception by adding an intermediate amount of chestnut flour. Gluten free bread with enhanced fibre contents was achieved by incorporating apple fibres and sugar beet fibres with hydroxypropyl methylcellulose (HPMC) (Djordjevic et al., 2019). The addition of rice bran in gluten free bread has been investigated by Phimolsiripol, Mukprasirt and Schoenlechner (2012). The investigations on the addition of fibres were mainly based on the materials with a defined processing procedure or commercial products but the influence of fibre treatment, such as fibrillation, is less understood.

The fibres added in the bread formulation can be enriched in cellulose, hemicellulose, and/or pectin. These materials, either soluble or insoluble, usually have high water absorbability which significantly alters the rheological properties of doughs. The insoluble cellulose enriched materials can also be expected to play a role as fillers.

Cellulose is one of the main components in the plant cell wall, which structures, with hemicellulose, pectin, and lignin, to maintain the mechanical property of cell walls. Microfibrillated cellulose (MFC) was first processed using high pressure and shearing by Herrick, Casebier, Hamilton and Sandberg (1983) and Turbak, Snyder and Sandberg (1983a), where the native cellulose fibres are physically unwound and highly entangled cellulose fibrils are generated with high surface area, liquid retention, and reactivity to chemical treatments. In food productions, MFC can be used as a thickener, compound carriers, and suspension & emulsion stabilisers (Turbak, Snyder & Sandberg, 1982, 1983b). Currently, the applications of high pressure homogeniser and microfluidiser as mechanical cellulose fibrillation treatments are widely investigated to produce MFC (López-Rubio et al., 2007; Nakagaito & Yano, 2004; Pääkkö et al., 2007; Stenstad, Andresen, Tanem & Stenius, 2008; Zimmermann, Pöhler & Geiger, 2004).

In this study, a colloid mill was used which has lower energy input. Therefore, the cellulose was fibrillated to a less extend comparing to the production of MFC. The water absorbability and fibre configuration and rigidity were greatly altered by the fibrillation process. The aim of this study was to investigate the starch/flour reduction of rice flour based gluten free bread by the addition of cellulose and fibrillated cellulose. A high replacement level of up to 20% was included in the investigation. Dough properties, proofing behaviours, cooking performance, and bread qualities were of the interests to achieve a comprehensive understanding.

2. Materials and methods

2.1. Materials

Rice flour was purchased from Doves Farm online store. Allinson Easy Bake Yeast (Allinson Flour, Peterborough, UK), sugar (Sainsbury's, UK), sunflower oil (Sainsbury's, UK), and salt (Sainsbury's, UK) were purchased from local supermarkets. Methylcellulose (Methocel® A4M) was supplied by The Dow Chemical Company (Bomlitz, Germany). Vitacel® Psyllium seed husk powder was kindly donated by the JRS (J. Rettenmaier & Söhne Group, Rosenberg, Germany). Pure amylose from potato and amylopectin from corn were purchased from Sigma-Aldrich (Dorset, UK). Pure powdered cellulose, Solka floc 900FCC, was supplied by International Fiber Corporation, US.

2.2. Biochemical analysis

Nitrogen content was measured using a Nitrogen Analyser NA 2000 (Fisons Scientific Equipment, Loughborough, UK) and the protein contents of rice flour and psyllium seed husk powder were converted with a factor of 6.25. Lipid was extracted with a chloroform-methanol mixture (2:1) and the contents were calculated. Moisture contents were obtained by drying at 105 °C and ash contents were measured using a muffle furnace at 550°C.

AACC method (61-03) and the method from Kaufman, Wilson, Bean, Herald and Shi (2015) were modified to determine the amylose content of rice flour. Rice flour was defatted using 85% methanol. Defatted rice flour, potato amylose and corn amylopectin (100 mg) were dispersed in 1 ml of 95% ethanol and then dissolved in 9 ml of 1 M NaOH in a boiling water bath for 10 min. After standing at room temperature for 3 h, the starch solutions were diluted to 100 ml. The standard curve was obtained from mixtures of potato amylose and corn amylopectin in a series of different ratios. Samples (5ml) were diluted by 20 times with additions of 1 ml of 1M acetic acid and 2 ml of iodine solution (0.2% I_2 and 2% KI w/v) and were incubated at room temperature for 20 min. The absorbance at 720 nm was determined using a spectrophotometer. The amylose contents were calculated against the regression equation determined from the standard curve and verified by a standard starch sample with 66% amylose content.

2.3. Cellulose fibrillation

Five grams of cellulose was dispersed in 500 ml of reverse osmosis (RO) water and fibrillated by a colloid mill (Winkworth, Basingstoke, UK) for 30 minutes (60 min L^{-1}) and labelled as FC60. The fibrillated cellulose was then centrifuged at 4000 g, 4 °C, for 15 min and the supernatants (residue less than 0.01%) were discarded. The concentration of sediments was verified by drying at 105 °C for each batch and stored at 4 °C, which were then diluted to the required concentrations for dough preparations.

Table 1. Addition levels of rice flour, FC0, and FC60.

Sample code	Rice flour	FC0	FC60
F(100) (control)	100	0	0
F(98)+FC60(2)	98	0	2
F(90)+FC0(10)	90	10	0
F(90)+FC0(8)+FC60(2)	90	8	2
F(80)+FC0(20)	80	20	0
F(80)+FC0(18)+FC60(2)	80	18	2
F(80)+FC0(16)+FC60(4)	80	16	4

2.4. Dough formulation and preparation

The basic formulation (control) was decided according to preliminary tests and included 100g of rice flour, 5g of sugar, 2g of salt, 1.5g of yeast, 5g of vegetable oil, 1g of methylcellulose (MC), 1g of psyllium, and 120g of water. Rice flour was partially replaced with pure cellulose powder (FC0) and/or fibrillated cellulose (FC60) as shown in Table 1. Apart from F(100) (control) and F(98)+FC60(2), all other FC0/FC60 incorporated formulas are expected to be claimed ‘high fibre’ according to Regulation (EC) No 1924/2006 based on a rough calculation that the fibre content is higher than 3 g per 100 kcal. FC0 was directed used to replace rice flour and mixed with other dry ingredients. FC60 was redispersed in water required in the formulation and added with oil into dry ingredients. Doughs were mixed based on 200g of rice flour or the mixture of flour and cellulose per batch by a stand mixer (Kenwood, UK) equipped with a CHEF flexible beater (AT501, Kenwood, Havant, UK). Dry ingredients were mixed thoroughly and then mixed with water/FC60 dispersion and oil for 7 min at speed 1. Doughs prepared for rheological measurements did not contain yeast and were allowed for hydration at room temperature for 1 hour.

2.5. Dough evaluation

2.5.1. Fundamental rheological measurements and thermal behaviour of doughs

Doughs without yeast, which were hydrated for 1 hour at room temperature, were subjected to shear stress ramp tests and small amplitude oscillatory shear tests by MRC 301 rheometer (Anton Paar, Austria) equipped with serrated parallel plate geometry (PP25/P2-SN15766, Anton Paar). The measuring gap was 2 mm. The temperature was controlled by a Peltier system with the assistance of a water bath (R1, Grant, Shepreth, UK). The extra sample was trimmed by a spatula during sample loading and the edge was covered by low viscosity mineral oil (Sigma, USA) to prevent drying. Doughs rested at 30 °C for 500 seconds before measurements. Logarithmic increase of shear stress from 0.03 to 30000 Pa in 18.5 min was performed to obtain the yield point or yield zone. Frequency sweep tests were performed in a logarithmic decrease from 600 to 0.06 rad s^{-1} with 0.02% strain (in the linear viscoelastic region). The slopes of lgG’ versus lgω in the middle frequency range (0.881 to 40.9 s^{-1}) were calculated. The dynamic oscillatory measurement data were also fitted into the ‘weak gel’ model (equation 1) proposed by Gabriele, de Cindio and D'Antona (2001) where z represents the number of the interacting rheological units of ‘weak gel’ structure and A refers to the interaction strength between the rheological units. $G^*(\omega)$ is the complex modulus.

$$G^*(\omega) = \pi A \omega^{\frac{1}{z}} \qquad (1)$$

Additionally, the obtained mechanical spectra were fitted to generalised Maxwell model as shown in equation (2) with individual relaxation time (λ_i) and individual relaxation moduli (G_i) describing the discrete relaxation time spectrum of the sample where $G(t)$ is the relaxation modulus at time t.

$$G(t) = \sum_{i=1}^{N} G_i e^{-t/\lambda_i} \qquad (2)$$

The relaxation times of each component (λ_i) were arbitrarily decided as shown in Table 2. The dynamic moduli G'(ω) and G''(ω) were calculated from angular frequency (ω) by equation (3) and (4) (Baumgaertel & Winter, 1992; Ferry, 1980; Laun, 1986). Individual relaxation moduli G_i, which is listed in Table 2, was varied to minimise the sum of square differences between calculated G'(ω), G''(ω) and experimentally obtained G', G''. G_e represents a pure elastic component connected in parallel with other Maxwell elements in the model.

$$G'(\omega) = \sum_i G_i \frac{\omega^2 \lambda_i^2}{1 + \omega^2 \lambda_i^2} + G_e \tag{3}$$

$$G''(\omega) = \sum_i G_i \frac{\omega \lambda_i}{1 + \omega^2 \lambda_i^2} \tag{4}$$

Temperature sweep tests were performed at a constant strain (0.02%) and angular frequency (10 rad s^{-1}) with the temperature increased from 20 °C to 98 °C with a heating rate of 2.6 °C min^{-1} mimicking the temperature profile during baking.

Table 2. Individual relaxation time and relaxation moduli for Generalised Maxwell Model fitting on the mechanical spectra shown in Figure 2.

individual relaxation time λ_i (s)	0.0001	0.001	0.01	0.1	1	10	100	1000	10000	100000	R^2
F(100) (control)	123755	0	1201	1146	724	1934	1237	0	0.010	0.001	0.991
F(98)+FC60(2)	0	14804	6066	3950	3113	1925	8856	0	0.012	0.001	0.996
F(90)+FC0(10)	255156	0	3575	3270	2264	6034	3748	0	0.011	0.001	0.989
F(90)+FC0(8)+ FC60(2)	279077	0	5451	6761	3917	11764	6455	0	0.013	0.002	0.980
F(80)+FC0(20)	354470	18597	23156	18045	12357	22615	25966	0	0.010	0.002	0.998
F(80)+FC0(18) +FC60(2)	904692	0	31077	29132	18275	37525	42383	0	0.010	0.000	0.994
F(80)+FC0(16) +FC60(4)	1271	88716	36796	29398	18524	32671	47364	0	0.010	0.017	0.998

2.5.2. Empirical rheological measurements

Doughs were also characterised on a TA-XT plus texture analyser (Stable Micro systems, Surrey, UK) equipped with a 5 kg loading cell. Backward extrusion tests and SMS/Chen-Hoseney Dough Stickiness tests (Stable Micro Systems, UK) were performed. For the backward extrusion tests, doughs were loaded into a container with a diameter of 42 mm to a height of 36 mm avoiding big air pockets. A disc with a diameter of 30 mm and thickness of 5 mm was centred and extruded into the dough by 22.5 mm and then returned to the height of 120 mm from the base at a speed of 1 mm s^{-1}. The positive peak force and positive area during downward stroke indicated firmness and consistency of doughs respectively. The negative peak force and area during upward stroke indicated cohesiveness and index of viscosity. The returning distance before the force reached a constant negative value indicated dough extensibility. As for the SMS/Chen-Hoseney Dough Stickiness tests (Chen & Hoseney, 1995), doughs were loaded into the equipped cell (A/DSC) and a uniform surface was generated. The surface was compressed by an aluminium cylinder probe (P/25) by 40 g for 0.1 s. The probe compressed the sample at the speed of 0.5 mm s^{-1} and withdrew at 10 mm s^{-1}. The maximum force, area, and travel distance during probe withdrawing indicated dough stickiness, work of adhesion, and strength individually. Data recording and analysis were performed by the equipped software Exponent 6.1.14.

2.6. Pasting properties of flour blends

The pasting profiles of flour blends were measured by Rapid Visco Analyser (RVA) (Newport Scientific Pty. Ltd., Warriewood, New South Wales, Australia) equipped with a water bath (Thermo scientific C10, Karlsruhe, Germany). The solid levels of rice flour, MC, psyllium, FC0, and FC60 are listed in Table 3, which were in the same ratio as in the dough formulation without sugar, salt, yeast, and oil. More specifically, flour, MC, psyllium and FC0 were mixed thoroughly. Flour blends (2.55 g) prepared according to the formula which does not include FC60 were dispersed in 24 ml of RO water. As for FC60-containing formula, FC60 stock suspensions were diluted to 24.05 g or 24.1 g with FC60 concentration of 0.208% and 0.415% respectively for the formulations of 2% and 4% replacement by FC60. Flour blends (2.5 or 2.45 g respectively) were dispersed into diluted FC60 suspensions. The test started with high shear rate mixing (960 rpm) for 60 seconds at 25 °C followed by 60 seconds mixing at 160 rpm. The temperature then increased to 95 °C in 350 s, held at 95 °C for 150 s, and decreased to 25 °C in 350 s. The pasting profiles were analysed by Thermocline where pasting temperature (the temperature of the onset of viscosity increase), peak (the highest viscosity), peak time (the time when the highest viscosity was reached), trough (lowest viscosity after the peak), trough time (the time when the trough occurred), final viscosity, breakdown (difference between peak and trough), and setback (difference between final viscosity and trough viscosity) were reported.

Table 3. Addition levels of rice flour, MC, psyllium, FC0, and FC60 in pasting property analysis.

Sample code	Rice flour	MC	psyllium	FC0	total dry blends	FC60
F(100) (control)	2.5	0.025	0.025	0	2.55	0
F(98)+FC60(2)	2.45	0.025	0.025	0	2.5	0.05
F(90)+FC0(10)	2.25	0.025	0.025	0.25	2.55	0
F(90)+FC0(8)+FC60(2)	2.25	0.025	0.025	0.2	2.5	0.05
F(80)+FC0(20)	2	0.025	0.025	0.5	2.55	0
F(80)+FC0(18)+FC60(2)	2	0.025	0.025	0.45	2.5	0.05
F(80)+FC0(16)+FC60(4)	2	0.025	0.025	0.4	2.45	0.1

2.7. Baking tests

Doughs prepared according to section 2.3 were loaded in a baking pan with a dimension of 7.5 x 7.5 x 10 cm (W×L×H). Each batch generated two doughs with weights of 200 to 210 g. Baking pan was shaken to expel big air pockets in doughs. The doughs were covered by cling film and proofed in an incubator (Binder, Tuttlingen, Germany) at 30 °C for 85 min and then baked in a deck oven (Tom Chandley, Manchester, UK) for 40 min at 230 °C. The loaves were cooled on a rack at the atmosphere environment for 1 hour before further evaluation.

2.7.1. Basic analysis and calculations

Proofing behaviour was monitored by incubating approximately 10 ml of doughs in a measuring cylinder at 30 °C for 85 min during which the volume was recorded. Doughs were weighted before baking and the loaves were weighted after baking and cooling. Loaf volumes were measured by rapeseed replacement. The baking loss was calculated by $baking\ loss = (dough\ weight\ before\ proving\ -\ loaf\ weight\ after\ cooling)/dough\ weight\ before\ proving.$ Specific volume was the ratio of loaf volume to loaf weight. The moisture content of the loaf centre was measured by drying a piece of crumb cut from the centre at 105 °C.

The loaves were sliced perpendicularly to a thickness of 1.25 cm and the middle surface was scanned using a C-Cell imaging system (Calibre Control International LTD, Warrington, UK). The obtained imaged were analysed using C-Cell software version 2.0.

2.7.2. Bread textural evaluation

The textural property of gluten free bread was evaluated by texture profile analysis (TPA) on a TA-XT plus texture analyser (Stable Micro systems, Surrey, UK) equipped with 30 kg loading cell. A cylinder piece of bread crumb was cut from the centre of every loaf slice by a diameter of 3 cm and two pieces from the middle four slices of each loaf were stacked together. Therefore, two pieces of the sample were obtained from the middle of each loaf with the shape of a cylinder with a height and a diameter of 2.5 cm and 3 cm individually. The samples were 65% compressed by a 100 mm plate twice at a speed of 1 mm s^{-1} with 5 s between two compresses. Hardness, springiness, cohesiveness, chewiness, and resilience were obtained from the TPA profiles.

2.8. Statistical analysis

All measurements were repeated at least three times. Plots are shown as representative curves. Results are shown as the mean ± standard deviation in bar charts and tables. The C-Cell parameters were analysed and compared using one-way analysis of variance (ANOVA) and Turkey's test at a significance level of $p < 0.05$ using IBM SPSS statistics version 26 (IBM Corp., Armonk, NY, USA).

3. Results and discussion

3.1. Biochemical properties

The moisture content, protein content, ash content, and lipid content of rice flour were 11.1 %, 7.23 %, 0.42 %, and 2.8 % respectively. That of PSY was 7.23 %, 3.40 %, 2.89 %, and 3.30 % respectively (Ren, Yakubov, Linter, MacNaughtan & Foster, Unpublished results-b). The amylose content of rice flour was 28.8 %.

3.2. Dough rheological properties

3.2.1. Fundamental rheological analysis of dough properties

The stress ramp data are shown in Figure 1. It can be seen (Figure 1a) that the shear strain increased with shear stress with different rates in different stress ranges. The replacement of rice flour by FC0 and FC60 significantly decreases the shear strain caused by a certain applied shear stress in the higher stress range (>10 Pa). The control dough (F100) shows a relatively short transition from the linear-elastic deformation behaviour (shear strain increases linearly with shear stress) to flow behaviour and a yield point can be defined by calculating the deviation from the linearity at low shear stress range. However, when FC0 or FC60 is added, it shows a long yield zone and yield point cannot be defined by this method. The longer yield zone could due to the long cellulose fibres with various sizes. The viscosity from the same data sets is plotted versus shear stress in Figure 1b. The viscosity plateau at low shear stress which is similar to the Newtonian plateau is, however, possibly due to the fact that the doughs did not reach a steady state. However, the start of viscosity decrease also indicates yield stress (Walls, Caines, Sanchez & Khan, 2003). Although it is still difficult to define the yield point due to a long yield-flow transition, it can be seen that the replacement by FC0 and FC60 increased the stress at which doughs start to flow (significant decrease in viscosity). Similar enhancing effects on viscosity and yield stress by the addition of chestnut flour, which has high fibre content, in gluten free rice bread has been reported by Demirkesen et al. (2010) which can be assigned to the high entanglement and water binding ability of fibres.

The mechanical spectra of doughs are shown in Figure 2. In the mechanical spectra, it can be seen that all doughs show solid like property with G' higher than G''. The additions of FC0 or FC60 increased both moduli and decreased tanδ suggesting a more solid like property, which is in accordance with the strengthening effects of doughs evidenced by yield stress and viscosity as discussed above.

The similarities in the general structure and rheological behaviours of different foods has been concluded and a weak gel model has been proposed by Gabriele et al. (2001). Foods can be described to be structured by strands constituted by weakly interacting flow units, and strongly interacting topological points, which is analogous to the classic 'true gel' with permanent cross-linked three dimensional network (Gabriele et al., 2001). The complex moduli (G^*) of gluten free doughs and angular frequencies (ω) were fitted with the weak gel model (equation (1)) with R^2 higher than 0.95 and two model parameters, A and z, were obtained and are listed in Table 4. It can be seen that A increased significantly with the addition of FC0 and FC60 which is in accordance with the increase of moduli. However, there is no significant difference in the value of z between all flour-replaced doughs, which suggests that there are no significant effects on the extension of the network of strands and rheological units. The increase in both moduli and parameter A and the decrease of tanδ are more likely to be attributed to the increase in the strength of the topologically interactions and strand (rheological units) upon the additions of FC0 or FC60 fibres. In other words, it can be speculated that both flour particles and FC fibres are rigid fillers regardless of the difference in shapes. FC fibres are more rigid than the hydrated flour particles and they compete for water with starch which further increases the rigidity of the flour particles. Therefore, the doughs are strengthened by the addition of FC0 or FC60.

The slopes of lgG' versus lgω have a similar value of approximately 0.105 in all cases regardless of the addition of FC0 or FC60, which is in accordance with the uninfluenced z values. However, it is lower compared with most gluten free and wheat doughs which range from 0.11 to 0.37 (Georgopoulos, Larsson & Eliasson, 2004; Ronda et al., 2013; Tanner, Qi & Dai, 2008; Upadhyay, Ghosal & Mehra, 2012; Villanueva, Harasym, Muñoz & Ronda, 2019). The low lgG' versus lgω slope suggests approaching a true gel at the angular frequency range studied. It was less influenced by FC0 and FC60 at the addition levels studied.

FC60 has more pronounced influence than FC0 even with very low addition levels (2 or 4% replacement). In fact, the effect of 2% replacement by FC60 is similar to 10% replacement by FC0 shown as similar shear strains and viscosities plotted versus shear stress, and similar mechanical spectra and parameter A in the weak gel model. This could be explained by the higher water holding ability of FC60 due to fibrillation treatment (Agarwal, Hewson & Foster, 2018a; Ren, Linter & Foster, 2020). Additionally, an increase in fibre entanglement can also be expected as an outcome of fibrillation and FC60 appears as flocculates or aggregates (Ren et al., 2020). It is likely that FC60 exists as highly entangled fibre aggregates which can be considered as individual rheological units with enhanced properties. However, considering that there was no significant difference in the value of the weak gel model parameter z between FC0 and FC60 doughs, the interaction and entanglement between FC60 aggregates and FC60 aggregates and flour particles are limited therefore the influence on the extension of the structural network of doughs are limited.

Table 4. Weak gel model parameters.

	A (Pa.s$^{1/z}$)	z
F(100) (control)	3216±538^{a}	8.90±1.12^{a}
F(98)+FC60(2)	10915±2055^{a}	9.06±1.04^{a}
F(90)+FC0(10)	11486±493^{a}	10.47±0.10^{a}
F(90)+FC0(8)+FC60(2)	21971±1733^{ab}	10.85±0.46^{a}
F(80)+FC0(20)	52447±4287^{bc}	9.34±0.35^{a}
F(80)+FC0(18)+FC60(2)	82416±11993^{c}	9.69±0.34^{a}
F(80)+FC0(16)+FC60(4)	80984±18218^{c}	9.08±0.27^{a}

Data are shown as mean ± standard deviation. Values with different letters in the same column were significantly different ($p < 0.05$).

The generalised Maxwell model was applied to estimate the dough rheological behaviours at a longer time scale (slow deformation) (Figure 2). The arbitrarily decided λ_i and calculated corresponding relaxation moduli G_i are listed in Table 2. Ten pairs of λ_i and G_i along with a G_e representing a single pure elastic component were used to calculate $G'(\omega)$ and $G''(\omega)$ by equation (3) and (4). The value of G_e of each sample is either 0 or very low therefore it is not listed in Table 2. A zero value of G_e is typical for viscoelastic liquid of uncross-linked polymers (Ferry, 1980). Therefore, gluten free bread doughs are structurally and rheologically analogous to viscoelastic fluids instead of a solid.

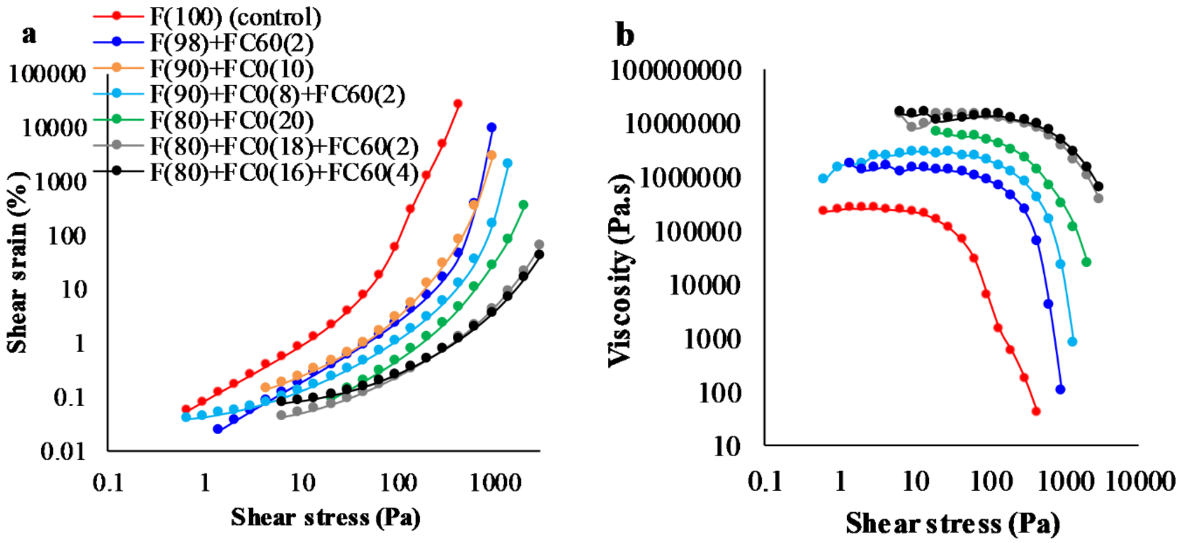

Figure 1. Shear stress ramp data (a) and viscosities plotted versus shear stress (b) of gluten free doughs. Plots are shown as representative curves from experiments run in triplicates.

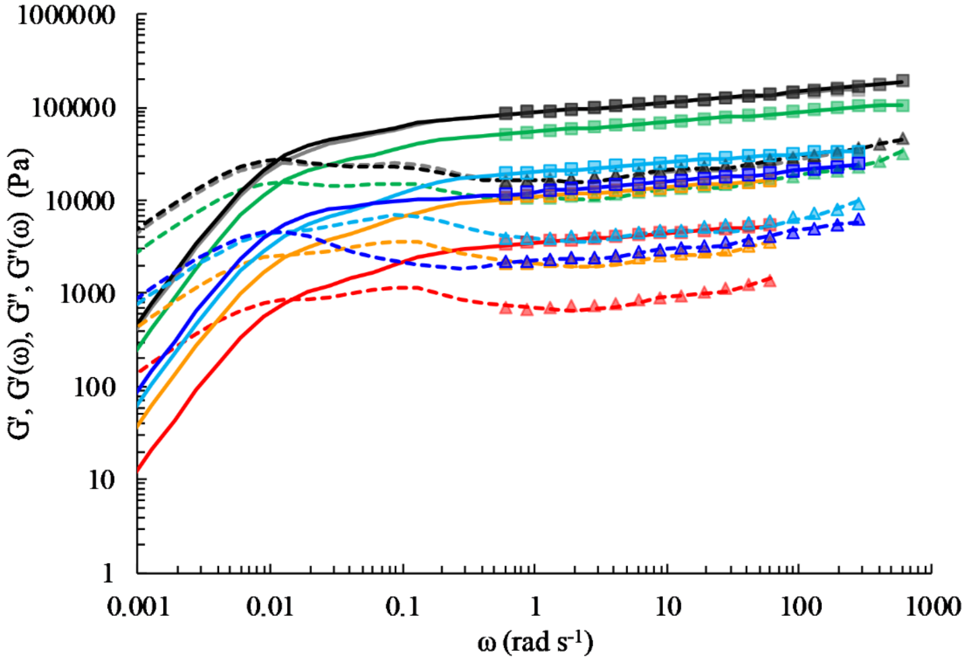

Figure 2. Mechanical spectra and curves calculated from generalised Maxwell model of F(100) (control) (red), F(98)+FC60(2) (dark blue), F(90)+FC0(10) (yellow), F(90)+FC0(8)+FC60(2) (light blue), F(80)+FC0(20) (green), F(80)+FC0(18)+FC60(2) (grey), and F(80)+FC0(16)+FC60(4) (black). Experimental storage moduli (G') and loss moduli (G'') are shown by squire and triangle symbols respectively and calculated G'(ω) and G''(ω) are presented by solid lines and dashed lines respectively. Plots are shown as representative curves from experiments run in triplicates.

The generalised Maxwell model expands the evaluable mechanical spectra to a lower frequency range with R^2 higher than 0.98 (Table 2). It can be seen in Figure 2 that all doughs have a relaxation frequency of nearly 0.01 rad s^{-1}. The influences of FC0 and FC60 additions are insignificant which is in accordance with the uninfluenced z value in the weak gel model that FC0 and FC60 do not significantly alter the extension of the rheological network. The porous structure of loaves mainly forms, develops, and sometimes collapses during proofing with low strain rates (10^{-4} to 10^{-3} s^{-1} as reported by Babin et al. (2006) and Grenier, Lucas and Le Ray (2010)). The time scale is longer than the relaxation time (reciprocal of relaxation frequency) of gluten free. In other words, doughs behave like fluids (G'' > G') in this low strain rate range as found during proofing, which could be beneficial to the development of the loaf structure. However, this also implies that only strengthening doughs might not be able to provide efficient stability to maintain the porous structure. This could explain from, one aspect, why air pockets and big voids in crumb structure is a common and difficult issue in the gluten free bread production which have been widely reported (Haque & Morris, 1994; McCarthy, Gallagher, Gormley, Schober & Arendt, 2005; Nishita et al., 1976; Paciulli, Rinaldi, Cirlini, Scazzina & Chiavaro, 2016; Schober, Bean & Boyle, 2007).

3.2.2. Empirical rheological analysis of dough properties

The influence of FC0 and FC60 addition on the behaviour of doughs under large deformation were analysed by backward extrusion experiments. Dough stickiness was measured using an SMS/Chen-Hoseney Dough Stickiness rig. As shown in Figure 3, the addition of FC0 and FC60 significantly increased dough firmness, cohesiveness, consistency, and index of viscosity, which is in agreement with the increasing effects on yield stress and dynamic moduli. Similar to what was observed in the fundamental rheological analysis, 2% replacement by FC60 and 10% replacement by FC0 show similar values for these four parameters which indicate that fibrillated cellulose is more efficient in strengthening doughs than untreated cellulose. The additions of FC0 and FC60 generally decreased dough extensibility by approximately 20 mm while the effects of FC0 fibres tend to be quadratic as 20% replacement doughs have higher extensibility than 10% replacement doughs by 6 mm. Extensibility describes the flowability of doughs in extensional flow. The negative effects of FC0 and FC60 are likely to be caused by their strengthening effect which leads to higher resistance to flow. However, the slight increase in extensibility by FC0 at higher addition level might because of its fibrous structure where the fibres are much longer (intact) than FC60 (fibrillated).

The addition of both FC0 and FC60 increased dough strength and work of adhesion while further increasing the addition levels decreased these two parameters. The stickiness was decreased by the addition of both FC0 and FC60. The reduction of dough stickiness is in agreement with the additions of fibres in wheat doughs (Collar, Santos & Rosell, 2007; Sangnark & Noomhorm, 2004). Minimised stickiness is a desirable dough textual property in the manufacture of wheat bread (Collar et al., 2007), where it is also beneficial to the handling property. Generally, stickiness of a material is affected by both adhesive force and cohesive (rheological) force which could oppose each other (Hoseney & Smewing, 1999). The SMS/Chen-Hoseney Dough Stickiness method minimises the interference from the bulk rheology (Chen & Hoseney, 1995). Therefore, it can be expected that the bulk rheology property have less influence on dough stickiness measured in this experiment. Additionally, the adhesive force is influenced by water surface tension (Hoseney & Smewing, 1999). The influence of water absorption on the stickiness of wheat doughs has also been highlighted

(Armero & Collar, 1997; Heddleson, Hamann, Lineback & Slade, 1994). The influence on dough stickiness is contrary to the influences on dough firmness and cohesiveness. In fact, they show close negative correlations when fitted by power equations with R^2 of 0.97 and 0.94 respectively. Therefore, the high water absorbability of FC0 and FC60 contribute both to the strengthening of doughs, in addition to the contribution of the fibrous structure, and reduction of stickiness. However, dough strength and work of adhesion reflect both the adhesive force (stickness measured in this experiment) and cohesive force (bulk rheological, dough strengthening effect). Therefore, they increased with the replacements by FC0 and FC60 at lower addition levels but decreased coincidently with stickiness with further additions. A lower stickiness and work of adhesion suggests better workability and handling properties of the doughs.

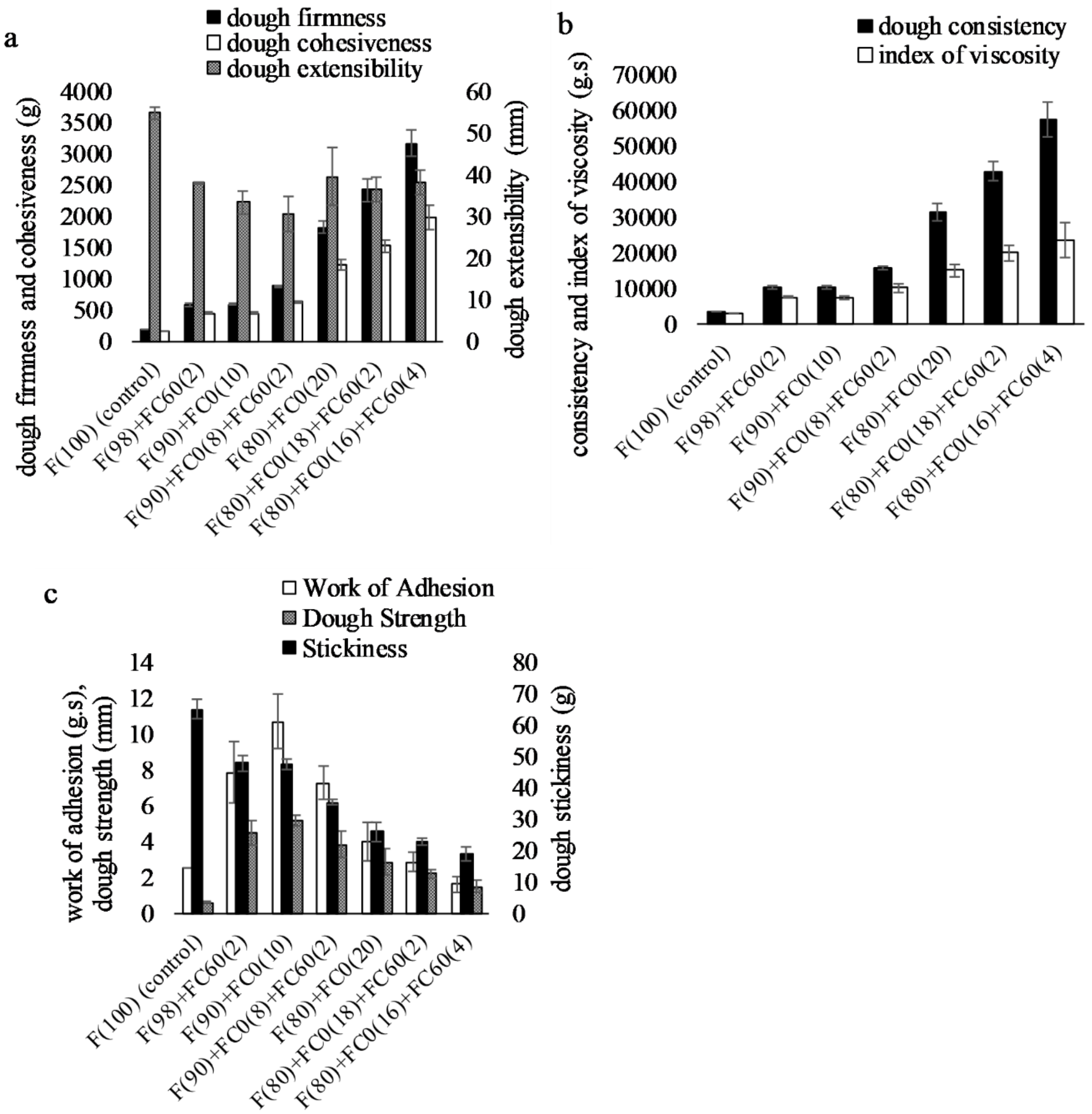

Figure 3. Effects of cellulose and FC fibres additions on dough firmness, Cohesiveness, extensibility (a), consistency, and index of viscosity (b) in backward extrusion measurements, and work of adhesion, dough strength, and dough stickiness (c) measured by SMS/Chen-Hoseney Dough Stickiness rig. Error bars represent the standard deviations averaging at least five replicates.

3.3. Proofing behaviours

The proofing behaviour of doughs are monitored by recording the volume increase of a piece of dough sample in a measuring cylinder and the proofing profiles are shown in Figure 4. The differences in volume between gluten free doughs are less significant during the first 30 minutes. However, with increasing addition of FC0 or FC60, volume growth stopped earlier during proofing which leads to lower final volume. Considering the correlations between low molecular weight sugars, yeast activity, and porosity kinetics (Romano, Toraldo, Cavella & Masi, 2007; Sahlström, Park & Shelton, 2004), sucrose in the formula might promote the volume increase during the early stage of proofing but be less influential after being consumed by yeasts during the later stage. The reduction of flour, instead, limits the further fermentation by yeast during the later stage of proofing. However, the difference between F(100) (control), F(90)+FC0(10), and F(98)+FC60(2) are less pronounced although the flour was replaced by only 2% and 10% respectively. Hence, the answer could also lie in rheological properties. As shown by the generalised Maxwell model, doughs behave like fluids during proofing and dough strengthening by FC0 and FC60 is less influential on the relaxation time. In addition, according to the model derived by Shah, Campbell, McKee and Rielly (1998), dough rheology is less influential during the early stage of proofing while it becomes critical during the later stage (Mills, Wilde, Salt & Skeggs, 2003). A rigid dough is highly resistant to deformation which limits the expansion of gas cells during proofing (Lazaridou et al., 2007; Van Vliet, Janssen, Bloksma & Walstra, 1992). Therefore, the influences of FC0 and FC60 on proofing behaviour during different stages can be also assigned to their strengthening effects on dough rheology. There is no difference between maximum volume increase and final volume increase indicating that doughs did not collapse during the proofing process even for F(80)+FC0(16)+FC60(4), whose volume did not change after 40 min of proofing. This stabilising effect is atributed to the dough strengthening by FC0 and FC60 additions as seen by the rheological measurements.

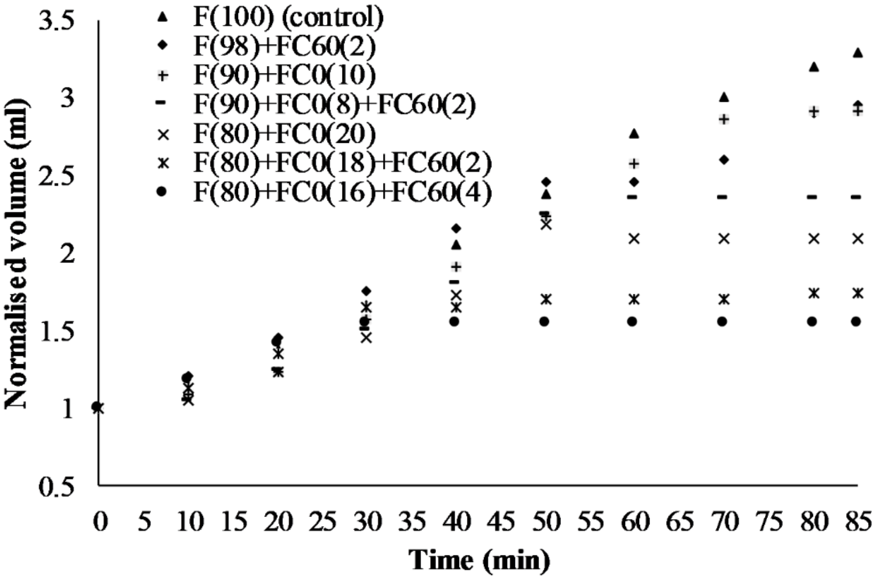

Figure 4. Proofing profiles (normalised) of gluten free doughs. Plots are shown as representative profiles from experiments run in triplicates.

3.4. Rheological properties during heating and cooling

Pasting profiles describe viscosity changes of flour blends during heating and cooling which typically describe the gelatinisation and retrogradation. The profiles are shown in Figure 5a. A secondary peak at 85 °C during cooling is observed at all curves due to the addition of psyllium, and possible to a balance between interaction with amylose (James M. Cowley, personal communication), formation of weak PSY gel particles, PSY particle interactions, and PSY particle breakdown (Ren, Linter & Foster, Unpublished results-a). The addition of FC0 and FC60 both significantly decreased the pasting temperature (the onset of viscosity increase) shown as a small shoulder before the main viscosity peak. The reduction of pasting temperature is widely observed for most starch/hydrocolloid mixtures (BeMiller, 2011; Naruenartwongsakul, Chinnan, Bhumiratana & Yoovidhya, 2004; Sullo & Foster, 2010). It is also observed for starch and bacterial cellulose mixtures (Díaz-Calderón et al., 2018). In the mixture of MC and starch, the thermal gelation of MC, which occurs at a lower temperature than the significant swelling of starch granules, leads to an increase in starch concentration and, therefore, enhanced starch granule interactions and increase in viscosity (Sullo & Foster, 2010). Naruenartwongsakul et al. (2004) suggested that the concentration increase of starch is due to water competition with MC. Therefore, water and volume competition might also explain the reduction of pasting temperature of FC0(FC60)/flour mixtures. When 20% of flour was replaced by FC0, the further addition of FC60 lead to another smaller shoulder at a lower temperature, which suggests that FC60 is more powerful than FC0 in the competition with starch for volume and water due to its higher abilities to hold water and to occupy volume (Agarwal et al., 2018a; Ren et al., 2020).

Replacement by FC0 decreased the overall viscosity, peak time and setback while it increased breakdown. The decrease of overall viscosity, including peak, and setback is, due to the decrease in starch concentration, as it is replaced by FC0 which does not swell as starch granules nor retrograde as amylose. The reduction of swellable starch granules leads to a reduction of the ability to be closely packed which appears as early onset of breakdown. The increased breakdown is likely to be due to the enhancement of shear force exerted on starch granules by the fibrous structure of FC0. However, the substitution by FC60 result in an increase in the overall viscosity and decrease in breakdown. Fibrillated cellulose (FC60) appears as flocculates or aggregates (Agarwal et al., 2018b; Ren et al., 2020) which can be considered similar to swollen starch granules or granule fragments with freed fibrils similar to leaked amylose. The increase of viscosity indicates that FC60 is similar to or even more efficient than starch in increasing viscosity while it does not breakdown as starch granules. Therefore, the functionality of fibrillated cellulose has an effective enhancement of overall composite properties beyond the effects of the ‘inert’ unfibrillated filler.

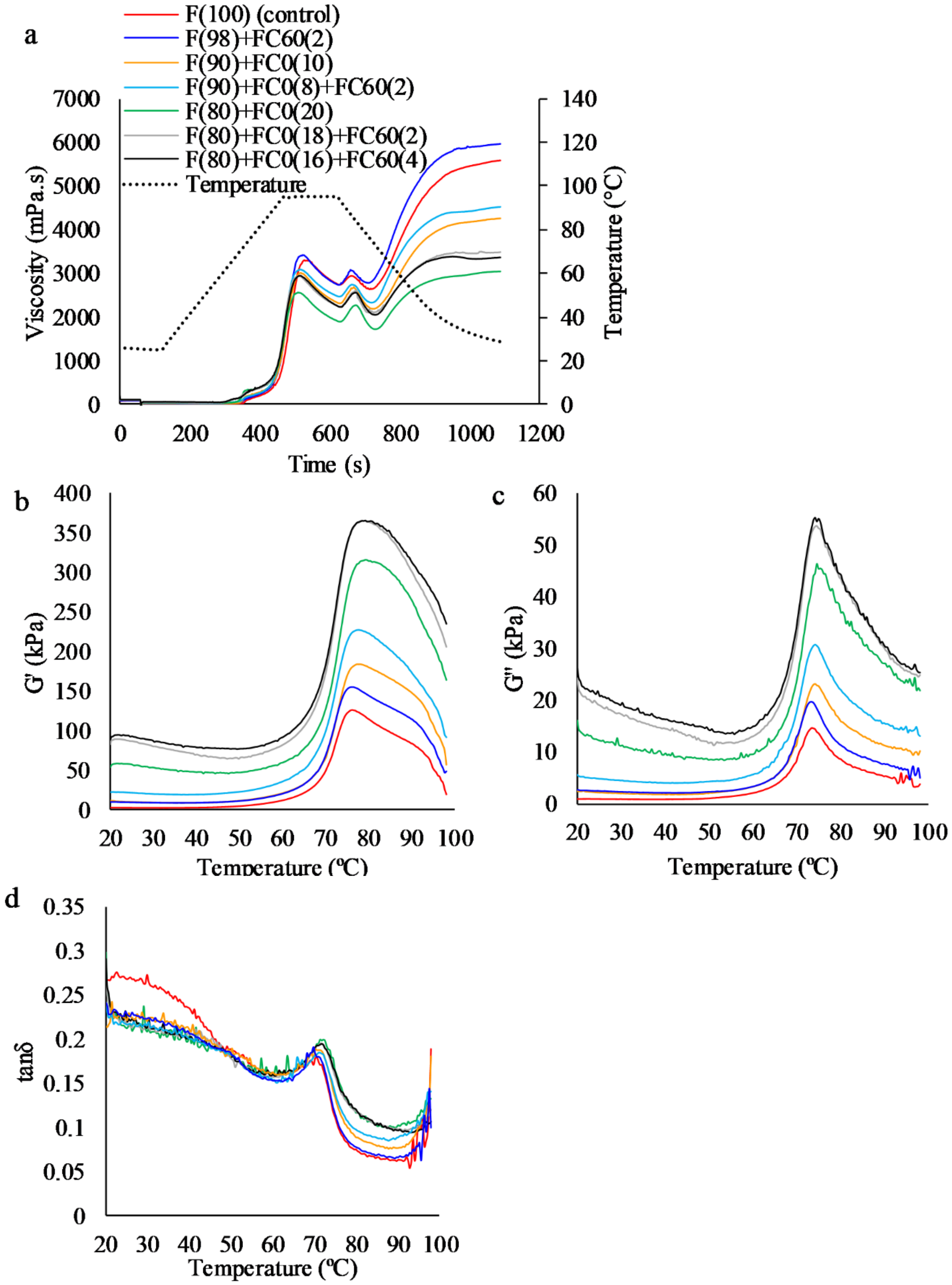

Figure 5. Pasting profiles (a), storage moduli G' (b), loss moduli G'' (c), and loss factor tanδ of flours blends. G' and G'' were recorded with a heating rate of 2.6 °C min^{-1}.

The rheological property of doughs during cooking was monitored by temperature sweep tests (Figure 5b, c, and d) with a temperature profile mimicking the temperature changes during baking. It can be seen that G’ and G’’ decreased slightly with the temperature increasing up to approximately 50 °C, especially with 20% replacement of flour by FC0 and FC60, which could be attributed to the softening induced by the increase of temperature. The initial G’ and G’’ decrease was accompanied by a moderate decrease in tanδ until approximately 60 °C, which can be attributed to slow swelling of starch granules which was not influenced by the replacement by FC0 and FC60. It can be speculated that the slow granule swelling overcomes the softening effect due to heating, therefore the temperature-induced softening (G’ and G’’ decrease) became more pronounced when flour was replaced by FC0 and FC60. With further increase in temperature, G’ and G’’ significantly increased to a peak because of a dramatic swelling and volume filling of starch granules which are eventually close-packed at the peaks in moduli. In contrast to pasting temperature, the onsets of G’ and G’’ increase shifted to a higher temperature when FC0 and/or FC60 were added. FC0/FC60 and flour were already closely packed with the higher concentration in doughs compared to the experimental condition of RVA. When the starch content was reduced, the starch granules needed to swell to a larger volume at a higher temperature to overcome the initial temperature-controlled softening (moduli decrease) and to contribute to the overall rigidity (G’ increase). It was also different from pasting properties that both the addition of FC0 or FC60 increased the peaks of G’ and G’’. This could also be due to the closely packed structure where the swelling and rigidity changes of starch granules can be detected and, as demonstrated by the rheological properties in both fundamental and empirical experiments, the fibrous structure and high water binding ability of FC0 and FC60. FC0 and FC60 competed for water with starch, which restrained the swelling of the granules, increased their rigidity, and, hence, further increased moduli. As for tanδ, it showed a peak at about 70 °C. Baldino et al. (2018) also observed the peaks of phase angle at 60.9 °C when gluten free doughs contained HPMC which is related to the phase separation of HPMC. However, comparing to their observation, tanδ peaks in Figure 5d had lower value and occurred at a higher temperature but MC has a lower gelation temperature than HPMC. Moreover, phase separation between amylose and amylopectin in baked wheat bread crumb has been reported (Hug-Iten, Handschin, Conde-Petit & Escher, 1999). Therefore, the tanδ peaks are possibly attributed to the phase separation between amylose and amylopectin. The reasons for G’ and G’’ decreasing after peaks might be the melting of remaining crystallites, separation of amylose and amylopectin, amylopectin matrix breakdown, and disentanglement of amylopectin chains, which lead to granule softening (Keetels, vanVliet & Walstra, 1996).

3.5. Bread qualities

The influence of the addition of FC0 and/or FC60 on specific volumes and baking loss of starch reduced gluten free bread are shown in Figure 6a. The influence on the moisture content of the centre crumb is insignificant which is not shown. The baking loss is significantly reduced upon the additions of FC0 and/or, especially, FC60, which could be attributed to their high water holding ability. It could also because of the denser crumb structure where water evaporation within the gas cells was restrained but water diffusion in the crumb matrix became more dominant. The FC0/FC60 additions significantly reduced the specific volume, which is the same as their influences on final proofing volume. The close correlation between loaf volume and final proofing volume is also documented in a previous study (Ren et al., Unpublished results-a), which suggests that loaves are stable during both the later stage of proofing and oven rising. The detrimental effect on specific volume of fibre addition and generation of denser crumb structure have also been observed in studies on both wheat bread and gluten free bread (Demirkesen et al., 2010; Gómez, Oliete, Caballero, Ronda & Blanco, 2008; Gomez, Ronda, Blanco, Caballero & Apesteguia, 2003).

The crumb was evaluated by C Cell and images are shown in Figure 7. Six C Cell parameters are chosen to describe the crumb structures (Table 5). The area of cells indicates the percentage of the cells of the total slice area and the number of cells is the number of discrete gas cells. The addition of FC0 and/or FC60 increased the number of cells but decreased the area of cells and wall thickness, and, consequently, reduced cell diameters. With the fact that the specific volume was decreased, which led to a smaller slice area, a finer crumb structure was obtained. Comparing to the addition of FC0, FC60 was more effective in altering these parameters. It can be attributed to the fibrous structure of FC0 and, especially, FC60, which formed a fine fibrous framework. The fibrous framework stabilised the porous structure of doughs and loaves and allowed the gelatinised starch to attach on and, therefore, reinforce the starch matrix. Additionally, area of holes and top concavity, which reflect the structural instability are not significant among all loaves.

The texture properties of bread were evaluated by TPA. Hardness, springiness, and cohesiveness are shown in Figure 6b. Specific volume is negatively correlated with hardiness due to a denser crumb structure. As expected, loaves with FC0 and FC60, which have lower specific volume, have a harder crumb. They also show lower springiness and cohesiveness, which indicates that the crumb is less resistant to the applied large deformation. Good wheat bread is expected to have a thinner cell wall and uniform cells which, therefore, has softer and more elastic texture providing good mouth feel (Scanlon & Zghal, 2001). However, due to the absence of gluten and the more compact crumb structure, the starch reduced gluten free loaves show the opposite that they have smaller cells, thinner cell walls, but harder and less springy crumb.

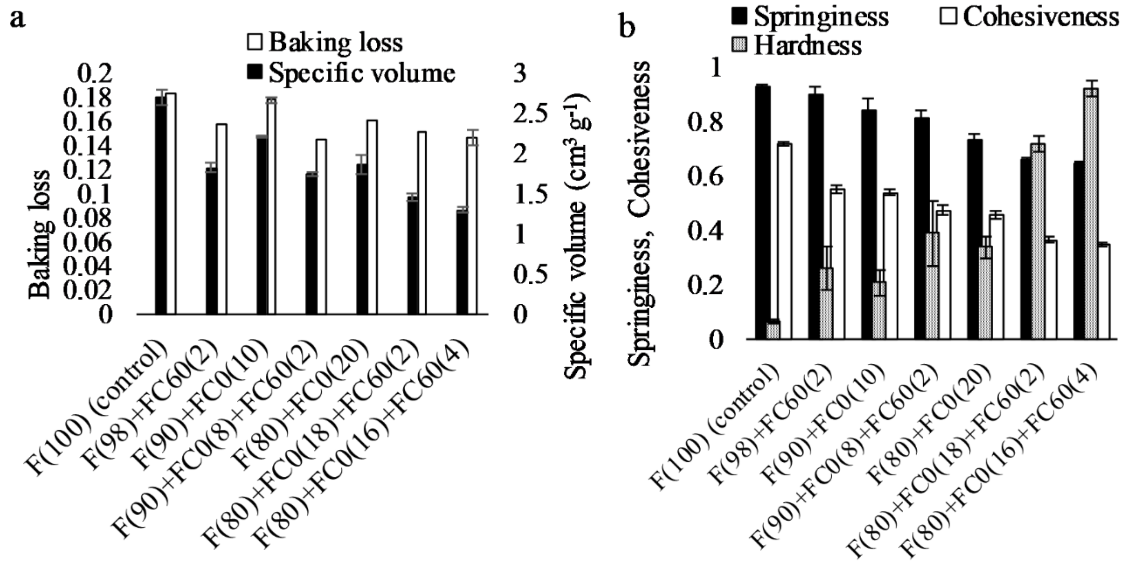

Figure 6. Baking lose, specific volume (a), and textural properties (b) of starch replaced gluten free breads. Error bars represent the standard deviations averaging four replicates.

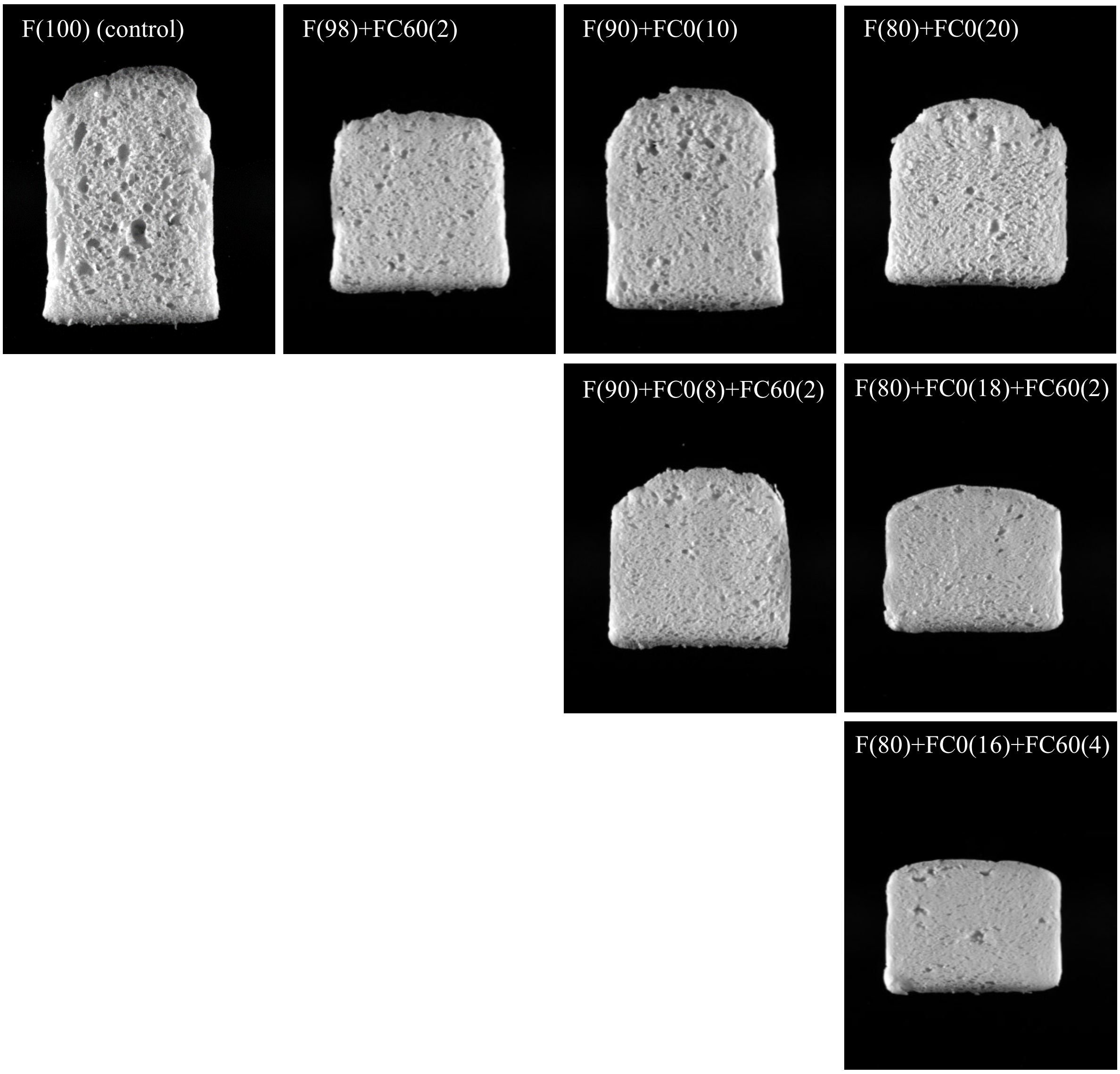

Figure 7. Images of starch replaced gluten free breads.

Table 5. C-Cell parameters of starch redplaced gluten free bread.

	Top Concavity (%)	Number of Cells	Area of Cells (%)	Area of Holes (%)	Wall Thickness (mm)	Cell Diameter (mm)
F(100) (control)	0.070.02^{a}	3523.25±27.90^{a}	50.83±0.96^{a}	2.41±0.39^{ac}	0.50±0.01^{a}	2.61±0.07^{a}
F(98)+FC60(2)	0.78±0.38^{b}	3986.75±91.51^{ab}	44.73±0.84^{b}	1.33±0.88^{abc}	0.44±0.01^{cd}	1.53±0.07^{b}
F(90)+FC0(10)	0.45±0.14^{bc}	3781.50±84.82^{ab}	48.20±0.57^{c}	1.43±0.81^{abc}	0.46±0.01^{b}	1.88±0.09^{c}
F(90)+FC0(8)+FC60(2)	0.56±0.07^{bc}	4033.50±69.00^{b}	45.00±0.43^{b}	0.85±0.48^{ab}	0.43±0.00^{d}	1.44±0.02^{b}
F(80)+FC0(20)	0.47±0.07^{bc}	3559.75±246.75^{a}	47.08±0.49^{c}	0.26±0.28^{b}	0.46±0.01^{bc}	1.76±0.13^{c}
F(80)+FC0(18)+FC60(2)	0.27±0.04^{ac}	4084.50±362.24^{b}	43.88±0.96^{bd}	1.29±0.82^{abc}	0.39±0.01^{e}	1.14±0.10^{d}
F(80)+FC0(16)+FC60(4)	0.21±0.04^{ac}	4065.00±274.07^{b}	42.73±0.78^{d}	2.56±0.94^{c}	0.38±0.01^{e}	1.09±0.06^{d}

Dara are shown as mean ± standard deviation. Values with different letters in the same column were significantly different ($p < 0.05$).

4. Conclusion

The conducted study aimed to evaluate the starch reduction of gluten free doughs and bread by FC0 and FC60 fibres. The fundamental dough rheological properties were analysed using the weak gel model and the generalised Maxwell model. The addition of FC0 and/or FC60 significantly increased the dough strength shown as increased viscosity, yield zone, and storage and loss moduli measured by fundamental rheological measurements, and dough firmness, cohesiveness, consistency and index of viscosity measured by empirical measurements. As the outcome of the fibrillation treatment, with high water absorbability and entangled fibrous aggregate structure, FC60 is more efficient than FC0 in altering the rheological properties. Further analysis of the mechanical spectra by generalised Maxwell model suggests that gluten free doughs are structurally and rheologically analogous to a viscoelastic fluid instead of a solid. It also suggests that doughs are flowable at the time scales of structure developing during proofing which is longer than their relaxation times. Generally, a comprehensive design of experiments including both fundamental and empirical analysis would be necessary to maximise the characterisation of doughs. Rheological properties of doughs influence the proofing behaviour where the addition of FC0/FC60 mainly restrained the volume increase during the later stage of proofing (after approximately 30 min). The fibrillated cellulose has an effective enhancement of overall composite properties beyond the effects of the 'inert' unfibrillated filler. Fibrillation of cellulose increased its similarity of pasting properties to flour in a cellulose/flour blend. Volume and water competition of FC0 and FC60 restricted the hydration and swelling of starch granules significantly influencing the pasting and thermal-mechanical behaviours of the blends. The additions of FC0 and FC60 decrease the specific volume of gluten free loaves, which is correlated with the decrease of final volume during proofing. The additions of FC0 and FC60 generate denser and harder but finer crumb. FC0 and, especially, FC60, play a role as a framework which stabilise the porous dough/crumb structure and reinforce the starch matrix. The further improvement of cellulose enriched gluten free bread might rely on the structuring of added hydrocolloids and fibrillated cellulose and optimisation of formulation including water addition levels to increase specific volume by obtaining desired rheological properties of doughs.

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