Heliyon 9 (2023) e18545

Contents lists available at ScienceDirect

Heliyon

journal homepage: www.cell.com/heliyon

Effects of sodium carboxymethyl cellulose on storage stability and qualities of different frozen dough

Tong-Chao Su *, Wen-Kai Du, Bing-Yan Deng, Jie Zeng, Hai-Yan Gao, Hai-Xu Zhou, Guang-Lei Li, Hao Zhang, Ya-Meng Gong, Jin-Yu Zhang

School of Food Science, Henan Institute of Science and Technology, Xinxiang, 453003, China

A R T I C L E I N F O

Keywords:
Hydrocolloids
Sodium carboxymethyl cellulose
Frozen dough
Rheology
Microstructure

A B S T R A C T

Hydrocolloids as Additives have been used for improving the quality of frozen dough for a long time. In this work, the effects of sodium carboxymethyl cellulose (CMC) on quality changes of frozen dough in storage were studied. The water loss rate of the dough and water holding capacity were measured. Rheological and texture properties of the frozen dough were measured by a rheometer and a texture analyzer, respectively. Scanning electron microscopy (SEM) was used to characterize surface network structure and protein structure changes of the frozen dough. Our results reveal that the addition of CMC can inhibit the formation of ice crystals and recrystallization, thus effectively stabilizing the molecular structure of starch, and resulting in more uniform moisture distribution in the frozen dough. When 3% addition of CMC, the water holding capacity of the two kinds of dough reached the best, and the water loss rate of corn dough reached the lowest. The cohesion of the two kinds of dough reaches the maximum with 3 wt% addition of CMC, while the hardness and chewiness of wheat and corn multigrain dough reaches the maximum with 3 wt% and 4 wt% addition of CMC, respectively. The results show proper CMC addition (3 wt% and 4 wt%) finally improves the stability and qualities of the frozen dough. The research concerning the effects of CMC on quality of frozen dough provides better understanding for the frozen food industry.

1. Introduction

Sodium carboxymethyl cellulose (CMC) as a kind of hydrocolloids is always used for improving the quality of food industry for a long time. The molecular chain of CMC is abundant in hydroxyl and carbonyl groups, which is insoluble in organic solvents and has strong hygroscopicity [1]. CMC is also widely used as a thickener in the food industry, a drug carrier in the pharmaceutical industry, a binder or an *anti*-redeposition agent in the daily chemical industry, and so on [2]. Due to its special chemical structure, CMC has a variety of unique physical and chemical properties, such as hydrophilicity, film formation ability, thickening, antibacterial properties, adhesion, and adsorption [3]. Many researchers have reported quality improvement of wheat flour and its end products by adding hydrocolloids. Studies have shown that addition of hydrocolloids can strengthen the gluten protein structure, increase the water holding performance and improve the stability of the dough, thus improving texture properties, appearance, frozen storage stability, and nutritional quality of related products, which is a relatively simple and effective method [4,5].

* Corresponding author.
E-mail address: sutongchao66@163.com (T.-C. Su).

https://doi.org/10.1016/j.heliyon.2023.e18545
Received 19 February 2023; Received in revised form 15 July 2023; Accepted 20 July 2023
Available online 23 July 2023

Many researchers have reported the effects of hydrocolloids on the properties of dough and flour products. Aravind et al. reported the effects of soluble fibers: guar gum (GG) and carboxymethyl cellulose (CMC) on the technological, sensory, and structural properties of durum wheat pasta. Their study revealed that addition of soluble fibers (CMC and GG) can significantly reduce in vitro digestibility of starch [6]. Tang et al. reported the effects of CMC combined with salt on the structural properties of wheat gluten protein. The study showed that CMC can protect the energy-stable disulfide bond in gluten protein from being destroyed by salt, and that combination of CMC and salt can improve thermal stability of gluten protein [7]. Chen et al. [8] reported the effects of CMC on the rheological properties and microstructures of wheat gluten protein under different pH conditions, showing that CMC-gluten exhibits stronger solid-phase behavior under alkaline conditions than under acidic conditions. Hu et al. [9] explored the effects of wheat gluten and CMC on the rheological properties and gluten network of potato-wheat compound flour, revealing that the interaction of CMC and gluten can reduce discontinuities of gluten matrix and aggregation of gluten proteins caused by the addition of potato flour, which makes the gluten network more closely branched, and improves gas storage capacity and stability of dough. Sim et al. reported the effects of non-starch polysaccharides on the quality and function of steamed bread and dough. The results showed that CMC can improve the quality of steamed bread, thus preventing spoilage and ensuring longer shelf life [10].

These studies mainly focus on the effects of CMC on the quality of pasta staple products. Few studies, however, have been conducted on the effects of hydrocolloids on the changes of protein network structure, moisture and microstructure of dough in frozen storage. The regulation mechanism of hydrocolloid on wheat dough and corn mixed grain dough during freezing was further studied from microscopic aspect. Based on previous research, in this work, we add CMC at different concentrations (0–5 wt%) into wheat flour and corn multigrain flour, and treat the dough under frozen storage. We study the rheological properties, moisture distribution, texture properties and microstructure of dough with CMC at different concentrations, which enriches the theoretical research concerning the effects of CMC on the quality changes of dough in frozen storage. Our study is innovative and practical compared with previous studies, which, we hope, can provide a theoretical basis for the effects of hydrocolloids on the quality changes of frozen dough in storage process, and broaden applications of hydrocolloids in food processing.

2. Materials and methods

2.1. Materials

Flour was prepared using commercial wheat and corn multigrain flour (Xinxiang Xinliang Grain and Oil Processing Co., Ltd.), yeast (Angel Yeast Co., Ltd.), sodium carboxymethyl cellulose (CMC) was prepared from Zhengzhou Yuhe Food Additives Co., Ltd. and the CMC had a degree of substitution of 0.9 carboxymethyl groups per anhydroglucose unit, white granulated sugar (Shanghai Sugar Tobacco (Group) Co., Ltd.), distilled water, glutaraldehyde (Wuxi Yaodexin Chemical Products Co., Ltd.).

2.2. Preparation of dough

Dough was made according to Wang's method [11] with some modifications: wheat dough is flour 100 g (corn multi-grain dough is 10 g corn flour + 90 g flour), 1 g of highly active dry yeast, 50 g of distilled water, 1 g of white granulated sugar, 0–5 g of CMC (0, 1, 2, 3, 4, 5 g). The specific production process of dough is as follows: all raw materials were weighed at room temperature of 25 °C according to the formula, mix well in a dough mixer (DL-CO3, Dongling Electric Appliance Co., Ltd.). Then kneaded vigorously for 10 min to form dough which was fermented at 37 °C for 30 min in a constant temperature chamber (HYC-TH-80DH, Dongguan Hongjin Testing Instrument Co., Ltd.). After the fermentation was completed, the fermented dough was divided into a number of 5 g small pieces wrapped them separately in plastic wrap. The remaining dough was stored in a fresh-keeping bag as fresh dough. The fresh dough was frozen at −30 °C for 1.5 h, then transferred to a polyethylene sealing bag, and stored at −18 °C for 8 h (BC/BD-106DT Meiling freezer, Changhong Meiling Co., Ltd.) for further use. The frozen dough was defrosted at 25 °C for 30 min while measurements were made. A total of twelve groups of samples were prepared shown in Table S1 and Table S2.

2.3. Frozen temperature curves

The frozen temperature curves for dough were determined using the method described by Ban's method with some modifications [12]. Put the prepared wheat dough (from WF-0 to WF-5) and corn dough (from CF-0 to CF-5) was placed in a freezer, and the sensor of a temperature recorder (L93-1, Hangzhou Luge Technology Co., Ltd.) was placed in the center of dough to record temperature every 2s. The recording stopped when the temperature dropped to −18 °C, based on which frozen temperature curves of the dough were plotted.

2.4. Water loss rate of frozen dough

During the freezing process, moisture of dough will gradually change with increase of time, so the quality of dough will change accordingly. The water holding rate of dough can be measured by weight changes of dough before and after freezing. The fermented dough was taken out and cut into pieces, three small pieces of which were weighed and recorded. Three pieces of the well-divided fermented dough were weighed and recorded before freezing. After freezing, the frozen dough was thawed at room temperature of 25 °C for 30min, weighed and recorded again. The water loss rate of the dough was calculated according to the following formula:

$$\text{Water loss rate} / \% = (m_1 - m_2) / m_1 \times 100\% \tag{1}$$

In the formula, m_1 = mass of dough before freezing (g); m_2 = mass of dough after freezing (g).

2.5. Water holding capacity of frozen dough

The measurement was conducted according to Cao's method [13]. Briefly, the thawed frozen dough was taken and three 5.00 g samples from each group weighed and placed in centrifuge tubes. All samples underwent centrifugation at a speed of 8000 r/min for 20 min in a centrifuge (X1R, Thermo Corporation). The lost water after centrifugation was removed with filter paper and the remaining samples were weighed. The water holding capacity was calculated based on the following formula:

$$\text{Water holding capacity (WHC)} / \% = W_2/W_1 \times 100\% \tag{2}$$

In the formula, W_1 is the mass/g of the sample before centrifugation; W_2 is the mass/g of the sample after water removal.

2.6. pH changes

The measurement of pH changes was conducted based on Li's method [14]. The frozen dough was removed from the refrigerator and defrosted at 25 °C for 30 min 5 g of the thawed sample was transferred into a conical flask containing 45 mL of CO_2-free distilled water, homogenized for about 1 min at 8000–10000 r/min in a high speed dispersator (XHF-D, Ningbo Xinzhi Biotechnology Co., LTD.), and ground into a homogenate which was measured by a pH meter (PHS–3C, Shanghai Shengci Instrument Co., Ltd.). Three parallel experiments were performed for each sample in order to obtain accurate pH values.

2.7. Texture analysis of frozen dough

The texture characteristics were studied by a TA-XT Plus Texture Analyzer (Stable Micro Systems, UK). Texture analysis was conducted according to Luo's and Witek's methods with slight modifications [15,16]. The frozen dough was taken out, thawed, and made into a spherical shape with a radius of 3 cm. After thawing at room temperature of 25 °C, the texture properties of the dough including hardness, elasticity, gumminess, resilience, cohesion and chewiness can be obtained by using a texture analyzer (P36R probe). Test was conducted under the following parameters: 2 mm/s of test speed before the test, 1 mm/s of test speed, and 1 mm/s of test speed after the test, 70% of compression ratio, 5 g of trigger force, and 5 s of compression time interval. Textural parameters, such as the hardness, springiness, adhesiveness, and gumminess, were obtained.

2.8. Low-field nuclear magnetic relaxation time (LF-NMR)

The water distribution and migration was studied by low-field nuclear magnetic resonance (NMI20-040 V–I, Suzhou Niumai Analytical Instrument Co., Ltd.), and the tests were carried out according to Meng's method [17]. The resonance central frequency was adjusted by the FID test, and the spin relaxation time (T_2) of the sample was measured by the CPMG pulse sequence. The frozen dough was removed and thawed completely, then wheat and corn dough with different concentrations of CMC were weighed (5.00 ± 0.01) g, transferred into test tubes, and placed at the center of the RF coil located in the center of a permanent magnetic field to perform the scanning test of the CPMG pulse sequence. CPMG test was conducted under the following parameters: 20 MHz of main frequency, 628049.19 Hz of offset frequency, number of sampling points TD = 40014, number of repeated scans NS = 4, 2000 ms of sampling interval time TW, 6.52μs of half echo time τ, and 32 °C of testing temperature. The relaxation spectrum and T_2 can be obtained by inverting the CPMG relaxation decay curve using T_2 inversion fitting software.

2.8.1. Scanning electron microscopy (SEM)

The profiles of the dehydrated dough were measured by a scanning electron microscopy (Quanta 200, FEI Co., Inc., Hillsboro, OR, USA) according to Gong's method [18]. 10 g of dough with different CMC concentrations frozen was thawed at room temperature for 30 min. All samples received pretreatment: slicing and fixation, dehydration, freezing, freeze-drying in vacuum and observation under scanning electron microscopy.

The samples were cut into 3 mm × 3 mm × 5 mm of blocks, and placed in test tubes containing 2.5% glutaraldehyde for fixation for 2 h. After fixation, the samples were transferred onto clean plates (each plate was evenly divided into three areas), and washed with phosphate buffer for 10 min three times. Dehydration was carried out by treating the dough with ethanol at increased concentrations (30%, 50%, 70%, 90%, and 100%) twice for 20 min each time. The ethanol was replaced with isoamyl acetate, and the samples were placed in a vacuum freeze-drying oven (Aiphal-2LDPlus, CHRIST Lyophilizer Co., Ltd.) and dry at −40 °C for 36 h. After that, the samples were sprayed with gold by ion sputtering. Then SEM was used to characterize the samples at 1200× and 2400× magnifications.

2.9. Rheological properties of dough

The rheological properties of dough were studied by a rotary rheometer (HAAKE MARS, Thermo scientific, Germany). Measurement was conducted according to Huang's method [19]. 5 g of all samples with various concentrations of CMC stored were thawed for 30 min, and subjected to dynamic rheological tests. The frequency sweep test in the oscillation mode was used with the following parameters: 40 mm of the plate in diameter, we selected the defrosted back dough, peeled the plastic wrap and placed it between parallel plates, then first adjusted the gap to 1.005 mm, then pressed it to 1 mm, and placed the sample between two pans for 5 min to eliminate residual stress., 0.1–10 Hz of sweep frequency, 25 °C of testing temperature. Measurements were made in the linear viscoelastic region with a target strain of 0.1%.

2.10. Statistical analysis

Excel 2010 was used to process experimental data. Origin 2017 was used for plotting. The data were average values of at least three repeated measurements and were analyzed by one-way analysis of variance and Duncan's tests (SPSS 16.0; SPSS Inc., Chicago, IL, USA).

3. Results and discussion

3.1. Frozen temperature curves

The central temperature of wheat dough and corn multigrain dough was measured and recorded every 2 s in a freezing environment and the results are shown in Fig. 1. Fig. 1A and B shows the changes of the central temperature of wheat dough and corn multigrain dough, respectively. It can be seen that the frozen temperature curves of wheat dough and corn multigrain dough are roughly the same. The entire curves drop sharply at first, then tend to be flat, and then gradually decrease, which can be roughly divided into three stages. The initial stage: from the initial temperature to the freezing point. In this region, the frozen temperature curves of the two kinds of dough declined fastest and the curves were steepest. In middle stage: from freezing point to −5 °C, the freezing process passes through zone of maximum ice crystal formation, and it can be seen that the decreasing speed of the frozen temperature curve gradually slows down [12]. In the case of recording the temperature every 2s, we can see that the freezing curves of both WF-0 to WF-3 and CF-0 to CF-3 show temperature recovery in this region, followed by further decline, which can be explained as follows: 80% of water in the dough turns into ice at this time, and grows to form ice crystals gradually. Heat released in this process warms up the dough, leading to the slow and flat curves. However, the frozen temperature curve of corn multigrain dough is relative flat compared with those of wheat dough. In final stage: from −5 °C to −18 °C, the curves decline more slowly, approaching the final temperature from ice formation. In this region heat release is partly from cooling of the ice and partly from freezing of remaining water. Generally, since the ambient temperature is set at −30 °C, the freezing curve declines more slowly as the freezing time increases [20]. The result, however, reveals that the curves in final stage are not as steep as those in the initial stage. The curves tend to be flat and decline is relatively smaller compared to the initial stage because there is still residual water to be frozen and gives off heat. When 5 wt%CMC is added, the freezing curve of wheat dough will reach −18 °C faster, possibly because CMC has good hydrophilicity, which will increase the water holding capacity of dough and make water freeze faster.

Overall, the addition of CMC can reduce the time for wheat dough and corn multigrain dough in passing through zone of maximum ice crystal formation during freezing process and the increase of CMC concentration gives rise to decreasing time in passing through zone of maximum ice crystal formation, indicating that addition of CMC during freezing process can effectively inhibit the formation of large ice crystals, prevent damages of dough network structure, and is beneficial to quality maintenance of dough.

3.2. Water loss rate of frozen dough

The water loss rate of frozen dough is obtained according to formula 1. Figure S1 shows the effects of CMC at different concentrations on the water loss rate of dough. It can be seen from Figure S1 that the addition of CMC can effectively reduce the water loss rate of corn multigrain dough after freezing. Particularly, the water loss rate of wheat dough reaches lowest level of 0.63% with the

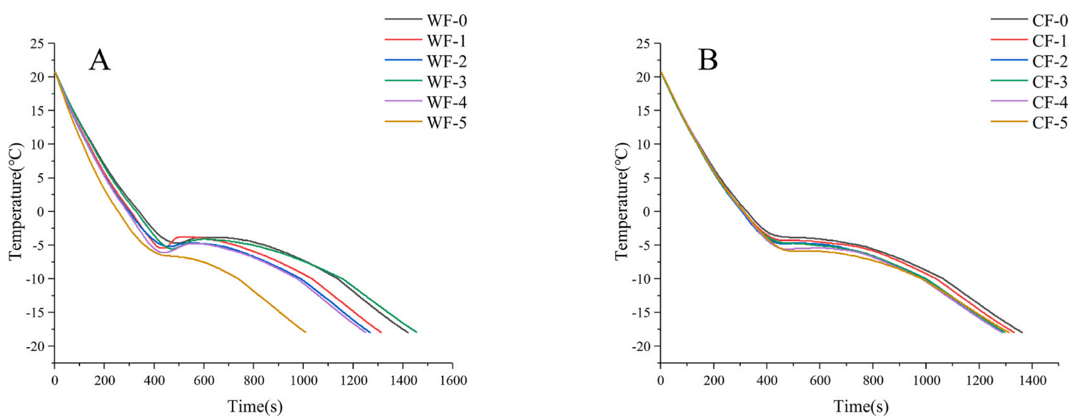

Fig. 1. Frozen temperature curves of frozen dough: (A) wheat dough from WF-0 to WF-5, (B) corn multigrain dough from CF-0 to CF-5..

addition of 2 wt% CMC and then slightly increased while the corn multigrain dough shows a gradually decreasing trend. The results show that, during the frozen storage of dough, the vapor pressure difference between freezing chamber and the frozen dough, and the continuously decreased surface moisture of the frozen dough by sublimation of ice crystals lead to continuous outward migration and decrease of the moisture inside the frozen dough [21]. The water loss rate of both the frozen dough with the addition of CMC and control groups during the frozen storage is lower, indicating that the addition of CMC can effectively improve the water holding capacity of both frozen wheat dough and frozen corn multigrain dough and prevent the frozen dough from loss of moisture to a large extent, which is more effective for frozen corn multigrain dough.

3.3. Water holding capacity of frozen dough

The water holding capacity of the frozen dough is obtained according to formula 2. Figure S2 shows the results of water holding capacity of frozen dough with addition of CMC at different concentrations. It can be seen from Figure S2 that water holding capacity of both wheat and corn multigrain dough increase after addition of CMC and the rising trend of water holding capacity for frozen corn multigrain dough is similar regardless of the amounts of CMC while a surge of water holding capacity occurs for WF-2 and WF-4. The results reveal that the water holding capacity of frozen wheat dough increases fastest with 0–3 wt% addition of CMC, and tends to increase slowly with increasing CMC. The water holding capacity shows slow increase for WF-3, probably because with the increase of CMC its thickening and stabilizing functions have different effects on the quality of frozen dough [6]. It can be seen from the results above that CMC significantly enhances the water holding capacity of corn multigrain dough, mainly because CMC itself can be used as a water humectant in food industry, so the addition of CMC will inevitably improve the water holding capacity of corn multigrain dough. Overall, in both frozen wheat and corn multigrain dough, the addition of CMC increases significantly the water holding capacity, indicating that during the frozen storage process, CMC can effectively prevent the water loss of frozen dough, reduce the formation of ice crystals, and protect the gluten network structure, thereby maintaining the water holding capacity of the frozen dough.

3.4. Changes of pH

Figure S3 shows the pH changes of frozen dough with addition of CMC at different concentrations. As can be seen in Figure S3, the pH values of the two kinds of frozen dough with the addition of CMC show negligible difference. CMC can affect the pH values of the dough to a certain extent, which will lead to different degrees of increase in pH values of the dough. But it does not necessarily mean that the higher addition of CMC will cause the higher pH value. At initial stage with 0–2 wt% addition of CMC (from WF-0, CF-0 to WF-2, CF-2), the pH value gradually increases, showing a uniform rising curve. But for WF-4 and CF-4, the pH value decreases to the lowest. With 4 wt% and 5 wt% addition of CMC (WF-4, WF-5 and CF-4, CF-5), the pH value gradually increases with the increase of CMC and the increase is higher than that in the initial stage.

With the addition of CMC, the pH value of the dough changes obviously in the initial stage. With increasing addition of CMC, the pH value shows a trend of rising, falling and rising again. With 3 wt% addition of CMC (WF-3 and CF-3), the pH value reaches the lowest, the strongest acid state of frozen dough in our study. The results show that CMC can inhibit the activity of related hydrolase, and reduce the acidification rate of frozen dough during the storage process [22]. In general, the pH value of the two kinds of dough does not change significantly. For both wheat and corn multigrain dough under different CMC conditions, the pH value changes within 1 in range, so CMC has a limited impact on the pH of frozen dough.

3.5. Texture analysis of frozen dough

Table 1 show the effects of CMC at different concentrations on texture properties of wheat and corn multigrain dough, respectively,

Table 1
Texture properties of wheat and corn multigrain dough.

Ingredients	Hardness (g)	Chewiness (g)	Gumminess (g)	Elasticity (−)	Cohesion (−)	Resilience (−)
Wheat dough						
WF-0	5268.46 ± 2068.76^{bc}	216.42 ± 132.89^{a}	1274.76 ± 342.72^{bc}	0.166 ± 0.092^{a}	0.252 ± 0.065^{ab}	0.073 ± 0.003^{bcd}
WF-1	12290.65 ± 2386.10^{a}	638.20 ± 323.16^{a}	2911.02 ± 410.94^{a}	0.231 ± 0.147^{a}	0.239 ± 0.024^{b}	0.087 ± 0.008^{ab}
WF-2	12937.66 ± 1285.46^{a}	615.53 ± 140.14^{a}	3179.45 ± 299.51^{a}	0.193 ± 0.037^{a}	0.246 ± 0.019^{ab}	0.094 ± 0.004^{a}
WF-3	2612.82 ± 1680.32^{c}	155.41 ± 61.74^{a}	803.38 ± 290.84^{c}	0.201 ± 0.087^{a}	0.342 ± 0.086^{a}	0.068 ± 0.004^{d}
WF-4	8037.99 ± 3441.97^{b}	1498.81 ± 2164.62^{a}	2584.80 ± 1491.21^{a}	0.409 ± 0.463^{a}	0.309 ± 0.047^{ab}	0.086 ± 0.018^{abc}
WF-5	7297.83 ± 1129.47^{b}	495.95 ± 267.65^{a}	2142.33 ± 497.29^{ab}	0.220 ± 0.084^{a}	0.291 ± 0.027^{ab}	0.070 ± 0.006^{cd}
Corn multigrain dough						
CF-0	4074.89 ± 475.03^{b}	124.37 ± 2.95^{b}	891.96 ± 112.92^{bc}	0.141 ± 0.017^{b}	0.219 ± 0.002^{d}	0.088 ± 0.004^{a}
CF-1	796.39 ± 85.77^{c}	85.21 ± 44.18^{b}	355.96 ± 92.21^{c}	0.231 ± 0.062^{a}	0.289 ± 0.025^{c}	0.067 ± 0.014^{b}
CF-2	2952.54 ± 815.73^{b}	159.41 ± 85.92^{b}	905.55 ± 331.20^{bc}	0.171 ± 0.038^{ab}	0.301 ± 0.036^{bc}	0.087 ± 0.010^{a}
CF-3	3146.16 ± 253.06^{b}	195.55 ± 21.26^{b}	1075.79 ± 181.20^{bc}	0.186 ± 0.041^{ab}	0.340 ± 0.031^{ab}	0.093 ± 0.008^{a}
CF-4	10410.83 ± 2662.88^{a}	484.22 ± 186.13^{a}	2815.40 ± 855.54^{a}	0.169 ± 0.017^{ab}	0.268 ± 0.016^{c}	0.101 ± 0.016^{a}
CF-5	3891.02 ± 633.60^{b}	257.78 ± 70.42^{b}	1379.36 ± 302.12^{b}	0.187 ± 0.028^{ab}	0.352 ± 0.022^{a}	0.091 ± 0.011^{a}

Different letters in the same column indicate significant difference ($P < 0.05$).

and which includes elasticity, gumminess, resilience, hardness, chewiness and cohesion. Results show that with addition of CMC at different concentrations, the elasticity of the two kinds of frozen dough increases. Particularly, the elasticity of the frozen wheat dough reaches the maximum with 4 wt% addition of CMC and approaches the average value with the increase of CMC while for the frozen corn multigrain dough the elasticity reaches the maximum with 1% addition of CMC. Frozen corn multigrain dough is less elastic than frozen wheat dough. It could be that corn flour has more dietary fiber, making the difference. The cohesion of the frozen wheat dough decreases slowly at first, increases later and finally decreases slowly, while the cohesion of the frozen corn multigrain dough increases gradually at first, and then decreases slowly. The cohesion of the two kinds of dough reaches the maximum with 3 wt% addition of CMC. Compared with control groups, however, it still shows an increasing trend because CMC maintains the water activity in the frozen dough during the storage process and increases the water content [23], thus increasing the cohesion. Meanwhile, the stability and adhesiveness of the frozen dough increase with the increase of CMC, leading to the stable increase of cohesion of the frozen dough. The results show no significant changes in resilience. The hardness and chewiness of the frozen wheat dough increase at first, then decrease to the minimum with 3 wt% addition of CMC, and finally tend to increase. After CMC was added to frozen corn multigrain dough, the hardness first showed a downward trend, and then gradually increased, indicating that adding appropriate amount of CMC can effectively inhibit water migration and maintain the stability of dough frozen storage [24]. For the frozen corn multigrain dough the gumminess decrease at first, reaching the minimum with 1 wt% addition of CMC, then gradually increase to the maximum with 4 wt% addition of CMC, and then decrease. No significant changes occur in terms of chewiness because the stable and thickening function of CMC can effectively reduce the water activity and increase the stability of the dough [25]. However, the excessive CMC will compete with the water originally bound to the dough to form a colloid and increase the hardness. It may also be attributed to the water holding capacity of CMC in the storage process, which keeps gluten protein in the dough increasing steadily, makes the dough tightly combined [26,27], and increases the cohesion.

3.6. LF-NMR analysis

Low-field nuclear magnetic resonance (LF-NMR) is a novel detection technology for food quality, which is used to study the state and migration of moisture content in food. The moisture content, distribution and the degree of its combination with other components play pivotal roles in the quality of frozen dough and related end products. Fig. 2 and Table 2 shows the effects of the addition of CMC at different concentrations on the distribution and migration of water in wheat dough and frozen corn multigrain dough. Fig. 2A and B show inversion spectrum of wheat frozen dough and corn multigrain frozen dough.

In Fig. 1, the different wave crests represent the different states of the water. In Table 2, The peak area reflects the proportion of bound water, immobile water and free water in the dough. P_{21} is the bound water content of dough, indicating the water tightly bound with starch or protein; P_{22} is the motionless water content, indicating that the water does not easily flow between starch or protein; P_{23} is the free water content. It can be seen that the moisture states of the two kinds of frozen dough are all fluidic water and multi-layered water combined with starch and protein molecules with fluidity between deep bound water and free water [28]. The water is wrapped by protein network structure and is distributed in the gaps of macromolecular particles such as starch [29]. Compared with the blank sample, the weakly bound water (P_{22}) in wheat and corn multigrain dough increased significantly when CMC was added by 5 wt%. It increased significantly from 67.61 $\pm$ 5.12% and 68.38 $\pm$ 1.92% to 82.42 $\pm$ 2.79% and 80.07 $\pm$ 3.54%. This indicates that appropriate amount of CMC can weaken the flow of water molecules, make the water in the dough more difficult to freeze, and protect the dough structure [30]. The P_{22} peak area of wheat dough with addition of CMC is larger than that without addition of CMC, indicating the decreased fluidity of water during the freezing process. The decrease might be caused by CMC enhanced binding ability of the frozen dough to water molecules during the freezing process, which increases the weakly bound water content and inhibits reduction of the moisture content in the dough to a certain extent [31]. It can be seen that the amount of weakly bound water shows a gradual upward trend with the increase of CMC, probably due to interaction between hydroxyl groups of sugar and water molecules to form hydrogen bonds, correspondingly reducing the "free water" in dough system. Meanwhile, polysaccharides and starch chains can form a strong

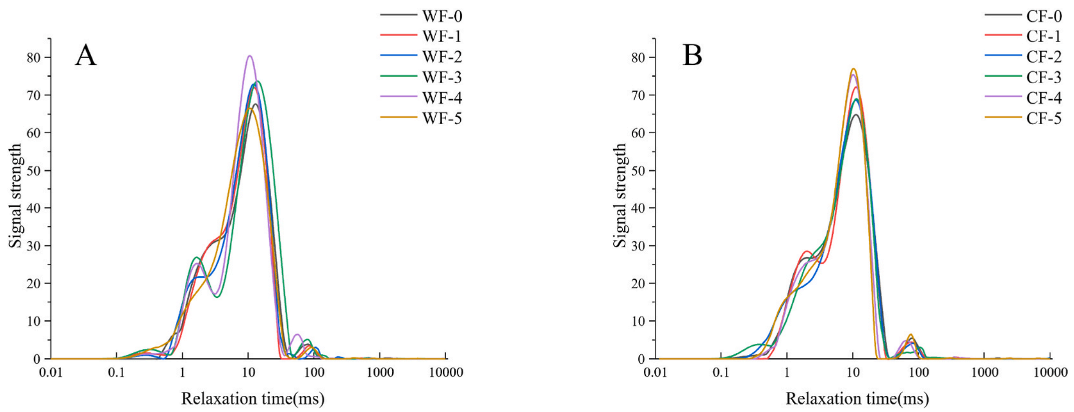

Fig. 2. LF-NMR spectrum of frozen dough: (A) wheat dough from WF-0 to WF-5, (B) corn multigrain dough from CF-0 to CF-5..

Table 2
Water distribution and migration of wheat dough and corn multigrain dough.

Ingredients	P_{21} (%)	P_{22} (%)	P_{23} (%)
Wheat dough			
WF-0	30.29 ± 4.92^{a}	67.61 ± 5.12^{b}	2.10 ± 0.16^{ab}
WF-1	24.81 ± 1.14^{ab}	73.78 ± 0.69^{ab}	1.41 ± 0.44^{c}
WF-2	23.61 ± 4.25^{ab}	75.00 ± 4.12^{ab}	1.39 ± 0.42^{c}
WF-3	22.31 ± 4.98^{ab}	76.08 ± 4.79^{ab}	$1.61 \pm 0.33b^{c}$
WF-4	21.07 ± 1.83^{ab}	76.31 ± 2.06^{ab}	2.62 ± 0.24^{a}
WF-5	16.47 ± 3.00^{b}	82.42 ± 2.79^{a}	1.11 ± 0.21^{c}
Corn multigrain dough			
CF-0	29.18 ± 2.04^{b}	68.38 ± 1.92^{b}	2.44 ± 0.08^{b}
CF-1	24.61 ± 2.28^{ab}	73.88 ± 0.39^{ab}	1.51 ± 0.46^{a}
CF-2	23.64 ± 3.89^{ab}	74.49 ± 4.19^{ab}	1.87 ± 0.30^{a}
CF-3	22.78 ± 7.00^{ab}	75.91 ± 7.23^{ab}	1.31 ± 0.29^{a}
CF-4	22.63 ± 2.44^{ab}	75.42 ± 3.00^{ab}	1.95 ± 0.56^{a}
CF-5	18.82 ± 3.16^{a}	80.07 ± 3.54^{a}	1.11 ± 0.37^{a}

P_{21}: the bound water content; P_{22}: immobile water content; P_{23}: free water content; Different letters in the same column indicate significant difference ($P < 0.05$).

sugar-starch interaction, which replaces sites originally occupied by water molecules and stabilizes the molecular structure of starch [32].

3.7. Rheological properties of frozen dough

Fig. 3 show the effects of CMC at different concentrations on the rheological properties of frozen wheat and corn multigrain dough. Fig. 3A and B show the storage modulus and loss modulus of wheat frozen dough. Fig. 3C and D show the storage modulus and loss modulus of corn multigrain frozen dough. The dynamic rheology of dough is related to the final quality and processing of the products. Dynamic rheology has three main parameters: storage modulus G′ which represents elastic nature of materials, loss modulus G″ which

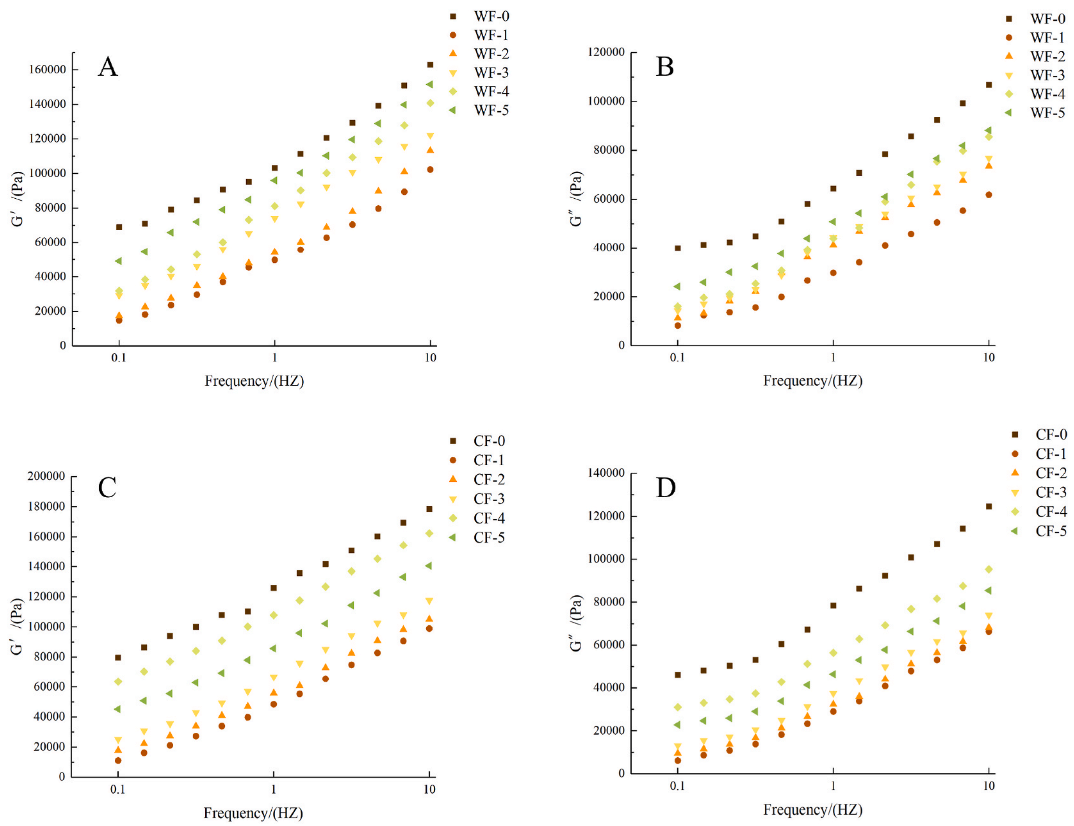

Fig. 3. Rheological properties of frozen dough：(A) storage modulus of wheat dough, (B) loss modulus, (C) storage modulus of corn multigrain dough, (D) loss modulus of corn multigrain dough.

represents viscous nature of materials and loss angle tanδ (tanδ = G"/G′) reflecting viscoelasticity ratio of systems [33]. It can be seen from Fig. 3 that G″ is smaller than G' (tanδ<1), indicating that the system is mainly characterized as solids [34,35]. Compared with control groups, G″ and G′ of the two kinds of frozen dough show significant decrease with 1% addition of CMC, probably because the addition of CMC weakens the cross-linking effect of gluten protein, and further affects the viscoelastic properties of the dough, leading to the loose network structure of both wheat and corn multigrain dough [36]. With the increasing addition of CMC, G″ and G′ of the dough gradually increase, probably because CMC has good hydrophilicity and can interact with gluten protein and starch particles to form complex, thereby strengthening the protein network structure of the dough [37,38]. When 5 wt% CMC was added, G′ and G″ were lower than 4 wt% CMC. This was because CMC has strong water absorption. When excessive CMC was added, it will compete with gluten protein for water, thus weakening the network structure of gluten protein and reducing the gumminess and elasticity of dough. Both of wheat and corn multigrain dough show a similar upward tendency of viscoelasticity, but G″ and G′ of corn multigrain dough are larger than those of wheat dough and corn multigrain dough has higher viscoelasticity than wheat dough with addition of CMC at certain amounts.

3.8. *Characterization of surface structure of dough protein*

The microstructure of frozen wheat dough and frozen corn multigrain dough is characterized by SEM. Fig. 4A - F and Fig. 5A - F show the SEM images of the two kinds of frozen dough with 0%, 1 wt%, 2 wt%, 3 wt%, 4 wt%, and 5 wt% of CMC at 600× (middle) and 1200× (top right) magnifications.

Dough is a system composed of starch and protein network structure. The round granules of different sizes in the dough are starch granules which adhere to each other, and are wrapped by gluten to form a gluten network structure [39,40]. The frozen process will increase diameter of pores, loosen protein network, and weaken structure of the dough. In freezing process the water in the dough turns into ice crystals, and the recrystallization of ice crystals can bring damages to the protein network. Fig. 5 shows that the gluten protein in the control group shows significantly smaller holes than that in the experimental groups with addition of CMC. The control group has a small number of giant pores, and looser gluten protein structure compared with the experimental groups. In the experimental groups, the size and distribution of pores of gluten protein is more uniform. Fig. 5 shows detachment of round particles from the gluten structure, which means the formation of a large number of ice crystals in the corn multigrain dough during the freezing process and destruction of gluten structure. With the increase of CMC, the microstructure of corn multigrain dough changes and starch granules are observed clearly. With 1 wt% addition of CMC, the gluten structure of wheat dough and corn multigrain dough was become sticky, and with the increase of CMC the starch granules fill slowly the gluten structure. This is because the strong water holding capacity of CMC can maintain the microstructure of gluten protein during the freezing process, which effectively inhibits the formation of ice crystals and the migration of water. Compared with the control groups, the formed ice crystals in the experimental groups are smaller and more uniform in size, which reduces the destruction of the protein network structure by the ice crystals during the freezing process, further demonstrating that the addition of CMC into the dough can inhibit recrystallization.

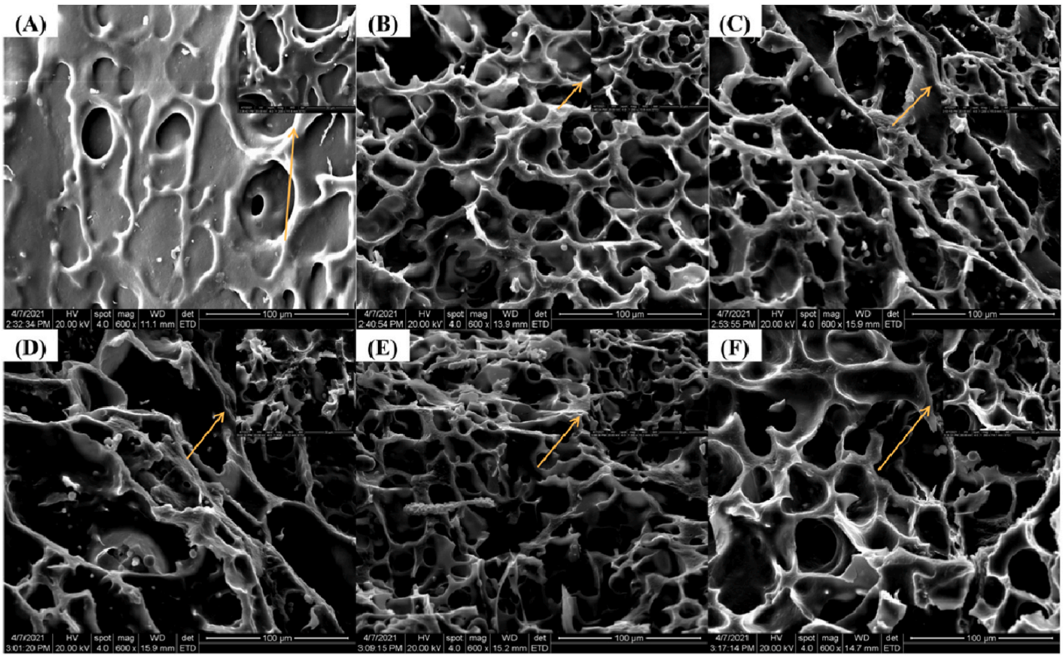

Fig. 4. SEM images of wheat dough: (A) WF-0, (B) WF-1, (C) WF-2, (D) WF-3, (E) WF-4, (F) WF-5.

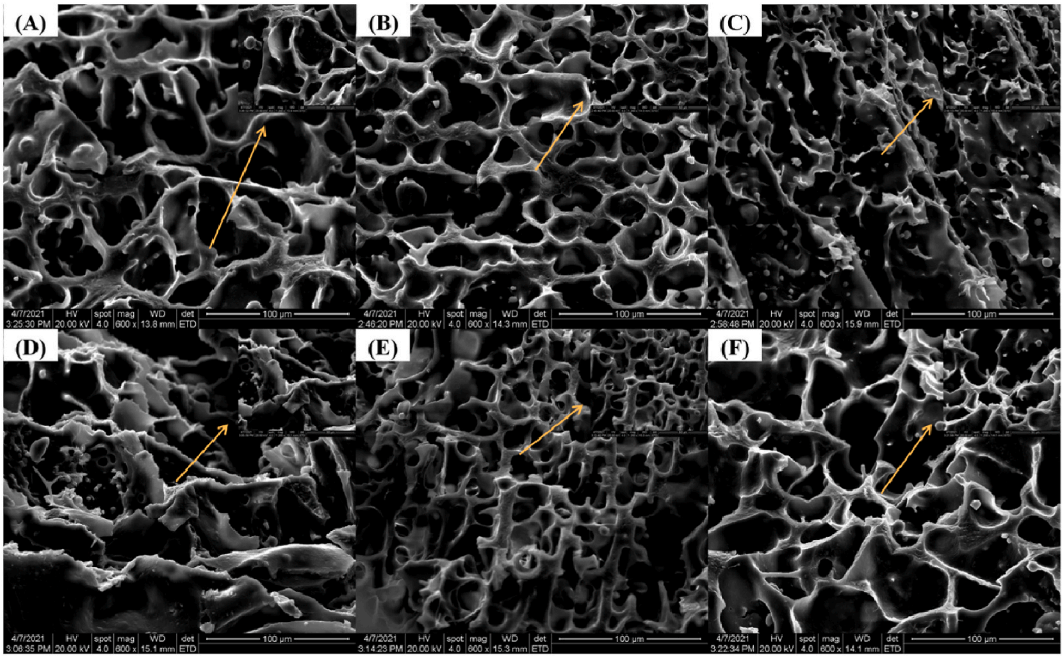

Fig. 5. SEM images of corn multigrain dough: (A) CF-0, (B) CF-1, (C) CF-2, (D) CF-3, (E) CF-4, (F) CF-5.

4. Conclusions

In this work we have investigated the effects of CMC on the quality changes of frozen dough with 0–5 wt% CMC addition. The results reveal that the addition of CMC can effectively inhibit the water loss, while increasing the water holding capacity and has a significant effect on corn multigrain dough during the freezing process. Certain amounts of CMC can keep moisture inside the dough to inhibit the recrystallization, while excessive CMC has an undesired effect on the dough. What's more, the addition of CMC stabilizes the molecular structure of starch and weakens the cross-linking effect of gluten protein. G'' and G' of the two kinds of frozen dough show significant decrease with 1 wt% addition of CMC, and the cohesion of the two kinds of dough reaches the maximum with 3 wt% addition of CMC, while the hardness and chewiness of wheat and corn multigrain dough reaches the maximum with 3 wt% and 4 wt% addition of CMC, respectively. CMC at different concentrations can change the quality of the frozen dough, and our study could provide beneficial clues for the frozen food industry by analyzing the quality of both wheat and corn multigrain frozen dough with addition of CMC at different concentrations.

Author contribution statement

Tong-Chao Su: Conceived and designed the experiments; Analyzed and interpreted the data; Wrote the paper.
Wen-Kai Du: Performed the experiments; Analyzed and interpreted the data; Wrote the paper.
Bing-Yan Deng: Performed the experiments; Analyzed and interpreted the data.
Jie Zeng, Hai-Yan Gao, Hai-Xu Zhou, Guang-Lei Li: Contributed reagents, materials, analysis tools or data.
Hao Zhang, Ya-Meng Gong: Performed the experiments; Analyzed and interpreted the data.
Jin-Yu Zhang: Performed the experiments.

Data availability statement

Data will be made available on request.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgments

This work was financed by Natural Science Foundation of Henan Province (NO.212300410142), Central government guiding local scientific and technological development projects (No.105020221021), and the Program of Xinxiang Major Scientific and Technological Project (No.ZD2020003), Innovation Training Program for college students of Henan Institute of Science and Technology (2023CX068), and the High-Level Talent Research Project of Henan Institute of Science and Technology (grant number 2017017, 2017020).

Appendix A. Supplementary data

Supplementary data related to this article can be found at https://doi.org/10.1016/j.heliyon.2023.e18545.

References

[1] J. Li, M.P. Yadav, J.J. Li, C.S. J. o, Effect of different hydrocolloids on gluten proteins, starch and dough microstructure, J. Cereal. Sci. 87 (2019) 85–90, 10.1016/j.jcs.2019.03.004Get rights and content.

[2] C. Ferrero, Hydrocolloids in wheat breadmaking: a concise review, Food Hydrocolloids 68 (2017) 15–22, https://doi.org/10.1016/j.foodhyd.2016.11.044.

[3] X. Liu, T. Mu, H. Sun, M. Zhang, J. Chen, M.L. Fauconnier, Influence of different hydrocolloids on dough thermo-mechanical properties and in vitro starch digestibility of gluten-free steamed bread based on potato flour, Food Chem. 239 (2018) 1064–1074, https://doi.org/10.1016/j.foodchem.2017.07.047.

[4] A.F. Ammar, A. Siddeeg, F.M. Aqlan, S.M. Howladar, M.Y. Refai, M. Afifi, H.A. Ali, D. Hajjar, M.G.M. Sulamain, M.V.M. Chamba, B.G. Kimani, A. Saleh, M. Baeshen, Shelf life extension of wheat bread by alhydwan flour and Carboxymethylcellulose and improvement of their quality characteristics, dough rheological and microstructure, Int. J. Biol. Macromol. 156 (2020) 851–857, https://doi.org/10.1016/j.ijbiomac.2020.04.023.

[5] R. Numfon, Effects of different hydrocolloids on properties of gluten-free bread based on small broken rice berry flour, Food Sci. Technol. Int. 23 (4) (2017) 310–317, https://doi.org/10.1177/1082013217690064.

[6] N. Aravind, M. Sissons, C.M. Fellows, Effect of soluble fibre (guar gum and carboxymethylcellulose) addition on technological, sensory and structural properties of durum wheat spaghetti, Food Chem. 131 (3) (2012) 893–900, https://doi.org/10.1016/j.foodchem.2011.09.073.

[7] Y. Tang, Y. Yang, Q. Wang, Y. Tang, F. Li, J. Zhao, Y. Zhang, J. Ming, Combined effect of carboxymethylcellulose and salt on structural properties of wheat gluten proteins, Food Hydrocolloids 97 (2019), https://doi.org/10.1016/j.foodhyd.2019.105189.

[8] Y. Chen, Y. Tang, Q. Wang, L. Lei, J. Zhao, Y. Zhang, L. Li, Q. Wang, J. Ming, Carboxymethylcellulose-induced changes in rheological properties and microstructure of wheat gluten proteins under different pH conditions, J. Food Sci. 86 (3) (2021) 677–686, https://doi.org/10.1111/1750-3841.15646.

[9] X. Hu, L. Cheng, Y. Hong, Z. Li, C. Li, Z. Gu, Combined effects of wheat gluten and carboxymethylcellulose on dough rheological behaviours and gluten network of potato–wheat flour-based bread, Int. J. Food Sci. Technol. 56 (8) (2021) 4149–4158, https://doi.org/10.1111/ijfs.15043.

[10] S.Y. Sim, A.A. Noor Aziah, L.H. Cheng, Quality and functionality of Chinese steamed bread and dough added with selected non-starch polysaccharides, J. Food Sci. Technol. 52 (1) (2013) 303–310, https://doi.org/10.1007/s13197-013-0967-1.

[11] P. Wang, H. Tao, Z. Jin, X. Xu, Impact of water extractable arabinoxylan from rye bran on the frozen steamed bread dough quality, Food Chem. 200 (2016) 117–124, https://doi.org/10.1016/j.foodchem.2016.01.027.

[12] C. Ban, S. Yoon, J. Han, S.O. Kim, J.S. Han, S. Lim, Y.J. Choi, Effects of freezing rate and terminal freezing temperature on frozen croissant dough quality, Lwt 73 (2016) 219–225, https://doi.org/10.1016/j.lwt.2016.05.045.

[13] F.-H. Cao, X.-J. Li, S.-Z. Luo, D.-D. Mu, X.-Y. Zhong, S.-T. Jiang, Z. Zheng, Y.-Y. Zhao, Effects of organic acid coagulants on the physical properties of and chemical interactions in tofu, LWT - Food Sci. Technol. (Lebensmittel-Wissenschaft -Technol.) 85 (2017) 58–65, https://doi.org/10.1016/j.lwt.2017.07.005.

[14] M. Li, K. Zhu, X. Guo, W. Peng, H. Zhou, Effect of water activity (aw) and irradiation on the shelf-life of fresh noodles, Innovat. Food Sci. Emerg. Technol. 12 (4) (2011) 526–530, https://doi.org/10.1016/j.ifset.2011.06.005.

[15] L.-J. Luo, X.-N. Guo, K.-X. Zhu, Effect of steaming on the quality characteristics of frozen cooked noodles, LWT - Food Sci. Technol. (Lebensmittel-Wissenschaft -Technol.) 62 (2) (2015) 1134–1140, https://doi.org/10.1016/j.lwt.2015.02.008.

[16] M. Witek, I. Maciejaszek, K. Surowka, Impact of enrichment with egg constituents on water status in gluten-free rice pasta - nuclear magnetic resonance and thermogravimetric approach, Food Chem. 304 (2020), 125417, https://doi.org/10.1016/j.foodchem.2019.125417.

[17] K. Meng, H. Gao, J. Zeng, J. Zhao, Y. Qin, G. Li, T. Su, Rheological and microstructural characterization of wheat dough formulated with konjac glucomannan, J. Sci. Food Agric. 101 (10) (2021) 4373–4379, https://doi.org/10.1002/jsfa.11078.

[18] S. Gong, D. Yang, Q. Wu, S. Wang, Z. Fang, Y. Li, F. Xu, Z. Wang, J. Wu, Evaluation of the antifreeze effects and its related mechanism of sericin peptides on the frozen dough of steamed potato bread, J. Food Process. Preserv. 43 (8) (2019), https://doi.org/10.1111/jfpp.14053.

[19] W.N. Huang, Y.L. Yuan, Y.S. Kim, O.K. J.C.c. Chung, Effects of transglutaminase on rheology, microstructure, and baking properties of frozen dough, Cereal Chem. 85 (3) (2008) 301–306, https://doi.org/10.1094/CCHEM-85-3-0301.

[20] H. Kiani, D.-W. Sun, Water crystallization and its importance to freezing of foods: a review, Trends Food Sci. Technol. 22 (8) (2011) 407–426, https://doi.org/10.1016/j.tifs.2011.04.011.

[21] C.G. Arp, M.J. Correa, C. Ferrero, High-amylose resistant starch as a functional ingredient in breads: a technological and microstructural approach, Food Bioprocess Technol. 11 (12) (2018) 2182–2193, https://doi.org/10.1007/s11947-018-2168-4.

[22] M.C. Cortez-Trejo, M. Gaytán-Martínez, M.L. Reyes-Vega, S. Mendoza, Protein-gum-based gels: effect of gum addition on microstructure, rheological properties, and water retention capacity, Trends Food Sci. Technol. 116 (2021) 303–317, https://doi.org/10.1016/j.tifs.2021.07.030.

[23] S.A. Mir, M.A. Shah, H.R. Naik, I.A. Zargar, Influence of hydrocolloids on dough handling and technological properties of gluten-free breads, Trends Food Sci. Technol. 51 (2016) 49–57, https://doi.org/10.1016/j.tifs.2016.03.005.

[24] X. Ning, Z. Chen, Z. Luo, Z. Liu, X. Huang, X. Cao, X. Zheng, W. Du, X. Pan, K.J.I.J. Yu, E. o. F, Corn silk flour fortification as a dietary fiber supplement: evolution of the impact on paste, dough, and quality of dried noodles, Int. J. Food Eng. 18 (6) (2022) 479–487, https://doi.org/10.1515/ijfe-2021-0360.

[25] P.H. Nitcheu Ngemakwe, M. Le Roes-Hill, V.A. Jideani, Effects of carboxymethylcellulose, yoghurt and transglutaminase on textural properties of oat bread, J. Texture Stud. 47 (1) (2016) 74–84, https://doi.org/10.1111/jtxs.12163.

[26] Z.M. Kang, K.Y. Zhang, L.L. Gao, Q.Y. Wen, W.J.M.F.S. Sheng, Technology, Study on the correlation between water distribution and quality of frozen twisted dough- stick in freeze-thaw process, Modern Food Science and Technology 34 (2) (2018) 182–188, https://doi.org/10.13982/j.mfst.1673-9078.2018.2.029, 181.

[27] J. Xue, M. Ngadi, Effects of methylcellulose, xanthan gum and carboxymethylcellulose on thermal properties of batter systems formulated with different flour combinations, Food Hydrocolloids 23 (2) (2009) 286–295, https://doi.org/10.1016/j.foodhyd.2008.01.002.

[28] E.F. Esselink, H. van Aalst, M. Maliepaard, J.P. J.C.c. van Duynhoven, Long-term storage effect in frozen dough by spectroscopy and microscopy, Cereal Chem. 80 (4) (2003) 396–403, https://doi.org/10.1094/CCHEM.2003.80.4.396.

[29] V. Kontogiorgos, H.D. Goff, S. Kasapis, Effect of aging and ice-structuring proteins on the physical properties of frozen flour–water mixtures, Food Hydrocolloids 22 (6) (2008) 1135–1147, https://doi.org/10.1016/j.foodhyd.2007.06.005.

[30] C. Xin, L. Nie, H. Chen, J. Li, B.J.F.H. Li, Effect of degree of substitution of carboxymethyl cellulose sodium on the state of water, rheological and baking performance of frozen bread dough, Food Hydrocolloids 80 (7) (2018) 8–14, https://doi.org/10.1016/j.foodhyd.2018.01.030.

[31] M.J. Correa, E. Ferrer, M.C. Añón, C. Ferrero, Interaction of modified celluloses and pectins with gluten proteins, Food Hydrocolloids 35 (2014) 91–99, https://doi.org/10.1016/j.foodhyd.2013.04.020.

[32] J. Ahmed, L.T. Joseph, A.J.J. Al-Hazza, o.F.M. Characterization, Effects of frozen storage on texture, microstructure, water mobility and baking quality of brown wheat flour/β-glucan concentrate Arabic bread dough, J. Food Meas. Char. 15 (2021) 1258–1269, https://doi.org/10.1007/s11694-020-00725-5.

[33] B. Peng, Y. Li, S. Ding, J. Yang, Characterization of textural, rheological, thermal, microstructural, and water mobility in wheat flour dough and bread affected by trehalose, Food Chem. 233 (2017) 369–377, https://doi.org/10.1016/j.foodchem.2017.04.108.

[34] L.S. Sciarini, P.D. Ribotta, A.E. León, G.T. Pérez, Incorporation of several additives into gluten free breads: effect on dough properties and bread quality, J. Food Eng. 111 (4) (2012) 590–597, https://doi.org/10.1016/j.jfoodeng.2012.03.011.

[35] J. Xiong, Q. Li, Z. Shi, J. Ye, Interactions between wheat starch and cellulose derivatives in short-term retrogradation: rheology and FTIR study, Food Res. Int. 100 (Pt 1) (2017) 858–863, https://doi.org/10.1016/j.foodres.2017.07.061.

[36] Z. Xu, F. Zhong, Y. Li, C.F. Shoemaker, W.H. Yokoyama, W.J. J.o. a. Xia, f amp; chemistry, Effect of polysaccharides on the gelatinization properties of cornstarch dispersions, Food Chem. 60 (2) (2012) 658–664, https://doi.org/10.1021/jf204042m.

[37] M. Azeem, T.-H. Mu, M. Zhang, Effects of hydrocolloids and proteins on dough rheology and in vitro starch digestibility of sweet potato-wheat bread, Lwt 142 (2021), https://doi.org/10.1016/j.lwt.2021.110970.

[38] L. Das, U. Raychaudhuri, R. Chakraborty, Effects of hydrocolloids as texture improver in coriander bread, J. Food Sci. Technol. 52 (6) (2015) 3671–3680, https://doi.org/10.1007/s13197-014-1296-8.

[39] T. Li, X.-N. Guo, K.-X. Zhu, H.-M.J.F.C. Zhou, Effects of alkali on protein polymerization and textural characteristics of textured wheat protein, Food Chem. 239 (2018) 579–587, https://doi.org/10.1016/j.foodchem.2017.06.155.

[40] C.G. Arp, M.J. Correa, C. Ferrero, Improving quality: modified celluloses applied to bread dough with high level of resistant starch, Food Hydrocolloids 112 (2021), https://doi.org/10.1016/j.foodhyd.2020.106302.