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Effects of psyllium seed husk powder, methylcellulose, pregelatinised starch, and cold water swelling starch on the production of gluten free crackers

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The production of gluten free crackers is challenging because the formation of a gluten network is required. This study investigated the effects of psyllium seed husk powder (PSY), methylcellulose (MC), pregelatinised starch (PGS), and cold water swelling starch (CWSS) on gluten free crackers made of rice flour. The evaluations of pasting properties, dough rheological properties, textural properties, acoustic emissions, and structures were included in this study. Gluten free cracker doughs were more solid-like compared to wheat doughs based on their frequency dependence shown in the mechanical spectra. However, PGS significantly increased the fluid-like property and shapeability. The addition of MC at a high level significantly modified the pasting profile and a secondary swelling and breakdown might occur. As for the crackers, PSY and PGS crackers had comparable textural properties and sound release to wheat crackers, while CWSS crackers were slightly weaker. However, MC did not improve the textural properties compared to rice crackers because the interaction between the MC molecules was limited at the low water addition level, which limited its functionality in cracker making.

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1. Introduction

Crackers are a type of baked product occupying a substantial segment of the snack food market. They are also sometimes consumed as part of meals. Although the definition and classification can be complicated, crackers are usually characterised to be dry, thin, flaky and crispy and they can be produced to be plain, sweet, or savoury. The production of most types of crackers on the market, such as cream crackers and soda crackers, usually requires the addition of leavening agents. Compared to other biscuits, the formulation of crackers usually contains less sugar and fat but more water.[1] Therefore, a three-dimensional gluten network is formed in wheat cracker doughs, which is critical to the formation of the laminar structure of crackers.[2] The gluten network endows the doughs with extensibility; so the doughs can be laminated with less breakage. Furthermore, the gluten and starch matrix coagulates and gels at high temperature during baking and forms a structure that can survive, at least partially, the coalescence of gas cells, gas release and drying during baking.[3] However, gluten free foods need to be developed due to gluten intolerance such as coeliac disease. Celiac disease (CD) is an autoimmune enteropathy, which leads to self-perpetuating damage to the small intestinal mucosa and malabsorption of nutrients.[4] It can be triggered by the gliadin fraction of proteins in wheat, barley and rye. Currently, a life-long gluten-free diet is the only treatment for the disease.[5] Because of the important role of gluten in the manufacture of wheat crackers, the production of gluten free crackers is more challenging than the production of gluten free high sugar/short dough biscuits where the formation of gluten network is limited.

Gluten free crackers have been prepared using buckwheat flour, which had comparable sensory quality to wheat crackers.[6] The levels of antioxidant compounds and antioxidant activity were also found to be improved.[6] Gluten free crackers were also made from brown rice with the addition of chia seeds to improve the dough texture.[7] The nutritional quality was improved by adding hemp flour and green tea leaves.[7] Han *et al.*[8] produced gluten free crackers made from different pulse flours and both bench-top scale and commercial-scale processing were investigated. Most studies on gluten free crackers focus on nutritional quality and sensory profiles with characterisation of the physical properties of the products. However, investigations on the rheological properties of

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cracker doughs and the structures of crackers are limited compared to the published investigations on gluten free bread.

The addition of various hydrocolloids and dietary fibres in gluten free bread and doughs has been widely investigated and gluten free bread with improved qualities has been produced.[9–14] Dietary fibres can be either soluble or insoluble. Most of them significantly modify the rheological properties of doughs and influence the structure and texture of food products.[10,13–15] Additionally, dietary fibres have many health benefits such as improving intestinal peristalsis, eliminating intestinal toxins, helping weight control, and helping to prevent breast cancer, several chronic diseases, type II diabetes and cardiovascular diseases.[16–20]

Starch and modified starch are also widely utilised in the food industry, among which pre-gelatinised starch (PGS) is made from cooked starch and dried using a drum dryer or prepared in an extruder. The degree of gelatinisation of PGS ranges from 44% to 100% with the degree of crystallinity ranging from 14% to 18% depending on the processing procedures and measuring methods.[21] Without the starch granular structure, which is broken down during the process, most PGS forms a smooth paste in an aqueous system. PGS is widely used as a thickener in beverages, soups and toppings, a moisture retainer in cakes and a filler in puddings and cheese.[21,22] It is also widely used in the production of low-moisture snacks.[21,22] Being different from PGS, cold-water-swelling starch (CWSS) has intact starch granules. Several methods and conditions have been applied to prepare CWSS including increased temperature, increased pressure, aqueous alcohol, alcohol/alkaline solution, and/or spray-drying with a special nozzle.[23–25] CWSS made from waxy maize at high temperature in aqueous ethanol was reported showing an amorphous diffraction pattern.[26] However, the influences on the crystallinity and the overall starch granules depend on the types of starch and the process applied. CWSS is applied as a thickener, stabiliser, and fat replacer to improve the texture and mouth-feel and is used to make gum candies.[21,27,28] Although CWSS can also be classified as pre-gelatinised starch, CWSS and PGS are used in this study to identify these two different types of modified starch.

This study investigated the applications of methylcellulose (MC), powder of psyllium (*Plantago ovata*) seed husk (PSY), PGS and CWSS in the production of gluten free crackers. The influences of different gums and modified starch on the pasting properties of flour blends, dough rheological properties, textural properties of crackers, and structures of doughs and crackers were evaluated and compared. Additionally, acoustic emission was simultaneously recorded with mechanical measurements.

2. Materials and methods

2.1. Materials

Rice flour containing 11.08% moisture, 7.23% protein, 0.42% ash, and 2.8% lipid with an amylose content of 28.79% as measured in our previous study[29] was purchased from Doves Farm. Dr Oetker Gluten Free Baking Powder, caster sugar (Sainsbury's, UK), salt (Sainsbury's, UK), sunflower oil (Sainsbury's, UK), and unsalted butter (Morrisons, UK) were purchased from local supermarkets. Psyllium husk powder (Vitacel®) was kindly donated by the J. Rettenmaier & Söhne Group (JRS, Rosenberg, Germany). Methylcellulose (Methocel® A4M) was supplied by The Dow Chemical Company (Bomlitz, Germany). Pregelatinised waxy maize starch (Merigel®) was supplied by Tate & Lyle (UK). Cold water swelling waxy maize starch (Ultra-Tex® 4) was kindly donated by Ingredion (UK). Nile red (Sigma-Aldrich), fluorescein isothiocyanate (FITC, Acros Organics, New Jersey, USA), methyl blue (Sigma-Aldrich, UK), fast green (Sigma-Aldrich) and rhodamine B (Sigma-Aldrich) were of analytical grade.

The water absorption capacity of rice flour was 133% (118% after being corrected for the moisture content of flour), which was determined using time-domain nuclear magnetic resonance (NMR). Flour was hydrated overnight before measurements. The Carr–Purcell–Meiboom–Gill (CPMG) sequence[30] was applied to obtain the transverse relaxation curves using an R4 Benchtop NMR System (Advanced Magnetic Resonance Ltd, Abingdon, UK) at 20 °C, which was controlled using a thermal controller (Advanced Magnetic Resonance Ltd). The initial 90° pulse for all the sequences was approximately 2.6 μs and the signals were recorded after 10 μs (dead time). There were 4096 echoes for the following 180°-pulses with 64 μs (TAU) between every two pulses. The results were obtained averaging 32 scans with a recycle delay of 10 s. The distributed exponential using the Lexus fixed method was applied to calculate the T2 distribution spectra. The water absorption capacity was determined at the water addition level when the T2 peak at approximately 700 ms for the unabsorbed bulk water was absent.

2.2. Formulation and cracker preparation

The formulation of gluten free crackers was optimised singly for rice, MC, PSY, PGS, CWSS and wheat crackers by trial and error and is shown in Table 1. For rice, MC, and PSY crackers, the water addition levels were determined as required to form the doughs under the applied mixing conditions. The water addition levels for wheat, PGS and CWSS crackers were determined by trial and error and kept constant. Doughs were mixed using a Kenwood stand mixer (UK) equipped with a Chef K beater. To prepare the cracker doughs, all powdered ingredients were mixed and combined with butter (stored at 10 °C) thoroughly. Sunflower oil and water were mixed and gradually added over 1 min at speed 2. The dough was then mixed at speed 4 for 2 min. After resting at 5 °C for 10 min, the doughs were laminated to 16 layers and sheeted to 1.6 mm thickness (2.6 mm for rice and MC cracker doughs because they were very fragile and difficult to handle). The dough sheets were cut into 36 × 36 mm squares on a piece of baking parchment, transferred to a preheated baking tray and baked in a fan oven (Hotpoint, UK) at 250 °C for 6 min and 105 °C for 10 min. The crackers were cooled at room temperature for 1 h and stored in polyethylene bags for further analysis.

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Table 1 Addition levels of the ingredients in the cracker formulation

	Rice crackers (negative control)	Wheat crackers (positive control)	MC crackers	PSY crackers	PGS crackers	CWSS crackers
Rice flour	100	0	100	100	80	70
Wheat flour	0	100	0	0	0	0
Baking powder	5	5	5	5	5	5
Sugar	5	5	5	5	5	5
Salt	1	1	1	1	1	1
Butter	30	30	30	30	30	30
Oil	10	10	10	10	10	10
Water	52	50	71	71	50	50
MC	0	0	4	0	0	0
PSY	0	0	0	4	0	0
PGS	0	0	0	0	20	0
CWSS	0	0	0	0	0	30

2.3. Pasting properties of the flour blends

The influences of the addition of MC, PSY, PGS and CWSS on the pasting profiles of the flour blends were evaluated using a Rapid Visco Analyser (RVA) (Newport Scientific Pty Ltd, Warriewood, New South Wales, Australia) and the temperature control was assisted by a water bath (Thermo Scientific C10, Karlsruhe, Germany). To prepare the hydrocolloid blends, 0.1 g of MC or PSY was mixed with 2.5 g of rice flour and dispersed in 24 g of reverse osmosis (RO) water in an aluminium canister. The PGS blend and CWSS blend were prepared by substituting 20% of the rice flour with starch and 3.125 g of the flour blends were added into 21.875 g of RO water. During the measurements, the flour or flour blends were dispersed thoroughly at a high shear rate (960 rpm) for 60 s, followed by a low rate shearing (160 rpm) for another 60 seconds at 25 °C. Keeping the shear rate constant at 160 rpm, the temperature profile started with an increase from 25 °C to 95 °C in 350 s, maintained at 95 °C for 150 s, and decreased back to 25 °C in 350 s. The measurements were repeated three times for each sample.

2.4. Dough rheology

Cracker doughs were prepared as described in section 2.2 without the addition of baking powder. The doughs were sheeted to 2.5 mm and loaded on an MRC 301 rheometer (Anton Paar, Austria) equipped with a Peltier system controlling the temperature. The measurements were performed using a serrated parallel plate geometry (PP25/P2-SN15766, Anton Paar) and the measuring gap was maintained at 2 mm. After being loaded, the extra doughs were trimmed by a spatula and the edge was covered with low viscosity mineral oil (Sigma, USA) to prevent drying. After resting at 20 °C for 500 s, the doughs were subjected to oscillatory shear tests with a logarithmic decrease of angular frequency at a constant shear strain of 0.02%, which was within the linear viscoelastic regions for all the doughs. The measurements were performed in triplicate.

2.5. Cracker evaluation

The thickness was measured using a vernier caliper by averaging the thickness measured at the different positions (4 to 8 positions) on a piece of cracker. The water activity of crackers (powdered using a coffee grinder) was measured using an Aqua Lab water activity meter (Aqua Lab, 3TE, Decagon, Pullman, WA, USA). The moisture contents of the crackers were measured by drying at 105 °C.

The crackers were further evaluated by three-point bending tests on a TA-XT plus texture analyser (Stable Micro Systems, Surrey, UK) equipped with 5 kg loading cell and the sound emitted during testing was recorded using an Acoustic Envelope Detector (Stable Micro Systems). The distance between the two lower supporting points was 18 mm. The upper-middle point descended at a speed of 2 mm s^{-1} to a distance of 5 mm after the surface of the crackers was detected. The trigger force was 5 g. The Acoustic Envelope Detector was equipped with a microphone located close to the fracture positions (about 2 cm). The microphone was calibrated to 94 and 114 dB SPL using a sound level calibrator. The gain was set to four. Sixteen crackers were evaluated for each formulation. The maximum force was reported as hardness of the crackers and the travelling distance of the probe to the first force peak was reported as fracturability. The intensity of the emitted sound was described by the maximum sound peak, the average peak decreases, and the number of sound peaks. A decrease in the sound pressure level to 8.5 dB was set as the threshold to identify the peaks and calculate the average peak decreases.

2.6. Confocal laser scanning microscopy

The microstructure of the cracker doughs was observed using a Zeiss LSM880 confocal laser scanning microscope (Carl Zeiss Microscopy GmbH, Jena, Germany). Stock solutions of the stains were prepared, including 1 g L^{-1} Nile red in acetone, 1 g L^{-1} FITC in acetone, 1 g L^{-1} methyl blue in water, 1 g L^{-1} rhodamine B in water and 1 g L^{-1} fast green in water. The stock solutions were diluted and used to make the working solutions containing different stains specific for different doughs with a final concentration of 0.1 g L^{-1} for each stain. The working solutions were added to the doughs replacing the water in the formulation. A working solution containing fast green and rhodamine B was used to stain gluten and starch/lipid, respectively, in the wheat doughs. FITC and Nile red were used to stain starch and lipid, respectively, in the rice doughs, PGS

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doughs and CWSS doughs. FITC, Nile red and methyl blue stained starch, lipid and PSY in the PSY cracker doughs. Fast green was excited at 633 nm and the signal was collected at 641 to 710 nm. Rhodamine B was excited at 488 nm and the signal was collected at 545 to 590 nm. FITC and Nile red were both excited at 488 nm, while the signal collection wavelengths were 510–540 nm and 570–600 nm, respectively. Images were acquired at 20× magnification using an EC Plan Neofluar® objective.

2.7. Scanning electron microscopy

The powdered ingredients including PSY, MC, PGS, and CWSS and the crackers were evaluated using scanning electron microscopy (JEOL JSM-6060LV, JEOL (UK) Ltd, Welwyn Garden City, UK). The powdered ingredients were mounted onto aluminium stubs using carbon tapes. The crackers were broken and a piece of cracker was mounted onto the aluminium stub using carbon tapes with the exposed surface facing up. The powdered ingredients and cracker pieces were coated with gold using a Leica EM SCD005 Sputter Coater at an argon current rate of 27 mA for 3 min. The images were acquired under high vacuum conditions at 10 kV.

2.8. Statistical analysis

Representative curves and images were selected from the replicates for each sample and are shown in the figures, while the means and standard deviations were calculated and are shown in the tables. Additionally, the results were analysed and compared using one-way analysis of variance (ANOVA) with Turkey's test using IBM SPSS Statistics (version 26, IBM Corp., Armonk, NY, USA) at a significance level of $p < 0.05$.

3. Results and discussion

3.1. Microstructures of the raw ingredient powders

The microstructures of rice flour, PSY, MC, PGS, and CWSS were scanned using a scanning electron microscope and the microscopic images are shown in Fig. 1. Fig. 1a shows that rice flour contains both small and big flour particulates. Rice starch exists as compound granules which consist of small individual starch granules[31] and Fig. 1b shows that the granules are enclosed in proteinaceous materials and other cell contents.[32] Fig. 1c and d show PSY at different magnifications. It can be seen in Fig. 1d that PSY particles show a lamellar structure, which is in agreement with the results of the study by Mariotti *et al.*[33] and the observation is dependent on the positions of observation. The functional polymer of PSY, which is heteroxylan, is likely to be expelled from this lamellar structure when hydrated in water and swells into 'weak-gel' particles.[34] Fig. 1e and f show that the MC powder is composed of fibrous particles. In the production of methylcellulose, native cellulose is only activated in heated alkaline solutions without being fully dissolved; therefore, the product is fibrous like native cellulose.[35,36] The fibrous structure of the MC powder could be a clue to the distribution of the MC molecules in cracker doughs and crackers as they might not be fully hydrated due to the low moisture content in the formulation. Fig. 1g and h show the microstructure of PGS and Fig. 1i and j show that of CWSS. As expected, CWSS shows individual starch granules which are not observed for PGS.[37,38] These modified starch granules of CWSS absorb water and swell at room temperature.

3.2. Pasting properties

The influences of the addition of PSY and MC and the replacement by PGS and CWSS on the pasting properties of the flour blends were evaluated using RVA 2. As shown in Fig. 2a, the addition of PSY significantly increased the overall viscosity of the paste. However, the MC flour blend showed an atypical pasting profile with three viscosity peaks possibly due to the high addition level of MC which led to high viscosity. The decrease of pasting temperature with the addition of MC and PSY was observed, which was also reported in one of our previous studies due to water and volume competition between starch and the added hydrocolloids.[12] Although the addition level of MC in the current study was higher than our previous study, the pasting temperature was decreased to a similar value of 70 °C. The pasting temperature was also less influenced by the molecular weight of MC and different types of MC or hydroxypropylmethylcellulose.[39] Therefore, it can be speculated that the decreased pasting temperature is predominantly controlled by the swelling behaviour of starch granules. It is known that MC gelation occurs at 50 to 55 °C and it forms a rigid gel at high temperature. Therefore, the system of the MC/starch mixture under RVA conditions (high temperature with shearing) can be described as a dispersion of MC gel particles and swollen starch granules.[40] According to the Krieger–Dougherty model,[41] viscosity significantly increases when the volume fraction of a suspension reaches the packing fraction, which explains the significant increase in viscosity after the pasting temperature due to the swelling of starch granules. Because the MC particles occupied a certain volume fraction, a lower swelling degree of starch granules could lead to a volume fraction which triggers a significant viscosity increase at a temperature that is identified as pasting temperature. With a lower addition level of MC, viscosity developed a shoulder after this pasting temperature before the significant increase caused by the temperature-trigged, significant swelling of the starch granules,[12] which was also observed from the PSY blend (Fig. 2a). In a typical pasting profile of starch or flour, the starch granules are closely packed at peak viscosity and the following breakdown is accompanied by a viscosity decrease. Following the initial viscosity increase, the starch granules keep swelling and growing in size, which increases their volume fraction and leads to an increase in viscosity. The increased viscosity causes a higher shear force applied to the starch granules. At the same time, the granules lose their rigidity during swelling due to hydration, melting of the helical conformation, and loss of crystallinity. Therefore, the granules eventually cannot resist the increased shear force and start to lose the granule integrity, which is observed as the decrease in viscosity and identified as the breakdown stage. Hence, the packing at the viscosity peak and breakdown of starch granules can be influenced by the balance

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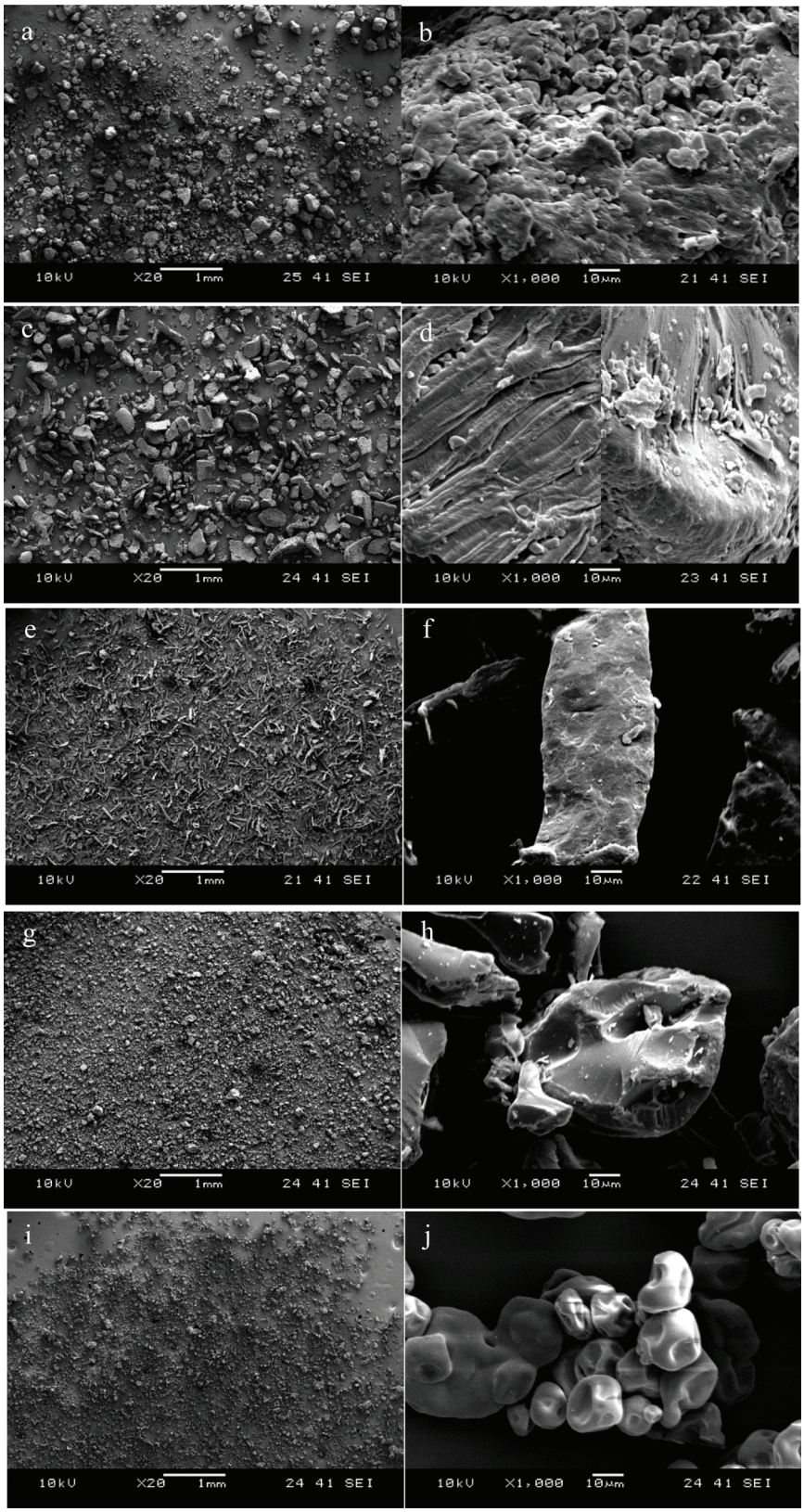

Fig. 1 Scanning electron microscopy images of rice flour (a and b), PSY powder (c and d), MC (e and f), PGS (g and h) and CWSS (i and j) used in the gluten free cracker formulation.

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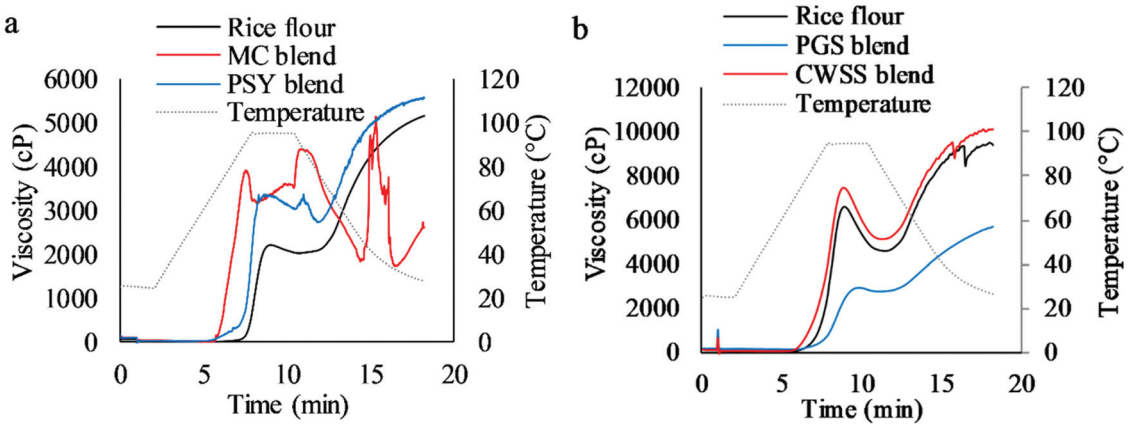

Fig. 2 Pasting profiles of (a) MC blend and PSY blend compared with rice flour (control), where 0.1 g of MC or PSY was mixed with 2.5 g of rice flour in 24 g of water, and (b) PGS blend and CWSS blend compared with rice flour (control), where 3.125 g of rice flour or flour blends were added into 21.875 g of water. The flour blends were prepared by 20% replacement of rice flour by PGS or CWSS. The pasting profiles are shown as representative curves from the experiments run in triplicate.

between the shear force applied on the starch granules due to the high viscosity at high volume fractions and starch granule rigidity. In the case of the MC blend in this study, the significant increase in viscosity due to the MC gel particles caused a higher shear force on the starch granules and they broke down earlier compared to the flour only sample, which was shown as a lower temperature for the peak viscosity (Fig. 2a). As the starch granules in the MC blend broke down earlier, there might be a secondary swelling of the granules and broken granule fragments, which was shown as the slight increase in viscosity from 8 min to 10 min. However, this small increase was not reproducible and a slight decrease of viscosity was sometimes observed (data not shown) due to the following breakdown instead of swelling. For both MC and PSY blends, the obvious viscosity increase led to a second peak occurring at a slightly earlier time than the end of the breakdown stage of the flour only sample. It is possible because of the continuous granule swelling, the formation of granule ghosts[42] and the cessation of granule breakdown, and the increased viscosity led to the secondary breakdown after the second peak. It is proposed in addition to what has been proposed in a separate study from our group that the second peak of the PSY blend is due to a balance between the interaction with amylose (James M. Cowley, personal communication), the formation of weak PSY gel particles, PSY particle interactions, and PSY particle breakdown.[12] Compared to the rice flour and PSY blends, MC significantly decreased retrogradation, which was seen as low viscosity during cooling. An irreproducible peak appeared during this stage because of the disruption of the structure formed by the alignment and association of amylose molecules.

As shown in Fig. 2b, the replacement by CWSS slightly increased the overall viscosity of the pasting profiles compared to the flour only sample. However, the replacement by PGS significantly decreased the overall viscosity due to the absence of the granular structure. The low viscosity was also observed in other studies when the starch granules were disrupted to a high degree during preparation.[43] Additionally, PGS is soluble in cold water and quickly absorbs water. Therefore, it is likely that PGS was quickly hydrated in cold water and formed a barrier outside the rice flour particulates, which restrained the hydration and swelling of the native rice starch and further decreased the viscosity.

3.3. The microstructure of the cracker doughs

The microstructures of the cracker doughs are shown in Fig. 3. As shown in Fig. 3a, the strands of the gluten network were seen in the wheat cracker doughs. The wheat starch granules were shown as dark circles, which indicates that the stain did not penetrate the granules. It suggests that the granules were not fully stained as the hydration of starch was highly limited in the wheat cracker doughs. The rice starch in the rice cracker doughs and PSY cracker doughs (Fig. 3b and c), in contrast, was fully stained in green, which indicates that they were more hydrated. It was possibly due to the higher water addition level, smaller granule size of the rice starch, and the difference in water-binding abilities between gluten and PSY. Although PSY did not form a network like gluten, it formed a matrix which holds the flour particles, starch granules, and other insoluble ingredients together and allows the formation of a cohesive dough. In the PGS and CWSS doughs, both rice starch from rice flour and the added PGS or CWSS were stained in green. Although the starch cannot be distinguished in colour, they were different in shape, size, and distribution. As shown in Fig. 3d, PGS formed a relatively continuous matrix in which the rice flour and starch were embedded. In contrast, swollen starch granules from CWSS can be observed in the CWSS dough (Fig. 3e). Being different from the PSY and rice doughs, the rice starch, especially the starch granules within the flour particles, in the PGS and CWSS doughs was less hydrated (less stained and showed lower colour intensity). It was because of the lower water addtiion levels in the formulation which reduced the hydration of rice flour. Additionally,

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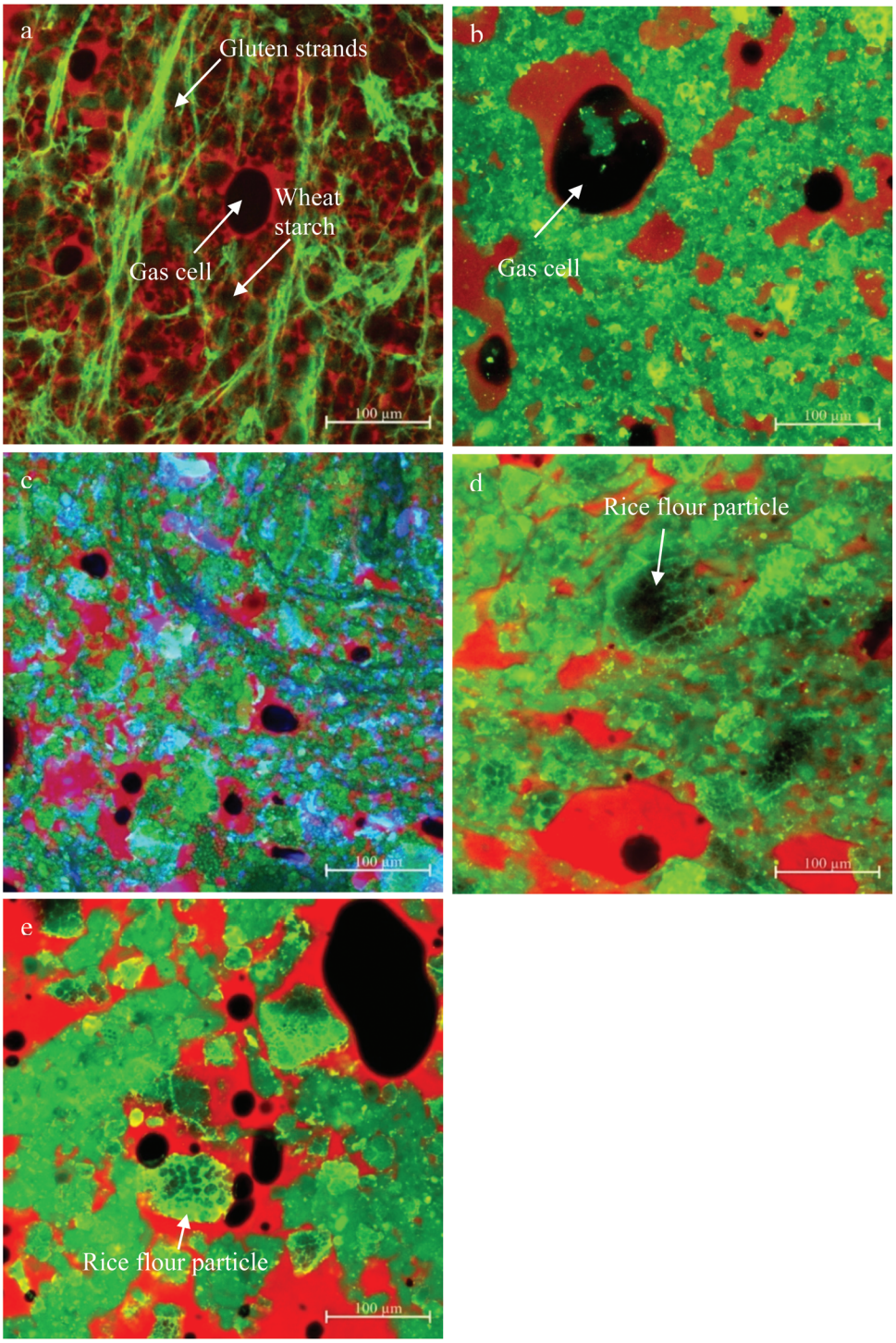

Fig. 3 Confocal laser scanning microscopy images of the wheat cracker doughs (a), rice cracker doughs (b), PSY cracker doughs (c), PGS cracker doughs (d) and CWSS cracker doughs (e). Gluten and starch/lipid in the wheat doughs were stained using fast green and rhodamine B, and are shown in green and red, respectively. In the rice, PSY, PGS and CWSS doughs, starch and lipid were stained using FITC and Nile red, and are shown in green and red, respectively. PSY in the PSY cracker doughs was stained using methyl blue, and is shown in blue.

PGS and CWSS have high water-binding abilities. The additions of PGS and CWSS at high levels further restrianed the hydration of rice flour by competing for water. Lipid, which was stained in red, was distributed in the cracker doughs and accumulated surrounding the gas cells.

3.4. Rheological properties of the cracker doughs

The rheological properties of the cracker doughs were measured using oscillatory shear tests. The amplitude sweep spectra are shown in Fig. 4a. Both wheat cracker doughs and

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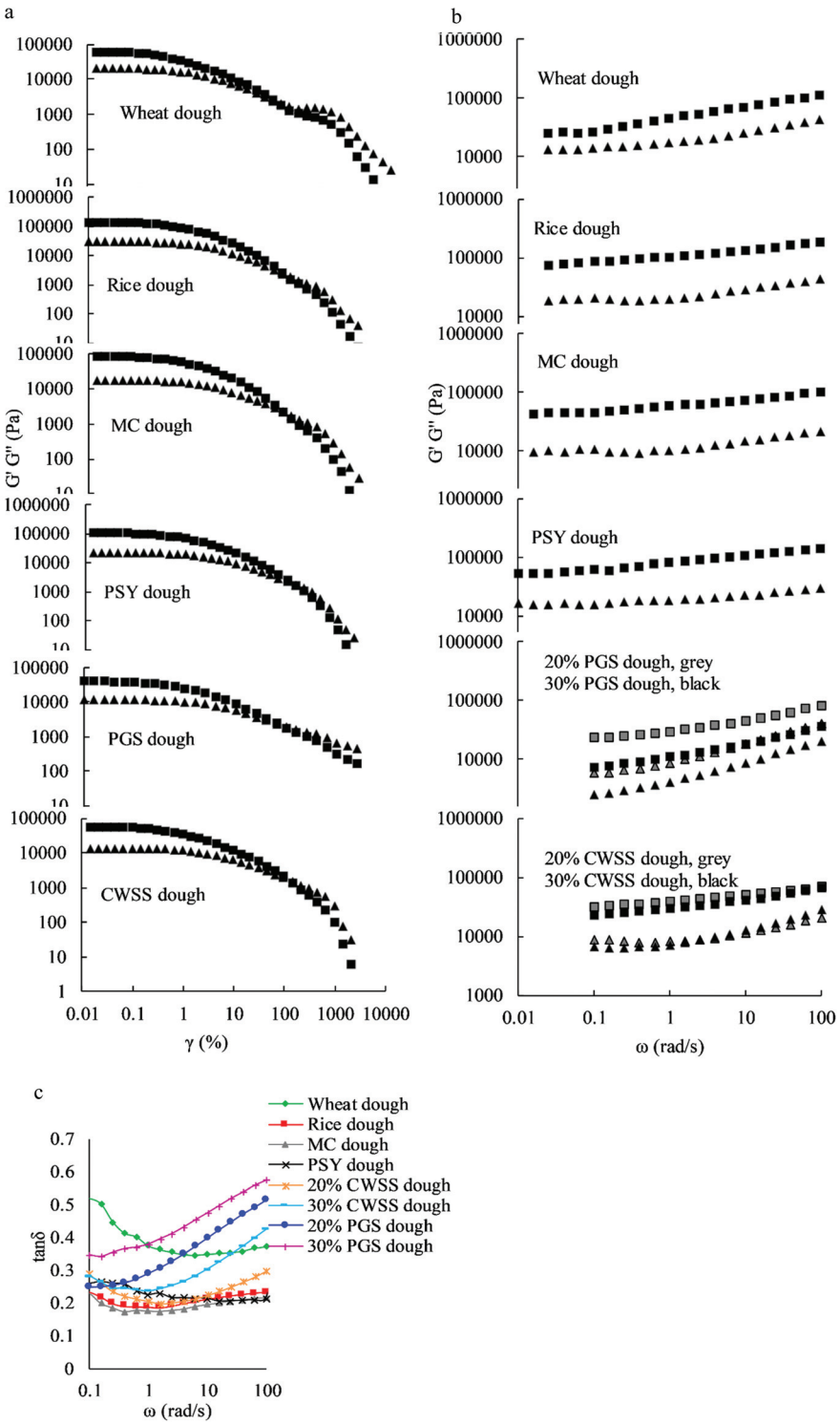

Fig. 4 Storage and loss moduli (G' square and G'' triangle) measured in the strain amplitude sweep tests (a), the mechanical spectra (b), and tan δ measured in the frequency sweep tests (c) of wheat and all gluten free cracker doughs.

gluten free cracker doughs had similar lengths of linear viscoelastic regions of about 0.1%. As complex concentrated suspensions, they had very short linear viscoelastic regions compared to the polymer solutions and melts which usually have LVE regions higher than 10% [44] and gels. The gradual decrease of the moduli with the increase of shear strain outside the LVE region could be due to the breakdown and reformation of the weak network of the rheological units that are composed of flour particles, starch granules and other soluble and insoluble flour components, which weakly interact with the neighbour-

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ing rheological units to different levels. It was evidenced that starch predominantly contributes to the non-linearity of wheat doughs.[45] However, it is noticed that wheat doughs showed a secondary G' and G'' plateau at high strain (between 100 and 1000%), which might be attributed to the gluten network, although the evaluation at large stain must be careful and wall slips need to be taken into consideration.

The mechanical spectra of the cracker doughs are shown in Fig. 4b. As expected, all doughs had higher G' than G'' with $\tan\delta$ lower than 0.6 (Fig. 4c), suggesting solid-like properties. As shown in Fig. 4b, the storage moduli of the gluten free cracker doughs tended to be less dependent on the angular frequency (with $\log G'$ *versus* $\log\omega$ of 0.11, 0.11, 0.13, 0.11, 0.14, 0.18, and 0.23 for the rice doughs, MC doughs, PSY doughs, 20% CWSS doughs, 30% CWSS doughs, 20% PGS doughs, and 30% PGS doughs, respectively) than those of the wheat doughs ($\log G'$ *versus* $\log\omega$: 0.21) as a result of close interactions between the packed rigid flour particles in the absence of the gluten network. Compared to the rice doughs, the addition of CWSS and, especially, PGS significantly increased the frequency dependence, while the PSY and MC doughs remained less influenced. As shown in Fig. 4c, the lower $\tan\delta$ of the rice, PSY and MC doughs than the wheat doughs suggests that they were more solid-like. However, the addition of CWSS and, especially, PGS significantly increased $\tan\delta$ at high angular frequencies, which were even higher than those of the wheat doughs. It indicates that PGS and CWSS doughs were more fluid-like at shorter time scales. This was consistent with the observation during dough handling that PGS doughs were more shapeable.

3.5. Cracker properties

The physical properties of the gluten free crackers are shown in Table 2. As the dough sheets were cut to a dimension of 36 × 36 mm, it can be seen that both wheat crackers and all gluten free crackers had lengths shorter than 36 mm, which suggests that they all contracted in the lateral direction during baking. The shrinkage of the wheat crackers was partially due to the elastic properties of the gluten network which caused shrinkage after cutting. It was also due to the elastic shrinkage of the gluten network which displays hardening behaviour above its glass transition temperature.[46] The shrinkage of the crackers was also caused by water loss under the temperature gradient and moisture gradient during baking. Although the difference in lengths between the MC, CWSS and PSY crackers was insignificant, the PSY crackers contracted to the same length as the wheat crackers. It suggests that heteroxylan from PSY possibly manifested a similar elastic shrinkage behaviour to gluten in the wheat crackers during baking. In contrast, the rice crackers and PGS crackers contracted less. It is possibly because they were predominantly plasticised by water and hydrated PGS, which tended to spread during baking, while the shrinkage due to water loss led to the final shrinkage.

The wheat crackers also featured a higher thickness, which increased by 156% compared to the thickness of the dough sheets (1.6 mm). Among the five gluten free crackers, the PGS crackers had the highest thickness, which increased to 2.77 mm by 73%, followed by the CWSS crackers and PSY crackers. However, the rice and MC crackers showed lower increases in thickness.

The PSY crackers had a relatively high moisture content but low water activity. The increase in the moisture content of the crackers and biscuits when gums were added was also observed previously,[47] which is assigned to the water retention ability of gums. The high water retention ability of PSY also explains the low water activity of the crackers. Additionally, a porous structure is expected to increase the water loss during baking, but increase the water absorption during storage.

3.6. Microstructure of the crackers

The microstructures of the wheat crackers, PSY crackers, PGS crackers and CWSS crackers are shown in Fig. 6. A laminar and plate-like structure was observed in the wheat crackers (Fig. 5a and b). It indicates that the wheat doughs were successfully laminated and the wheat crackers maintained the structure during the gas cell growth, cell ruptures and gas release during baking. Starch and other components existed as intact granules and particles embedded in the gluten matrix. It was also reported by Kulp *et al.*[48] that starch in cookie doughs and end products does not swell, gelatinise, and form a continuous network which, however, is usually observed in high moisture products. However, the PSY, PGS, and CWSS crackers did not show an obvious laminar or plate-like structure compared to the wheat crackers. As shown in Fig. 5c and d, the small rice starch granules were embedded in a continuous matrix in the PSY crackers. The PSY powder hydrated into a gel-like soft particle in water with an insoluble and unhydrated core rich in cellulose.[34] The PSY core was also seen in the matrix. The con-

Table 2 Physical properties of the wheat and gluten free crackers

	Length (mm)	Thickness (mm)	Thickness increase (%)	Moisture content (%)	Water activity
Wheat	32.13 ± 1.12^{c}	4.10 ± 0.09^{c}	156.09 ± 5.74^{b}	3.19 ± 0.00^{bc}	0.586 ± 0.010^{c}
Rice[a]	34.70 ± 0.57^{ab}	2.94 ± 0.14^{a}	13.08 ± 5.38^{a}	4.08 ± 0.05^{ab}	0.394 ± 0.007^{a}
MC[a]	33.13 ± 1.28^{c}	2.71 ± 0.19^{ab}	4.04 ± 7.34^{a}	2.46 ± 0.43^{c}	0.344 ± 0.016^{b}
PSY	32.31 ± 1.16^{c}	2.14 ± 0.08^{d}	33.91 ± 4.69^{c}	4.58 ± 0.51^{a}	0.248 ± 0.002^{d}
PGS	35.77 ± 1.12^{b}	2.77 ± 0.04^{ab}	73.13 ± 2.60^{d}	3.42 ± 0.03^{bc}	0.213 ± 0.002^{e}
CWSS	33.35 ± 0.69^{ac}	2.55 ± 0.10^{b}	59.06 ± 6.24^{e}	3.90 ± 0.14^{ab}	0.193 ± 0.003^{e}

[a] Rice and MC crackers were sheeted to 2.6 mm, and the other crackers were sheeted to 1.6 mm. Data are shown as mean ± standard deviation. The different letters associated with the values in the same column indicate significant differences ($p < 0.05$).

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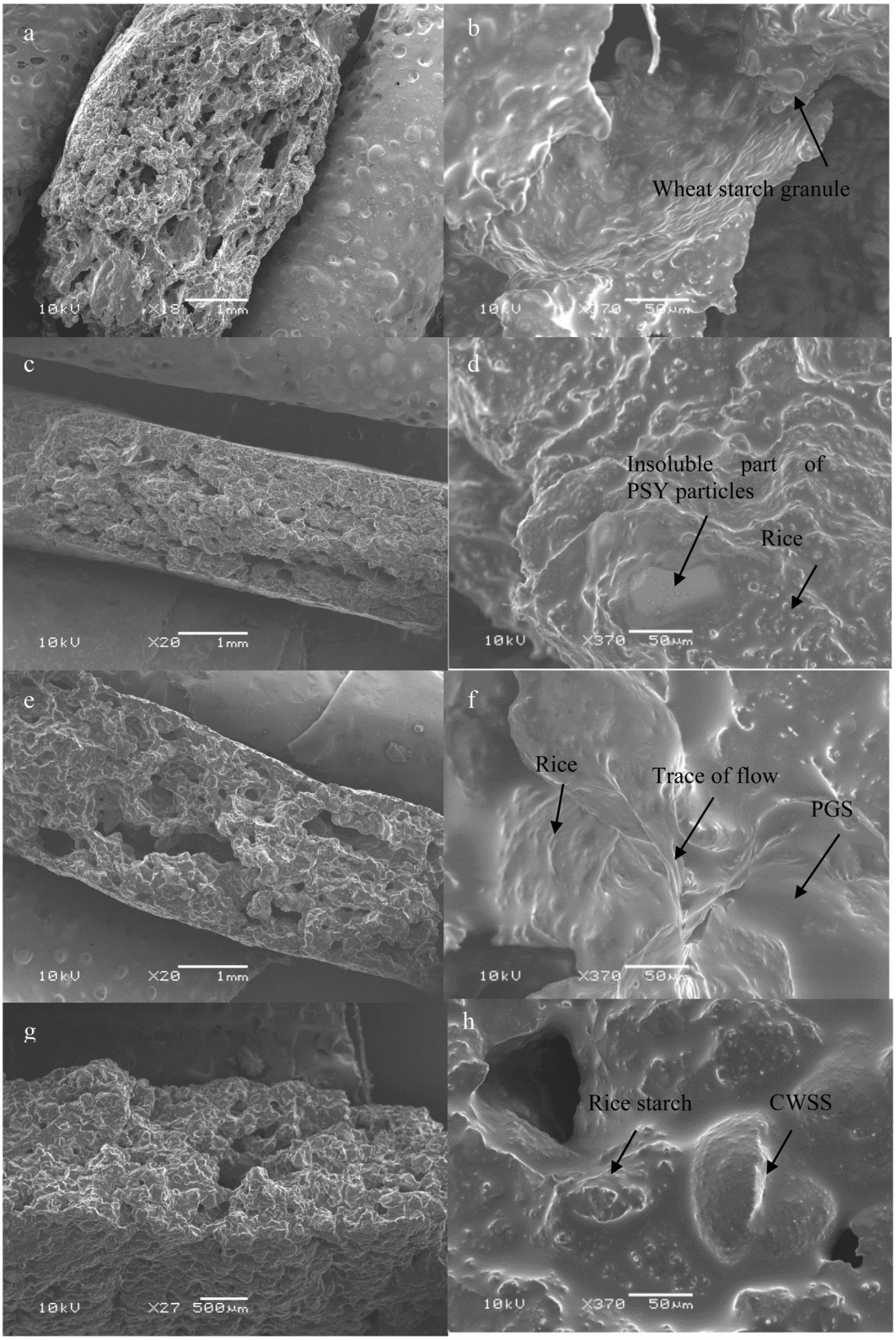

Fig. 5 Scanning electron microscopy images of the wheat crackers (a and b), PSY crackers (c and d), PGS crackers (e and f) and CWSS crackers (g and h).

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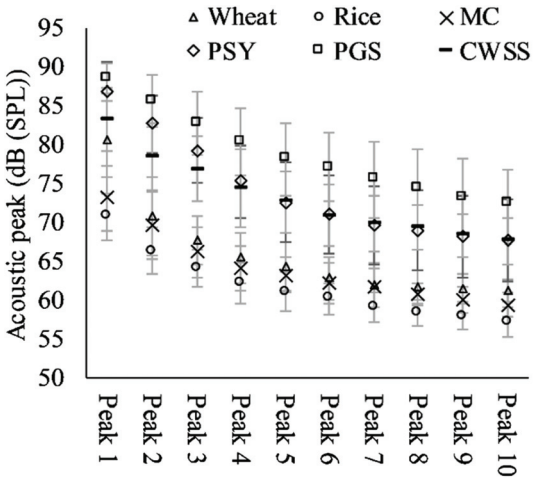

Fig. 6 The highest ten acoustic peaks of the sound pressure levels of the wheat and gluten free crackers. The acoustic peaks from peak 1 to peak 10 are shown from the highest to the lowest values from left to right.

tinuous matrix of the PSY crackers, therefore, predominantly consisted of the hydratable and gel-like part of PSY, which is highly branched heteroxylan. As for the PGS crackers (Fig. 6e and f), there were fewer rice starch granules in the matrix as 20% of rice flour was replaced by PGS, although some starch fragments and aggregates from PGS were observed. In addition to the observation that the PGS doughs were more fluid-like (Fig. 4), it was also flowable during baking as the trace of the extensional flow was observed. The flowability of the PGS doughs during baking allowed the formation and increase of gas cells, which contributed to the highest thickness increase as shown in Table 2. Nevertheless, some large swollen starch granules were observed in the CWSS crackers in addition to the rice starch, which is consistent with what is shown in Fig. 1j.

3.7. Textural properties and acoustic emission

The textural properties of the crackers were evaluated and the results are shown in Table 3. The wheat crackers showed the highest hardness with a relatively high fracturability. As for the gluten free crackers, the PSY crackers had hardness and fracturability closest to those of the wheat crackers, followed by the PGS crackers and CWSS crackers. The rice crackers and MC crackers, however, had very low hardness but high fracturability. It has been hypothesised that the interactions between the carbohydrate polymers at a low moisture level allow the formation of a crystalline-like zone which is critical to hardness and perception of the crispiness of snack products.[49] However, the texture of the wheat cracker is highly controlled by gluten,[50] while starch exists as intact granules embedded in the matrix of the gluten network and other ingredients as shown in Fig. 5. The high fracturability of the wheat crackers could be assigned to their higher thickness and flaky structure (Fig. 5a and b). The flaky layers were bent and broken gradually, which contributed to the initial force increase before the first peak, during which the distance indicated fracturability. The force increase with the gradual deformation and collapse of the walls of the gas cells has been observed in cellular products like bread.[51] However, a hard matrix was absent in the rice crackers, therefore they had the lowest hardness. The high fracturability of the rice crackers is also due to their weak structure. The gradual collapse of the structure caused the force to increase before the first decrease (fracturability). It could be speculated that the high hardness and relatively high fracturability of the PSY crackers are attributed to the self-interaction between the PSY polymers and interactions with other ingredients. Similarly, a matrix was formed by PGS, which played a similar role to the matrix or film of (pre)gelatinised starch in extruded cereal snacks.[52] However, the interaction between CWSS was limited to the level of that between granules because the integrity of starch granules was reserved. Hence, CWSS was less efficient than PGS in the formation of the continuous matrix. The low hardness and high fracturability of the MC crackers, which were similar to the rice crackers, indicate that the functionality of MC was limited possibly due to the low moisture level. The molecular mobility of MC was limited at the low water addition level, which inhibited the self-interaction between the MC molecules and the formation of a matrix to bind other ingredients.

When dry food is deformed under an applied force, strain energy is stored. Structural destruction occurs after a critical point is reached, which causes vibration and generates sound pressure waves, during which the stored energy is released as acoustic energy. The sound emission is correlated to the perception of crispiness or crunchiness.[53,54] The acoustic emis-

Table 3 Textural properties and acoustic emission of the crackers

	Hardness (g)	Fracturability (mm)	Maximum sound peak (dB(SPL))	Average peak decrease (dB(SPL))	Number of sound peaks (dB(SPL))
Wheat crackers	1005 ± 135^{d}	0.634 ± 0.088^{ab}	80.65 ± 4.95^{b}	20.74 ± 5.90^{b}	8.81 ± 3.75^{a}
Rice crackers[a]	150 ± 26^{a}	0.748 ± 0.204^{a}	70.72 ± 3.05^{a}	15.55 ± 2.70^{a}	11.57 ± 3.08^{ab}
MC crackers[a]	326 ± 50^{b}	0.727 ± 0.133^{a}	73.12 ± 4.13^{a}	18.69 ± 5.58^{ab}	11.88 ± 4.87^{ab}
PSY crackers	801 ± 166^{c}	0.568 ± 0.117^{b}	86.87 ± 3.73^{c}	19.78 ± 5.51^{ab}	15.27 ± 5.18^{b}
PGS crackers	667 ± 129^{c}	0.330 ± 0.087^{c}	88.41 ± 1.86^{c}	23.27 ± 3.28^{b}	30.31 ± 11.29^{c}
CWSS crackers	461 ± 163^{b}	0.329 ± 0.129^{c}	83.20 ± 4.06^{b}	23.08 ± 4.60^{b}	22.00 ± 8.49^{d}

[a] Rice and MC crackers were sheeted to 2.6 mm, and the other crackers were sheeted to 1.6 mm. Data are shown as mean ± standard deviation. The different letters associated with the values in the same column indicate significant differences ($p < 0.05$).

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sion during the three-point bending tests was recorded, and the maximum sound peak, average peak decrease and number of sound peaks are shown in Table 3. The first ten acoustic peaks recorded for each cracker were ordered from high to low and are shown in Fig. 6. As shown in Table 3, the rice crackers and MC crackers had lower maximum sound peaks and average peak decreases compared to the wheat crackers. The observation is consistent with hardness. However, the addition of CWSS, PSY, and PGS in the crackers significantly increased the maximum sound peak and average peak decrease compared to the wheat crackers, which is also shown in Fig. 6, suggesting that the highest ten acoustic peaks of the PGS, CWSS, and PSY crackers were generally higher than those of the wheat, rice, and MC crackers. The observations indicate that higher levels of sound were emitted during the breaking of these three types of crackers. The gluten free crackers also had a higher number of sound peaks than the wheat crackers, which was opposite to hardness. During each structural destruction, there is a force decrease accompanied by an acoustic event.[53,55] Saeleaw *et al.*[56] observed the decrease of hardness with a lower number of sound peaks due to the increase in the pores in the products. Therefore, the high maximum sound peak accompanied by the high hardness of the wheat crackers is assigned to the gluten network, while the low number of sound peaks indicates that there might be weak points in the flaky structure which do not contribute to the strength of the structure and sound emission. In contrast, the gluten free crackers did not show a flaky structure. The breaking and sound emission during tests were caused by multiple fractures across the whole cracker due to inhomogeneity and weak points instead of predominantly at the contacting position with the probe. It is also noticed from Fig. 6 that compared to the wheat crackers, there was less difference between the highest acoustic peak and the second highest peak of all gluten free crackers, which indicates that the break and release of the acoustic energy were less sharp than those of the wheat crackers. It also evidences the multiple fractures of the gluten free crackers.

4. Conclusion

The production of gluten free crackers was developed by the incorporation of PSY, MC, PGS, and CWSS and compared to wheat crackers. PSY and PGS form matrices which hold flour particles together and form cohesive doughs. The PSY and PGS matrices in the crackers contribute to comparable hardness and sound release to wheat crackers during breaking. Multiple fractures occur across gluten free crackers during breaking, while there is a shape break of wheat crackers due to their laminar structure. PSY increases the moisture content and lowers the water activity of gluten free crackers due to its high water retention ability. PGS significantly increases the shapeability of gluten free doughs and allows them to flow during baking. MC increases the viscosity and leads to significant effects on the swelling and breakdown behaviours of starch granules which dramatically modify the pasting profile. However, the low water addition level of crackers inhibits self-interaction of MC molecules and limits their functionality; therefore MC crackers have a poorer quality similar to rice crackers. CWSS crackers also show certain levels of hardness, fracturability, and sound release comparable to wheat crackers but the interaction is mainly between their starch granules which limit their ability to form a strong matrix. Therefore, further studies will be based on PSY and PGS and focus on the modification of the process.

Conflicts of interest

Bruce R. Linter is employed by PepsiCo, Inc. The views and opinions expressed in this manuscript are those of the authors and do not necessarily reflect the position or policy of PepsiCo, Inc.

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