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Effect of carboxymethyl cellulose and xanthan gum on the quality of gluten-free cookies from rice varieties with different amylose contents

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Abstract

Amylose and hydrocolloids may influence the rheology of dough and quality parameters of gluten-free cookies. The present study addresses an aspect of the effect of rice varieties (IR50404, Song Hau and Jasmine) and hydrocolloids (hydrocolloids (carboxymethyl cellulose, CMC, and xanthan gum, XG) with concentrations from 1.0 to 3.0% on rice cookie quality parameters (moisture content, water activity, density, expansion ratio, hardness and sensory value). The results noted that amylose content of rice varieties mainly influenced the hardness, expansion and thickness of cookies. The CMC was strongly and positively correlated with hardness while XG had a sharply positive relation with hardness, expansion ratio and water activity of cookies. Their interaction contributed mainly to the density of the rice cookie. The most appropriate hydrocolloid contents were 2% of CMC and 1% of XG for making good quality cookies from the Song Hau rice variety with medium amylose content.

1. Introduction

A cookie is a low moisture content food, one of the most popular snacks around the world, prepared with wheat flour, egg, fat, milk, salt, sugar and a leavening agent. A micro-baking simulation showed the expansion of the bubbles in the batter (Diez-Sánchez *et al*., 2020). These ingredients have many functional properties and serve to improve the colour, flavour, texture, density and consistency. They are characterized by having a well-developed gluten structure, but with increasing amounts of sugar, fat or gluten become more extensible and less elastic (Diez-Sánchez *et al*., 2020).

However, around 1% of individuals worldwide have been suffering from a Celiac disorder - an autoimmune enteropathy caused by gluten proteins found in wheat, barley, and rye. It affects people who are genetically predisposed to it. Gluten causes atrophy of the small intestine mucosa, significantly weakened intestinal absorption and extreme malnutrition. Therefore, a gluten-free diet for the whole life is the only recovery option for Celiac patients (Green and Cellier, 2007; Niewinsky, 2008; Husby *et al*., 2012; Garnier-Lengline *et al*., 2015).

Among the wheat substitutes, rice flour is extensively used in the preparation of gluten-free products, including cookie due to its hypoallergenic properties and large amount of easily digestible carbohydrates (Demirkesen *et al*., 2010). Besides, rice flour is ground from broken rice with a huge source and different amylose varieties in Vietnam for high demand to increase the value of rice-based food products for markets. Amylose is an important component of starch granules, affecting the physical properties of starch and starch gel such as gelatinization, retrogradation, swelling power, hardness, viscosity and viscoelasticity (Piyarat, Sandra, Imad *et al*., 2005; Nhan and Copeland, 2014). However, rice dough was unable to maintain the gas released during baking due to its lack of a viscoelastic network formed by protein, resulting in a crumbly product.

Therefore, carboxymethyl cellulose (CMC) and xanthan gum (XG) have been used commonly in food to improve the physical and sensory characteristics of gluten-free products. Both have a cellulose backbone of (1,4)-D-glucose units and substituents that protrude from the main chain. The chain of XG, a microbial heteropolysaccharide, is replaced with a trisaccharide side chain on alternative glucose residues (Lee and

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Brandt, 2002), while carboxymethyl groups are the substituents in CMC. XG is used to enhance the texture and moisture preservation in the dough and strengthen the effectiveness of other hydrocolloids (Lee *et al.,* 2002). CMC is known for its ability to regulate rheological properties and improve moisture preservation in cereal doughs (Sindhu and Bawa, 2000).

Therefore, the present work aimed to evaluate cookies in terms of chemical, physical and sensory characteristics by influences of amylose contents from Vietnamese rice flours (Song Hau, Jasmine and IR50404 varieties) and hydrocolloid (CMC and XG) contents. Furthermore, strong relationships were identified between the variability of hydrocolloids and amylose contents with the characteristics of cookies.

2. Materials and methods

2.1 Rice varieties

Rice flours of different varieties (IR50404, Song Hau and Jasmine), egg, salt, margarine (Meizan), icing sugar (Bien Hoa) were purchased from Mega market in Can Tho. Vanilla extract (Rayner's), sodium bicarbonate (Arm and Hammer), XG (Bob's Red Mill), CMC and citric acid were purchased from Southern Chemicals Joint Stock Company (Vietnam).

2.2 Cookie formula

The dough formula consisted of 30 g rice flour, 22 g icing sugar, 20 g margarine, 10 g whole egg liquid, 0.6 g sodium chloride, 0.5 g vanilla extract, 0.5 g water, 0.9 g sodium bicarbonate. XG and CMC were combined each of 1, 2 and 3% (as partial replacement for rice flour).

2.3 Cookie preparation

Icing sugar, margarine, vanilla and whole egg were stirred for 2 mins with a handy mixer (Philips). Rice flour, sodium bicarbonate, XG and CMC were blended with the mixture for 2 mins. Then citric acid was dissolved in water and mixed in the dough. The dough wrapped in a plastic bag was rested in the fridge for the internal temperature to be 15°C. It was sheeted and using the cylinder frame of 30 mm diameter and 5 mm height (or thickness). After baking at 165°C for 25 mins, the cookies were cooled for 30 mins at room temperature about 25°C, packed in sealed polyacrylamide (PA) bags, and stored at room temperature for further analysis.

2.4 Amylose content determination

Rice starch was isolated using the method of Wang and Wang (2011) with some modifications. Rice grains (100 g) were soaked in 200 mL of 0.1% NaOH for 18 hrs, and ground using a blender (Philips) for 2 mins. The starch slurry was passed through a micro-sieve 63 µm and centrifuged at 1400×*g* for 10 mins. The supernatant was removed. The starch layer was washed with distilled water and centrifuged. Then the starch was re-slurried and neutralized with 1.0 M HCl to pH 6.5 and centrifuged at 1400×*g* for 10 mins. Starch was washed with distilled water at least 3 times before being dried at 45°C until 12% moisture content and stored at room temperature.

Amylose of rice starch was determined using the method of Chrastil (1987). Starch (20 mg) was solubilised by adding 4.0 mL of deionised water and 2.0 mL of 1.0 M NaOH solution and heated for 30 mins in a boiling water bath with occasional mixing. An aliquot (0.1 mL) of this solution was added to 5 mL of 0.5% trichloroacetic acid in a separate test tube, the tubes were vortexed and 0.05 mL of an iodine solution (1.27 g I_2 and 3.0 g KI/L) was added and mixed in immediately. Potato amylose and amylopectin from Sigma Chemical Co. (St. Louis, MO, USA) were used to make a calibration curve with different amylose: amylopectin ratios. Total amylose values were obtained from iodine binding with the starch solution in hot ethanol. The absorbance was read at 620 nm after 30 mins at 28°C against water blank. Amylose content (AC) was determined by reference to a standard curve and expressed as g/100 g dry weight.

2.5 Moisture content and water activity determinations

Moisture content was determined by using the standard methodology of the Association of Official Analytical Chemists (2010). Water activity (a_w) was measured with the method according to Suzanne (2010).

2.6 Dimension measurement

Cookies were evaluated for the weight (g), thickness (mm) and diameter (mm). Diameter and thickness were measured using a Vernier Caliper. To determine the diameter, six cookies were placed edge to edge and the total diameter was measured. The cookies were rotated at 90° for duplicate reading. The average diameter was calculated. To determine the thickness, six cookies were placed on top of one another. The total thickness was measured. This process was repeated twice, and the average thickness was calculated.

2.7 Volume expansion measurement

The volume of the cylinder dough (*V*) was calculated using the following equation:

$$V = \pi.r^2.h$$

Where, *p*, 3.14; *r*, the radius of the dough (15 mm); *h*, the height (or thickness) of the dough (5mm).

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The volume of baked cookies was measured by the rapeseed displacement method (American Association of Cereal Chemists (AACC) Method 10-05.01). The ratio of expansion (ER) was defined as the ratio of the baked cookie volume and the dough volume (*V*).

2.8 Texture measurement

The texture properties of baked cookies were determined using a dual-range force sensor of Vernier (USA). The pre-test, test and post-speeds were 1.75 mm.s^{-1}. Texture determination was repeated 3 times. The hardness of the cookies was the maximal force to break the cookies by the cylinder prober with a diameter 2 mm.

2.9 Sensory evaluation of cookies

Baked cookies were organoleptically evaluated for their sensory characteristics. Cookie samples were served on white, odourless and disposable plates, and water was provided for rinsing between samples for ten panellists. Samples were scored for colour, flavour, texture and overall acceptability according to the method of Poste *et al.* (1991).

2.10 Statistical analysis

The data obtained were statistically treated by means of a one-way ANOVA, while the means were compared by Fisher's least significant difference test at a significance level of 0.05. These analyses were performed using Statgraphic IV software.

3. Results and discussion

3.1 Effect of rice varieties on cookie quality

The amylose content (AC) of rice starches in this study ranged from 12.04 to 25.25% (Table 1). According to Prisana *et al*. (2007), based on amylose content, these varieties can be classified into low amylose (8-20%) for Jasmine and Song Hau and high amylose (>25%) for IR50404. The Jasmine rice starch AC (12.04%) in this study is similar to Jasmine (15.3%) from Huynh *et al*. (2016) and Jasmine (15.1%) from Piyarat, Sandra, Pasawadee *et al*. (2005). The AC of IR50404 rice starch (25.25%) in this study was a little lower than the one (26.9%) of IR50404 from Huynh *et al*. (2016).

3.1.1 Moisture content

The cookie moisture content (MC) varied considerably among samples, ranging from 2.63% to 3.69% (Table 1). The MC of cookies were lower if they were made from higher AC rice flour. The cookies with the lowest (2.63%) MC were made from the highest AC flour (IR50404). High amylose starch had a low capacity to bind water (Varavinit *et al*., 2003).

3.1.2 Water activity (a_w)

Jasmine has the highest water activity value (0.42±0.03) compared to the 2 other samples with higher AC (Table 1). Despite their variation in amylose content, Song Hau and IR50404 flours gave cookies with a similar a_w. However, no statistically different values were found among the samples.

3.1.3 Expansion ratio

The expansion ratio (ER) values were differently significant between cookie samples (Table 1). The ER was correlated strongly (p<0.001) and negatively with amylose content (Table 2). Cookies obtained the largest ER (3.14 times) by adding Jasmine rice flour but the lowest ER (2.51 times) with IR50404 rice flour. The addition of rice flour with a lower amylose content resulted in cookies with a higher ER. A thin and tender membrane in baked rice-amylopectin products holds gases which expand product volume by heating, whereas the heterogeneous and hard gel in amylose-rich starch persisted without expanding, leading to a smaller expansion (Gimeno *et al*., 2004).

3.1.4 Thickness

Cookie thickness varied significantly among samples and was shown to be negatively associated with amylose content but positively correlated with the expansion ratio (Table 2). The thickness (11.57±0.16) was the highest for cookies by adding Jasmine rice flour, and the lowest (10.64±0.14) by adding IR50404 rice flour (Table 1). The Jasmine flour with low amylose content may have more viscosity than the IR50404 with high amylose. Because the rice flour with low amylose had a higher viscosity than the one with high amylose (Kemashalini *et*

Table 1. Starch amylose and the characteristics of cookies made from the flours of different rice varieties.

Rice varieties	Characteristics of cookies from flour of the rice varieties						
	AC of starch (%)	MC (%)	a_w	Expansion ratio (time)	Thickness (mm)	Density (g/mL)	Hardness (N/mm^2)
IR50404	25.25±1.56^a	2.63±0.15^c	0.37±0.03^a	2.51±0.02^c	10.64±0.14^b	0.44±0.03^a	4.03±0.15^c
Song Hau	18.69±2.24^b	3.11±0.34^b	0.37±0.03^a	2.71±0.03^b	10.96±0.35^b	0.46±0.03^a	4.88±0.14^b
Jasmine	12.04±1.45^c	3.69±0.25^a	0.42±0.03^a	3.14±0.05^a	11.57±0.36^a	0.46±0.01^a	7.11±0.16^a

Values are presented as mean±SD of triplicates. Values with different superscripts within the same column are statistically significantly different at 5% significant level (p<0.05). AC: amylose content, MC: moisture content.

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Table 2. Correlation between the characteristics of cookies from multiple rice varieties.

	AC	Moisture	ER	Hardness	Density	Thickness
Moisture	-0.96***					
Expansion ratio	-0.98***	0.92***				
Hardness	-0.95***	0.95***	0.97***			
Density	0.35	-0.5	-0.57	-0.62		
Thickness	-0.90***	0.93***	0.88**	0.92***	-0.51	
a_w	0.52	0.51	0.43	0.54	-0.22	0.51

***p ≤ 0.001, **0.001 < p ≤ 0.01, *0.01 < p ≤ 0.05.

al., 2018). With the more viscous paste, the higher vertical growth cookies rise while with the lower viscosity, the gases escape from the starch network more easily during baking and expand less volume of the cookies (Brites *et al*., 2019). Therefore, the rice Jasmine flour with the lowest amylose and the highest viscosity traping high gas bubbles produced the baked cookies with the largest thickness.

3.1.5 Hardness

Cookies made with Jasmine rice flour reached the strongest hardness (7.11±0.16N/mm^2) but cookies from IR50404 with hardness (4.03±0.15N/mm^2) (Table 1). The amylose content had a strong (p<0.001) and negative correlation to the hardness of baked cookies (Table 2). Since the amylose-rich cookies lost free water without contributing to the formation of a continuous structure resulted in the product's poor tolerance to force application or deformation. The crystalline regions of high amylose rice starch have low hydration and swelling (Park *et al*., 2007; Chung *et al*., 2011).

3.1.6 Sensory evaluation

A consumer acceptability test was carried out to evaluate the effect of rice flour on rice-based cookies. Results show that, in general, cookies from the Song Hau rice flour obtained the highest acceptability and texture scores among the samples (Figure 1). However, no statistical variation was found in the colour and taste of cookies from different varieties. From the sensory results, it can be concluded that the Song Hau rice variety should be chosen as the suitable flour to make cookies.

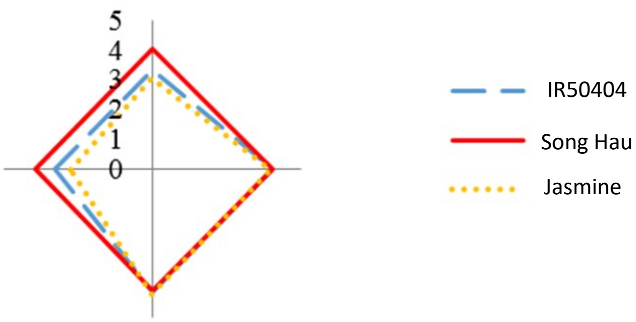

Figure 1. Effect of the flours from different rice varieties on the sensory values of baked cookies.

3.1.7 Correlation between amylose contents of rice starch and characteristics of rice cookies

Pearson correlation coefficients to test for relationships between starch amylose content and cookie characteristics are shown in Table 2. Some of the significant correlations observed between starch characteristics across the full data set were as expected that amylose content is related strongly (p<0.001) and inversely with characteristics (MC, ER, hardness and thickness). However, some other significant correlations (p<0.01) were noted, including positive relationships between ER and thickness. The MC had strongly (p<0.001) positive correlations with ER and thickness. It may be that the cookies made from rice flour containing lower amylose contents (or higher amylopectin contents) have higher MC resulting to create more water vapour pressure during baking and an increase in expansion ratio and thickness.

3.2 Effect of carboxymethyl cellulose and xanthan gum concentration on cookie quality

Xanthan gum content accounted for the majority of the variance in water activity (58%), moisture content (50%) and expansion ratio (41%). CMC content accounted for a large part of the total variability of hardness (61%) and density (32%), but contributions to the variability of these rice cookies' properties from factors related to the thickening agents were also significant (p<0.001, Figure 2).The physicochemical and sensory properties of gluten-free biscuits prepared from buckwheat flour with the incorporation of various gums such as carboxymethyl cellulose (CMC), hydroxypropyl methylcellulose (HPMC) and xanthan gum (Kaur *et al*., 2015). Xanthan gum has been used to improve the network and structure of gluten-free products (Lazaridou *et al*., 2007).

3.2.1 Moisture content

The positive relationship of the moisture contents was strongly related (p<0.001) with XG and moderately related (p<0.01) with CMC (Table 3). Significant variations in cookie moisture were recorded due to the addition of CMC and XG, ranging from 2.31% to 5.36%

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Table 3. Correlation coefficients (r) of CMC, xanthan gum and the characteristics of cookies.

	CMC	XG	Hardness	Density	ER	MC
Hardness	0.43*	0.77***				
Density	0.33	0.2	0.42*			
ER	0.44*	0.63***	0.68***	-0.49		
MC	0.69***	0.56**	0.85***	0.47*	0.60***	
Water activity	0.33	0.76***	0.69***	0.01	0.76***	0.64***

***p ≤ 0.001, **0.001 < p ≤ 0.01, *0.01 < p ≤ 0.05, AC: amylose content, MC: moisture content, CMC: carboxymethylcellulose, XG: xanthan gum, ER: expansion ratio

(Figure 3). Cookies with 3% CMC, 3% XG had the greatest MC value (5.36%). A positive correlation between cookie moisture and gum content, is similar to previous findings (André *et al*., 2005; Thejasri *et al*., 2017). Water molecules orient toward the hydroxyl (or carboxyl) groups of the hydrocolloid molecules during hydration, improving water holding capacity (Dickinson, 2003; Nammakuna *et al*., 2015). The hydrocolloid structure affects the water absorption of baking dough. As shown in Figure 2, XG had a greater effect on the cookie moisture (accounted for 50.33% of the variance) compared with CMC (accounted for 30.67%). Significant variations in water retention were observed in the samples at various gum concentrations, with more apparent in samples containing XG (Andrés *et al*., 2005). The difference was explained by a larger number of free carboxyl groups presented in XG molecular to have a higher water absorption ability than CMC (Andrés *et al*., 2005).

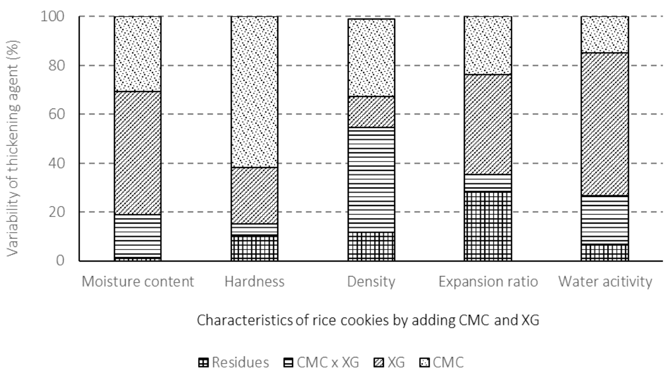

Figure 2. Contributions to the variance of characteristics of rice cookies from CMC and Xanthan gum.

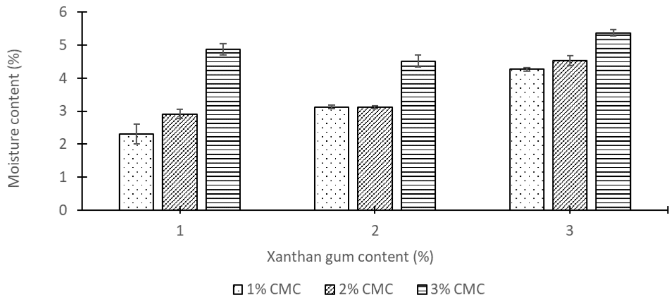

Figure 3. Effect of CMC and xanthan gum on the moisture content of cookies.

3.2.2 Water activity (a_w)

The addition of hydrocolloids had a positive association with cookie water activity (a_w), with the higher the amount of added CMC and xanthan, the higher the a_w value (Figure 4). The various XG contents made significant differences in the a_w between the cookies. Adding 3% of XG in the dough resulted in cookies with the highest a_w (0.43). Benkadri *et al*. (2018) also found that water holding capacity and viscosity significantly increased when xanthan gum added in dough resulting in baked cookies that have high moisture content and high a_w. The water activity was positively related with the water content of the baked cookies (Table 3).

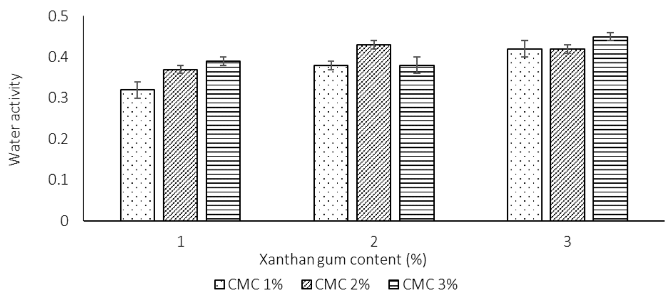

Figure 4. Effect of CMC and xanthan gum on the water activity of cookies.

3.2.3 Expansion ratio

The positive correlation of the cookies' ER was strong (p<0.001) with adding XG content; and weakly (p<0.05) with adding CMC content (Table 3). The addition of CMC and XG ratios led to significant variations in the cookie expansion ratio, ranging from 2.40 to 3.14 times (Figure 5). When 3% of CMC and 3% of XG were mixed in the dough, baked cookies obtained the largest ER. A positive correlation was found between hydrocolloid addition and the expansion rate. Both XG and CMC can keep dough viscous at elevated temperatures, due to starch–hydrocolloid reorganization, which leads to a higher number of crosslinks being formed during the gel's conformation process (Andrés *et al*., 2005). Therefore, a high number of water molecules bound in starch–hydrocolloid networks generate a large vapour pressure increasing the baking cookie volume (Mali *et al*., 2003; Gimeno *et al*., 2004; Lazaridou *et al*.,

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2007).

The expansion ratio of cookies reached 3 times with the addition of 3% XG but only 2.8 times with the addition of 3% CMC. Gimeno *et al.* (2004) found a similar pattern in the study on corn pellets. The difference in water binding capacity of the two hydrocolloids led to variation in gas retention of the cookie dough.

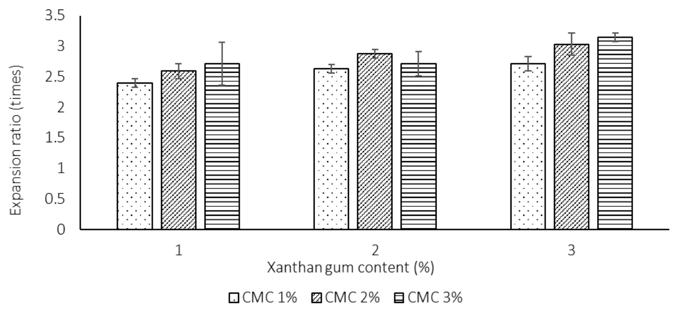

Figure 5. Effect of CMC and xanthan gum on the expansion ratio of cookies.

3.2.4 Density

Cookies had the lowest density if the dough was mixed with 2% CMC and 1% XG (Figure 6). The cookies' densities increased by adding CMC and XG with more concentrations than these. Viscosity was higher and the specific volume of bread was small if the dough was added with a high concentration of XG (Turkut *et al*., 2016).

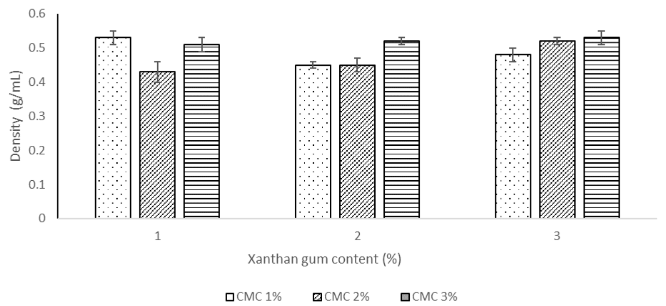

Figure 6. Effect of CMC and xanthan gum on the density of cookies.

3.2.5 Hardness

The addition of CMC and XG increased cookie hardness (Figure 7). The cookie added with 3% CMC, 3% XG had the highest hardness value (8.44 N/mm^2). It may be that a film or a network in the hydrocolloid-added samples (Nammakuna *et al*., 2015; Rajesh *et al*., 2015). When these films dehydrate in the baking process, tough skin forms on the cracker's surface, enhancing the cracker's resistance to breakage and hence the fracture force and hardness values. The hardness of cookies had a strong ($p<0.001$) positive relationship with XG concentration and weakly ($p<0.05$) positive correlation with CMC concentration (Table 3).

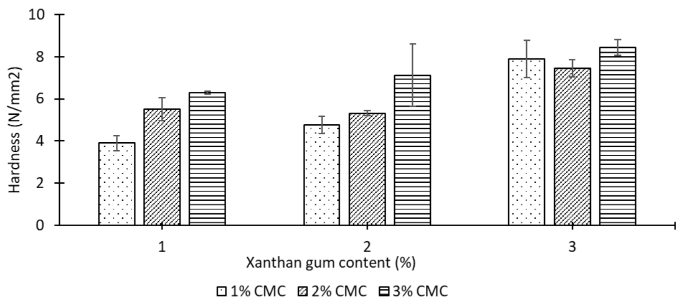

Figure 7. Effect of CMC and xanthan gum on the hardness of cookies.

3.2.6 Correlation between carboxymethyl cellulose, xanthan gum and characteristics of rice cookies

Pearson correlation coefficients to test for relationships between hydrocolloids and properties of cookies from the Song Hau rice variety are presented in Table 3. The results show that strongly positive correlations ($p<0.001$) were noted between XG content with hardness, expansion ratio and water activity of cookies. CMC had a strongly positive correlation ($p<0.001$) with moisture content, a weakly positive correlation ($p<0.1$) with hardness and ER. The ER was strongly and positively correlated ($p<0.001$) with MC and water activity. CMC and XG have high water holding capacity which contributes to high MC and water vapour pressure in baked cookies resulting increase in ER. On the other hand, XG has a strongly positive relationship with the hardness of baked cookies. The strong cookie texture may result from XG enhancing the texture in the dough and strengthening the effectiveness of other hydrocolloids (Lee *et al.,* 2002) contributing in improving the network and texture of the baked cookies.

3.2.7. Sensory evaluation

Baked cookies achieved the best sensory values (colour, odour, taste and texture) when 2% of CMC and 1% of XG were mixed in the dough (Table 4). Adding CMC and XG higher than these concentrations increased water content and water activity in baked cookies (Benkadri *et al*., 2018) which caused lighter colour and less sweet taste in the cookies. The cookies had a little odour of the gums. The cookies containing higher CMC and XG concentrations had a relatively harder and stickier texture. The reason may be explained that higher concentrations of CMC and XG cause smaller pore volume resulting in higher hardness; and more viscosity resulting in stronger stickiness.

4. Conclusion

Amylose content had inverse and strong correlations

Table 4. Effect of CMC and xanthan gum on the sensory value of cookies.

	CMC (%)	XG (%)			Average
		1	2	3	
Color	1	4.23±0.25	4.00±0.10	4.41±0.36	4.21[b]
	2	4.52±0.42	4.51±0.30	4.40±0.56	4.47[ab]
	3	4.59±0.20	4.50±0.10	4.63±0.32	4.54[a]
	Average	4.42[a]	4.33[a]	4.48[a]	
Texture	1	3.00±0.07	4.00±1.00	2.33±1.52	3.11[a]
	2	4.33±0.58	4.67±0.55	2.33±0.58	3.78[a]
	3	3.33±0.52	3.67±0.58	2.00±1.00	3.00[a]
	Average	3.56[a]	4.11[a]	2.22[b]	
Taste	1	4.50±0.50	4.50±0.87	3.12±0.29	4.06[a]
	2	4.33±0.58	4.83±0.29	3.67±0.61	4.28[a]
	3	2.83±0.30	3.75±0.25	2.80±0.75	3.12[b]
	Average	3.89[a]	4.36[a]	3.21[b]	

Values are presented as mean±SD of triplicates. Values with different superscripts within the same column are statistically significantly different at 5% significant level ($p<0.05$).

with the hardness, expansion ratio, and thickness of rice cookies. The Song Hau rice with a medium amylose content was suitable for making cookies with more favourable sensory attributes than high amylose rice varieties. The CMC was strongly and positively correlated with hardness while XG had a strongly positive relation with hardness, expansion ratio and water activity of cookies. Their interactions contributed mainly to the density of the rice cookies. Medium amylose rice can be used for making free gluten products (cookies) with good quality by adding hydrocolloid contents (CMC and XG). Gluten-free products (cookies) can obtain good quality when it made from rice flour (the Song Hau variety) with medium amylose and added with hydrocolloids (1% of XG and 2% of CMC).

Conflict of interest

The authors declare no conflict of interest.

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