Journal of Food Engineering 364 (2024) 111812

Contents lists available at ScienceDirect

Journal of Food Engineering

journal homepage: www.elsevier.com/locate/jfoodeng

Development of a novel fat reduction system with quercetin-loaded annealed wheat starch for enhanced emulsifying and oxidative stability in low-fat mayonnaise

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ARTICLE INFO

Keywords:
Mayonnaise
Fat replacement
Annealed starch
Encapsulation
Stability

ABSTRACT

This study developed a novel encapsulation system, a natural antioxidant-loaded annealed wheat starch (AS), as a fat replacer with high antioxidative properties. Quercetin (QUE)-loaded AS (QAS) showed the highest antioxidant activity among the other antioxidants-loaded AS. Physicochemical properties of native wheat starch (NS), AS, and QAS were examined. Mayonnaise properties were assessed after substituting fat with AS and QAS. Annealing treatment increased pasting viscosity and decreased swelling power and water solubility index of AS and QAS compared to NS. Low-fat mayonnaise (LF-M) surpassed full-fat mayonnaise (FF-M) in viscoelastic properties and had brighter color, but overall color properties of LF-M were similar to FF-M. LF-M prepared with AS and QAS by replacing fat content at 50% demonstrated 1.767- and 1.765- times higher emulsifying stability than FF-M, respectively. Moreover, replacing fat content with QAS at 30 and 50% decreased lipid hydroperoxide levels by 2.57 and 3.35 times, respectively, compared to FF-M. This study suggests that AS can effectively encapsulates QUE, serving as a promising fat replacer in clean-label LF-M formulations and providing a healthier alternative to conventional mayonnaise.

1. Introduction

The global food industry is currently undergoing a transformative movement as consumers increasingly demand transparency and healthier options in their dietary choices (Vermeulen et al., 2020). This trend has led to the demand for clean-label food, emphasizing natural, minimally processed ingredients, and no synthetic additives (Asioli et al., 2017). Thus, the manufacturing of clean-label food has gained prominence in response to evolving consumer preferences. Concurrently, certain foods like mayonnaise, an oil-in-water emulsion with semi-solid properties and 70–80% fat content, have gained attention due to health concerns associated with excessive fat consumption (Commission, 1989). This has prompted the exploration of strategies to reduce fat content in mayonnaise while maintaining appearance, flavor, shelf life, and texture (Wang et al., 2022).

Recent studies have explored using chemically modified and enzyme-treated starch as fat substitute in low-fat mayonnaise (LF-M) owing to their high viscosity and emulsion stability (Ma et al., 2006; Ansari et al., 2017). However, challenges arise from complex treatment and chemical hazard, leading to a search for safe, simple, and cost-effective modification methods that align with clean-label food principles. Among physical modification methods, the annealing process is safe, inexpensive, and environmentally friendly option (Wang et al., 2017). During annealing, starch granules absorb water, rearranging their structure between gelatinization onset temperature and glass transition temperature, influencing various physicochemical properties (Seow and Teo, 1993; Majzoobi et al., 2012).

Mayonnaise's high oil content leads to lipid oxidation during storage, shortening shelf life and causing undesirable odors (Gorji et al., 2016). Synthetic antioxidants like butylate hydroxy anisole and butyl hydroxy-toluene are commonly added to prevent lipid oxidation but raise concerns due to their toxicity (Gorji et al., 2016; Xu et al., 2021a; Roshandel et al., 2023). Therefore, identifying natural alternatives to synthetic antioxidants is crucial for producing stable and safe mayonnaise in the food industry. Previous studies demonstrated that natural antioxidants effectively reduce lipid oxidation in mayonnaise (Li et al., 2014; Raikos et al., 2016; Khalid et al., 2021). However, the distinct colors and aromas of natural antioxidants could negatively affect

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https://doi.org/10.1016/j.jfoodeng.2023.111812
Received 10 July 2023; Received in revised form 19 October 2023; Accepted 27 October 2023
Available online 28 October 2023

mayonnaise's sensory qualities. Moreover, low stability and solubility of natural antioxidants in oil often restrict their application in the food industry, demanding strategies to overcome their limitations (Pokorný, 2007; Lourenço et al., 2019).

Recently, encapsulation systems gained interest for preventing bioactive compounds from environmental stress, thereby increasing the stability (Munin and Edwards-Lévy, 2011). These systems also potentially mask bioactive compounds with distinct colors or flavors, mitigating negative sensory impacts on mayonnaise (Gökmen et al., 2011). Previous studies on LF-M predominantly have focused on individual aspects like improving emulsion stability or resistance to lipid peroxidation with antioxidant (Li et al., 2014; Park et al., 2020; Werlang et al., 2021). Moreover, there is a lack of research that simultaneously have explored an integrated approach encompassing both functions. This study aims to bridge this research gap by proposing the use of annealed starch (AS) as a novel encapsulation system for natural antioxidants. This hypothesis is based on the starch molecules' porous characteristics, which may facilitate antioxidant encapsulation by restructuring starch molecules during annealing (Tester and Debon, 2000). To the best of our knowledge, this research is the first to comprehensively investigate AS's dual impact as a fat substitute and an encapsulation system for natural antioxidants in LF-M, enhancing both emulsification and oxidation stability.

Therefore, this study sought to 1) compare the antioxidant activity of quercetin (QUE)-, curcumin (CUR)-, and resveratrol (RES)-loaded AS (QAS, CAS, and RAS, respectively) and choose the one with the highest antioxidant activity; 2) evaluate physicochemical properties of AS and antioxidant-loaded AS, including morphology, pasting property, swelling power, and water-soluble index; 3) examine the influence of AS and antioxidant-loaded AS in replacing fat content (30 and 50%) in mayonnaise, evaluating viscoelastic properties, colorimetric properties, emulsifying stability, and oxidative stability.

2. Materials and methods

2.1. Materials

Starch from wheat, quercetin (QUE), and 2,2-diphenyl-1-picrylhydrazyl (DPPH) were obtained from Sigma-Aldrich (St. Louis, MO, USA). Iron sulfate heptahydrate, 2-propanol, n-butanol, and ammonium thiocyanate were acquired from Daejung Chemical & Metals Co., Ltd. (Incheon, Korea). Curcumin (CUR) was sourced from Acros Organics (Morris Plains, NJ, USA), while resveratrol (RES) and hydrogen peroxide were purchased from TCI (Tokyo, Japan). The vinegar, sugar, salt, canola oil, and eggs used in the preparation of mayonnaise were procured from a local market in Seoul, Korea.

2.2. Preparation of AS

According to Wang et al. (2017), the authors identified 40 °C as the ideal temperature for annealing wheat starch (Wang et al., 2017). Thus, NS was dispersed in distilled water and incubated at 40 °C in a water bath for 24 h to produce AS. The sample procedure was followed for QAS, CAS, RAS, except that QUE, CUR, and RES were dissolved in 99.9% ethanol (2 mg/mL) and mixed with the NS dispersion. Afterward, the dispersions of AS, QAS, CAS, and RAS were freeze-dried and then sieved using a 75 μm sieve. Samples were stored at room temperature until further analysis.

2.3. Encapsulation properties of QAS, CAS, and RAS

2.3.1. Entrapment efficiency

Based on the method previously reported by Jafari et al. (2016), the entrapment efficiency (EE) was assessed (Jafari et al., 2016). QAS, CAS, and RAS solution (1 mL) were subjected to centrifugation at 2400×*g* for 15 min. The resulting supernatants underwent centrifugation again at 13,500×*g* for 10 min. Following this, the absorbance of QUE, CUR, and RES was measured at 370, 425, and 306 nm, respectively, using a Synergy HT Multi-microplate reader (Bio Tek Instruments, Winooski, VT, USA). The EE was determined using the subsequent equation:

$$\textit{Entrapment efficiency } (EE)\ (\%) = \frac{\textit{Total amount of antioxidant} - \textit{amout of free antioxidant}}{\textit{Total amount of antioxidant}}\ X\ 100 \quad (1)$$

2.3.2. Antioxidant activity

The antioxidant activity of QAS, CAS, and RAS were evaluated using a DPPH radical scavenging assay (Bai et al., 2018). Each sample was mixed with a 0.36 mM DPPH solution (1:1, v/v) and allowed to incubate in darkness for 45 min. The absorbance of the mixture was measured at 517 nm using a Synergy HT Multi-microplate reader. The DPPH radical scavenging activity was determined using the following equation:

$$\text{DPPH radical scavenging activity } (\%) = \frac{(\textit{Absorbance of control} - \textit{Absorbance of sample})}{\textit{Absorbance of control}} \times 100 \quad (2)$$

2.4. Physicochemical properties of NS, AS, and QAS

2.4.1. Morphology

The microstructure of starch samples was analyzed using a field emission scanning electron microscopy instrument (SEM) (Verios G4UC, FEI Company, Hillsboro, OR, USA). The samples were mounted onto aluminum specimen stubs using double-sided adhesive tape and subsequently coated with a thin layer of gold. The microstructure imaging was conducted with an accelerating voltage of 15 kV.

2.4.2. Pasting properties

Starch samples were analyzed for their pasting properties using a rheometer (DHR 2, TA Instruments, New Castle, DE, USA) (Bae and Lee, 2018). Starch suspensions (12%, w/w) were maintained for 1 min at 50 °C before raising to 90 °C with 12 °C/min, kept for 2 min at 90 °C. Then samples were cooled with 12 °C/min to 50 °C, followed by being kept for 4 min at 50 °C. For the first 10 s, samples were swirled at 960 rpm, then 160 rpm for the remaining period.

2.4.3. Swelling power and water solubility index

Starch samples were analyzed for their swelling power and water solubility index with slight modifications (Lee and Inglett, 2006). Starch suspensions (1:60, w/v), incubated at 80 °C, were centrifuged for 30 min

Table 1
Ingredient composition of full- and low-fat mayonnaise (wt%).

Ingredient	Full-fat[1)]	AS30[2)]	AS50[3)]	QAS30[4)]	QAS50[5)]
Canola oil	70	49	35	49	35
Vinegar	15	15	15	15	15
Egg yolk	12	12	12	12	12
Salt	2	2	2	2	2
Sugar	1	1	1	1	1
Starch pastes	–	21	35	21	35
Total	100	100	100	100	100

[1)] Full-fat mayonnaise.
[2)] Low-fat mayonnaise in which oil was substituted by an annealed wheat starch at 30%.
[3)] Low-fat mayonnaise in which oil was substituted by an annealed wheat starch at 50%.
[4)] Low-fat mayoonasie in which oil was substituted by quercetin-loaded annealed wheat starch at 30%.
[5)] Low-fat mayonnaise in which oil was substituted by quercetin-loaded annealed wheat starch at 50%.

at 1952×*g* after being cooled. Subsequently, supernatant dried for 4 h and residues were weighed and calculated using Equations (3) and (4):

$$Swelling\ power = \frac{Weight\ of\ sedimental\ paste\ (g)}{Weight\ of\ the\ sample\ (dry\ basis)\ (g) x\left(1-\frac{WSI}{100}\right)} \quad (3)$$

2.5. Mayonnaise preparation

Oil used in full-fat mayonnaise (FF-M) was substituted with AS and QAS at 30 and 50% level (AS30-, AS50-, QAS30, and QAS50-M, respectively) to prepare LF-M with minor modifications (Table 1) (Teklehaimanot et al., 2013). Using a homogenizer (HG-15A, DAIHAN Scientific, Co., Ltd., Wonju, Korea), AS and QAS paste, gelatinized at 90 °C for 30 min, egg yolk, vinegar, sugar, and salt were combined. The slurry was gradually infused with canola oil, followed by being homogenized at 8000 rpm. The substitution of oil with AS and QAS was fixed at 30 and 50% of the oil content in the formulations AS30-, AS50-, QAS30-, and QAS50-M, respectively. The FF- and LF-M were kept at refrigerated temperatures until further analysis.

2.6. Viscoelastic properties of mayonnaise

Mayonnaise samples were analyzed for their viscoelastic properties using a rheometer (Puelles-Román et al., 2021). The dynamic frequency sweep test was run on samples at 25 °C with a gap distance of 1 mm and a constant strain of 1.0% at 0.1–10 Hz. The storage modulus (G′) and loss modulus (G″) were estimated.

2.7. Colorimetric properties of mayonnaise

The colorimetric properties of mayonnaise samples were analyzed using a Minolta Chromameter (Model CR-400, Minolta Co., Ltd, Osaka, Japan). The L*, a*, and b* values of the samples were measured against a standard white plate. The L* value indicates the level of whiteness (100) or blackness (0), the a* value signifies the presence of red (+a) or green (-a), and the b* value represents the presence of yellow (+b) or blue (-b) within the samples.

2.8. Storage characteristics of mayonnaise

2.8.1. Emulsifying stability

According to a previous study, FF-, AS30-, AS50-, QAS30-, and QAS50-M were analyzed for their emulsifying stability with minor modifications (Park et al., 2020). Briefly, all mayonnaise samples (2 g) were incubated at 50 °C for 15 days. Each sample was placed in the centrifuge at 1952×*g* for 10 min every three days until the end of the experimental period. The emulsifying stability of each mayonnaise sample was calculated using equation:

$$Emulsifying\ stability = \frac{Emulsified\ layer\ weight\ (g)}{Total\ sample\ weight\ (g)} \times 100 \quad (5)$$

2.8.2. Lipid oxidation stability

According to a previous study, FF-, AS30-, AS50-, QAS30-, and QAS50-M samples underwent analysis to determine their resistance to lipid oxidation with slight modification (Hu et al., 2003). Each sample (0.3 g) was mixed with a 1.5 mL solution of isooctane/2-propanol (3:1, v/v), and then subjected to centrifugation at 2000×*g* for 5 min. The resulting organic solvent phase (200 μL) was combined with a 2.8 mL solution of methanol and n-butanol (2:1, v/v), and supplemented with 3.94 M ammonium thiocyanate/ferrous iron solution (1:1, v/v). The samples were incubated at room temperature for 20 min, after which the absorbance was measured at 510 nm using a Synergy HT Multi-microplate reader.

2.9. Statistical analysis

Data were expressed as means ± standard deviation and all experiments performed in three times. One-way analysis of variance was utilized to assess data. Statistical significance, which was set at $p < 0.05$ was confirmed by SPSS version 25.0 (SPSS Inc., Chicago, IL, USA) and Duncan's multiple range test.

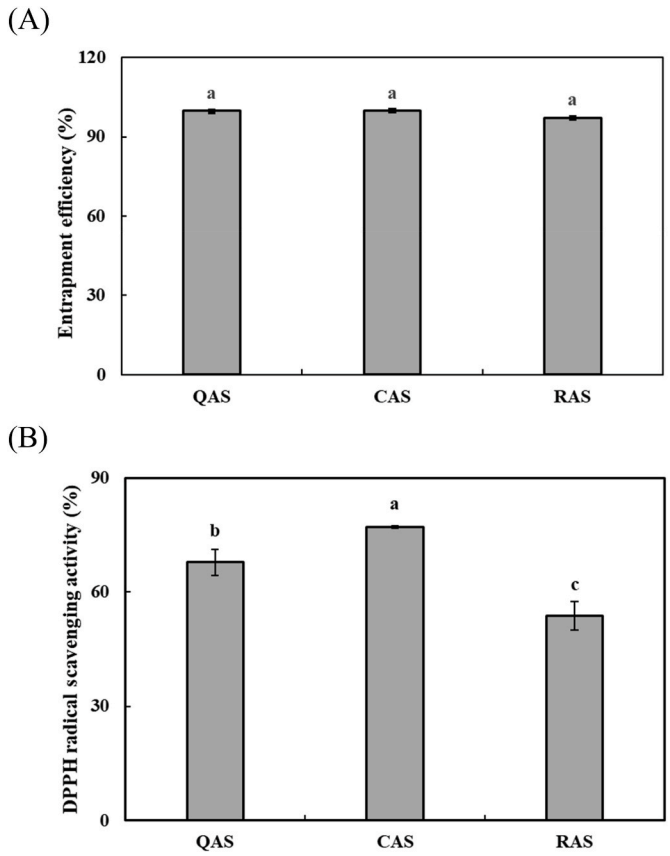

Fig. 1. The entrapment efficiency (A) and antioxidant activity (B) of quercetin (QUE)-loaded annealed wheat starch (AS) (QAS), curcumin (CUR)-loaded annealed wheat starch (AS) (CUR), and resveratrol (RES)-loaded annealed wheat starch (AS) (RAS). [a-c] Different letteres represent significantly different from each other ($p < 0.05$).

3. Results and discussion

3.1. Encapsulation properties of QAS, CAS, and RAS

3.1.1. Entrapment efficiency

The characteristics of QAS, CAS, and RAS were demonstrated in Fig. 1, where the EE of QAS, CAS, and RAS were determined as 99.84, 99.96, and 97.24%, respectively (Fig. 1A). Notably, there were no significant differences in EE observed among antioxidant-loaded AS samples. This might be due to AS's porous structure and starch molecule rearrangement during annealing, allowing antioxidant encapsulation (Tester and Debon, 2000). Thus, these findings affirmed AS's potential as a delivery system for natural antioxidants with a high EE.

3.1.2. Antioxidant activity

QAS, CAS, and RAS exhibited antioxidant activity of 77.18, 67.82, and 53.79%, respectively (Fig. 1B). Interestingly, despite the similar EE among antioxidant-loaded AS samples without significant differences, their DPPH radical scavenging activity displayed significant differences ($p < 0.05$). Particularly, QAS demonstrated the highest antioxidative activity. The different antioxidative activity in QAS, CAS, and RAS could be attributed to their quantity of hydroxyl groups (Nimse and Pal, 2015). QUE contains five phenolic hydroxyl groups, while CUR and RES have 2 and 3, respectively (Lund and Pantuso, 2014). It is well-known that DPPH radical scavenging activity is influenced by the presence of hydroxyl groups, especially phenolic hydroxyl groups, which are closely associated with the antioxidant activity against free radicals (Chen et al., 2020). Therefore, the greater number of phenolic hydroxyl groups in QUE contributes to superior antioxidant activity compared to CUR and RES. Based on these findings, QAS, high EE and the highest antioxidant activity, was chosen for further analysis.

3.2. Morphology

The morphology of NS, AS, and QAS was examined using SEM (Fig. 2). The obtained results were consistent with the findings reported by Wang et al. (2017), who characterized wheat starch as comprising of macroscopic A-type and B-type granules, displaying distinct surface features such as indentations and grooves on the A-type granules (Wang et al., 2017). Notably, no morphological differences were observed among the different samples, exhibiting an irregularly spherical shape. This result could be attributed to annealing treatment which modifies the starch's physicochemical properties while preserving its granular structure (da Rosa Zavareze and Dias, 2011). These align with Wang et al. (2017), in which demonstrated that annealing treatment at 40 °C did not influence the wheat starch's granular structure, while its granular structure was destroyed at 50 °C (Wang et al., 2017). Furthermore, this present study verified that QUE encapsulation within AS during the annealing process did not alter morphological properties, thereby affirming that encapsulation of antioxidant within starch molecule during annealing treatment did not influence the structural integrity of the starch granules.

3.3. Pasting properties

The pasting properties of different starch samples, including NS, AS, and QAS, were analyzed to explore the physicochemical characteristics following the annealing process (Fig. 3). AS and QAS showed higher peak, trough, and final viscosity than NS. This result indicates that the annealing process at 40 °C could enhance the pasting viscosity. Moreover, QUE incorporation into AS did not alter its pasting properties. Increased pasting viscosity of starch after the annealing process could be ascribed to the reconfiguration of starch molecules and enhanced crystalline structure, making starch more organized structure (Wang et al., 2017; Su et al., 2020). The ordered arrangement contributes to the increased rigidity of starch granules, hindering their hydration and

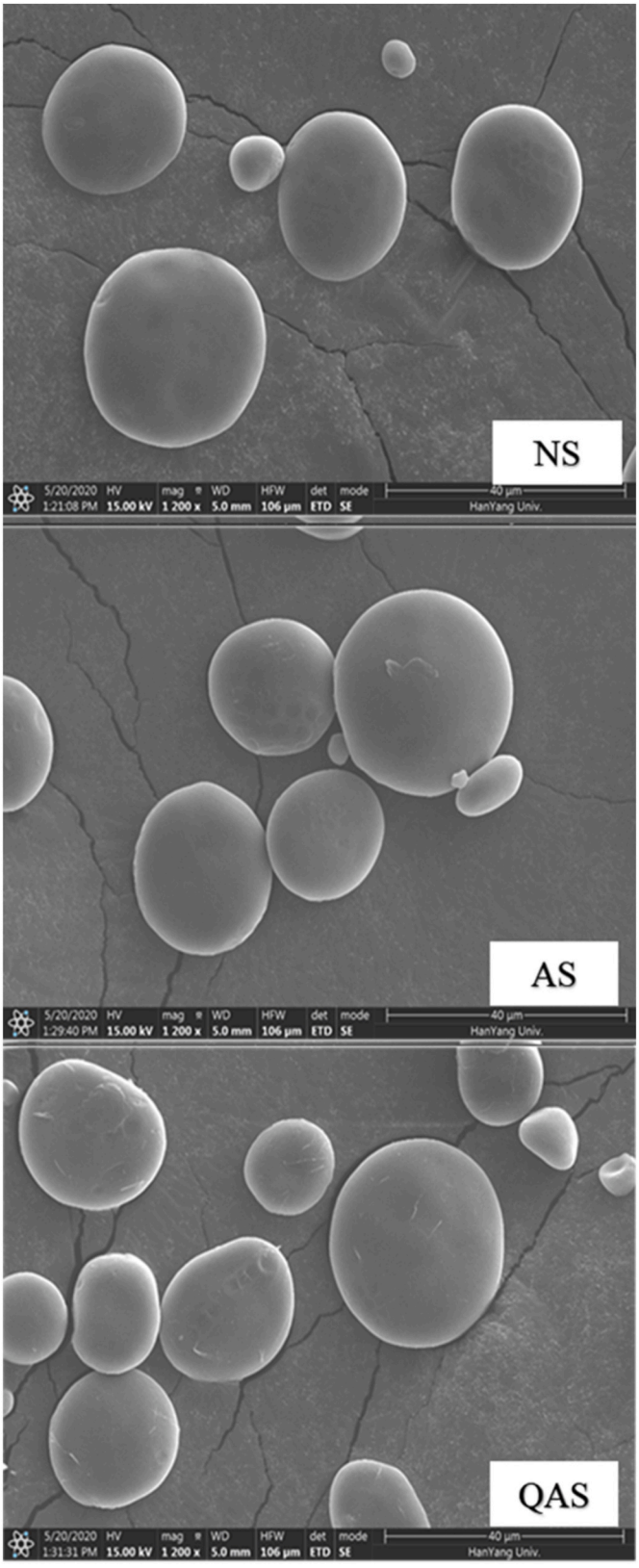

Fig. 2. Morphology of native wheat starch (NS) and annealed wheat starch (AS), and quercetin (QUE)-loaded annealed wheat starch (AS) (QAS).

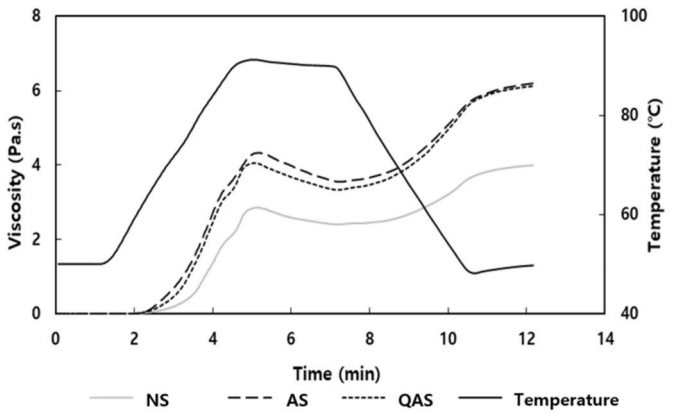

Fig. 3. Pasting properties of native wheat starch (NS), annealed wheat starch (AS), and quercetin (QUE)-loaded annealed wheat starch (QAS).

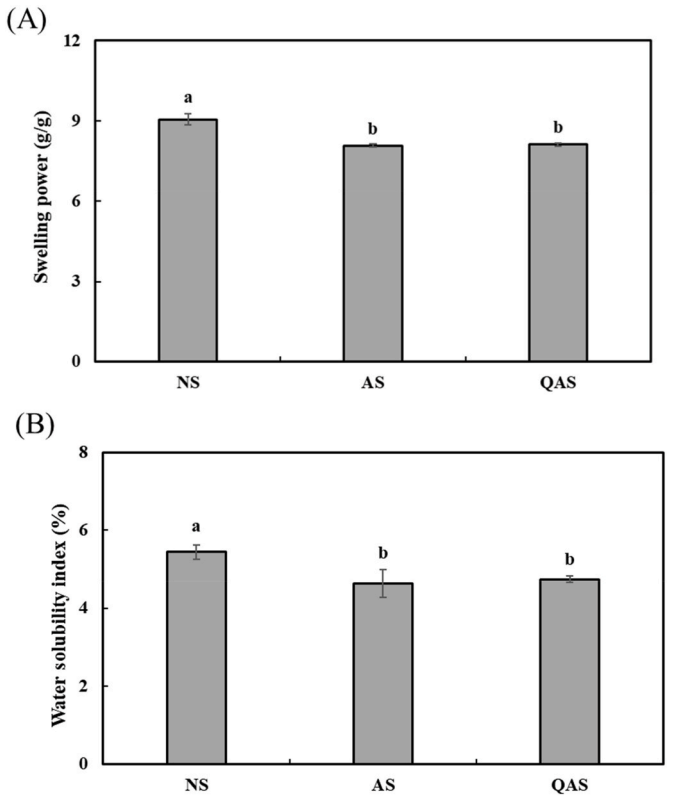

Fig. 4. Swelling power (A) and water solubility index (B) of native wheat starch (NS), annealed wheat starch (AS), and quercetin (QUE)-loaded annealed wheat starch (AS) (QAS). [a-b] Different letters represent significantly different from each other.

dispersion in water. Consequently, the incomplete hydration in water may form a more viscous starch gel, increasing pasting viscosity (Xu et al., 2021b). A previous study also confirmed that AS with increased crystallinity enhanced pasting properties when compared to NS (Su et al., 2020). Interestingly, AS and QAS showed no differences in pasting viscosity, suggesting that QUE incorporation within AS's crystalline structure did not impact the pasting properties of AS during the annealing process.

3.4. Swelling power and water solubility index

The swelling power and water solubility index of NS, AS, and QAS were illustrated in Fig. 4A and B, respectively. NS exhibited significantly higher swelling power and water solubility index than AS and QAS, indicating that annealing process could reduce those properties of starch ($p < 0.05$). Additionally, no significant differences were observed between AS and QAS in terms of swelling power and water solubility index. The decline in the swelling power may be due to structural changes of AS after the annealing treatment. As aforementioned, the annealing treatment increases crystallinity and the rearrangement of starch molecule, leading to a more ordered structure (Wang et al., 2017; Su et al., 2020). Consequently, its enhanced structural order hinders water molecule penetration, decreasing the swelling power of AS (Dias et al., 2010). This finding algins with the previous study, noting a significant decrease in the swelling power of AS compared to NS ($p < 0.05$) (Güllich et al., 2023). The authors attributed this to the enhanced interactions between amylose-amylose, amylopectin-amylopectin, and amylose-lipid, which might restrict the hydration capacity of the amorphous region within the starch granules.

The water solubility index indicates the release of soluble components, particularly amylose, from starch when it encounters water (Lan et al., 2015). Thus, amylose leaching is crucial in determining AS's water solubility index. AS's reduced water solubility index could be attributed to the limited water accessibility resulting from its compact structure, impeding the release of amylose. The lack of significant difference in those properties between AS and QAS could be elucidated by that QUE incorporation within AS has no impact on its structural characteristics, as discussed previously.

3.5. Viscoelastic properties of mayonnaise

The viscoelastic characteristics, including storage modulus (G′) and loss modulus (G″), for FF-, AS30-, AS50-, QAS30-, and QAS50-M were illustrated in Fig. 5. In the case where G′ exceeds G″, it indicates solid-like behavior; conversely, if G″ surpasses G′, it suggests liquid-like behavior (Siwatch, et al., 2022). All mayonnaise samples demonstrated frequency-dependent increases in both G′ and G″, indicating weak gel properties (Ahmed and Ramaswamy, 2005). Furthermore, all mayonnaise samples consistently exhibited higher G′ values than G″, indicating solid-like behavior. Specifically, AS30-, AS50-, QAS30-, and QAS50-M exhibited higher G′ and G″ values than FF-M. It is noteworthy that these viscoelastic properties of LF-M improved as AS and QAS content increased. Generally, reducing fat content in mayonnaise could

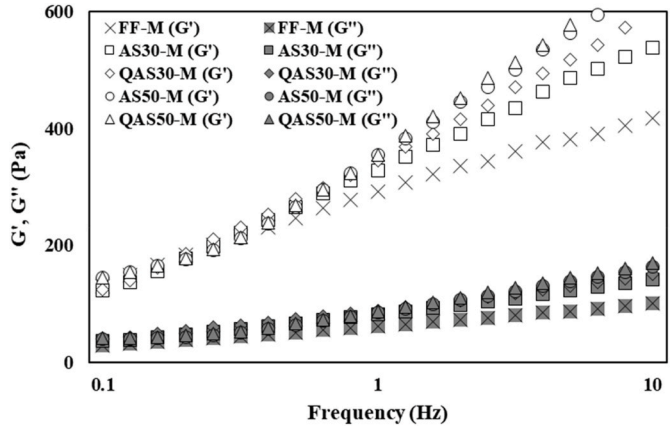

Fig. 5. Influence of frequency on the storage modulus (G′) and loss modulus (G″) of mayonnaise samples. FF-M, full-fat mayonnaise; AS30-M, low-fat mayonnaise in which oil was substituted by an annealed wheat starch at 30%; AS50-M, low-fat mayonnaise in which oil was substituted by an annealed wheat starch at 50%; QAS30-M, low-fat mayonnaise in which oil was substituted by quercetin (QUE)-loaded annealed wheat starch (AS) (QAS) at 30%; QAS50-M, low-fat mayonnaise in which oil was substituted by quercetin (QUE)-loaded annealed wheat starch (AS) (QAS) at 50%.

Table 2
Colorimetric value of different mayonnaise samples.

Sample	Color value		
	L*	a*	b*
FF-M[1)]	67.05 ± 0.92[b]	−2.45 ± 0.32[NS]	18.43 ± 1.67 [NS]
AS30-M[2)]	68.43 ± 1.57[ab]	−2.33 ± 0.38 [NS]	17.19 ± 1.53 [NS]
AS50-M[3)]	70.38 ± 1.92[a]	−2.37 ± 0.36 [NS]	17.13 ± 0.94 [NS]
QAS30-M[4)]	68.12 ± 0.93[ab]	−2.20 ± 0.30 [NS]	17.08 ± 1.41 [NS]
QAS50-M[5)]	70.10 ± 0.48[a]	−2.20 ± 0.20 [NS]	16.66 ± 2.36 [NS]

[a-b] Means with different letters are significantly different at $p < 0.05$.
[NS], not significant ($p > 0.05$).
[1)] Full-fat mayonnaise.
[2)] Low-fat mayonnaise in which oil was substituted by an annealed wheat starch at 30%.
[3)] Low-fat mayonnaise in which oil was substituted by an annealed wheat starch at 50%.
[4)] Low-fat mayoonasie in which oil was substituted by quercetin-loaded annealed wheat starch at 30%.
[5)] Low-fat mayonnaise in which oil was substituted by quercetin-loaded annealed wheat starch at 50%.

shift it from a solid-like state to a more liquid consistency, affecting mouthfeel. However, incorporating AS and QAS improved the viscoelastic characteristic of LF-M, possibly due to increased crystallinity of annealed starch granules, creating a more ordered structure (Wang et al., 2017). This leads to the formation of stable gel network that enhances the viscoelastic properties of LF-M. A previous study also found that the addition of annealed arrowroot starch as a fat substitute improved LF-M's viscoelastic properties (Park et al., 2020). Importantly, the consistent trends in the viscoelastic properties of AS- and QAS-M align with results from analyzing pasting properties, swelling power, and water solubility index, indicating that QUE incorporation within AS did not affect the ordering structure of AS.

3.6. Colorimetric properties of mayonnaise

The L*, a*, and b* values of FF-, AS30-, AS50-, QAS30-, and QAS50-M were demonstrated in Table 2. AS50- and QAS50-M demonstrated significantly higher L* value than FF-M, indicating increased lightness ($p < 0.05$). The perception of mayonnaise's visual aspect is profoundly influenced by its lightness attribute. Previous study has illustrated that adding xanthan gum and starch to LF-M can augment the L* value. This confirmed that the fat content in mayonnaise has a notable impact on the L* value, as higher fat content enhances the light refraction (Mun et al., 2009; Thaiudom and Khantarat, 2011). Regarding the a* and b* values, all samples exhibited negative and positive values, respectively, without any significant differences. These findings indicate that all LF-M samples displayed greenish and yellowish coloration similar to FF-M. Previous study has shown that fillets wrapped in plain film had significantly lower b* values than those wrapped in film containing QUE (Giteru et al., 2017). This suggests that incorporating QUE into a food product could enhance the b* value due to its distinctive yellow color. In contrast, our current study revealed no significant differences in b* values between AS and QAS incorporated LF-M. This indicates masking of yellow color when QUE is encapsulation within AS, thereby having no impact on the overall color of LF-M.

3.7. Storage characteristics of mayonnaise

3.7.1. Emulsifying stability

As illustrated in Fig. 6A, emulsification stabilities of FF-, AS30-, AS50-, QAS30-, and QAS50-M were assessed at 50 °C during 15-day storage period. Initially, at day 0, all mayonnaise samples exhibited high emulsifying stability. However, starting from day 3, FF-M's emulsifying stability notably decreased, while AS30-, AS50-, QAS30-, and QAS50-M maintained consistently high emulsifying stability throughout

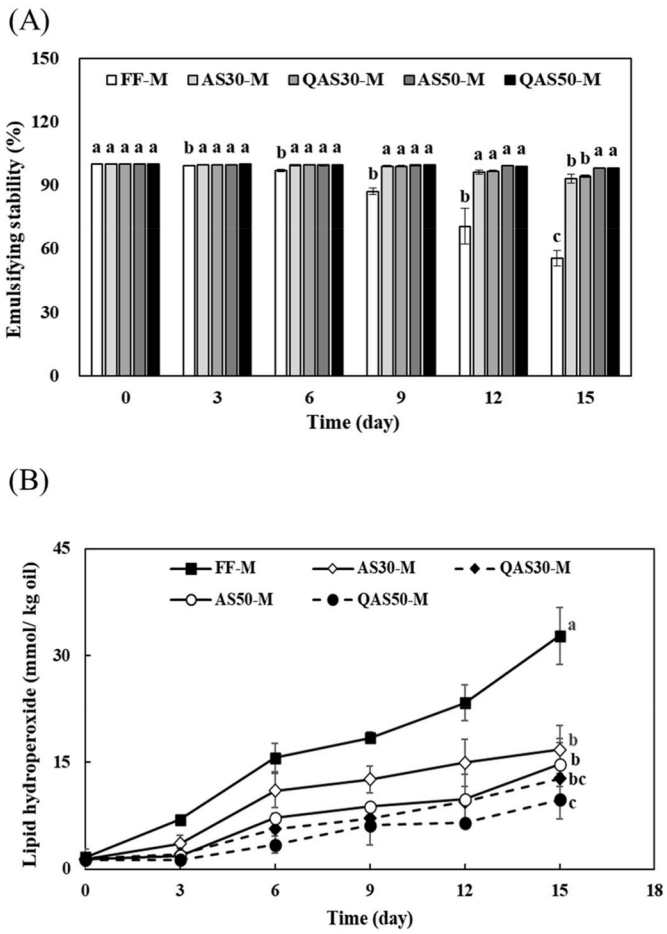

Fig. 6. Emulsion stability (A) and lipid oxidation stability (B) of mayonnaise samples during 15-day storage. [a-c] Columns with different letters mean significantly different from each other. FF-M, full-fat mayonnaise; AS30-M, low-fat mayonnaise in which oil was substituted by an annealed wheat starch at 30%; AS50-M, low-fat mayonnaise in which oil was substituted by an annealed wheat starch at 50%; QAS30-M, low-fat mayonnaise in which oil was substituted by quercetin (QUE)-loaded annealed wheat starch (AS) (QAS) at 30%; QAS50-M, low-fat mayonnaise in which oil was substituted by quercetin (QUE)-loaded annealed wheat starch (AS) (QAS) at 50%.

the entire storage period. Particularly on day 15, the emulsifying stability of FF-M reduced by approximately 1.676, 1.767, 1.698, and 1.765 times compared to AS30-, AS50-, QAS30-, and QAS50-M, respectively. In general, egg yolk enhances mayonnaise stability by adsorbing at the oil-water interface (Anton et al., 2000). However, since both FF- and LF-M formulations contain an identical amount of egg yolk, its effect on their stability considered to be minimal. Thus, these results indicate that incorporating AS and QAS into the formulation of LF-M significantly enhances its stability ($p < 0.05$). Furthermore, AS50- and QAS50-M demonstrated significantly higher emulsifying stability than AS30- and QAS30-M ($p < 0.05$).

In general, emulsifying stability, a crucial property in food systems, might be influenced by steric repulsion (Ma and Chatterton, 2021). Steric repulsion arises from the repulsive forces between dispersed droplets, facilitated by absorbed particle coatings on droplet surfaces. Reduced steric repulsion leads to droplet aggregation, coalescence, and phase separation, resulting in unstable emulsions (Park et al., 2020). In this study, AS and QAS incorporation may envelope emulsion droplets, impeding their contact and inhibiting coalescence. Likewise, Xu et al. (2020) reported that starch could enhance the interfacial thickness, improving stability of emulsion via increased steric repulsion (Xu et al., 2020). Moreover, as previously mentioned, annealed treatment

increased starch granule rigidity, reducing hydration and dispersion in water, resulting in denser starch gel with high viscosity. Additionally, a previous study has affirmed that low swelling power of AS is related to less syneresis and retrogradation of starch gel owing to decreased re-association between amylose and amylopectin chain (Werlang et al., 2021). These characteristics, high viscosity and low potential of syneresis and retrogradation, could also enhance emulsifying stability of AS- and QAS-M compared to that of FF-M. Importantly, no significant differences in emulsifying stability between AS- and QAS-M implied that the QUE incorporation did not compromise the emulsification properties of AS, further supporting its potential as a fat substitute in mayonnaise formulation.

3.7.2. Lipid oxidation stability

The oxidative stability of FF-, AS30-, AS50-, QAS30-, and QAS50-M was assessed over a 15-day storage period (Fig. 6B). FF-M consistently exhibited higher concentrations of lipid hydroperoxides compared to LF-M with varying levels of AS and QAS. At the end of the 15-day storage period, AS30, AS50, QAS30, and QAS50-M reduced lipid hydroperoxide formation by 1.95, 2.23, 2.57, and 3.35 times, respectively, compared with FF-M. Notably, among the LF-M samples, QAS50-M showed significantly lower lipid hydroperoxides levels than AS30-, AS50-, and QAS30-M ($p < 0.05$). These results indicated that QAS incorporation effectively enhances the oxidative stability of mayonnaise and effectively mitigates the formation of lipid hydroperoxides during storage.

In general, FF-M, which contains a large amount of unsaturated lipids, is prone to oxidation. This leads to elevated levels of lipid hydroperoxide during storage and negative changes in sensory qualities, including unpleasant taste (Merkx et al., 2021). In contrast, the inclusion of AS30, AS50, QAS30, and QAS50 in LF-M delayed peroxide formation. Notably, the remarkable antioxidant potency of QUE, attributed to its hydroxyl group, contributed to the reduction in lipid hydroperoxide levels in QAS50-M (Lund and Pantuso, 2014). This strong antioxidative capacity dramatically enhances protection against peroxide generation, ultimately resulting in a significant delay in the increase of lipid hydroperoxide values in LF-M. Similarly, Feng et al. (2020) encapsulated tocopherol to decrease lipid oxidation in fish sausages. The researchers reported that encapsulation of tocopherol served as an efficient delivery system, enhancing the stability and antioxidant properties of tocopherol, consequently reduce the rate of lipid oxidation in fish sausages (Feng et al., 2020). Previous studies have utilized various natural antioxidants, such as beetroot powders, purple corn extracts, and apple peel extract, to enhance the oxidation stability of mayonnaise; however, these negatively affected its color (Li et al., 2014; Raikos et al., 2016; Khalid et al., 2021). On the other hand, the QAS50-M formulation exhibited similar color value to FF-M as demonstrated in section 3.6. Therefore, QAS serves as an excellent fat replacer, improving both the emulsifying stability and lipid peroxidation stability in LF-M while maintaining similar viscoelastic properties and color to that of FF-M.

3.8. Conclusion

This study successfully developed an innovative encapsulation system, AS loaded with natural antioxidants, specifically QUE as a fat replacer in LF-M formulations. We examined the physicochemical properties of AS and QAS and evaluated their impact on LF-M. QAS exhibited the highest antioxidant activity among the analyzed antioxidants-loaded AS. Annealing process increased the pasting viscosity of AS and QAS while decreasing their swelling power and water solubility index compared to NS. LF-M with AS and QAS showed enhanced viscoelastic properties and increased brightness in color compared to FF-M, while maintaining similar overall color properties. It also demonstrated the highest emulsifying stability, while LF-M incorporating QAS exhibited the highest oxidative stability, effectively preventing lipid oxidation during storage. It is important to note that QUE encapsulation within AS improved antioxidant stability without compromising AS's properties, making it a safe and natural alternative to synthetic antioxidants in LF-M. Overall, AS, as an innovative encapsulation system for natural antioxidants in LF-M, offers a healthier and clean-label mayonnaise alternative. This present research contributes to the development of low-fat food products addressing health concerns associated with excessive fat intake while maintaining desirable characteristics and stability.

CRediT authorship contribution statement

Yuan Dan: Conceptualization, Investigation, Methodology, Data curation, Visualization, Writing – original draft. **Youjin Baek:** Conceptualization, Methodology, Data curation, Writing – review & editing. **Eun Woo Jeong:** Conceptualization, Data curation. **Hyeon Gyu Lee:** Conceptualization, Supervision, Project administration, Funding acquisition.

Declaration of competing interest

The authors declare no competing financial interest.

Data availability

Data will be made available on request.

Acknowledgement

This work was conducted by the generous financial support of Samyang Igeon (以建) Scholarship Foundation in Korea and supported by the National Research Foundation of Korea (NRF) grant funded by the Korea government (MSIT) (No.2021R1A2C2013460).

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