



# Diacylglycerol pre-emulsion prepared through ultrasound improves the gel properties of golden thread surimi

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## ABSTRACT

This study determined the influence of diacylglycerol (DAG) pre-emulsion on the gel properties and microstructure of golden thread surimi gels. DAG emulsion stabilized using sodium caseinate was pre-emulsified through ultrasound. The average particle size of DAG pre-emulsion decreased from 1324.15 nm to 41.19 nm, with notable improvements in apparent viscosity and storage stability. The surimi gels with different amounts (0%, 1%, 3%, 5%, and 7% w/w) of DAG pre-emulsion were prepared under heat induction. The whiteness of the composite gels markedly increased with the incorporation of DAG pre-emulsion. The peak  $T_{22}$  value of immobilized water, the gel strength, and water-holding capacity increased gradually, but it slightly decreased with the addition of 7% pre-emulsion. The curve of  $G'$  and  $G''$  kept climbing as the concentration of pre-emulsion, and the microstructure of the gel network tended to become denser and more orderly. Principal component analysis (PCA) of electronic nose results showed that the surimi gels containing pre-emulsion could be clearly distinguished from the control group. In conclusion, the addition of 5% DAG pre-emulsion to surimi not only improved gel properties to the highest extent but also be compensated for lipid loss during the rinsing of surimi.

## 1. Introduction

Rinsing is a two-edged part of the surimi production process: Although it facilitates the removal of lipids and other substances inconducive to the gel properties and storage stability of surimi products, it also results in the loss of some essential nutrients [1,2]. Therefore, the nutritional enhancement of surimi products has become a significant research direction [3,4]. In response to this problem, the research thus far has been focused on add exogenous lipids, mainly vegetable oil [3–7] and animal fat [2,8,9], which can not only enhance the nutrition but also improve the color and flavor of surimi products [3,10].

Various sources of lipids also exert different effects on gel quality and storage characteristics of surimi products [4]. Zhou et al. [1] found that the breaking force, water-holding capacity (WHC), rheological properties, and whiteness of the myofibrillar protein-lipid composite gels containing pre-emulsified camellia oil were better than those containing emulsified lard. Fang et al. [8] concluded that with the increase in the content of emulsified lard, the breaking force and hardness of surimi gels continued to decline, mainly due to the lipid interference on the 3D gel network. Wang et al. [11] investigated the storage characteristics of

surimi gels with the addition of different lipids and found that surimi gels containing fish oil were more prone to lipid oxidation than those gels with flaxseed oil or soybean oil added. Based on existing literature, vegetable oils or lipids with a higher proportion of unsaturated fatty acids may be more suitable for the fortification of surimi products [12,13].

Diacylglycerol (DAG) derived from linseed oil is considered as a good source of *n*-3 fatty acids and has been used in food production [11,14]. It has been reported that DAG can effectively inhibit the accumulation of fat in the gut and reduce serum triacylglycerides (TAGs) [15]. Compared with TAGs, DAG has better emulsification and hydrophilic properties thanks to the free hydroxyl group presented in its structure [15,16], and the addition of DAG instead of TAG in meat products can improve food texture and WHC [16,17]. According to the report of Diao et al. [17], myofibrillar protein composite gel containing DAG had higher gel strength and WHC than that containing ordinary oil, with a more compact and orderly microstructure. Nevertheless, DAG activity will be reduced due to oxidation; however, its stability can be improved through pre-emulsification to ensure its function in the food matrix [18,19].

Pre-emulsification, mainly using high-speed homogenization and

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high-intensity ultrasound, is the most effective approach to improve the stability of oils [3,5,20–22], and it is usually combined with the addition of emulsifiers such as sodium caseinate (SC) [23–25]. The effects of ultrasound emulsification are mainly based on the cavitation effect [5,26]. Specifically, the physical effects are produced at the moment when the cavitation bubble bursts, such as physical shearing, pressures, shock waves and turbulence, which can destroy larger droplets [25–27]. Li et al. [23] used high-intensity ultrasound to improve the interfacial properties and oxidation stability of soybean oil pre-emulsion prepared with SC. Gani et al. [3] prepared a nanoscale virgin coconut oil emulsion through ultrasound with SC as the emulsifier and then added to surimi.

Research on the application of DAG extracted from vegetable oil to surimi products has been scant. In this study, ultrasound was used to prepare an SC-stabilized DAG pre-emulsion, and the effects of different amounts of the DAG pre-emulsion on the gel properties and microstructures of golden thread surimi were investigated.

## 2. Materials and methods

### 2.1. Materials

Frozen golden thread surimi (AA grade) was purchased from Qingdao Baiteng International Trade Co., Ltd. (Qingdao, Shandong, China) and stored at  $-20^{\circ}\text{C}$ . Sodium caseinate (SC) was obtained from Henan Wan Bang Industrial Co., Ltd. (Zhengzhou, Henan, China). DAG (linseed oil source) were supplied by Xian Zeilan Biotech Co., Ltd. (Xian, Shanxi, China). All the chemicals were of analytical grade.

### 2.2. DAG pre-emulsion preparation through ultrasound

An emulsion consisting of DAG and 2% (w/v) SC solution, at a ratio of 3:7 (w/w), was prepared; here, SC and deionized water were mixed for 1 h with a magnetic stirring to ensure complete dissolution. To obtain a coarse emulsion (C), the emulsion was initially emulsified using a high-speed homogenizer two times (30 s each) at 10,000 rpm [23]. The ultrasonic emulsion (U) was further prepared by a low-frequency (20 kHz) ultrasonic cell disruptor (JY98-IIIN, Scientz Biotechnology Co., Ltd., China) at the output power of 300 W for 6 min [24], and the emulsion was kept in an ice bath during this process. The ultrasound intensity in our study was  $66.88\text{ W cm}^{-2}$ , which was determined referring to the method described by Hu et al. [28].

### 2.3. Characterization of DAG pre-emulsion

#### 2.3.1. Optical microscopy

An optical microscope (Niko80i, Nikon Corporation, Tokyo, Japan) was used to observe and photograph the microstructures of the freshly prepared DAG pre-emulsion [29]. 10  $\mu\text{L}$  of pre-emulsion was dropped onto a slide microscope slide and observed under a  $400\times$  microscope.

#### 2.3.2. Particle size distribution

The particle size of the fresh DAG pre-emulsion was measured by Zeta potentiometer (NanoBrook 90 Plus Zeta, Brookhaven Instrument Co., Ltd., USA) referred to the method of Li et al. [23]. The particle distribution and mean particle diameter of the emulsion were recorded for analysis.

#### 2.3.3. Viscosity

The apparent viscosity of freshly prepared DAG pre-emulsion was measured by a Discovery HR-1 dynamic rheometer (TA Co. Ltd., Manchester, England) according to Jiang et al. [21]. The emulsion was evenly spread on the rheometer and no bubbles were ensured. The shear rate rose from  $0.1$  to  $100\text{ s}^{-1}$ , and the test temperature was  $25^{\circ}\text{C}$ .

#### 2.3.4. Appearance and storage stability

The freshly prepared DAG pre-emulsion (1 mL) was dropped onto a

slide to observe its appearance and record images, and another 10 mL of fresh emulsion was injected into a glass bottle with a volume of 15 mL and stored at  $4^{\circ}\text{C}$  for 7 days. The macro changes of the emulsion were regularly observed and recorded by a mobile phone camera.

### 2.4. Surimi gel preparation

The semi-thawed surimi was chopped three times in a Stephan UMC5 chopper mixer, each time for 3 min. During the first chopping, no substances were added, and salt (2.5%, w/w) was added to promote the dissolution of more salt-soluble protein during the second chopping. Finally, the DAG pre-emulsion was separately added to five equal parts of surimi at 0% (control), 1%, 3%, 5%, and 7% (w/w). At the same time, different amounts of ice water were used to replenish and uniformly adjust the moisture content to 78% (w/w). The surimi paste was loaded into plastic casing with a folding diameter of 32 mm after bubbles were drained, and both ends were tied tightly. The surimi samples were cooked at  $40^{\circ}\text{C}$  for 30 min and then transferred to  $90^{\circ}\text{C}$  for another 20 min. The prepared surimi gels were quickly cooled and stored at  $4^{\circ}\text{C}$ .

### 2.5. Determination of whiteness

The  $L^*$  (lightness),  $a^*$  (redness/greenness) and  $b^*$  (yellowness/blueness) values of surimi gels were measured by a colorimeter (CR-400, Konica Minolta, Tokyo, Japan), each sample was set in 6 parallel. The whiteness was calculated by Eq (1) [30]:

$$\text{Whiteness} = 100 - [(100 - L^*)^2 + a^{*2} + b^{*2}]^{1/2} \quad (1)$$

### 2.6. Gel strength and texture profile analysis (TPA)

The gel strength and TPA of surimi gel samples were determined using TA-XT Plus texture analyzer (Stable Micro Systems Ltd., Godalming, UK) [31,32]. The gel samples were equilibrated at room temperature for 30 min and cut into cylinder with a height of 20 mm. The gel strength was determined by a P/5s spherical plunger probe and operated at a constant speed of  $1.00\text{ mm/s}$ , a trigger force of 5.0 g, and a compression strain of 75%. The product of the measured breaking force and deformation was recorded as gel strength. TPA were measured using a P/50 cylindrical probe, and the samples were compressed for two cycles at a speed of  $1.00\text{ mm/s}$  and a compression ratio of 40%. The results were repeated for 6 times.

### 2.7. Determination of water-holding capacity (WHC)

To determine the WHC of the gel samples, the method of Meng et al. [33] was referenced with some modification. The slices of gel samples (about 1 g) were accurately weighed as  $M_1$ , and wrapped with 3 layers of filter paper, then placed in 50 mL centrifuge tubes and centrifuged at  $5,000\times g$  for 15 min at  $4^{\circ}\text{C}$ . The samples after centrifugation were weighed as  $M_2$ , and WHC was calculated according to the Eq (2):

$$\text{WHC}(\%) = M_2/M_1 \times 100 \quad (2)$$

### 2.8. Low field nuclear magnetic resonance (LF-NMR)

The spin-spin relaxation time ( $T_2$ ) of the gel samples was determined by Carr-Purcell-Meiboom-Gill (CPMG) sequence carried by an NMR analyzer (NMI20, Niumag Electric Co. Ltd., Shanghai, China) according to the method of Mi et al. [34] with minor modification. The gel samples ( $10\text{ mm} \times 10\text{ mm} \times 20\text{ mm}$ ) were obtained with a sampler and placed in NMR tubes with a diameter of 20 mm. The resonance frequency SF1 and sampling frequency were set to 22 MHz and 100 kHz, respectively. The data from 3500 echoes were scanned at 8 repetitions. Finally,  $T_2$  relaxation time maps were obtained by the inversion of CPMG exponential attenuation curve.

## 2.9. Determination of rheological properties

The dynamic rheological properties of surimi paste with or without DAG pre-emulsion were measured on a Discovery HR-1 dynamic rheometer immediately after the third chopping in reference to Zhou et al. [1]. The parallel plate (40 mm in diameter) was lowered to a 0.5 mm clearance from the sample stage, and paraffin wax was used for sealing. The changes in storage modulus ( $G'$ ) and loss modulus ( $G''$ ) were recorded as the sample was heated from 20 °C to 90 °C at a rate of 2 °C/min with a scanning frequency of 0.1 Hz and a strain of 2%.

## 2.10. Microstructure

The microstructure of surimi gels was observed by optical microscopy. The gel samples were frozen at −80 °C for 20 min, and then cut into 10 μm thick with a freezing microtome (CM-1850, Leica Instrument Co. Ltd., Germany) [35]. The slices were fixed on the slides, stained by HE staining, observed and photographed under a 40 × objective lens.

## 2.11. E-nose analysis

The flavor profile of surimi gels was analyzed by a portable e-nose system (PEN3, Win Muster Airsense Analytics Inc., Germany) according to the method of Xu et al. [36] with some modification. The surimi gel (5 g) was cut into cubes and packed into a 50 mL centrifuge tube sealed with three layers of clingfilm, and then equilibrated in water bath at 40 °C for 30 min. There was a flushing time of 120 s before each test to restore the sensor to its initial state, and each sample was detected for 100 s with a flow rate of 300 mL/min.

## 2.12. Statistical analysis

All reported data were expressed as the mean ± standard errors and all determinations were carried out in triplicate unless otherwise noted. Significant differences were achieved via one-way ANOVA provided by SPSS 19.0 software (SPSS Inc., Chicago, IL, USA). The results were considered significant as  $P < 0.05$ .

# 3. Results and discussion

## 3.1. DAG pre-emulsion characterization

### 3.1.1. Microstructure

Optical microscopy was used to observe the distribution of droplets in the DAG emulsion; the microscopic images are presented in Fig. 1. We observed that round oil droplets surrounded by clear interfacial membranes were formed in the emulsion, with SC acting as an emulsifier. The droplets in the C emulsion were larger, with more uneven distribution. In contrast, the U emulsion demonstrated a smaller particle size with uniform distribution. Similar results were observed in a previous study [37]. The lipid globules were emulsified to a suitable size such that they

could be trapped and wrapped by the protein after infiltration into the protein matrix [25].

### 3.1.2. Particle size distribution

The particle size distribution of the C and U emulsions is shown in Fig. 2. The main peak shifted significantly to the left under the action of ultrasound, and the average particle size also decreased dramatically ( $P < 0.05$ ), from 1324.15 nm in the C emulsion to 41.19 nm in the U emulsion. Li et al. [23] reported that the particle size of SC-stabilized soybean oil decreased to varying degrees after ultrasound treatment for different durations, where the peak shifted from 2304.7 to 295.3 nm at most. These findings were largely due to the physical force brought by the cavitation effect, which intensified the shearing and destruction of the emulsion particles during the emulsification process using ultrasonic [27]. Notably, the smaller particle size was conducive to the stability of emulsion [37], as verified by the storage stability results (Fig. 4).

### 3.1.3. Viscosity

To evaluate the rheological properties of the DAG pre-emulsion, the apparent viscosity was measured, as presented in Fig. 3. The emulsions exhibited the shear-thinning behavior of the pseudoplastic fluid; that is, the apparent viscosity decreases with the increase in the shear rate [5]. The viscosity of the U emulsion was significantly higher than that of the C emulsion at low shear rate; this result may be related to the particle size of the emulsion. Smaller droplet sizes lead to a closer spacing, thereby enhancing hydrodynamic interactions and improving viscosity [22]. Moreover, the increase in emulsion viscosity can decelerate the movement and collision frequency of droplets and ultimately effectively

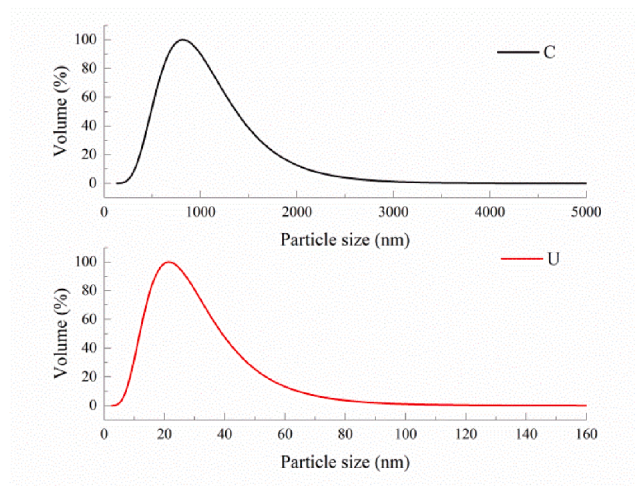


Fig. 2. Particle size distribution of coarse emulsion (C) and ultrasonic emulsion (U).

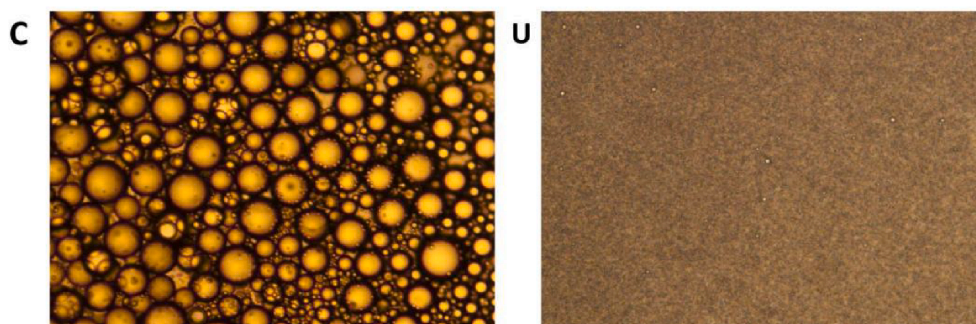


Fig. 1. Images of coarse emulsion (C) and ultrasonic emulsion (U).



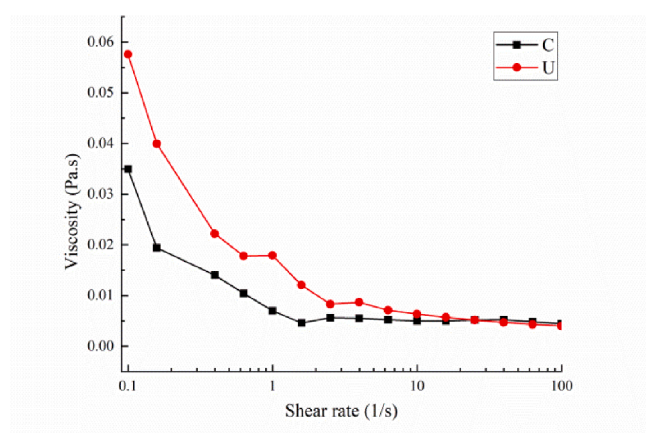


Fig. 3. Apparent viscosity of coarse emulsion (C) and ultrasonic emulsion (U).

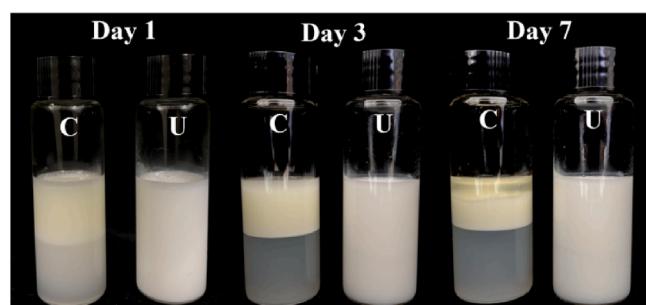


Fig. 4. The image of DAG pre-emulsion emulsion in different periods of storage.

resist flocculation and coalescence between droplets [21].

#### 3.1.4. Storage stability

Good stability is a prerequisite for an emulsion to be used in food products, which is actually determined by the interface protein content and droplet size [38]. Ultrasonic emulsification has been recognized as an effective approach to obtain a stable emulsion by reducing droplet size and enhancing the oil–water interface contact to accelerate emulsification [37,39]. The images of the C and U emulsions stored at 4 °C for 1, 3, and 7 days are shown in Fig. 4. For the C emulsion, creaming caused by gravitational separation was difficult to inhibit. Even on the 7th day of storage, crystal fat appeared in the upper layer of the emulsion. In contrast, ultrasonic emulsion always provided good stability during all 7 days of storage.

#### 3.2. Effect of DAG pre-emulsion on whiteness of surimi gel

Color is an important sensory indicator for evaluating surimi product quality, which can directly affect consumer acceptance [9]. The changes in the color parameters of surimi gels containing different amounts of DAG pre-emulsion are listed in Table 1. The  $L^*$  and whiteness of the surimi gel improved significantly with the increase in the content of DAG pre-emulsion ( $P < 0.05$ ). This improvement was generally thought to be due to light scattering caused by emulsion addition [40]. The appearance of the U emulsion was more milky white than that of the C emulsion, as presented in Fig. S1, which was beneficial to the whiteness of the composite gels. In addition, with the smaller particle size caused by ultrasound, DAG pre-emulsion could be more evenly dispersed in the gel network and thus enhancing the light scattering effect on the gel surface [3]. According to Fang et al. [8] and Gani et al. [3], the whiteness of surimi gel could be improved by adding emulsified animal oil

Table 1

Whiteness of surimi gels added with DAG pre-emulsion at different levels.

	Control	1%	3%	5%	7%
$L^*$	82.71 ± 0.48 <sup>c</sup>	83.08 ± 0.28 <sup>d</sup>	83.72 ± 0.24 <sup>c</sup>	84.70 ± 0.16 <sup>b</sup>	85.14 ± 0.15 <sup>a</sup>
$a^*$	−1.75 ± 0.04 <sup>ab</sup>	−1.87 ± 0.04 <sup>c</sup>	−1.88 ± 0.04 <sup>c</sup>	−1.79 ± 0.03 <sup>b</sup>	−1.73 ± 0.04 <sup>a</sup>
$b^*$	8.59 ± 0.14 <sup>c</sup>	8.59 ± 0.24 <sup>c</sup>	8.34 ± 0.12 <sup>d</sup>	8.94 ± 0.20 <sup>b</sup>	9.17 ± 0.17 <sup>a</sup>
W	80.61 ± 0.40 <sup>e</sup>	80.93 ± 0.17 <sup>d</sup>	81.61 ± 0.17 <sup>c</sup>	82.19 ± 0.10 <sup>b</sup>	82.45 ± 0.15 <sup>a</sup>

Error bars represent mean ± standard deviations, values with different letters in the same row are significantly different at  $P < 0.05$ .

(5% lard) and vegetable oil (5% virgin coconut oil).

#### 3.3. Effect of DAG pre-emulsion on gel strength and TPA of surimi gel

Table 2 indicates that the addition of DAG pre-emulsion improved the gel strength of surimi gels significantly ( $P < 0.05$ ). The pre-emulsified fat globules stabilized using SC in combination with ultrasound were filled in the fish protein matrix and participated in gel formation during the heating process [25]. In particular, the combination between emulsified fat and fish protein through hydrophobic interactions and disulfide bonds led to a dense gel network, and thus, the gel strength of the composite gel was enhanced [15,20]. Moreover, free hydroxyl groups exist in the structure of DAG, which can bond with myofibrillar proteins through the hydrogen bond and further improve the gel network [15]. Zhao et al. [15] and Diao et al. [17] have also confirmed that compared with ordinary lard, the addition of DAG significantly enhanced the strength of the composite gels. Nevertheless, when 7% emulsion was added, the strength of the composite gel did not continue to increase, indicating that the wrapping and binding of the pre-emulsion by the fish protein was close to saturation [1,7]. However, some studies have also reported the adverse effects of oil without pre-emulsification on surimi gel strength, possibly because of oil supplementation reducing the protein content under the fixed moisture content, which was positively correlated with gel strength [4,6]. In the current surimi gel preparation process, the moisture content in each group was adjusted to the same level by using oil, considered to be part of water content. Therefore, the oil dilution effect on protein concentration could be ignored [4].

The texture properties of surimi gels were improved obviously to varying degrees with the continuous addition of DAG pre-emulsion, whereas the difference between the two groups with 5% and 7% additions was not significant. The increase in hardness and springiness might be influenced by the reasons related to the enhancement of gel strength promoted by various factors [25]. The emulsion droplets were coated by the protein matrix and tightly combined, which provided the composite gels with better performance to resist external forces, thereby improving the hardness and springiness [20,41]. On the other hand, the interaction between the emulsion droplets and the protein continuous phase was affected by the emulsifier distributed on droplet surface [25]. Under cavitation effect caused by ultrasound, emulsion oil droplet particle size decreased and the surface area increased, and more SC could be exposed on the surface of the oil droplets, which was conducive to combining with protein matrix after mixing with the surimi paste [25]. In the study of Jiménez-Colmenero et al. [12], the addition of oil-in-water emulsion containing transglutaminase-modified proteins (i.e., soybean protein or casein) improved the hardness and chewiness of Frankfurters significantly. Furthermore, the changes in  $G'$  (Fig. 7A) among different groups could also explain the improvement of springiness in composite gel. However, the enveloping of emulsion droplets by finite proteins approached limit when the addition of the emulsion reached 5%, thus the further addition of DAG pre-emulsion to 7% did not cause a significant change in the textural properties. This was consistent with the

**Table 2**

Gel strength and TPA of surimi gels added with DAG pre-emulsion at different levels.

	Gel strength	Hardness/g	Springiness	Cohesiveness	Gumminess/g	Chewiness/g
control	3530.57 ± 128.91 <sup>c</sup>	1516.24 ± 36.15 <sup>b</sup>	0.93 ± 0.00 <sup>b</sup>	0.81 ± 0.00 <sup>b</sup>	1177.67 ± 89.62 <sup>c</sup>	1145.54 ± 37.90 <sup>b</sup>
1%	3777.03 ± 45.52 <sup>b</sup>	1524.60 ± 40.64 <sup>b</sup>	0.94 ± 0.01 <sup>a</sup>	0.81 ± 0.00 <sup>b</sup>	1203.67 ± 51.42 <sup>bc</sup>	1158.11 ± 42.27 <sup>b</sup>
3%	3891.69 ± 45.94 <sup>b</sup>	1578.65 ± 32.72 <sup>ab</sup>	0.94 ± 0.01 <sup>a</sup>	0.81 ± 0.00 <sup>b</sup>	1263.99 ± 24.39 <sup>ab</sup>	1185.44 ± 18.04 <sup>b</sup>
5%	4090.07 ± 198.43 <sup>a</sup>	1597.66 ± 8.60 <sup>a</sup>	0.94 ± 0.01 <sup>a</sup>	0.82 ± 0.00 <sup>a</sup>	1279.56 ± 37.20 <sup>a</sup>	1227.73 ± 39.80 <sup>a</sup>
7%	4051.58 ± 91.23 <sup>a</sup>	1608.27 ± 43.47 <sup>a</sup>	0.95 ± 0.01 <sup>a</sup>	0.82 ± 0.00 <sup>a</sup>	1297.72 ± 47.92 <sup>a</sup>	1245.82 ± 65.12 <sup>a</sup>

Error bars represent mean ± standard deviations, values with different letters in the same column are significantly different at  $P < 0.05$ .

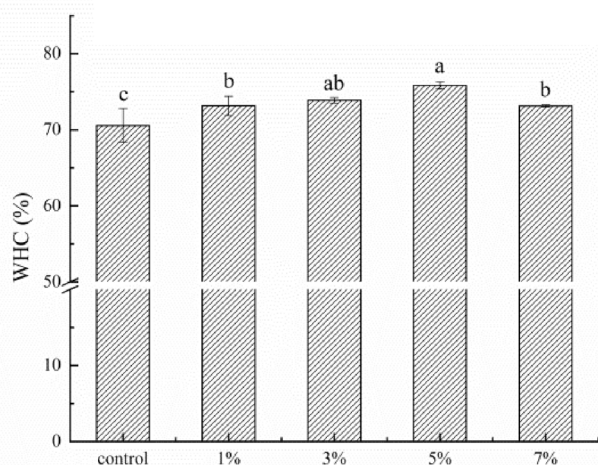
results of the gel strength.

### 3.4. Effect of DAG pre-emulsion on WHC of surimi gel

The WHC of surimi gel denotes the binding ability of its gel network to water molecules, which is closely related to the texture of the gel [32]. The dense and ordered gel network could provide usable space for water retention and restrict the fluidity of water molecules in the gel [1,42]. As shown in Fig. 5, the WHC of the surimi gels was significantly improved ( $P < 0.05$ ) as the content of the DAG pre-emulsion increased; however, it decreased slightly with the addition of 7% DAG pre-emulsion. This corresponded with the aforementioned results for gel strength. Gani et al [3], also reported that the expressible moisture content (EMC) of croaker surimi gel decreased as the virgin coconut oil content increased from 5% to 15% and then plateaued. Both the increase in WHC and the decrease in EMC reflected that the 3D network structure of surimi gel tended to be dense and orderly. As a result, more water molecules were trapped and retained in the gel network to avoid leakage [22]. In addition, the fat globules dispersed in the emulsion interacted with protein; the resulting interfacial protein film was also conducive to restricting the flow of fat and water in the mixed gel network [5,20]. However, the ability of proteins in the gel network to wrap the emulsion was also limited. Therefore, the excess emulsion was potentially lost from the gaps and interfered with the composite gel structure [1].

### 3.5. Effect of DAG pre-emulsion on $T_2$ time of surimi gel

LF-NMR has been considered as an effective method to determine moisture distribution and migration of surimi gel [34]. The binding force of hydrogen protons in the gel system was inversely proportional to the  $T_2$  relaxation time, indicating that the longer the  $T_2$  relaxation time corresponding to water molecules, the more easily it flows [33].



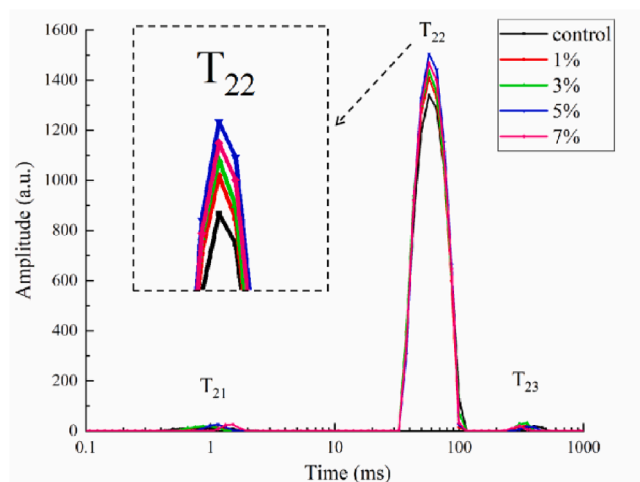
**Fig. 5.** WHC of surimi gels added with DAG pre-emulsion at different levels. The different letters on the bars indicate significant difference ( $P < 0.05$ ).

Therefore, there were usually three peaks in the relaxation time ( $T_2$ ) spectrum ranging from 0 to 1000 ms:  $T_{21}$  (0–10 ms) for bound water,  $T_{22}$  (10–100 ms) for immobilized water, and  $T_{23}$  (100–1000 ms) for free water. As shown in Fig. 6 immobilized water ( $T_{22}$ ) with the maximum peak area was the main form of water in surimi gels. With the continuous addition of the DAG pre-emulsion (1%–5%) in surimi gels, the peak value of  $T_{22}$  increased dose-dependently. In theory, this was because the emulsified fat globules filled the protein-gel matrix and bound with the protein to form a more compact gel network, allowing the free water molecules in the gel matrix to be immobilized [15,33]. Studies have shown that the denser and more orderly the gel network, the better restricted was the flow of water [31,34]. However, overfilling of the emulsion might also exert pressure on the gel network, thereby breaking its integrity and leading to partial water loss.

### 3.6. Effect of DAG pre-emulsion on rheological properties of surimi gel

Rheological behavior of surimi paste containing different amounts of DAG pre-emulsion during heating process from 20 to 90 °C is illustrated in Fig. 7. A consistent trend was noted with increasing temperature between the different treatment groups, regardless of whether it was  $G'$  or  $G''$ . The  $G'$  value increased gradually, and the first peak appeared around 30 °C, which could be explained as the formation of gel network between actomyosin molecules through hydrogen bonding [3,8]. Then, the  $G'$  value declined and reached the lowest value around 50 °C. The reason for the lower value included protein aggregate dissociation and hydrogen bond destruction as well as protein degradation caused by endogenous proteolytic enzymes [3,4,8]. Subsequently, the  $G'$  value increased again and tended to be relatively stable after reaching approximately 70 °C. This process represented the formation of stable gel networks through protein aggregation and crosslinking under thermal induction [3,15,22].

Regarding the influence of the DAG pre-emulsion, the  $G'$  and  $G''$  values at the same temperature were gradually increased with the



**Fig. 6.** The relaxation times ( $T_2$ ) of surimi gels added with DAG pre-emulsion at different levels.



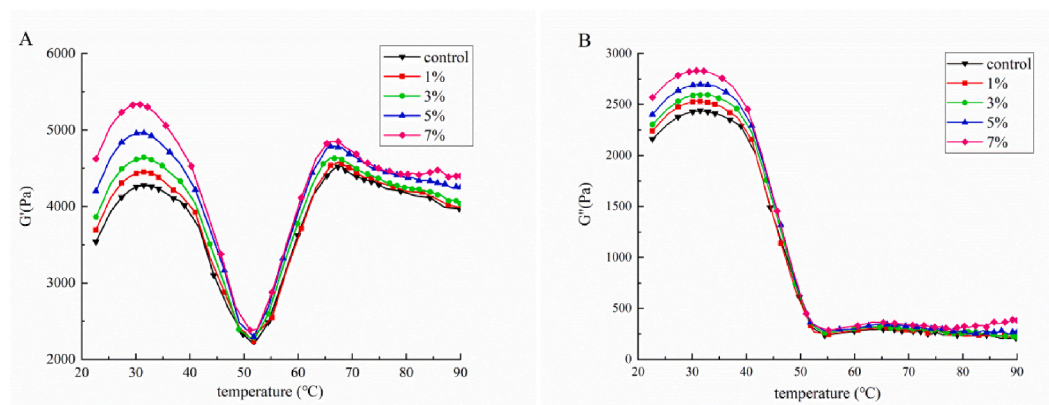


Fig. 7.  $G'$  (A) and  $G''$  (B) of surimi paste added with DAG pre-emulsion at different levels.

increasing amount of emulsion added, which was strongly related to the springiness of the gels [3]. Álvarez et al. [43] stated that in a continuous protein matrix, the aggregation mode of myofibrillar protein can be affected by the incorporation of oils, thereby changing the viscoelastic properties of the composite gels. In particular, the interaction between the fat globules of the pre-emulsion and the proteins of the continuous phase induced a more viscoelastic network structure [15]. Zhao et al. [25] also found that a more viscoelastic composite gel network was formed in the heating process with the addition of soybean oil pre-emulsion to myofibrillar protein sols. In the whole heating process, the  $G'$  value remained greater than the  $G''$  value, indicating that elastic gel occupied a dominant position in the gel system [15,16].

### 3.7. Surimi gels microstructure

The microscopic images of stained surimi gel slices are illustrated in Fig. 8. Here, the gel network structure can be observed clearly. The holes in pure surimi gel are large and unevenly distributed, which decreased its WHC and gel properties [44]. Compared with the control group, the holes within the gel networks were significantly shrunk due to the increasing amount of the DAG pre-emulsion filling the protein-gel gap. The gradually dense gel network could also support the improvement in the gel strength and WHC, as mentioned above [1,7]. The aforementioned advantages of the DAG pre-emulsion were largely owing to the smaller particle size obtained through ultrasound treatment [20]. In the study of Gani et al. [3], the structure of the surimi gels containing large

virgin coconut oil droplets had larger holes, leading to reducing breaking force. However, the addition of 5% ultrasound-treated virgin coconut oil nano-emulsion led to no adverse effects.

### 3.8. E-nose analysis

E-nose analysis with PCA was used to describe the difference in the

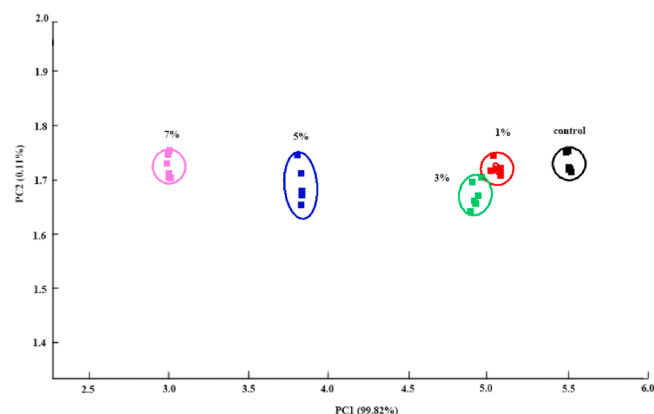


Fig. 9. PCA by e-nose of surimi gels added with DAG pre-emulsion at different levels.

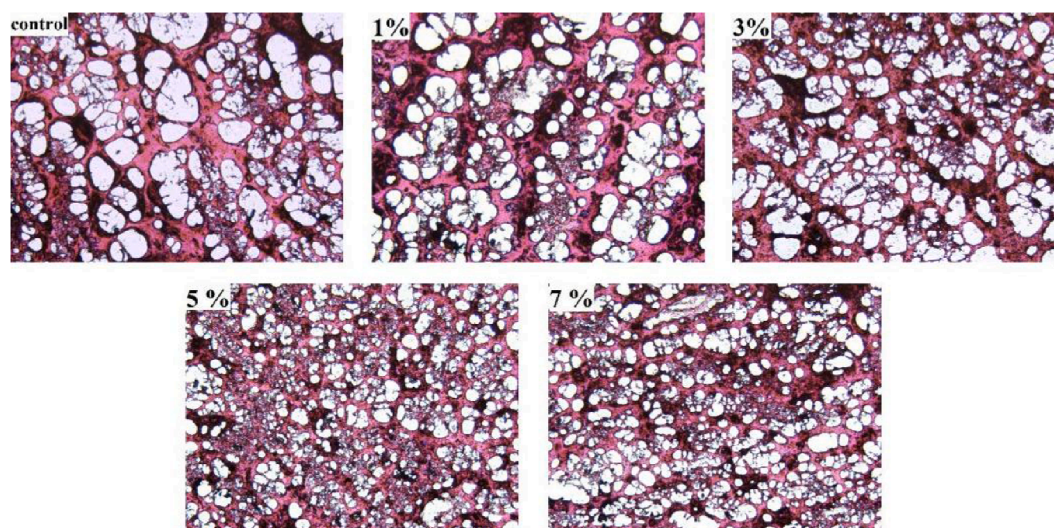


Fig. 8. Microstructure of surimi gels added with DAG pre-emulsion at different levels.

odor profiles of surimi gels containing varying amounts of the DAG pre-emulsion [45]. As shown in Fig. 9, the total contribution rate of the two principal components was 99.93%. As the amount of DAG pre-emulsion increased, the difference in PC1 between each treatment group and the control group gradually expanded. These results indicated that the odor characteristics of the surimi gels were changed due to the incorporation of DAG pre-emulsion, especially the sample with the addition of 5% emulsion, which can be clearly separated from the samples of other treatment groups. The PCA results revealed that appropriate DAG pre-emulsion can modify the flavor characteristics of surimi gels.

#### 4. Conclusion

DAG emulsion obtained through ultrasonic pre-emulsification demonstrated good stability. As the amount of added DAG pre-emulsion was increased, the whiteness of the surimi gels improved significantly, and odor characteristics were modified. The microscopic results indicated that the pores distributed in the network structure shrunk and the gel network tended to be dense with the continuous addition of DAG pre-emulsion. In addition, the gel strength, hardness, and WHC of the composite gels were also improved considerably. However, excessive addition produced some adverse effects. In summary, DAG pre-emulsion supplementation can be a feasible method to compensate for lipid loss in the surimi-rinsing process and enhance the surimi gel properties.

#### CRedit authorship contribution statement

**Yongxia Xu:** Conceptualization, Writing – review & editing. **Yanan Lv:** Data curation, Investigation, Writing – original draft. **Honglei Zhao:** Writing – review & editing. **Xueli He:** Methodology, Data curation. **Xuepeng Li:** Supervision. **Shumin Yi:** Validation. **Jianrong Li:** Project administration, 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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#### Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.ultsonch.2022.105915>.

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