



Effect of carboxymethyl cellulose and/or wheat gluten on the pasting, rheological and quality properties of wheat starch-based batter for deep-fried products

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ABSTRACT

This study aimed to investigate the effects of the individual and synergistic addition of wheat gluten (WG) and carboxymethyl cellulose (CMC) on the wheat starch (WS)-based batter characteristics to determine the molecular basis of texture formation in the actual batter system. Results showed that adding WG and/or CMC significantly increased the viscosity of WS during pasting. The rheological behavior showed that the WG-treated and CMC-treated group had the highest and lowest viscoelasticity. The addition of WG-CMC helped the WS-based batter obtain moderate viscoelasticity. These outcomes could be attributed to the enhancement of hydrogen bonding. The microstructure suggested that the addition of WG-CMC increased the density and integrity of the gel network. Overall, CMC competed for the binding sites of WG on WS, reducing the increase in viscoelasticity caused by the interaction between WG and WS. This might alleviate the unwanted springiness of fried products.

1. Introduction

Deep-fried breaded food is enjoyed worldwide for its golden color, crispy texture, and unique flavor (Liberty et al., 2019). The original batter is usually a viscous liquid consisting of wheat flour and water. WS and a small amount of gluten protein are the main functional components of wheat flour (Li et al., 2024). Compared to individual starch, wheat flour has better functional properties, mainly due to the possibility of hydrogen bonding or covalent bonding between the hydroxyl group of WS and the amino acid side chain of protein, which can enhance the elasticity of starch-based products such as steamed bread, toast, croissants, and so on (Ghumman et al., 2016). However, for deep-fried breaded food, the high elasticity of the batter could result in an unwanted springiness of crust.

Hydrocolloid is widely used in fried batter products by food manufacturers because it has a good synergistic effect with starch and protein

and can adjust the viscosity, rheology and gelation of the system (Cui et al., 2023). As a hydrophilic long-chain polymer, CMC can play a role in thickening, emulsifying and coating in batter (Pirsa & Hafezi, 2023). Currently, the research on CMC in the batter system mainly focuses on its inhibiting properties on oil absorption (Cui et al., 2023; Pongsawatmanit et al., 2018). Clearly, excessive oil absorption of fried foods can lead to various health threats (i.e. hyperlipidemia, fatty liver and diabetes, etc.) and product quality problems (oxidation degradation and decreased crispiness) (Primo-Martín, 2012). Parimala and Sudha (2012) reported that hydrocolloid can form an oil-resistant gel film around fried food, thereby reducing the oil absorption rate. Usually, the formation of a water/oil migration barrier typically reduces the texture of fried batter due to water retention (Primo-Martín, 2012). However, our previous research found that the addition of CMC can increase the crispness of the WS-WG batter system under the condition that the coating pick up is consistent. The increase in crispness is related to the storage modulus

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(G') and loss modulus (G'') of the batter (Zhang et al., 2024). Currently, the effect of the interaction of the components in the ternary mixed system on the batter characteristics is still unclear. Clarifying the molecular basis is of great significance for improving the processing properties of batter. Additionally, existing research mainly focuses on exploring the interaction between hydrocolloid and WS, while ignoring the role of small amounts of gluten protein in wheat flour (Johansson et al., 2022; Lin et al., 2022). Generally, the hydroxyl group of WS and the amino acid side chain of protein can combine to form a complex through hydrogen bonding or covalent bonding. This cross-linking can reduce the cross-linking between starch molecules and change the gel structure (Johansson et al., 2022). If the formation of protein gel is preferred to that of WS gel, gluten protein will form a gel network during heating, and then the water-absorbing and expanded WS particles will fill in or outside the protein gel network, promoting the viscoelasticity of the gel (Li et al., 2024). In addition to cross-linking with WS, gluten protein can also form soluble complexes with CMC (anionic colloid) through electrostatic interactions which promotes the strength and integrity of the gel (Cui et al., 2023). Considering this, there is need for the further study on the effects of individual or synergistic addition of each component in the ternary system of starch, protein and CMC on the batter characteristics. A recent report suggested that hydrophilic CMC-Na can interact with WS and WG to form stable gels that enhances the water-binding ability and thus inhibit lipid penetration (Cui et al., 2023). The ratio of WS to WG in this work is 1:1, which is different from the composition of the batter in actual production. Since WS is the main component in wheat flour, accounting for about 70–80 %, while protein accounts for approximately 10 % (Li et al., 2024). Feng et al. (2023) proposed that the ratio of WS to gluten protein would affect the formation of gel in the system. They found that when the mass ratio of WS to protein was 11:1, the formed starch-protein gel was the most complete in the study. While, Wong et al. (2011) reported that high levels of protein can compete with WS for available water, weakening the gel formed by WS gelatinization. Additionally, at low concentrations, hydrocolloids did not cover WS particles, which promoted the free expansion potential of WS particles, resulting in an increase in the viscosity of the system (Chen et al., 2014). It can be seen that the content of components in the system also has a key influence on the interaction. However, based on the content of each component in the actual system, there is a dearth of research on the individual or synergistic effects of each component in the ternary system on the development of batter properties. It is necessary to establish an independent system to determine the role of each component.

Understanding the viscosity of batter and the structure of batter gel is helpful in predicting the processing properties and texture development of fried products (Bai et al., 2022). In this work, four batter systems of WS, WS-WG, WS-CMC, and WS-WG-CMC were prepared according to the content of WS, WG, and CMC in the actual optimized batter system. The viscosity of batter, rheological properties, structure and microscopic observation of batter gel were analyzed to reveal the intermolecular interaction. Finally, the texture characteristics of fried batter were evaluated to verify its application performance. This study clarifies the interaction between hydrocolloid, protein and starch molecules, and provides a feasible option for developing efficient and low-cost starch-based batter formulations in practical applications.

2. Materials and methods

2.1. Materials

Commercial native WS (starch content 88.5 %) was purchased from Xinliang Food Co. Ltd. (Beijing, China). CMC was purchased from Aladdin Biochemical Technology Co., Ltd. (Shanghai, China). WG (protein content 85.8 %) was obtained from Huafeng Powder Industry Co., Ltd. (Henan, China). All solutions were dissolved with Milli Q water.

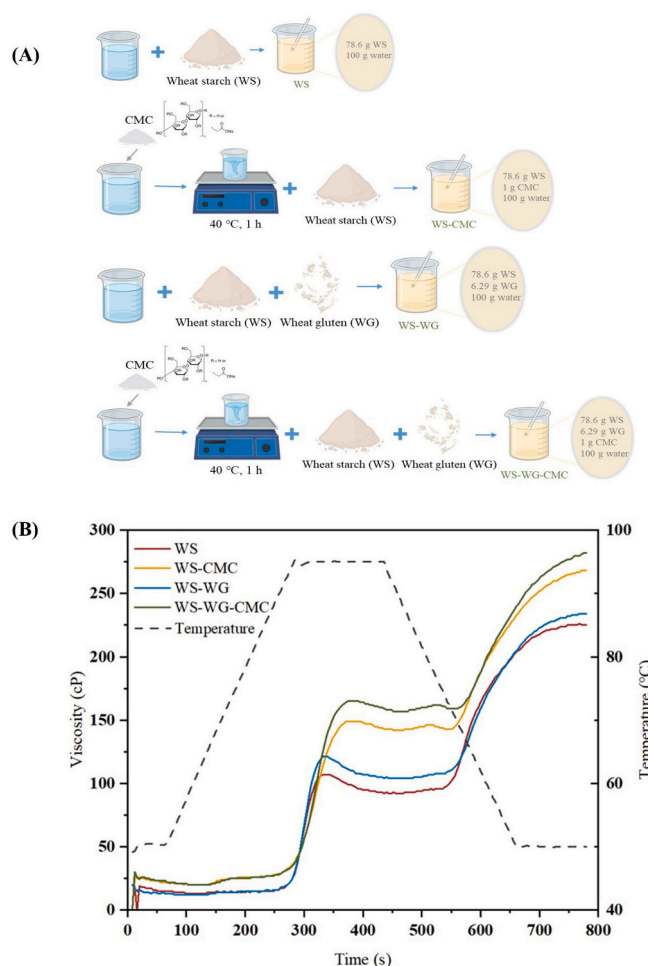


Fig. 1. Schematic diagram of sample preparation (A); Effect of the addition of WG, CMC and WG-CMC on the apparent viscosity of WS-based batter (B). WG: wheat gluten; CMC: carboxymethyl cellulose; WS: wheat starch.

2.2. Batter preparation

The batter formulations were according to Cui et al. (2022) with slight modifications. The optimized batter consisted of wheat flour (60.0 g, 9.0 % protein, 72 % starch), WS (40.0 g), water (100.0 g), and CMC (1.0 g). The wheat flour was replaced with WG and WS to simulate the actual batter system. By equivalent conversion, the system includes 78.6 g WS, 6.29 g WG, 1.0 g CMC and 100 g deionized water. CMC powder (1 g) was dispersed in deionized water (100 mL) and then heated to 40 °C with gentle stirring for about 1 h to ensure complete dissolution. In order to investigate the effects of wheat protein and/or CMC on the WS-based batter, four blends were prepared (WS, WS-CMC, WS-WG, WS-WG-CMC) according to the content of each component in the real system (Fig. 1A). The recipe for WS group was 78.6 g WS and 100 g deionized water. The recipe for WS-WG group was 78.6 g WS, 6.29 g WG, and 100 g deionized water. The recipe for WS-CMC group was 78.6 g WS, 1 g CMC, and 100 g deionized water. And the recipe for WS-WG-CMC group was 78.6 g WS, 6.29 g WG, 1.0 g CMC and 100 g deionized water.

2.3. Pasting properties

A rapid visco-analyzer (RVA, Perten Pty. Ltd., Sweden) was used to determine the pasting properties of WS-based batter incorporated with different adhesives. About 28 g of each sample was weighed in an aluminum canister and inserted into the instrument. The pasting

parameters were measured following the general pasting method (STD 1). After equilibration at 50 °C for 1 min, the dispersions were heated to 95 °C in 240 s, maintained at 95 °C for 120 s, subsequently cooled down to 50 °C within 240 s, and finally maintained at 50 °C for 120 s. The analysis was performed under constant stirring (160 rpm).

2.4. Determination of leached amyloses content and swelling power

The leached amyloses content and swelling power of mixtures was determined using the methodology proposed by Chen et al. (2014) with slight modifications. 5 % w/w of sample slurry was heated at 95 °C for 30 min in a shaking water bath. Then the sample was cooled down to room temperature and centrifuged at 16,000 ×g for 20 min. The supernatant was collected to measure the leached amyloses content by the iodine colorimetric reaction (Chen et al., 2014). The absorbance was determined at 620 nm. Wheat amylose was used as standards for calibration. The swelling power was expressed as the ratio of the wet weight of the precipitate to its dry weight.

2.5. Rheological properties

A controlled-stress rheometer (Physica MCR301, Anton Paar, Austria) was used to determine the rheological properties of pasting sample. The gelatinized samples were prepared following the pasting procedure described in Section 2.3, and then were immediately transferred to a cone-plate geometry (40 mm) with a gap of 1 mm for rheological test. Frequency sweep was conducted in the range of 0.1 to 100 rad/s at 1 % strain. The storage modulus (G'), loss modulus (G'') and loss tangent ($\tan \delta$) were recorded.

2.6. Fourier transform infrared spectroscopy (FTIR)

The molecular characteristics of gelatinized samples incorporated with different adhesives were measured by FTIR-ATR spectroscopy (SENSOR 27, Bruker, Germany). In this experiment, the freeze-dried samples were ground into powder and scanned between 4000 and 400 cm^{-1} . The spectra resolution was 4 cm^{-1} . The absorbance intensities at 1022 and 1047 cm^{-1} were measured relative to the baseline for each sample. The ratio of these absorbance intensities (1047/1022 cm^{-1}) was calculated to compare the short-range conformation of the samples (Warren et al., 2016).

2.7. X-ray diffraction and relative crystallinity

Different samples were characterized using X-ray diffraction (XRD) (D8, Bruker, Germany) at 40 kV and 15 mA using Cu $K\alpha$ radiation. The diffraction angle (2θ) was scanned over a range extending from 5° to 40°, with a constant scanning rate of 4°/min. In order to determine the relative crystallinity (RC), calculations were performed utilizing the Origin Lab software (Origin Lab Corp., Northampton, MA, USA.). The relative crystallinity of samples was calculated according to the methods of Ek et al. (2021), using Eq. (1).

$$\text{Relative crystallinity (\%)} = \frac{\text{Crystalline area}}{(\text{Crystalline} + \text{Amorphous area})} \times 100 \quad (1)$$

2.8. Scanning electron microscopy (SEM)

The gel samples prepared by the method described in Section 2.3 were freeze-dried, subsequent slicing into thin sections. Then the samples were fixed on the double-sided conductive tape. Afterwards the samples were then vacuum sprayed with gold and placed in SEM (S-570, HITACH, Japan). Images at 100 × and 200 × resolution were observed and recorded under an accelerating voltage of 10 kV.

Table 1

Changes in the pasting properties of WS-based batter incorporated with different adhesives.

Sample	Peak viscosity (cP)	Trough viscosity (cP)	Final viscosity (cP)	Breakdown value (cP)	Setback value (cP)
WS	107.50 ± 0.71 ^d	91.50 ± 2.12 ^d	225.50 ± 0.71 ^d	16.00 ± 2.83 ^a	134.00 ± 2.83 ^a
WS-CMC	148.50 ± 0.71 ^b	141.50 ± 0.71 ^b	267.50 ± 0.71 ^b	7.00 ± 0.00 ^b	126.00 ± 0.00 ^c
WS-WG	121.00 ± 0.00 ^c	104.00 ± 1.41 ^c	233.50 ± 0.71 ^c	17.00 ± 1.41 ^a	129.50 ± 0.71 ^{abc}
WS-WG-CMC	167.50 ± 3.54 ^a	159.00 ± 2.83 ^a	285.50 ± 4.95 ^a	8.50 ± 0.71 ^b	126.50 ± 2.12 ^{bc}

Each value is represented as mean ± SD. Values with different letters (a–d) in the same column are significantly different ($P < 0.05$). WS: wheat starch-based batter; WS-CMC: wheat starch-based batter incorporated with carboxymethyl cellulose; WS-WG: wheat starch-based batter incorporated with wheat gluten; WS-WG-CMC: wheat starch-based batter incorporated with carboxymethyl cellulose and wheat gluten. The same below.

2.9. Quality index

2.9.1. Frying process

About 10 g of batter was placed into a stainless-steel mold (8 cm × 0.7 cm × 0.7 cm, length × width × height) for 30 s before being deep-fried. The fryer (model ZG-BK-ZL-81, manufactured by CHIGO, China) was preheated for a duration of 0.5 h, following which the frying operation was carried out at 180 °C for 5 min. The frying time was evenly apportioned into two parts. Upon the conclusion of the initial half, the batter was inverted to facilitate the frying of its upper surface.

2.9.2. Textural properties

The samples were subject to the texture analyzer (TA-XT2i, Stable Micro System, UK) with a P/6 probe. The speed of the test was 0.5 mm/s, and both the pre-test and post-test speed were 1 mm/s. The test compression percentage was 40 % and the data acquisition rate was 400 pps. The hardness, springiness, gumminess and cohesiveness of samples was recorded afterwards.

2.10. Statistical analysis

All dates are presented as the mean ± standard deviation values of three independent experiments. The analyses of variances, means and standard deviation values were determined using SPSS software (v.26.0, IBM, Chicago, IL, USA). Means were compared using Duncan's test at a significance level of $P < 0.05$.

3. Results and discussion

3.1. Pasting properties

3.1.1. RVA characteristics

Table 1 shows that the WS-WG-CMC group has the highest peak viscosity, followed by WS-CMC group, WS-WG group and WS group. This result suggested that the synergistic addition of CMC and WG promoted the network entanglement of system (Díaz-Calderón et al., 2018). The addition of CMC promotes the viscosity of starch, which is consistent with the results reported by Sullo (2012). They confirmed that due to the depletion flocculation mechanism, the low concentration of xanthan promoted the interaction between the gelatinized starch particles, thus making them clump together to increase the viscosity of starch. However, Chen et al. (2014) reported that Pullulan increased the peak viscosity of starch at low concentrations (0.01–0.07 %) and decreased the peak viscosity of starch at high concentrations (0.07–0.5 %). This phenomenon might be attributed to the different distribution of

Table 2

The amount of leached amylose and swelling power of gelatinized starch granules in the presence of WG, CMC and WG/CMC.

Sample	Leached amylose(mg/g)	Swelling power(%)
WS	335.33 ± 2.60 ^a	16.50 ± 0.12 ^b
WS-CMC	274.66 ± 2.62 ^d	17.16 ± 0.01 ^a
WS-WG	288.22 ± 1.64 ^c	14.30 ± 0.19 ^d
WS-WG-CMC	293.50 ± 1.98 ^b	14.87 ± 0.22 ^c

Each value is represented as mean ± SD. Values with different letters (a–d) in the same column are significantly different ($P < 0.05$).

pullulan on the surface of starch granules. Generally, water molecules migrate into starch granules during heating, causing starch to swell and gelatinize, thereby increasing the viscosity of the system (Chen et al., 2014; Chen et al., 2015). The high concentration of Pullulan reduced the viscosity of the system by inhibiting the swelling and gelatinization of starch through two main paths: 1) Pullulan wrapped around the surface of starch granules, which could delay the swelling and gelatinization of starch granules by competing with starch granules for water molecules; 2) Pullulan could form a limiting network on the surface of starch granules to hinder the swelling of starch granules (Chen et al., 2014; Chen et al., 2015). Low concentration of pullulan had little effect on the free swelling of starch due to incomplete coverage of starch granules (Chen et al., 2014). Meanwhile, a small amount of pullulan interacted with amylose to form a polymer, which increased the viscosity of the system (Chen et al., 2014). Therefore, the increase in peak viscosity of the system in this study might be attributed to the incomplete coverage of starch granules by CMC. The addition of WG increased the pasting viscosity of WS, mainly due to the interaction between WG and WS in the gelatinization process to produce a complex dispersed in the starch gel (Zhang et al., 2021). In addition, the WG addition had no significant effect on the viscosity of the WS before 300 s, while the addition of CMC increased the viscosity of the sample. This result indicated that the initial viscosity increased when CMC was added before gelatinization, which could be attributed to the excellent thickening ability of CMC. This indicated that in the starch system where WG and CMC were co-involved, CMC played a major role in increasing viscosity.

The breakdown (BD) value and setback (SB) value are also essential for assessing the pasting qualities of starch. The BD value mainly reflects the stability of starch particles during heating, and the SB value represents the short-term regrowth capacity of starch (Hu et al., 2020). The addition of WG had no significant effect on the BD value of WS, while the addition of CMC or WG-CMC reduced the BD value of WS. The decrease in BD value suggested that the addition of CMC inhibited the disruption of starch particles' integrity. The addition of WG or/and CMC decreased the SB value of WS, which could suggest that WG or/and CMC had an inhibition of short-term retrogradation. This behavior might be due to the formation of hydrogen bonds between WS and CMC or WG during the pasting process, resulting in a decrease in the association of amylose molecules (Cui et al., 2018). Compared to WG, the addition of CMC resulted in lower SB value for starch. In addition, there was no significant difference in SB values between the WS-CMC group and the WS-WG-CMC group. These outcomes indicated that CMC had a higher binding ability to amylose than WG, and CMC could compete for the binding sites of WG on WS.

3.1.2. Leached amylose and swelling power

Existing report indicated that the content of leached amylose, the swelling power of starch particles, and the competition between leached amylose and remaining ungelatinized particles for free water all have an impact on the viscosity of the WS-based batter (Wang et al., 2020). The swelling power is caused by the starch particles absorbing a large amount of water during heating, resulting in the expansion of the particle volume. As shown in Table 2, the addition of WG decreased the swelling power of WS, which was in accordance with the findings

reported by Liu et al. (2021). This may reflect a competition between protein and starch for available water, resulting in the incomplete gelatinization of starch. In fact, due to its large number of hydroxyl and carboxymethyl groups, CMC usually has a stronger ability to bind with water than wheat protein. Interestingly, the swelling power of WS with the addition of CMC significantly increased from 16.50 % to 17.16 %. This might be attributed to the differences in the concentration of CMC and WG in the system. As speculated in the previous section, the low concentration of CMC did not completely cover the starch molecules, which promoted the permeation of moisture into the starch particles, thereby enhancing the free expansion of the starch molecules (Chen et al., 2014). The addition of CMC decreased the contents of leached amylose of WS, and the degree of reduction was greater than that of WG addition. This may be attributed to the fact that CMC and WG can form complexes with amylose, and CMC has a higher binding ability than WG (Cui et al., 2023). Moreover, the addition of WG-CMC increased the leached amylose of samples compared with the addition of WG or CMC, which might be attributed to CMC interacting with WG to reduce the attachment to starch particles. This finding also suggested that the content of leached amylose is not responsible for the increase in peak viscosity of WS when CMC or WG is added, but is more likely to be due to reasons such as competition for free water between leached amylose and remaining ungelatinized particles and friction between swollen particles.

Overall, the following conclusions can be obtained from the pasting property analysis: 1) WG and CMC synergistically increase the viscosity of WS-based batter, with CMC playing a major role in increasing viscosity; 2) CMC has a higher binding ability to amylose than WG, and can compete for the binding sites of WG on WS; 3) CMC mainly increases the viscosity of the system by promoting the expansion of starch particles induced by absorbing water, while the increase in viscosity of the WS-WG-CMC group might be due to the fact that CMC could not only form complexes with WG through electrostatic interaction, but also form complexes with WS, which increased the friction and water absorption between particles.

3.2. Rheological properties

The dynamic rheology is an important parameter that reflects viscoelasticity. The G' is due to the change of the conformational entropy of the coiled chain and is recoverable, while the G'' is the energy loss due to viscous deformation and internal friction due to relative movement of chain segments and molecular chains, which is irrecoverable (Qiu et al., 2015). Fig. 2A and Fig. 2B illustrates the variation of G' and G'' as a function of frequency at 25 °C. The G' and G'' of all systems were raised with the increased frequency, showing typical weak gel viscoelasticity (Rosell et al., 2011). The addition of CMC or WG significantly changed the elasticity and viscosity of the system, which indicated that the structure of WS gel was affected by CMC or WG. Adding WG increased the G' value of WS, while adding CMC slightly decreased the G' value of WS. This suggested that WG plays a greater role than CMC in forming elastic gel. It has been reported that protein and starch can form gels respectively, and can also interact to form complex gels, which could increase the gel elasticity and improving the stability of the system (Johansson et al., 2022). But CMC was attached to starch particles to prevent starch gelatinization, which reduced the elasticity of the gel. Notably, the elasticity and viscosity of WG-CMC group are between WG and CMC group. Compared with the WG treatment, the reduction of G' in the WG-CMC group might be due to the formation of hydrogen bonds between CMC and amylose molecules, occupying WG binding sites on amylose, thereby reducing the number of gluten-amylose interactions necessary for the elastic quality of the gel network (Cui et al., 2018).

To further evaluate the viscoelastic properties of starch samples, the loss angle ($\tan \delta = G''/G'$) was analyzed (Fig. 2C). $\tan \delta$ of all samples was less than 1, indicating that all samples exhibited the rheological behavior characteristic of weak gels (Zhou et al., 2017). The CMC group

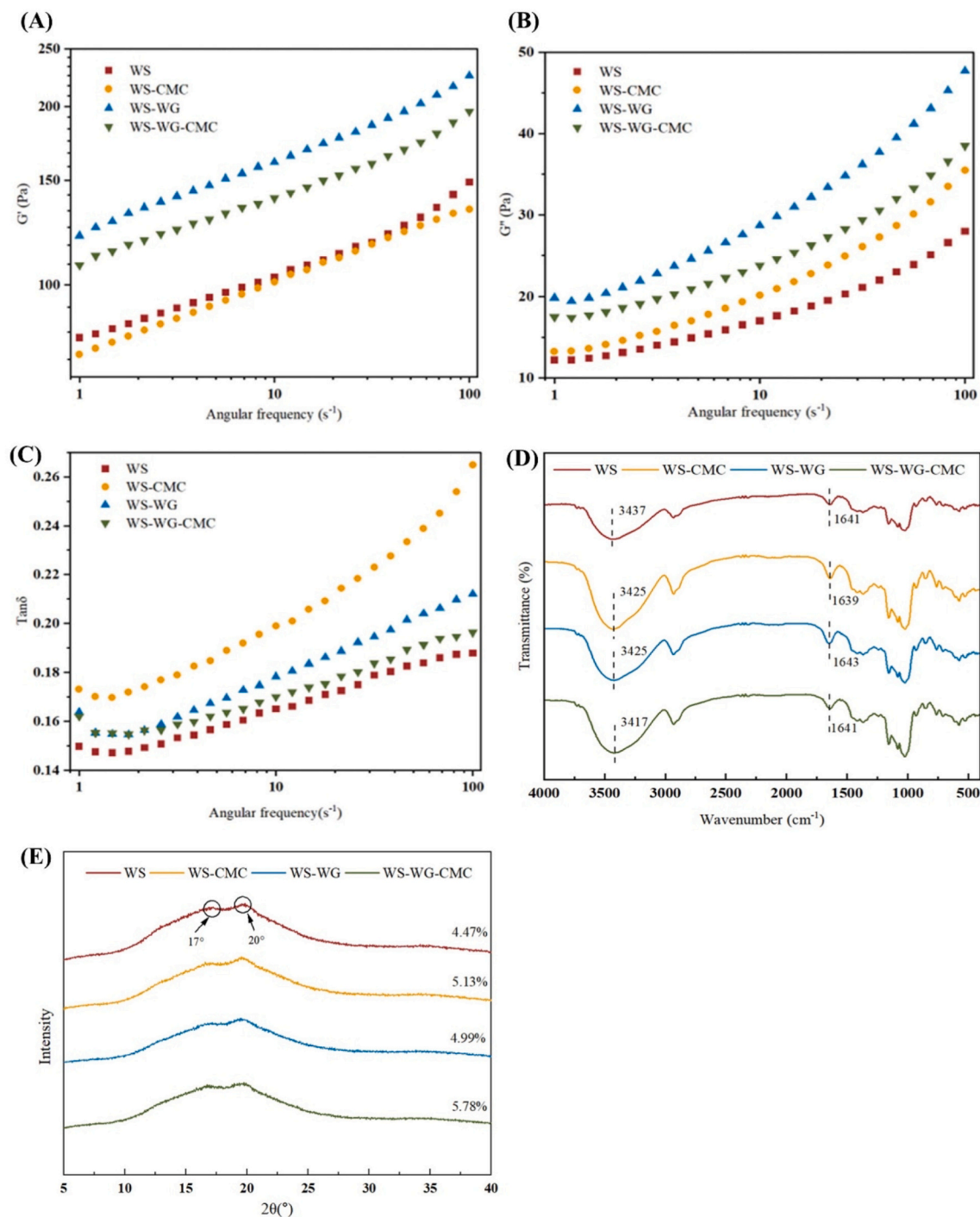


Fig. 2. Rheological properties of WS, WS-WG, WS-CMC and WS-WG-CMC pastes measured at 25 °C: (A) storage modulus G' , (B) loss modulus G'' , and (C) dynamic loss tangent ($\tan \delta$); FTIR spectra (D) and XRD curves (E) of WS, WS-WG, WS-CMC and WS-WG-CMC pastes. WS: wheat starch-based paste; WS-CMC: wheat starch-based paste incorporated with carboxymethyl cellulose; WS-WG: wheat starch-based paste incorporated with wheat gluten; WS-WG-CMC: wheat starch-based paste incorporated with carboxymethyl cellulose and wheat gluten. The same below.

has the highest $\tan \delta$ values, suggesting that the gels systems tended towards the liquid-like behavior. In addition, when the frequency raised to $10 s^{-1}$, $\tan \delta$ of the gel formed by WS containing CMC increased sharply, indicating that the gel at this stage had a tendency to change from solid to liquid. It might be attributed to the weakening effect of CMC on the rigidity of the starch gel network, which promotes its unable to resist the vibration of the starch gel at this high frequency (Liu et al.,

2020). Overall, compared to CMC, WG plays a major role in rheological properties.

3.3. Structural properties

3.3.1. FTIR analysis

To further explore the reason for the viscoelastic changes, the

Table 3

Characteristic absorption peak wavelength of WS incorporated with WG, CMC and WG/CMC.

Items	WS	WS-CMC	WS-WG	WS-CMC-WG
O-H stretching vibration peak (cm^{-1})	3437	3425	3425	3417
Water adsorption in amorphous regions	1641	1639	1643	1641
$R_{1047/1022}$	1.013	1.043	1.033	1.058

changes in structure and the functional groups were measured by FTIR (Fig. 2D). Compared with the WS group, no novel peaks were discernible in the infrared spectra of the WS-CMC, WS-WG, WS-WG-CMC gels, suggesting that no new covalent bond formed within these composite mixtures. The absorption peak located at $3100\text{--}3700\text{ cm}^{-1}$ were attributed to the O—H stretching vibration, which was related to the

formation and change of hydrogen bonds (Wang et al., 2024). The addition of CMC or WG shifted the wavenumber of hydroxyl stretching vibration peak of WS from about 3437 cm^{-1} to about 3425 cm^{-1} , while the addition of WG-CMC shifted that of WS from about 3437 cm^{-1} to about 3417 cm^{-1} , which indicated that the addition of WG and/or CMC could increase the formation of intramolecular or intermolecular hydrogen bonds (Fan et al., 2017). This outcome also explained the decline in SB values (Table 1). Additionally, with the addition of WG-CMC, the hydroxyl stretching vibration peak of WS has a greater shift, which indicated that the synergistic addition of WG and CMC had a strong effect on the formation of hydrogen bonds in the system than the addition alone. The absorption peak at $1525\text{--}1775\text{ cm}^{-1}$ is related to the water adsorption in the amorphous region of starch molecules (Ren et al., 2020). The wavenumber peak at $1525\text{--}1775\text{ cm}^{-1}$ of WS blended with the CMC system showed a blue shift, while WS blended with the WG system showed a red shift, which indicated that the addition of CMC could increase the content of residual water and improve water

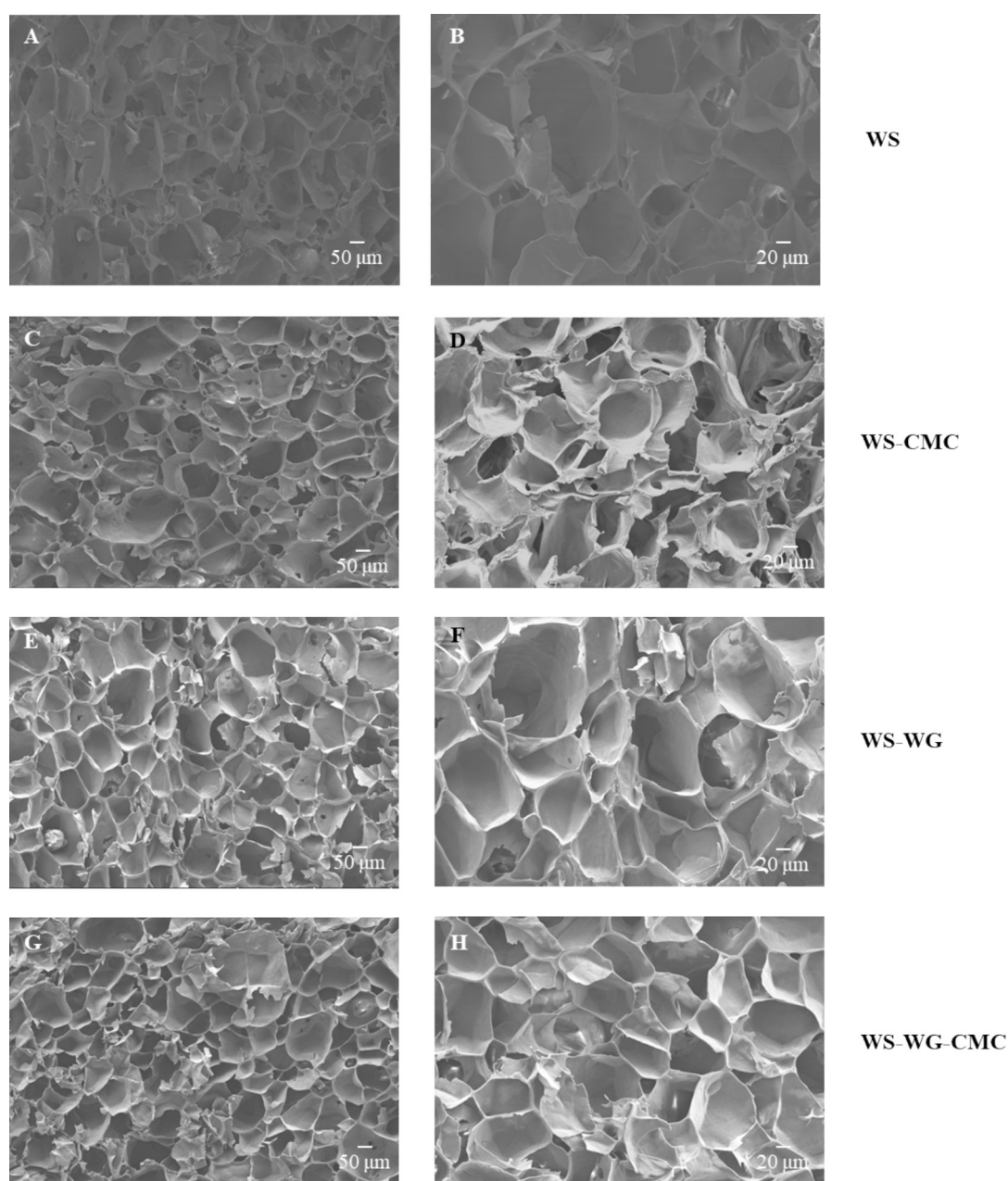


Fig. 3. Scanning electron micrographs of WS, WS-WG, WS-CMC and WS-WG-CMC gels (A, C, E, G: 100 ×; B, D, F, H: 200 ×).

retention. The typical crystal peaks of starch at 1022 cm^{-1} and 1047 cm^{-1} are indicative of the short-range ordered structure of starch (Yang et al., 2021). Zeng et al. (2015) reported that the DO value ($1047\text{ cm}^{-1}/1022\text{ cm}^{-1}$) can be used to describe the degree of order of starch. As shown in Table 3, the DO value of the WS increased with the addition of WG or CMC. According to the results in Section 3.1.2, it can be seen that the addition of WG or CMC decreased the leached amylose, indicating that WS was not completely gelatinized. The incompletely gelatinized starch still had solubilized starch particles to promote the crystalline regions of WS more orderly (Zhang et al., 2005). Moreover, the DO value of WS with the addition of WG-CMC rose from 1.013 to 1.058, which indicated that the order degree of starch molecules increased. In addition, the DO value increased to a greater extent with the addition of CMC than with the addition of WG, which suggested that CMC inhibited the gelatinization of WS more than WG. This finding suggests that CMC attachment to the starch surface inhibited starch pasting, to prevent cross-linking between starch and gluten, and reduced the elasticity of the WS-WG network structure.

3.3.2. XRD analysis

Native WS exhibits the characteristic A-type crystalline structure, characterized by prominent doublet diffraction maxima centered around 17° and 18° , along with singular diffraction peaks at 15° and 23° (Li et al., 2017). It has been reported that the majority of the diffraction peaks of WS disappeared after gelatinization, leaving only two weak peaks at 17° and 20° , which were consistent with B-type crystallization characteristics (Luo et al., 2017). This is consistent with our result (Fig. 2E), which suggests that the crystalline structure of starch was disrupted during gelatinization. The addition of either CMC or WG did not visibly alter the position of the diffraction peak, indicating that neither of them can change the crystal type of starch. Starch molecular chains underwent aggregation and established intermolecular associations via hydrogen bonding, which facilitated the recrystallization process of the starch paste upon cooling. The relative crystallinity (RC) value is an indicator for quantifying the extent of retrogradation and recrystallization of the starch (Ma et al., 2019). As shown in Fig. 2E, the RC value of WS exhibited an increase with the incorporation of CMC or WG. The increased RC value implied that the WG-CMC addition promoted the recrystallization and the long-range ordered structure of WS. The results were consistent with the findings discussed in Section 3.3.1, which indicated that both CMC and WG could increase the order of crystalline regions of WS-based batter (Table 3). In addition, the RC of starch increased to a greater extent when added CMC compared with the addition of WG. Although the addition of CMC and WG changed the crystallinity of starch molecules, the crystal type of starch did not change.

3.3.3. Microstructure

All of the samples exhibited an evident network structure with a “honeycomb” shape (Fig. 3), which is consistent with the characteristic structure of starch and polysaccharide (Luo et al., 2020). WS gel reticulation was disordered, with thin pore walls and uneven net pore sizes. Compared with WS gel, all composite gels displayed a continuous and relatively ordered lamellar network structure, characterized by parallel chains and thicker pore walls. The explanation might be the ability of CMC and WG to engage in hydrogen bonding interactions with the starch chains, promoting the rearrangement of the starch structure and enhancing the crystallinity of the starch (Fig. 2D and Fig. 2E). As shown in Fig. 3E, the gel network structure of the WS-WG group is more compact, showing a uniformly porous and interconnected mesh structure. This might be due to the interaction between WS and protein promoted the formation of gel network. The water molecules bound by the dense gel network leave pores after freeze-drying. It is consistent with the dynamic rheology results that WG increased the storage modulus of the system and enhanced the elasticity of the sample. The presence of larger pores within the gel networks of composites

Table 4

Texture of deep-fried batter prepared with WS, WS-WG, WS-CMC and WS-WG-CMC.

	Hardness (g)	Springiness (cm)	Gumminess (g)	Cohesiveness (N)
WS	875.36 ± 15.62^a	71.75 ± 0.09^d	681.25 ± 8.09^a	0.60 ± 0.01^c
WS-CMC	492.75 ± 5.43^c	82.57 ± 0.95^c	420.21 ± 10.43^c	0.78 ± 0.01^b
WS-WG	554.60 ± 25.81^b	89.86 ± 0.01^a	542.93 ± 31.62^b	0.85 ± 0.02^a
WS-WG-CMC	421.94 ± 7.09^d	86.00 ± 0.20^b	422.33 ± 22.65^c	0.84 ± 0.01^a

Each value is represented as mean \pm SD. Values with different letters (a–d) in the same column are significantly different ($P < 0.05$).

containing CMC (Fig. 3C) suggests that the system effectively immobilized more water molecules. This is mainly attributed to the low concentration of CMC which did not completely cover the starch molecules, which promoted the moisture permeated into starch particles (Chen et al., 2014). This is also the reason for the increase in viscosity and dilatancy of the WS-CMC group. As shown in Fig. 3G, the addition of WG-CMC made the gel network of the system denser and more orderly. This was mainly due to an increased probability of interaction between the three components, enhancing the cross-linking between molecules. Additionally, compared with other systems, the pore size of WG-CMC group is uneven. This might be attributed to the fact that CMC has a higher binding ability to amylose than WG, and can compete for the binding sites of WG on WS (Cui et al., 2018). This behavior would lead to the formation of network gels through cross-linking between different molecules, and different molecular interactions lead to different pore structures (Cui et al., 2023). Further magnifying the microstructure of the WS-WG-CMC group, we can observe that the pore walls are smooth and complete (Fig. 3H). The complete and orderly gel network has the potential to inhibit the oil absorption of batter during frying, which is crucial to the quality.

3.4. Quality properties

To further investigate the effect of CMC and/or WG on the final quality of deep-fried starch-based batter, the textural characteristics of the samples were also analyzed. After deep-frying, the WS group has the highest hardness and gumminess, as well as the lowest springiness and cohesiveness (Table 4). These properties may have resulted in the hard texture and poor toughness of the sample. Cui et al. (2023) reported that compared with individual starch, adhesive has a strong synergistic effect, which can improve the rheology, gel and thermal properties of starch, and ultimately enhances the texture of fried products. The addition of CMC or WG to the batter system significantly reduced the hardness of the samples, with the addition of CMC resulting in a more significant reduction ($P < 0.05$). This might be due to the fact that the adhesives promoted the formation of gel, thereby enhancing the retention of water. Meanwhile, Li et al. (2024) reported that the interaction between WG and adhesive will affect the structure, strength and integrity of gel network, thus affecting the texture of fried products. Compared with WG, CMC as a thickener could capture more water molecules, which could be reflected by the result of RVA and SEM (Fig. 1B and Fig. 3). In addition, the WS-WG-CMC group was observed to have the lowest hardness, which might be attributed to an increased probability of interaction between the three components, leading to improved water retention (Cui et al., 2023). Compared with the WS-CMC group, the WS-WG group has higher springiness and cohesiveness. To some extent, this could be attributed to the increase in WS storage modulus induced by WG (Fig. 2A). It is worth noting that the gelatinization of WS in an oil phase is different from the observed in water phase, so the viscoelasticity results could not absolutely reflect the

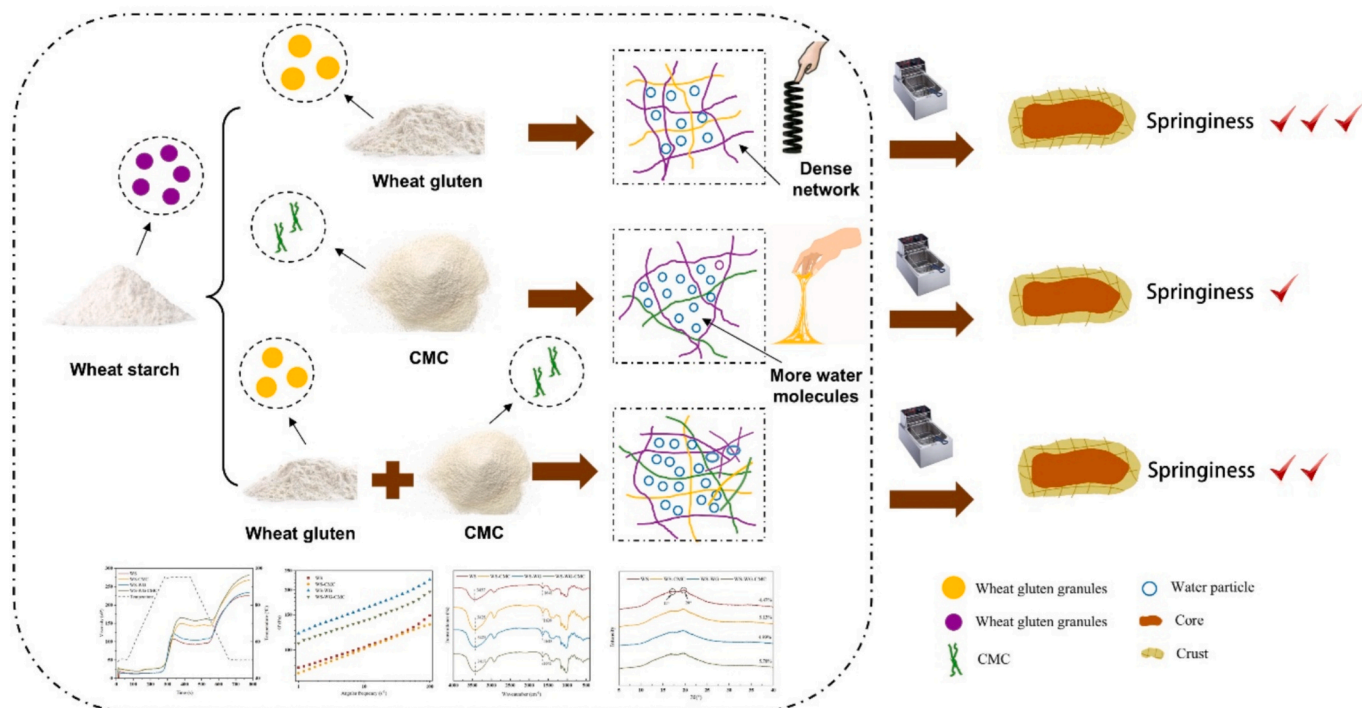


Fig. 4. Schematic description of the influence of CMC and/or WG on the gel structure and texture of WS-based batter for fried products.

springiness of fried products. However, the rheological properties reflect the structure of the batter, which still helps to better predict the processability and final texture of fried products (Bai et al., 2022). In fact, excessive enhancement of springiness can lead to the unwanted toughness of fried products. The above result showed that CMC plays a more significant role in reducing the hardness and gumminess in the batter system, whereas WG plays a more significant role in increasing the springiness and cohesiveness of the samples. The springiness value and cohesiveness value of WS-WG-CMC group are between the WS-CMC and the WS-WG group. Therefore, the synergistic addition of WG and CMC can neutralize the effect of their use alone on the texture of deep-fried products. Fig. 4 depicts a schematic illustration of WG-CMC alleviating the unwanted springiness of fried products prepared with WS-based batter. In other words, the texture of fried products prepared by starch-based batter can be target-controlled by adjusting the ratio of WG and CMC in practical applications.

4. Conclusion

A systematic study proved that WS, WG and CMC, as the main components of batter, can regulate the viscoelasticity and structure of gel network through intermolecular interaction. In the batter system, CMC mainly plays a major role in the viscosity characteristics, and WG mainly contributes to the elasticity of the batter gel. The increase in viscosity of WS-CMC group might be attributed to the incomplete coverage of WS particles caused by low concentration, which promoted the water absorption and expansion of WS. WG improved the viscoelasticity of the system by interacting with WS to form gel networks and enhancing the binding of water. CMC and WG can form complexes with amylose, and CMC has a higher binding ability than WG. The interaction between molecules is mainly a non-covalent interaction based on hydrogen bonds. Moreover, CMC can compete for the binding sites of WG on WS. This behavior might decrease the high springiness of gel caused by the interaction between WS and WG, thus reducing the unpopular toughness of fried products. This work reflected that the synergistic addition of WG and CMC neutralized the effect of their separate addition on the texture of the fried products, providing a theoretical

basis for the selection of batter formulations in WS-based fried foods. Clarifying batter properties in water phase can lay a foundation for further study of molecular interactions during frying. Further research will focus on the interactions between batter components and lipid molecules during the more complex frying process.

CRediT authorship contribution statement

Ying Xu: Writing – review & editing, Methodology, Formal analysis, Conceptualization. **Ruishu Zhang:** Writing – original draft, Methodology, Investigation. **Kangyu Wang:** Writing – review & editing, Visualization. **Prince Chisoro:** Writing – review & editing. **Feng Huang:** Resources, Project administration, Funding acquisition. **Jing Wang:** Supervision, Resources, Project administration. **Chunhui Zhang:** Resources, Funding acquisition.

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.

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Data availability

Data will be made available on request.

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