TITLE: Influence of buckwheat flour and carboxymethyl cellulose on rheological behaviour and baking performance of gluten-free cookie dough

AUTHORS: Tamara Dapčević Hadnađev, Aleksandra Torbica, Miroslav Hadnađev

NOTICE: This is the author's version of a work that was accepted for publication in *Food and Bioprocess Technology.* Changes resulting from the publishing process, such as peer review, editing, corrections, structural formatting, and other quality control mechanisms may not be reflected in this document. Changes may have been made to this work since it was submitted for publication. A definitive version was subsequently published in *Food and Bioprocess Technology*, Volume 6, Issue 7, July 2013, Pages 1770–1781. DOI: 10.1007/s11947-012-0841-6

Abstract

In the present study influence of buckwheat flour and carboxymethilcellulose (CMC) on the production of sheetable gluten-free cookie dough of acceptable rheological properties and subsequently their effect on the quality of gluten-free cookies was studied. The buckwheat flour was used to replace 10, 20 and 30% of rice flour in gluten-free formulations. Cookie doughs of 100% rice flour and 100% wheat flour served as control samples. The impact of CMC addition was examined on formulation containing 20% of buckwheat flour. Oscillatory and creep measurements were applied to test the effect of buckwheat flour and CMC on the viscoelasticity of gluten-free cookie dough. Frequency sweep results showed that all samples had solid elastic-like characteristics. Increase in the buckwheat flour addition led to decrease in storage modulus and zero shear viscosity and increase in tan δ and maximum creep compliance, while the addition of CMC led to increase in dough tenacity and resistance to deformation. Cookie dough containing 30% of buckwheat flour expressed the highest viscous properties, as revealed by relative viscous compliance value. The gluten-free dough containing CMC and buckwheat flour between 20 and 30% replacement level showed similar strength and extensibility as wheat cookie dough. The results of the physical and sensory evaluation of gluten-free cookies showed that buckwheat addition led to decrease in cookie hardness and fracturability and increase in eccentricity (deformation from regular shape) as well as the overall acceptability, as evaluated by untrained panellists.

Key words: gluten-free cookie, dough rheology, buckwheat, rice, carboxymethilcellulose

1. Introduction

In recent years, there is a trend of utilizing pseudocereals (amaranth, quinoa, buckwheat) in gluten-free bakery formulations in order to improve the nutritional profile of final products. The reason for that is greater awareness among the scientists and technologists regarding unbalanced diet of celiac patients. According a recently published study there is an unbalance in the intake of carbohydrates, fat and protein in the gluten-free diet (Alvarez-Jubete et al., 2010). Moreover, most of the commercially available cereal based gluten-free products contain lower levels of B-vitamins, iron and fiber in comparison to their gluten-containing counterparts (Alvarez-Jubete et al., 2010).

One of the pseudocereals which could be used as functional gluten-free alternative is buckwheat since it has proved to be a good source of essential amino acids, dietary fiber, vitamin B, vitamin E, calcium, magnesium and iron (Alvarez-Jubete et al., 2010). Moreover, ethanolic extracts of buckwheat flour exhibited better antioxidative properties than the ethanolic extracts of wheat flour (Sedej et al., 2011b). The special advantage of incorporating buckwheat flour into bakery products is its ability to maintain antioxidant capacity after thermal treatments (Sakač et al., 2011).

Therefore, a number of gluten-free bakery products containing buckwheat have been developed, such as gluten-free bread (Wronkowska et al., 2010; Torbica et al., 2010), biscuits (Schober et al., 2003), spaghetti (Verardo et al., 2011) and crackers (Sedej et al., 2011a). The gluten replacement in bakery products represents a major technological challenge due to its essential structure-building properties. Removal of gluten impairs dough structure to develop properly during kneading and baking. Therefore, its absence often results in a liquid batter rather than a dough, with a poor colour products and other quality defects after baking (Galagher et al., 2004). In order to provide structure and retain gas, substances which have the ability to imitate viscoelastic properties of gluten are commonly used. Gums and hydrocolloids are one of the most important ingredients in gluten-free formulation for improving the texture and appearance of the final products (Mariotti et al., 2009).

Unlike bread and pasta, gluten network in biscuits and cookies has to be only slightly developed (Schober et al., 2003), which allows greater diversification among nutritionally improved ingredients which could be used in gluten-free cookie manufacturing. Therefore, gluten-free cookies have the potential to be significant contributors of essential nutrients in the diet of celiac patients.

Cookies are baked products which are most commonly consisted of three major components, i.e. flour, sugar and fat and low final amount of water. These ingredients are mixed together with other minor components to form dough (Zucco et al., 2011). Dough making and handling, cookie baking and quality of the final product are thus largely influenced by cookie dough components (Pareyt & Delcour, 2008). Cookie spread, i.e. the extent to which the dough piece spreads during baking represents one of the major quality parameter (HadiNezhad & Butler, 2009). Generally cookie spread is associated to sugar, fat and protein content. Due to high fat and sugar content in cookie recipes, gluten network development is limited. However, proteins in wheat flour cookies are not functionally inert in cookie dough which is observed especially during the baking (Gaines, 1990). According to Pareyt et al. (2008) higher protein content results in decreased cookie spread. Cookie spread is also controlled by dough viscosity (HadiNezhad & Butler, 2009). Due to lower water content, cookie dough is generally more elastic and less extensible in comparison to bread dough. Since cookie dough rheology characterisation is related to dough handling properties and the dough tendency to contract, it is very important parameter in cookie quality evaluation (Pedersen et al., 2004).

Despite its importance, there is a lack in published research on rheological behaviour of gluten-free cookie dough. To the best of the Authors' knowledge, the only published papers concerning cookie or gluten-free dough rheology are those regarding wheat containing cookie dough (Pedersen et al., 2004) or gluten-free bread dough (Gural et al., 2003; Lazaridou et al., 2007).

The aim of this paper was to investigate the effect of substituting rice flour with buckwheat flour on the machineability of gluten-free cookie dough. Therefore, a step toward production of nutritionally improved gluten-free cookies which could be prepared in automated industrial processing systems was made. In order to achieve better sheetability, carboxymethilcellulose was also incorporated in formulation, since addition of hydrocolloids as gluten mimetics was already proven (Gallagher et al., 2004). The influence of buckwheat flour and carboxymethilcellulose on the production of gluten-free cookie dough of acceptable handling properties was evaluated by comparing their rheological behaviour to wheat-containing cookie dough properties. In order to study the structural aspects responsible for specific behaviour of rice flour/buckwheat flour/carboxymethilcellulose blends scanning electron microscopy was used for the integration of the information coming from rheological measurements. Subsequently, dimensions, texture, and sensory attributes of final products were evaluated.

2. Materials and methods

2.1. Materials

Rice flour, RF (moisture 10.6%, protein 7.7% db, starch 88.8% db, lipids 0.44% db and sugars 0.27% db), husked buckwheat flour, BF (moisture 11.3%, protein 12.3% db, starch 80.5% db, lipids 2.87% db and sugars 0.27% db) and wheat flour, WF (moisture 12.2%, protein 11.6% db, starch 84.1% db, lipids 1.31% db and sugars 1.44% db) were procured from Hemija Komerc, (Novi Sad, Serbia). Sodium carboxymethylcellulose, CMC was obtained from Alfa Aesar (Karlsruhe, Germany). Sodium bicarbonate, $NaHCO_3$ was purchased from Carl Roth (Karlsruhe, Germany) and DATEM - diacetyl tartaric acid esters of mono- and diglycerides of fatty acids (PANTEX DW 90) was purchased from Incopa (Illertissen, Germany). Vegetable fat-shortening (refined palm and sunflower oil) was obtained from Puratos (Groot-Bijgaarden, Belgium) while salt, granulated sugar and honey were purchased from local market.

2.2. Cookie-making procedure

In order to determine gluten-free cookie dough formulation, the ratio of principal ingredients was varied until the dough of good handling properties was made. The obtained recipe is presented in Table 1.

To examine the influence of buckwheat flour, rice flour was blended with 10, 20 and 30% of buckwheat flour. Cookie doughs of 100% rice flour and 100% wheat flour were prepared as control samples. Wheat containing cookies were produced without CMC. In order to get insight into the influence of carboxymethilcellulose, gluten-free formulation containing 20% of buckwheat flour without the addition of CMC was also prepared.

Cookie dough was made in a Farinograph mixing bowl (Brabender, Duisburg, Germany), at 30 °C. Flour, salt, sugar, sodium bicarbonate, DATEM and CMC were sifted together and mixed for 3 minutes. Subsequently, vegetable fat was added and mixed for additional 2 minutes. Finally, water containing honey was added to the resulting mass, and mixed for 25 min. The obtained cookie dough was rested for 24 h at 8 °C in order to achieve the hydratation of added CMC. Afterwards, the dough was tempered to room temperature and sheeted to a thickness of 4 mm with the help of a pilot scale dough sheeter (Mignon, Italy). Cookies were shaped using a cutter (60 x 55 mm) and baked at 170 °C for 12 min in a laboratory oven (MIWE gusto® CS, Germany). The baked cookies were cooled for 2 h and stored in polypropylene bags for further analysis.

2.3. Rheological testing of cookie dough

Rheological behaviour of cookie dough was determined immediately after the sheeting process, using a HAAKE Mars rheometer (Thermo Scientific, Karlsruhe, Germany). The rheometer was equipped with a 35 mm parallel plate measuring geometry. The plates were serrated in order to prevent the dough slippage. A dough sample was placed on the lower plate, and the upper plate was lowered until the gap of 1.0 mm was reached. The excess dough was trimmed and the edges were sealed with a paraffin oil to prevent the dough from drying during measurements. The sample was left to rest for 10 min before measurements, so that residual stresses could relax. All experiments were performed at 30 °C in triplicates.

Frequency sweeps test was carried out from 0.1 to 50 Hz, under a 5 Pa strain level, which was within a linear viscoelastic region of all cookie dough samples. The data of frequency sweeps were plotted as $G'(f)$ and $G''(f)$ in double logarithmic diagram and experimental data of G' vs. f were fitted using the following equation:

$$G'(f)=K'f^{n'}$$

where G' is storage (elastic) modulus, K' is coefficient which represent the storage modulus at 1 Hz (Peressini et al., 2000) and n' is coefficient which represents the slope of the curve in a log-log plot of G' versus the frequency (Sivaramakrishnan et al., 2004). The values of tan δ, which represents the ratio of energy lost or dissipated (G'') to energy stored in the material and recovered from it per cycle of sinusoidal deformation (G') were also reported.

Since frequency sweep test is a small deformation test which does not destroy the dough structure, creep-recovery test was conducted immediately after frequency sweep measurements on the same dough sample. Creep was measured at a shear stress of 50 Pa for 300 s, followed by a recovery phase of 900 s at a stress of 0 Pa. Namely, initial experiments confirmed that the tested cookie dough reached a steady viscous flow in this time range.

Creep data were described by Burgers model parameters, which can be expressed using the following equation:

$$J(t)=J_0(t)+J_r(t)+J_n(t)$$

or

$$J(t)=J_0+J_m(1-exp(-t/\lambda))+t/\eta_0$$

where J_0 is the instantaneous compliance, J_m is the viscoelastic compliance, λ is the mean retardation time and η_0 is the zero shear viscosity. Maximum creep compliance, J_{max}, elastic compliance, J_e and viscous compliance, J_v were extracted from recovery curve. The parameters are also presented in Figure 1.

2.4. Scanning electron microscopy (SEM)

In order to prepare cookie dough samples for scanning electron microscopy imaging, the procedure described by Ribotta et al. (2004) was followed. The samples were dried using CPD 030 BAL-TEC Critical Point Dryer (BAL-TEC AG, Liechtenstein) following coating with gold (180 s / 30 mA / 50 mm distance) in a SCD 005 BAL-TEC Ionic Sputter Coater (BAL-TEC AG, Liechtenstein). The images were obtained using the Jeol JSM 6460LV Scanning Electron Microscope (Tokyo, Japan) with a 25 kV acceleration voltage. The micrographs were taken using the magnification of 1000×.

2.5. Textural properties of cookies

Cookie break strength was measured by a TA.XTPlus Texture analyzer (Stable Micro Systems, UK) using a 3-Point Bending Rig (HDP/3PB) and 5 kg load cell in compression mode at a test speed of 3 mm/s and the gap distance of the base plate of 55 mm. Textural analyses were conducted after 24 h, at 20 °C, in nine replicates per batch.

2.6. Dimensional properties of cookies

Dimensional characteristics of cookies were described as % contraction in the direction of sheeting, % spread perpendicular to the direction of sheeting which represent changes in dimensions after cutting and baking; and width and length eccentricity which represent deformation from rectangular shape of the final product.

Cookies were evaluated for the contraction of the dough (% contraction) in the direction of sheeting, the spread (% spread) perpendicular to the direction of sheeting and the width and length eccentricity which were calculated as:

$$\% \text{ contraction} = \frac{W_m - W}{W_m} \cdot 100 \quad \text{and} \quad \% \text{ spread} = \frac{L - L_m}{L_m} \cdot 100$$

$$\text{width eccentricity} = \frac{\text{width of the cookie centre}}{\text{width of the cookie edges}}$$

$$\text{length eccentricity} = \frac{\text{length of the cookie centre}}{\text{length of the cookie edges}}$$

where W and L are the width and length of six randomly selected cookies W_m (55 mm) and L_m (60 mm) are the width and length of the mold used.

2.7. Sensory evaluation of cookies

The sensory evaluation of cookies was performed 24 h after baking by a panel of 20 consumers, both male and female. A 5-point hedonic rating scale was used to evaluate the overall acceptability of the cookie sample, with "1" being "dislike very much", "2" being "dislike moderately", "3" being "neither like nor dislike", "4" being "like moderately " and "5" being "like very much" (Moskowitz et al., 2004). Three coded samples per cookie formulation were tested. Cookies were considered acceptable if their mean scores for overall acceptability were above 3.

2.8. Statistical analysis

Data were analyzed by one-way analysis of variance with Tukey's test, which was performed using Statistica 8.0 (Statsoft, Tulsa, USA). The significance of differences among the mean values was indicated at the 95% confidence level.

3. Results and discussion

3.1. Properties of gluten-free doughs

According to mechanical spectra of analysed cookie doughs (Figure 2), storage modulus, G' was higher than loss modulus, G'' (tan δ <1) in the examined frequency range, indicating solid elastic-like behaviour of gluten-free cookie doughs.

The prevalence of elastic properties over viscous has been reported for gluten-free bread dough containing rice flour (Lazaridou et al., 2007; Sivaramakrishnan et al., 2004; Torbica et al., 2010), as well as for buckwheat flour dough (Han et al., 2011). However, since cookie dough is characterized by low moisture and high fat and sugar content compared to bread dough, it exhibited higher elastic modulus than gluten-free bread dough composed of rice and buckwheat flour (Torbica et al., 2010). According to previously conducted studies, addition of fat (Gujral et al., 2003) and reduction in water level in both gluten-free (Lazaridou et al., 2007) and wheat (Phan-Thien & Safari-Ardi, 1998; Edwards et al., 1999) dough led to increase in dough elastic modulus. Frequency sweep test has also showed a frequency dependence of both G' and G'' modulus. In order to express the magnitude of the dependence of storage modulus on oscillation frequency, the curves were fitted to power-low equation and the obtained coefficients are presented in Table 2. Mean values of tan δ (G''/G') from 1 to 10 Hz are also shown in Table 2. According to the results summarized in Table 2, rice cookie dough had higher values of K' (storage modulus at 1 Hz) and lower values of tan δ in comparison to other tested samples expressing the properties of rigid and stiff material (Weipert, 1990). Moreover, the value of n' was lower for rice dough than for wheat cookie dough indicating its frequency independent structural stability (Mohammed et al., 2011). Investigating the feasibility of rice dough for making rice bread, Sivaramakrishnan et al. (2004) have also revealed that rice dough exhibited higher dynamic moduli and lower frequency dependence than wheat flour. Addition of buckwheat flour in dough system decreased elastic modulus and increased tan δ, suggesting that presence of buckwheat flour lowered the strength and elasticity of gluten-free cookie dough. On contrary, addition of CMC resulted in a significant rise of storage modulus.

The creep curve analysis for both creep phase and the recovery phase are also given in Table 2. In general, under the applied stress of 50 Pa which did not exceed the linear viscoelastic range, rice cookie dough exhibited the greater resistance to deformation as shown by the reduction of maximum creep compliance. Partial replacement of rice flour with buckwheat flour led to rise in maximum creep compliance values, thus increasing dough extensibility. A significant increase in viscoelastic compliance and slight increase in instantaneous compliance were also noticed with the increase in buckwheat addition level, while the zero shear viscosity was lower. According to Edwards et al. (2001) zero or steady-state viscosity decreases with reduction in dough strength measured by Extensograph. Creep results were in accordance with oscillation results which also revealed the decrease in dough strength with increase in buckwheat flour content. Higher thermo-mechanical weakening of buckwheat flour dough in comparison to rice dough during dough kneading was reported earlier (Torbica et al., 2008; Dapčević Hadnađev et al., 2011). Results concerning the dough elastic properties (tan δ in frequency sweep test and relative elastic compliance in creep-recovery test) were also in accordance. During the recovery phase, recovered deformation which was presented as relative elastic compliance, was higher for pure rice dough than for buckwheat supplemented doughs. However, this difference in dough elastic character was only significant at 30% buckwheat replacement level. Moreover, among the tested samples, cookie dough containing 30% of buckwheat flour expressed the highest viscous properties, as revealed by relative viscous compliance value.

In order to determine the structural organization in cookie dough samples responsible for differences in dough strength and elasticity, SEM imaging was performed (Figure 3). Namely, Edwards et al. (2001) have revealed that strength of durum wheat dough, expressed as high steady-state viscosity and low extensibility, primarily depends on density of physical crosslinks present. Since in gluten-free dough gluten complex can not be formed, the differences in dough strength may be ascribed to differences in size and shape of their native starch granules (Singh et al., 2003).

Micro-structural observation of composite dough (containing 10 and 30% of buckwheat flour) revealed that buckwheat starch granules disturbed the structure of rice dough starch granule network which can influence the dough weakening determined during rheological tests. Gluten-free dough structures (Figure 3 a, b) were characterized by the regions of densely packed rice starch granules (2–9 µm) which were partially covered with proteins and CMC. This continuous phase was disrupted with the regions of irregular in shape small starch granules (< 7 µm) grouped in the clusters which is a characteristic of buckwheat flour (Mariotti et al., 2008; Hatcher et al., 2008). Furthermore, it can be observed that the clusters with the CMC network fragments and rice proteins fragments on the surfaces are more noticeable for gluten free-dough with 30% of buckwheat flour. In order to compare gluten-free doughs with gluten-containing dough, wheat cookie dough micrographs (Figure 3 c) were also taken. Wheat dough was characterized by the presence of small (< 10 µm) and large starch granules (>15 µm) which are densely distributed in partially formed protein matrix. These findings were in accordance with Kim et al. (2003) and Naruenartwongsakul et al. (2008). However, according to obtained micrographs starch granules in gluten-free doughs as well as in wheat flour dough were not completely covered with protein and CMC matrix due to less water content in cookie formulation in comparison to common bread formulation where the higher hydratation led to continuous protein network formation. This was in agreement with Létang et al. (1999) who reported that in bread dough, starch granules were covered with a continuous protein film and were less visible than in less hydrated doughs such as cookie dough.

Addition of CMC led to increase in dough firmness as revealed by decrease in maximum creep compliance and increase in zero shear viscosity and *G'*. The similar results were obtained for gluten-free bread dough (Lazaridou et al., 2007). Possible reasons for the increase in cookie dough firmness might be that (1) addition of CMC improved the starch granules cohesiveness, and (2) due to increase in water absorption level in CMC containing sample dough consistency increase.

In general, the gluten-free dough containing CMC and buckwheat flour between 20 and 30% showed similar storage modulus, instantaneous and maximum creep compliance, zero shear viscosity and recovered deformation values to that of wheat cookie dough. Therefore, it could be concluded that addition of buckwheat flour and CMC in rice dough resulted in gluten-free cookie dough of acceptable handling properties. Namely, inclusion of buckwheat flour into gluten-free cookie dough led to soft, viscous, deformable dough, easy to handle in comparison to rice dough, but due to presence of CMC, it was strong enough to resist sheeting without sticking to rollers and maintain acceptable shape.

3.2. Properties of gluten-free cookies

In order to determine the quality of gluten-free cookies, physical (texture, contraction and spread, eccentricity) and sensory (top surface cracking and overall acceptability) attributes of final products were evaluated.

Unlike the rheological experiments, quality of gluten-free cookies that did not contain CMC could not be evaluated. The absence of CMC resulted in cookie dough which was insufficiently cohesive for handling and shaping. Therefore, the dough without CMC addition, which was shaped with a rolling pin, resulted in cookies of irregular shape and more cracked surface than cookies containing CMC (Figure 4). Moreover, wheat containing cookies were also excluded from further evaluation although their dough was characterized with good machineability. Namely, in order to optimize the gluten-free formulations which will result in cookies of acceptable quality, amount of added water and fat were higher in comparison to standardized wheat dough cookie formulation, which made the comparison to wheat control cookie irrelevant. In addition, the consciously comparison of gluten-free to wheat cookies was avoided during hedonic test.

In general, the characteristics of a high quality cookie are the adequate hardness (enough to maintain its shape during transportation but able to fracture easily when chewed in the mouth), a high spread ratio (diameter/thickness), low eccentricity, brownish colour, attractive appearance (no surface cracks) and pleasant flavour.

The effect of buckwheat incorporation onto textural properties of gluten-free cookies is summarized in Table 3, whereas dimensional changes of cookies are presented in Figure 5. Textural properties of cookies were expressed as hardness/firmness and fracturability/brittleness. Maximum peak force recorded from force/distance curve (the maximum force required to break a cookie or maximum resistance of cookie when break) has been reported as hardness, firmness or breaking strength (Bourne, 2002; Mamat et al., 2010; Pareyt et al., 2009). Fracturability (Bourne, 2002) or brittleness (Wekwete & Navder, 2008) has been determined as peak distance which represents the distance travelled by the blade through the cookie in mm before the cookie will break or the distance the cookie will deform before breaking. According to the results presented in Table 3, partial replacement of rice with buckwheat flour led to decrease in cookie hardness and fracturability. However, no significant difference in cookie hardness and brittleness was observed with the increase in buckwheat flour replacement level. The experiments performed by Mamat et al. (2010), Pareyt et al. (2009) and Lee et al. (2005) revealed that wheat cookies containing optimal amount of fat and sugar have the hardness and brittleness values in the range 20 to 30 N and 0.65 to 0.9 mm, respectively, which is in agreement with the values obtained for buckwheat containing cookies. Therefore, it could be expected that cookies enriched with buckwheat flour would be more acceptable by consumers in terms of texture since high fracturability of rice cookies could have negative impact on cookie chewiness.

Dimensional characteristics of biscuits (Figure 5) were described as % contraction in the direction of sheeting, % spread perpendicular to the direction of sheeting which represent changes in dimensions after cutting and baking; and width and length eccentricity which represent deformation from rectangular shape of the final product. Similar dimensional properties were used by Pedersen (2004) in order to evaluate the baking characteristics of different wheat cultivars, with slight modification due to difference in cookie shape. As it can be seen from Figure 5, dimensions of cookies were strongly affected ($p < 0.05$) by buckwheat flour addition. Increase in buckwheat flour substitution level led to decrease in % contraction and increase in % spread and eccentricity.

The results obtained from textural and dimensional measurements of cookies were in accordance with the rheological properties of cookie dough. Rice dough which was stronger (higher elastic modulus, lower maximum creep compliance) and more elastic (lower tan δ, higher recovery) yielded harder cookies which were more shrunk and less deformed from regular shape than buckwheat containing cookies. Schober et al. (2003) have also revealed that rheological properties of cookie dough determine the quality of final product. Accordingly, a firm, non-sticky dough results in firm and thin round biscuits whereas soft, sticky dough yields soft and thick oval biscuits. Wani et al. (2010) have also reported that less extensible cookie doughs resulted in harder cookies. On the other hand, Maache-Rezzoug et al., (1998) have found that shrinkage after cutting increases with dough elasticity, which results in decreased biscuit length. Therefore, the addition of buckwheat flour, which influenced a decrease in dough elasticity, led to increase in cookie spread. Increased cookie spread of buckwheat enriched cookies can be also ascribed to the differences in rice and buckwheat starch gelatinization. As it was already shown, the addition of buckwheat to rice flour lowers the maximum peak torque during gelatinization (Torbica et al., 2010), which lead to decrease in dough viscosity and increase in cookie spread (Tsen et al, 1975).

In order to determine the overall acceptability of gluten-free cookies, the sensory evaluation was performed. The sensory characteristics of the cookies were screened by untrained panellists using a five point hedonic scale (Figure 6). Although the results obtained by untrained panellists are usually less precise and replicable (Barylko-Pikielna & Matuszewska, 1996) than those obtained by trained panellists, the judgment of untrained panellists, as future consumers, is certainly very valuable (Arifin et al., 2010) before launching a new food product into the market. According to untrained panellist evaluation, all gluten-free cookie formulations were acceptable, since they received scores much higher than 3 ranging from 3.5 to 4.2. Even though rice cookies were visually superior to those containing buckwheat flour in terms of eccentricity (Figure 4), buckwheat enriched cookies were rated higher for overall acceptability. Sensory characteristics influencing higher scores for overall acceptability were pleasant smell and taste of cookies prepared from buckwheat flour. Moskowitz & Krieger (1995) have shown that taste/flavour sensory attribute is more important than appearance. By ranking the importance for attribute liking Moskowitz & Krieger (1995) have obtained the following order: taste/flavour > texture > appearance.

It has been previously shown that incorporation of buckwheat flour in either gluten-free bread (Torbica et al., 2010) or wheat containing bread (Lin et al., 2009) and pasta (Jambrec et al., 2011) can increase the flavour and mouth feel sensory attributes of the product compared to controls containing only rice or wheat flour. However, during investigation of the influence of buckwheat flour on sensory properties of Turkish noodles, Bilgiçli (2009) revealed that taste scores increased up to the 20% buckwheat flour level while the further increase in buckwheat flour content led to a decrease in taste scores. The results presented in this study have also shown that increase in buckwheat flour content from 20 to 30% did not significantly influence ($p > 0.05$) the overall acceptability of gluten-free cookies.

4. Conclusions

Production of gluten-free cookies of acceptable processing properties and preferred sensory and textural characteristics was performed. The aim of the present study was to investigate the influence of buckwheat flour and CMC addition on rheological parameters and quality of gluten-free cookies.

As revealed by decrease in maximum creep compliance and increase in zero shear viscosity and *G'*, the addition of CMC resulted in increased dough strength. Namely, the absence of CMC led to cookie dough of insufficient cohesion for handling and shaping, which resulted in cookies of irregular shape and more cracked surface than cookies containing CMC. Rice cookie dough expressed higher storage modulus at frequency 1 Hz, lower values of tan δ and the greater resistance to deformation than the wheat and buckwheat containing doughs. The addition of buckwheat flour led to rise in maximum, viscoelastic and instantaneous compliances and increase in tan δ, while the zero shear viscosity and storage modulus were lower. Consequently, the inclusion of buckwheat flour into gluten-free rice dough decreased dough elasticity and extensibility. In general, the gluten-free dough containing CMC and buckwheat flour between 20 and 30% substitution level resembled the wheat cookie dough in terms of dough strength and resistance to deformation. Moreover it was found that creep results were in accordance with oscillation results which also revealed the decrease in dough strength with increase in buckwheat flour content. The addition of buckwheat flour and CMC in rice dough resulted in soft, viscous, deformable dough which was easy to handle and strong enough to resist sheeting and maintain acceptable shape. Microstructural observation revealed that gluten-free dough structures were characterized by the regions of densely packed rice starch granule structure which was disrupted with the regions of irregular in shape small starch granules grouped in the clusters.

This study also showed that partial replacement of rice with buckwheat flour led to decrease in cookie hardness, fracturability and % contraction and increase in % spread and eccentricity. Moreover the results from textural and dimensional measurements of cookies were in accordance with the rheological characteristics of cookie dough. Buckwheat supplemented dough which was softer and more viscous (lower elastic modulus, higher maximum creep compliance, higher tan δ, lower recovery) yielded softer and more brittle cookies which were more deformed from regular shape than control rice cookies. According to the sensory evaluation, performed by untrained panellists, buckwheat enriched cookies were rated higher for overall acceptability.

References

Alvarez-Jubete L, Arendt EK & Gallagher E (2010) Nutritive value of pseudocereals and their increasing use as functional gluten-free ingredients. Trends in Food Science & Technology, 21, 106-113.

Arifin N, Peng KS, Long K, Ping TC, Affandi Yusoff MS, Nor Aini I & Ming LO (2010) Relationship between textural properties and sensory qualities of cookies made from mediumand long-chain triacylglycerol-enriched margarines. Journal of the Science of Food and Agriculture, 90(6), 943-948.

Barylko-Pikielna N & Matuszewska I (1996) Progress in sensory analysis and consumer studies of food. Polish Journal of Food and Nutrition Science, 5/46(4), 3–18.

Bilgiçli N (2009) Effect of buckwheat flour on cooking quality and some chemical, antinutritional and sensory properties of erişte, Turkish noodle. International Journal of Food Sciences and Nutrition, 60(S4), 70-80.

Bourne MC (2002) Food texture and viscosity: concept and measurement (2nd edition). Academic Press, Elsevier Science, London, UK.

Dapčević Hadnađev T, Torbica A & Hadnađev M (2011) Rheological properties of wheat flour substitutes/alternative crops assessed by Mixolab, In: Procedia Food Science 1, 11th International Congress on Engineering and Food (ICEF11), 22-26 May 2011, Athens, Greece, 328 – 334.

Edwards NM, Dexter JE, Scanlon MG, Cenkowski S (1999) Relationship of creep-recovery and dynamic oscillatory measurements to durum wheat physical dough properties. Cereal Chemistry, 76, 638–645.

Edwards NM, Peressini D, Dexter JE & Mulvaney SJ (2001) Viscoelastic properties of durum wheat and common wheat dough of different strengths, Rheologica Acta, 40, 142–153.

Gaines CS (1990) Influence of chemical and physical modification of soft wheat protein on sugar-snap cookie dough consistency, cookie size, and hardness. Cereal Chemistry, 67, 73-77.

Gallagher E, Gormley TR & Arendt EK (2004) Recent advances in the formulation of gluten-free cereal-based products. Trends in Food Science and Technology, 15, 143–152.

Gujral HS, Guardiola I, Carbonell JV & Rosell CM (2003) Effect of cyclodextrinase on dough rheology and bread quality from rice flour. Journal of Agricultural and Food Chemistry, 51, 3814–3818.

HadiNezhad M & Butler F. (2009) Effect of flour type and dough rheological properties on cookie spread measured dynamically during baking. Journal of Cereal Science, 49 (2), 178-183.

Han L, Cheng Y, Qiu S, Tatsumi E, Shen Q, Lu Z & Li L (2011) The Effects of Vital Wheat Gluten and Transglutaminase on the Thermomechanical and Dynamic Rheological Properties of Buckwheat Dough. Food and Bioprocess Technology, DOI 10.1007/s11947-011-0738-9.

Hatcher DW, You S, Dexter JE, Campbell C & Izydorczyk MS (2008) Evaluation of the performance of flours from crossand self-pollinating Canadian common buckwheat (Fagopyrum esculentum Moench) cultivars in soba noodles. Food Chemistry, 107, 722–731.

Jambrec D, Pestorić M, Psodorov Đ, Sakač M, Nedeljković N, Mandić A & Sedej I (2011) Instrumental and sensory properties of buckwheat flour pasta. Food and Feed Research, 38(2), 45-50.

Kim H-J, Morita N, Lee S-H & Moon K-D (2003) Scanning electron microscopic observations of dough and bread supplemented with Gastrodia elata Blume powder. Food Research International, 36, 387–397.

Lazaridou A, Duta D, Papageorgiou M, Belc N & Biliaderis CG (2007) Effects of hydrocolloids on dough rheology and bread quality parameters in gluten-free formulations. Journal of Food Engineering, 79, 1033–1047.

Lee S, Warner K & Inglett GE (2005) Rheological Properties and Baking Performance of New Oat β-Glucan-Rich Hydrocolloids. Journal of Agricultural and Food Chemistry, 53(25), 9805–9809.

Létang C, Piau M & Verdier C (1999) Characterization of wheat four-water doughs. Part I: Rheometry and microstructure. Journal of Food Engineering, 41, 121-132.

Lin L-Y, Liu H-M, Yu Y-W, Lin S-D & Mau J-L (2009) Quality and antioxidant property of buckwheat enhanced wheat bread. Food Chemistry, 112(4), 987-991.

Maache-Rezzoug Z, Bouvier J-M, Allaf K & Patras C (1998) Study of mixing in connection with the rheological properties of biscuit dough and dimensional characteristics of biscuits. Journal of food engineering, 35, 43-56.

Mamat H, Abu Hardan MO & Hill SE (2010) Physicochemical properties of commercial semisweet biscuit. Food chemistry, 121, 1029-1038.

Mariotti M, Lucisano M, Pagani MA & Iameti S (2008) Macromolecular Interactions and Rheological Properties of Buckwheat-Based Dough Obtained from Differently Processed Grains. Journal of Agricultural and Food Chemistry 56, 4258-4267.

Mariotti M, Lucisano M, Pagani MA & Perry K (2009) The role of corn starch, amaranth flour, pea isolate, and Psyllium flour on the rheological properties and the ultrastructure of gluten-free doughs. Food Research International, 42, 963–975.

Mohammed I, Ahmed AR & Senge B (2011) Dynamic rheological properties of chickpea and wheat flour dough's. Journal of Applied Sciences, 11(19), 3405-3412.

Moskowitz HR & Krieger B (1995) The contribution of sensory liking to overall liking: An analysis of six food categories. Food Quality and Preference, 6(2), 83–90.

Moskowitz HR, Muñoz AM, Gacula MC (2004) Viewpoints and Controversies in Sensory Science and Consumer Product Testing, John Wiley & Sons, Connecticut, USA.

Naruenartwongsakul S, Chinnan MS, Bhumiratana S & Yoovidhya T (2008) Effect of cellulose ethers on the microstructure of fried wheat flour-based batters. LWT-Food Science and Technology, 41, 109–118.

Pareyt B & Delcour JA (2008) The role of wheat flour constituents, sugar and fat in low moisture cereal based products: a review on sugar-snap cookies. Critical Reviews in Food Science and Nutrition, 48 (9), 824-839.

Pareyt B, Wilderjans E, Goesaert H, Brijs K & Delcour JA (2008) The role of gluten in a sugar-snap cookie system: A model approach based on gluten–starch blends. Journal of Cereal Science, 48 (3), 863-869.

Pareyt B, Talhaoui F, Kerckhofs G, Brijs K, Goesaert H, Wevers M & Delcour JA (2009) The role of sugar and fat in sugar-snap cookies: Structural and textural properties. Journal of Food Engineering, 90, 400–408.

Pedersen L, Kaack K, Bergsøe MN & Adler-Nissen J (2004) Rheological properties of biscuit dough from different cultivars, and relationship to baking characteristics. Journal of Cereal Science, 39, 37–46.

Peressini D, Sensidoni A, Pollini CM & De Cindio B (2000) Rheology of wheat doughs for fresh pasta blends and salt content production: influence of semolina-flour blends and salt content. Journal of Texture Studies, 31 (2), 163-182.

Phan-Thien N & Safari-Ardi M (1998) Linear viscoelastic properties of flour–water doughs at different water concentrations. Journal of Non-Newtonian Fluid Mechanics, 74, 137–150.

Ribotta PD, Pérez GT, León AE & Añón MC (2004) Effect of emulsifier and guar gum on micro structural, rheological and baking performance of frozen bread dough. Food Hydrocolloids, 18, 305–313.

Sakač M, Torbica A, Sedej I & Hadnađev M (2011) Influence of breadmaking on antioxidant capacity of gluten free breads based on rice and buckwheat flours. Food Research International, 44(9), 2806-2813.

Schober TJ, O'Brien CM, McCarthy D, Darnedde A & Arendt EK (2003) Influence of gluten-free flour mixes and fat powders on the quality of gluten-free biscuits. European Food Reearch and Technology, 216, 369–376.

Sedej I, Sakač M**,** Mandić A**,** Mišan A**,** Pestorić M**,** Šimurina O**,** Čanadanović-Brunet J (2011a) Quality assessment of gluten-free crackers based on buckwheat flour. LWT - Food Science and Technology, 44(3), 694–699.

Sedej I, Sakač M, Mandić A, Mišan A, Tumbas V & Hadnađev M (2011b) Assessment of antioxidant activity and rheological properties of wheat and buckwheat milling fractions. Journal of Cereal Science, 54(3), 347-353.

Singh J, Singh N, Sharma TR & Saxena SK (2003) Physicochemical, rheological and cookie making properties of corn and potato flours. Food Chemistry, 83, 387–393.

Sivaramakrishnan HP, Senge B & Chattopadhyay PK (2004) Rheological properties of rice dough for making rice bread. Journal of Food Engineering, 62, 37–45.

Torbica A, Hadnađev M, Dokić P, Sakač M (2008) Mixolab profiles of gluten free products ingredients. Food Processing, Quality and Safety, 35(1), 19-26.

Torbica A, Hadnađev M & Dapčević T (2010) Rheological, textural and sensory properties of gluten-free bread formulations based on rice and buckwheat flour. Food Hydrocolloids, 24(6-7), 626-632.

Tsen CC, Bauck LJ & Hoover WJ (1975) Using surfactants to improve the quality of cookies made from hard wheat flours. Cereal Chemistry, 52, 629-637.

Verardo V, Arraez-Roman D, Segura-Carretero A, Marconi E, Fernandez-Gutierrez A & Caboni MF (2011) Determination of free and bound phenolic compounds in buckwheat spaghetti by RP-

HPLC-ESI-TOF-MS: effect of thermal processing from farm to fork. Journal of Agricultural and Food Chemistry, 59(14), 7700-7707.

Wani AA, Sogi DS, Singh P, Sharma P & Pangal A (2010) Dough-Handling and Cookie-Making Properties of Wheat Flour–Watermelon Protein Isolate Blends. Food and Bioprocess Technology, DOI 10.1007/s11947-010-0466-6.

Weipert D (1990) The benefits of basic rheometry in studying dough rheology. Cereal Chemistry, 67, 311–317.

Wekwete B & Navder KP (2008) Effects of avocado fruit puree and oatrim as fat replacers on the physical, textural and sensory properties of oatmeal cookies. Journal of Food Quality, 31, 131–141.

Wronkowska M, Zielińska D, Szawara-Nowak D, Troszyńska & Soral-Śmietana M (2010) Antioxidative and reducing capacity, macroelements content and sensorial properties of buckwheat-enhanced gluten-free bread. International Journal of Food Science & Technology, 45(10), 1993–2000.

Zucco F, Borsuk Y, Arntfield SD (2011). Physical and nutritional evaluation of wheat cookies supplemented with pulse flours of different particle sizes. LWT - Food Science and Technology, 44 (10), 2070-2076.

Figure captions:

Figure 1. Creep and recovery curve

Figure 2. Mechanical spectra of wheat and gluten-free cookie dough containing rice flour (RF) and buckwheat flour (BF) in different ratios, with and without CMC addition

Figure 3. Scanning electron micrographs, at 1000 magnification, of gluten-free and control cookie dough prepared with a) 90% rice flour and 10% buckwheat flour, b) 70% rice flour and 30% buckwheat flour and c) wheat flour

Figure 4. Photographs showing different shape and top surface cracking of gluten-free cookies prepared with a) 100% rice flour and CMC, b) 90% rice flour, 10% buckwheat flour and CMC, c) 80% rice flour, 20% buckwheat flour and CMC, d) 70% rice flour, 30% buckwheat flour and CMC and e) 80% rice flour and 20% buckwheat flour without CMC

Figure 5. Dimensional changes of gluten-free cookies containing different amount of buckwheat flour (BF)

Figure 6. Sensory panel overall acceptability scores of gluten-free cookies containing different amount of buckwheat flour (BF)

Table 1. Ingredients of gluten-free and control cookies

Ingredients	g	%
Flour	300	46.18
Deionised water	105	16.16
Vegetable fat	100	15.39
Granulated sugar	75	11.55
Honey	45	6.93
$NaHCO_3$	9	1.39
DATEM	9	1.39
CMC	4.5	0.69
Salt	2.1	0.32
Total	649.6	100

Table 2. Dynamic oscillatory and creep-recovery parameters of wheat (WF) and gluten-free cookie dough formulation containing rice (RF) and buckwheat flour (BF)[(a)]

Cookie dough formulation	100%RF/ CMC	90%RF/ 10%BF/ CMC	80%RF/ 20%BF/ CMC	70%RF/ 30%BF/ CMC	80%RF/ 20%BF	100% WF
Frequency sweep						
tan δ	0.256±0.007[a]	0.289±0.008[b]	0.290±0.002[b]	0.292±0.008[b]	0.271±0.013[ab]	0.441±0.007[c]
$K' \times 10^{-5}$ [(b)]	1.426±0.028[d]	1.090±0.045[c]	0.871±0.050[b]	0.833±0.040[b]	0.292±0.047a	0.848±0.023[b]
n' [(b)]	0.185±0.007[ab]	0.184±0.011[ab]	0.192±0.004[ab]	0.195±0.006[b]	0.169±0.004[a]	0.245±0.002[c]
Creep phase[(c)]						
J_0 (1/Pa)$\times 10^5$	1.02±0.14[a]	1.26±0.11[ab]	1.73±0.19[b]	1.73±0.13[b]	5.66±0.28[c]	1.23±0.14[ab]
J_m (1/Pa)$\times 10^5$	1.56±0.07[a]	3.77±0.11[b]	3.68±0.28[b]	4.20±0.28[b]	7.65±0.35[d]	5.97±0.21[c]
η_0 (Pa s)$\times 10^{-6}$	18.93±1.06[e]	13.70±0.28[d]	8.30±0.03[c]	5.20±0.14[a]	6.52±0.01[b]	5.57±0.02[a]
J_{max} (1/Pa)$\times 10^5$	4.15±0.28[a]	7.04±0.35[b]	8.92±0.06[c]	11.65±0.23[d]	17.89±0.42[e]	12.59±0.14[d]
Recovery phase[(d)]						
J_e/ J_{max} (%)	74.06±2.66[b]	69.75±3.40[ab]	70.57±0.27[ab]	63.86±4.00[a]	71.92±0.54[ab]	70.77±0.91[ab]
J_v/ J_{max} (%)	25.94±2.66[a]	30.25±3.40[ab]	29.43±0.27[ab]	36.14±4.00[b]	28.71±0.54[ab]	29.23±0.91[ab]

[(a)]Values in the same row followed by different letters are significantly different ($p < 0.05$)

[(b)]Coefficients of power-low equation

[(c)]J_0 - Instantaneous compliance, J_m - Viscoelastic compliance, η_0 - Zero shear viscosity, J_{max} – Maximum creep compliance

[(d)]J_e/J_{max} - Relative elastic part of maximum creep compliance, J_v/J_{max} - Relative viscous part of maximum creep compliance

Table 3. Textural properties of cookies in which rice flour was partially replaced with buckwheat flour (BF)[(a)]

Rice flour replacement with buckwheat	Hardness (N)	Fracturability (mm)
0% BF	36.3±3.7[b]	1.79±0.48[b]
10% BF	26.6±2.9[a]	0.67±0.09[a]
20% BF	29.0±5.7[a]	0.81±0.18[a]
30% BF	26.1±2.0[a]	0.62±0.08[a]

[(a)]Values in the same column followed by different letters are significantly different ($p < 0.05$)

Figure 1
Click here to download high resolution image

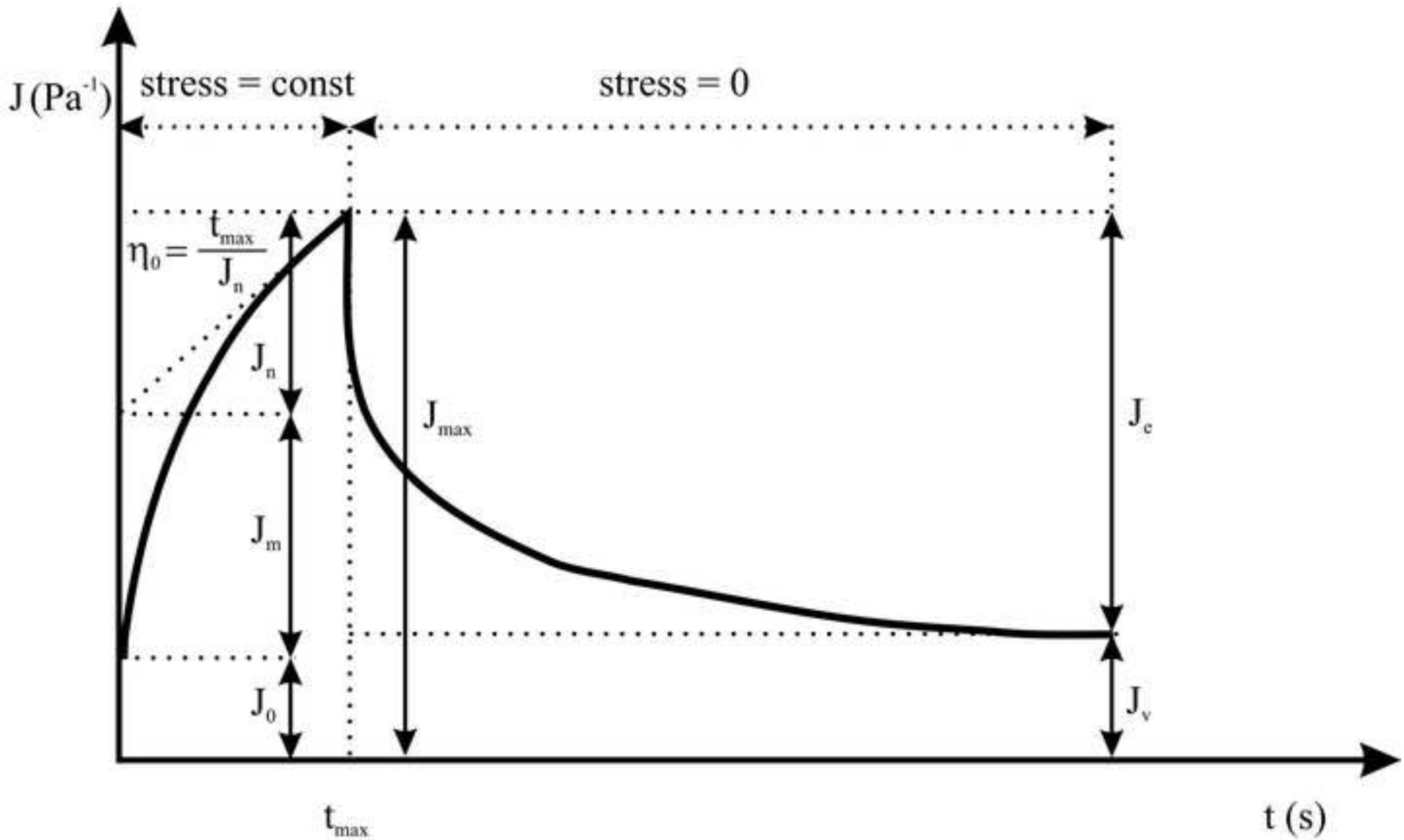

Figure 2
Click here to download high resolution image

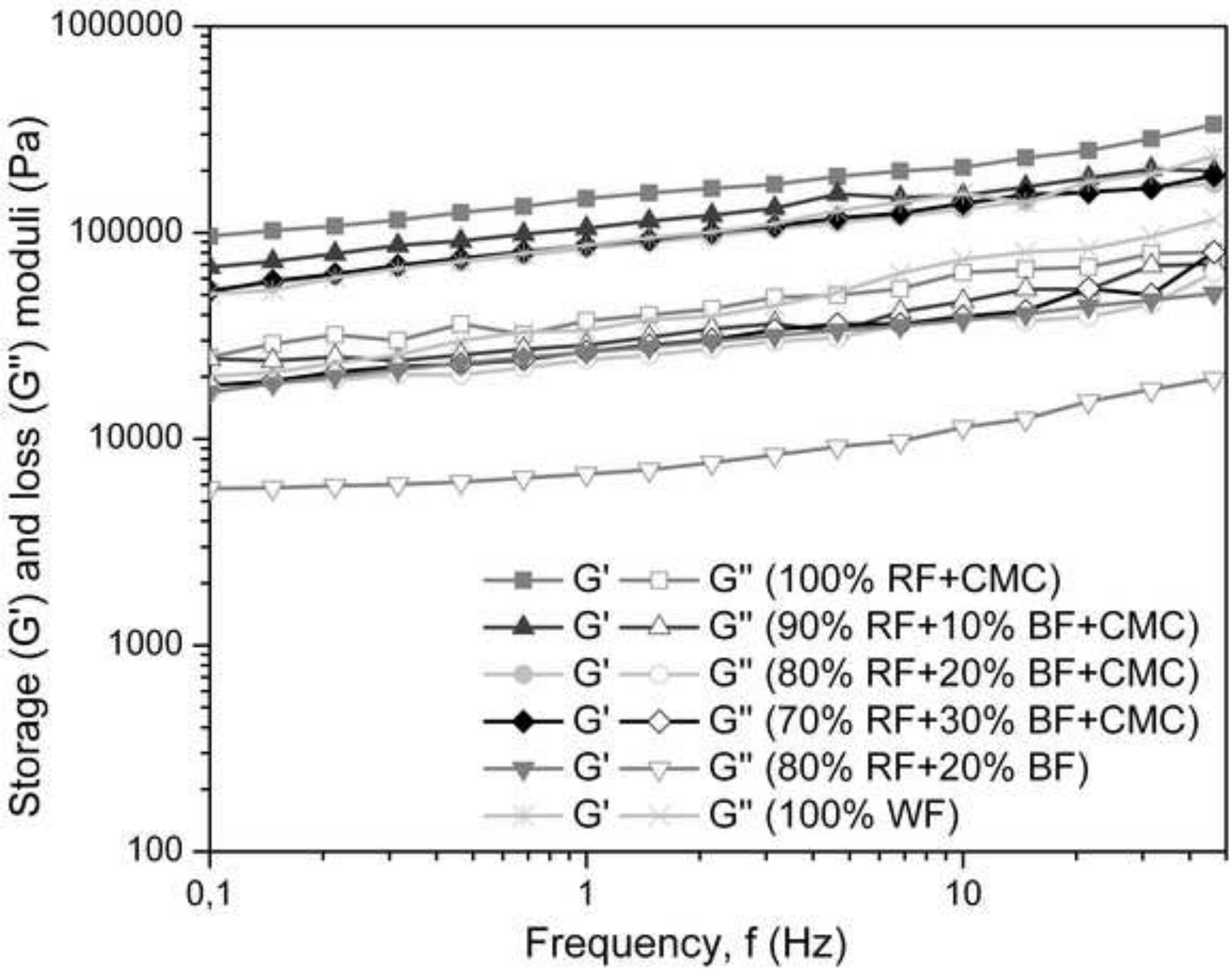

Figure 3
Click here to download high resolution image

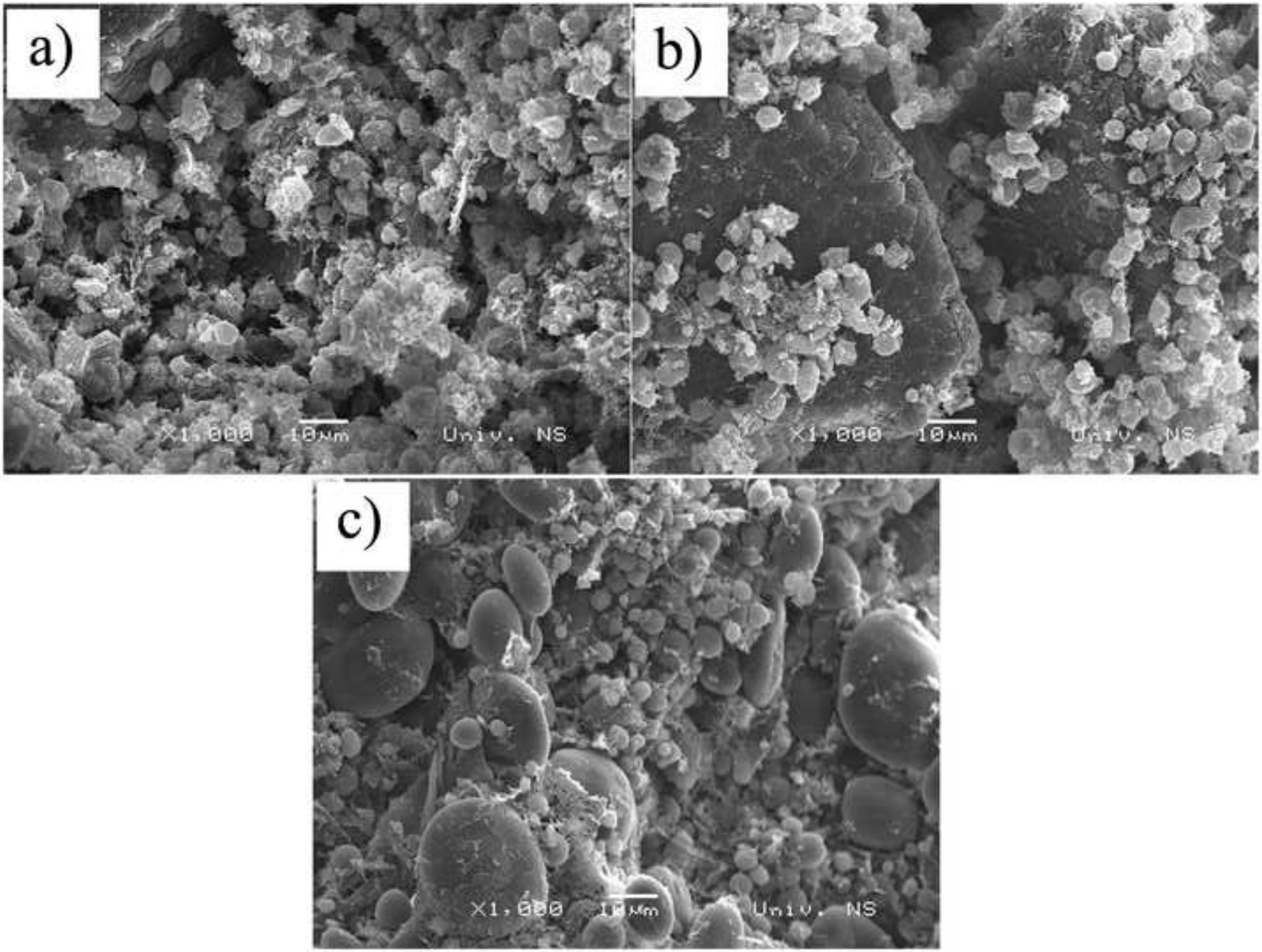

Figure 4
Click here to download high resolution image

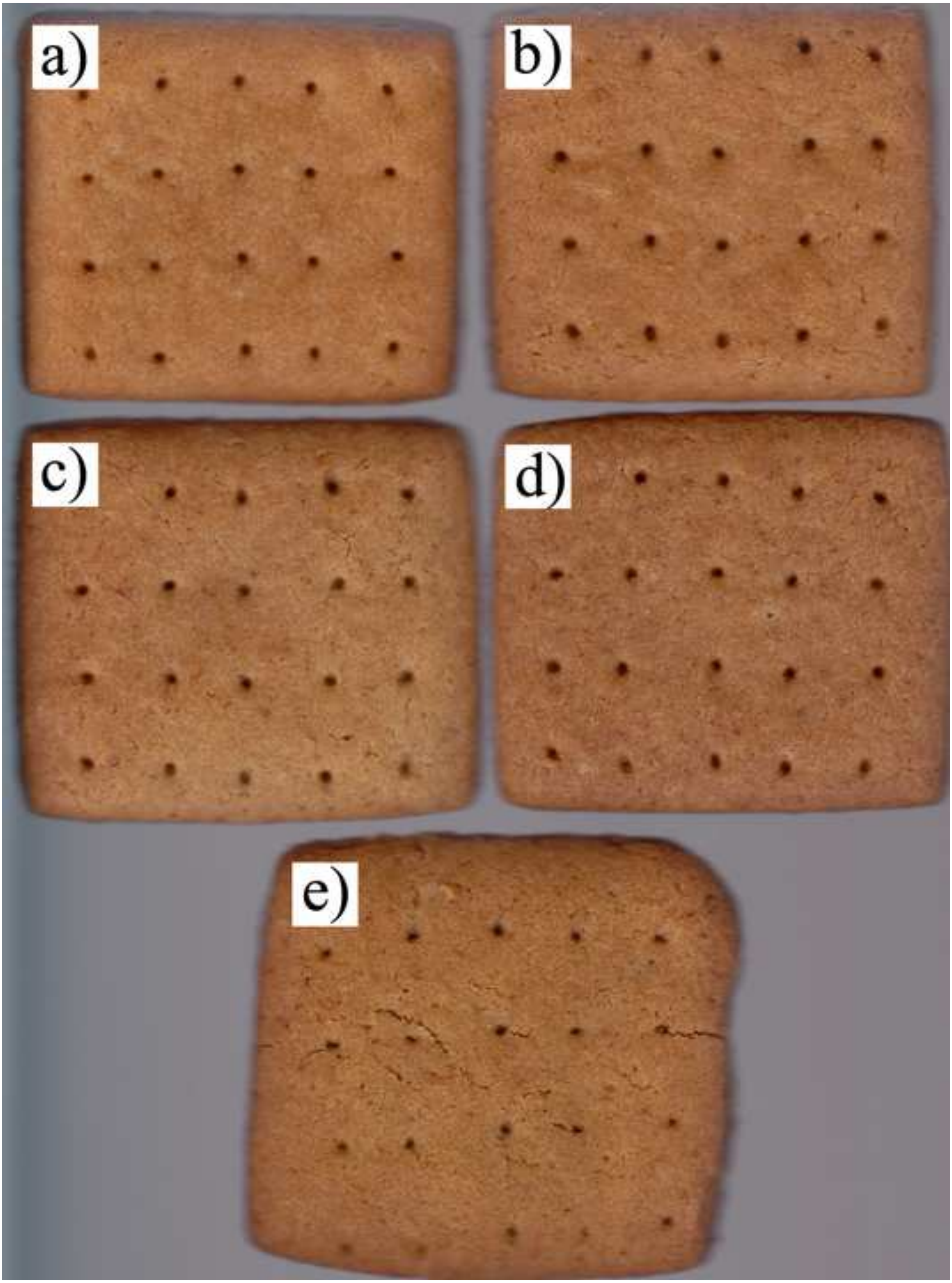

Figure 5
Click here to download high resolution image

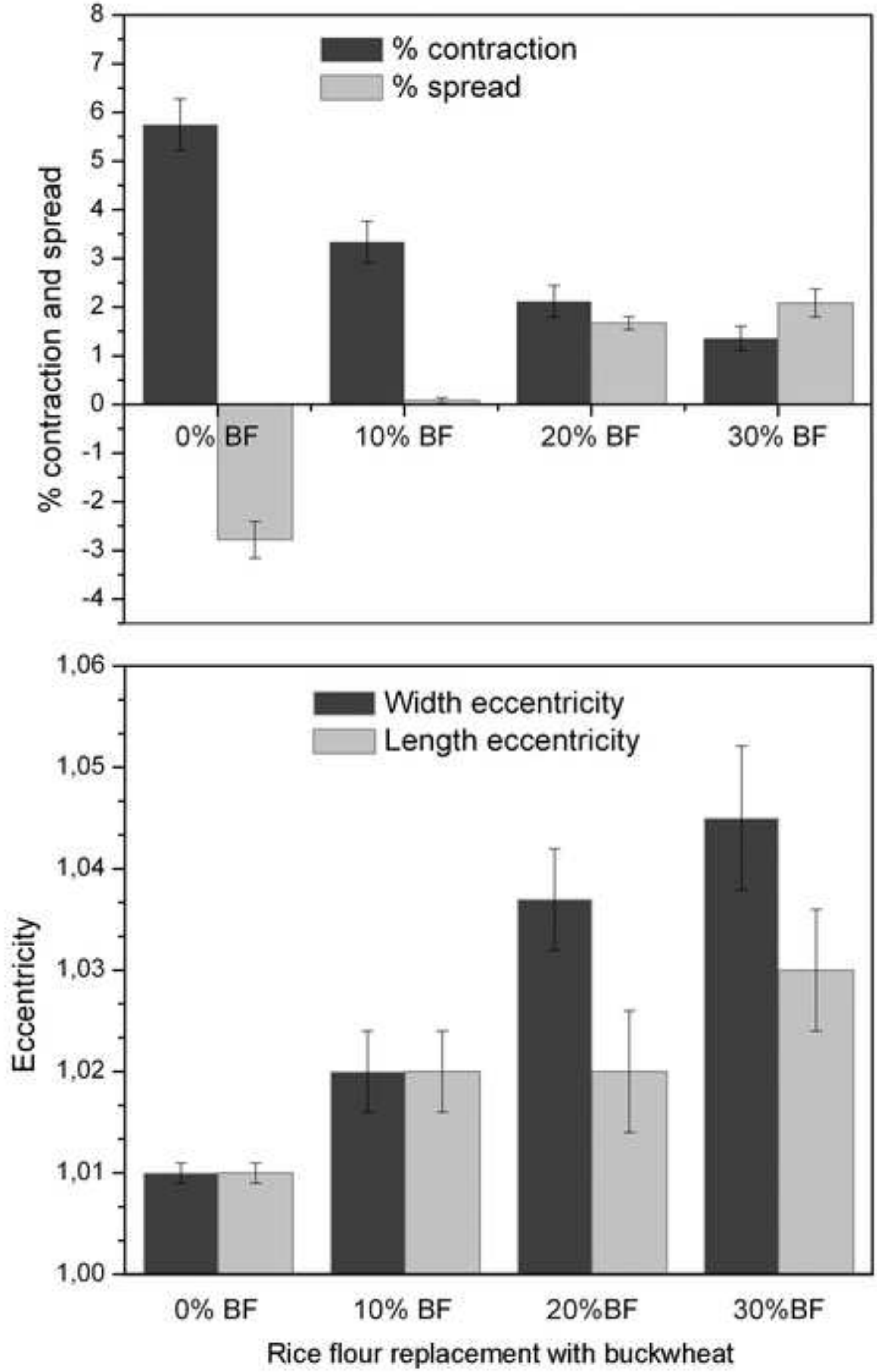

Figure 6
Click here to download high resolution image