Mass transfer enhancement of tuna brining with different NaCl concentrations assisted by ultrasound

Yao Yao\textsuperscript{a,b,c}, Rong Han\textsuperscript{a,b,c}, Feng Li\textsuperscript{a,b,c}, Juming Tang\textsuperscript{d}, Yang Jiao\textsuperscript{a,b,c,*}

\textsuperscript{a} College of Food Science and Technology, Shanghai Ocean University, Shanghai 201306, China
\textsuperscript{b} Engineering Research Center of Food Thermal-processing Technology, Shanghai Ocean University, Shanghai 201306, China
\textsuperscript{c} National R&D Branch Center for Freshwater Aquatic Products Processing Technology (Shanghai), Shanghai 201306, China
\textsuperscript{d} Department of Biosystems Engineering, Washington State University, Pullman, WA 99164-6120, USA

\textbf{ARTICLE INFO}

\textbf{Keywords:}
Fish
Ultrasound
Mass transfer
Salt diffusion

\textbf{ABSTRACT}

The influence of different NaCl concentrations (2.5, 5, 7.5 and 10\% (w/w)) on the mass transfer kinetics of tuna during brining process with and without ultrasound assistance was evaluated. Results showed that an increase in NaCl concentration and the application of ultrasound accelerated the salt diffusion in the tuna muscle, and the highest yield was obtained in 5\% brine concentration. Moreover, the kinetics parameters were significantly affected by the NaCl concentration and ultrasound application during brining. The values of the mass transfer kinetics parameters ($k_1$, $k_2$) for total and water weight changes decreased as NaCl concentration increased with and without ultrasound assistance during brining. In contrast, the higher the NaCl concentration, the higher the value of the salting kinetics parameters for salt weight changes. The application of ultrasound enhanced the salt effective diffusion coefficient ($D_e$) from 402.8\% to 653.21\% during the brining process, and the highest $D_e$ was also found at 5\% brine concentration. The application of ultrasound can improve the uniformity of salt distribution, enhance water holding capacity, reduce hardness and chewiness, but have no significant effect on color of tuna muscle.

1. Introduction

Brined fish, or called ‘fish in brine’, is popular in the mass market because of its distinct taste and convenience of consumption. Brining is to enhance sensory and organoleptic properties of food products by immersing them into brines [52]. In a brining process, water transfers from foods to brine and salt (NaCl) transfers from brine to foods simultaneously due to the concentration difference [14]. Brine contains a higher concentration of ions than the muscle fibers cells, allowing salt ions cross the membranes of the cells by diffusion until reaching equilibrium. Myogenic fibers absorb and retain large amounts of water due to ion concentration difference, and the osmosis and capillary actions result in a swelling to more than twice of their original volume [12,39]. Due to the limited permeability of salt in cell membrane, it usually takes hours for brined samples to reach a certain concentration. Therefore, there is a need of efficient brining process that reduces brining time and improves the uniformity of product.

Ultrasonic waves produce a series of effects which influence mass transfer when traveling across a medium. The most significant one is the cavitation effect caused by high intensity ultrasound, which leads to the growth and rupture of bubbles inside liquid. Asymmetric implosion of cavitation bubbles near the surface of a solid produces micro-jets in the direction of the surface, and these micro-jets can enhance the permeability of cell membranes [15,48]. Other ultrasonic effects that facilitate mass transfer processes include thermal effect [34], microstirring at the interface [15], and some structural effects, such as the ‘sponge effect’ and the generation of microchannels [37]. As a green and physical processing technology, ultrasound has been applied to many food products for accelerating the mass transfer process and altering food microstructure [40,32,36,28,15]. For example, González-González et al. [25] reported the effect of ultrasound-assisted marination with different durations (20, 40 and 60 min) on the uniformity of beef, and found the distribution of NaCl was more uniform with 60 min ultrasound assisted marination. Zhao and Eun [55] demonstrated that ultrasound-assisted brining of Chinese cabbage effectively reduced the brine immersion time and achieved more uniform salt distribution compared to conventional methods.

Characterizing the diffusion kinetics of a brining process and

\* Corresponding author.
E-mail address: yjiao@shou.edu.cn (Y. Jiao).

https://doi.org/10.1016/j.ultsonch.2022.105989
Received 15 December 2021; Received in revised form 21 March 2022; Accepted 23 March 2022
Available online 24 March 2022
1350-4177/© 2022 The Authors. Published by Elsevier B.V. This is an open access article under the CC BY-NC-ND license (http://creativecommons.org/licenses/by-nc-nd/4.0/).
quantitatively evaluating the diffusion rate of salt are desirable in improving mass transfer efficiency [50]. Fick’s second law of diffusion is commonly used to model a diffusion process and to determine the diffusion coefficients of food products during brining. Researchers have conducted extensive research on the mass transfer kinetics study of fish, pork and chicken in brines [19,1,33]. Telis et al. [50] studied the brining mass transfer kinetics of farmed caiman muscles and found the effective diffusion coefficient ranged from $0.47 \times 10^{-10}$ to $9.62 \times 10^{-10}$ m$^2$/s. Wang, He and Li [54] studied the mass transfer of rabbit meat in different concentrations of brine (5, 10, 15, 20 and 25%) and found that the highest yield was obtained with 5% salt content brine and the effective diffusion coefficient was the highest at the concentration of 25%.

The consumer’s demand for lightly-brined meat products has increased with the enhancement of dietary health awareness [45,41]. Moreover, light brining has many positive effects on fish muscle, including improving water-holding capacity, enhancing texture properties, and acquiring unique taste and high yield. Among the lightly-brined fish products, brined tuna (also known as ‘zuke’ in Japanese) is one of the most popular ready-to-eat raw fish products in Japan. However, limited information is available on salt diffusion kinetics of tuna in brines with ultrasound assistance. Therefore, in this study, the mass transfer kinetics of tuna in brine with different concentrations of NaCl solutions were investigated and the acceleration of brining process with ultrasound assistance was evaluated. Also, the brining uniformity in individual tuna sample with and without ultrasound was compared. Results in this study would provide fundamental data for convenient lightly-brined tuna product development and also provide an evaluation criteria of salt concentration uniformity.

2. Materials and methods

2.1. Raw materials and composition

Frozen yellowfin tuna fillets were used as raw material in this study. Frozen fish fillets were purchased from a local company (Shanghai Zhengrong Agricultural Products Co., Ltd.), packaged in polyethylene Ziploc bags individually, and then kept frozen at $-60 \degree$C before experiments. The water content of the tuna sample was determined using an electric blast drying oven (GZX-9076MBE, Shanghai Boxun Industrial Co., Ltd. Medical Equipment Corporation, Shanghai, China). The salt content of samples was determined using a hand-held salinity meter (PAL-ES1, Atago Co., Ltd., Japan). The moisture content of raw tuna fillet was measured as 76.62 ± 0.67 g water/g DW, and the salt content was 0.80 ± 0.08 g NaCl/g DW. The samples in polyethylene ‘Ziploc’ bags were thawed in a 4 \degree C refrigerator for 12 h, and then cut into pieces ($3 \times 3 \times 1$ cm) with an average sample weight of 9.5 ± 0.5 g. In this study, 452 tissue pieces in total were prepared, weighed, marked, and stored at 4 \degree C until brining experiment.

2.2. Brining treatment

Prepared fish fillet samples were divided into two equal groups and placed into NaCl solution with various concentrations (2.5, 5, 7.5, and 10%) at 4 ± 0.5 \degree C with a ‘fish: salt solution’ ratio of 1:3 (w/w) for brining experiments with and without ultrasound, respectively. Three replicates of experiments were conducted for each brining treatment.

For static immersion brining, six samples were placed vertically in a 200 mL glass jar filled with NaCl solution as a group, and the glass jar was placed directly above the ultrasonic outlet in the ultrasonic water bath cavity. Samples were separated manually to guarantee sufficient contact with brines. With continuously supplied ultrasound, one sample were taken out at each time interval (10, 20, 30, 60, 90 and 120 min) for determination of the kinetic parameters. The acoustic intensity of salt solutions was determined by calomel. This method involved the determination of the temperature increase in the first 90 s of ultrasound application [44].

$$P = MC \frac{dT}{dt}$$

where, $P$ is the ultrasonic power (W), $M$ is the mass of brine (kg), $C_p$ is its heat capacity ($J/kg \degree C$) and $dT/dt$ is the increase of temperature ($\degree C$/s). Each power level is repeated at least six times. Then the applied acoustic intensity ($W/cm^2$) was determined by dividing the ultrasonic power by the emitting surface area of the probe.

Three samples were randomly taken from four concentrations of brine solution at pre-set times for further analysis. The samples were weighed after draining with absorbent paper and grind with a meat grinder. The minced samples were used for moisture and salt content measurement.

2.3. Total, water, and NaCl weight changes of tuna meat

The total weight changes ($\Delta M_i$), water weight changes ($\Delta M_i^w$) and salt weight changes ($\Delta M_i^{NaCl}$) were calculated with Eqs. (2) – (4) [22,23]

$$\Delta M_i = \frac{(M_i - M_i^0)}{M_i^0} \times 100(\%)$$

$$\Delta M_i^w = \frac{(M_i^w - M_i^{0w})}{M_i^{0w}} \times 100(\%)$$

$$\Delta M_i^{NaCl} = \frac{(M_i^{NaCl} - M_i^{0NaCl})}{M_i^{0NaCl}} \times 100(\%)$$

where, $M_i^w$ and $M_i^{0w}$ are the weights (g) of the tuna sample at brining time $t$ and 0; $X_i^w$ and $X_i^{0w}$ are the moisture content (%) of the tuna sample at brining time $t$ and 0; $X_i^{NaCl}$ and $X_i^{0NaCl}$ are the salt content (%) of the tuna sample at brining time $t$ and 0.

2.4. Mass transfer kinetics of tuna during brining

In order to elucidate the mass transfer phenomena occurring during the brining process, a mathematical model of the mass changes was fitted with experimental data. In this model, the tuna sample weight changes were related to the square root of time and a pseudo-diffusive transport process was assumed. The weight changes during the brining process were calculated according to Eq. (5) [6,3,43].

$$\Delta M_i = 1 + k_1 + k_2 \times t^{0.5}$$

where, $\Delta M_i$ includes total weight changes ($\Delta M_i^0$), water weight changes ($\Delta M_i^w$) and salt weight changes ($\Delta M_i^{NaCl}$) as a function of the square root of time during the brining process. The independent term ($k_1$) is to describe the sample status before brining, which is mainly influenced by the salt concentration, water activity gradient and pressure gradient. The slope term ($k_2$) is related to the mass transfer kinetics of diffusion. The $k_1$ and $k_2$ for each experiment can be obtained by a linear fit of $\Delta M_i^0$ to $t^{0.5}$.
2.5. The salt concentration of the tuna meat liquid phase

The salt content in the aqueous phase ($Z_{\text{NaCl}}$) of tuna sample was calculated by measuring the water content ($X_w$) and sodium chloride content ($X_{\text{NaCl}}$) in the samples. $Z_{\text{NaCl}}$ was calculated from Eq. (6) [10]:

$$Z_{\text{NaCl}} = \left( \frac{X_{\text{NaCl}}}{X_{\text{NaCl}} + X_w} \right) \times 100\% \quad (6)$$

where, $X_w$ and $X_{\text{NaCl}}$ are the moisture content (%) and salt content (%) of tuna meat during brining, respectively.

2.6. Salt equilibrium equation

When the brining process reaches equilibrium, the salt content in the aqueous phase of the muscle ($Z_{\text{NaCl}}^e$) should theoretically be equal to the NaCl content in the brine ($y_{\text{NaCl}}^e$). $Z_{\text{NaCl}}^e$ and $y_{\text{NaCl}}^e$ can be obtained by the mass ratio of tuna meat to brine solution ($M_{\text{SD}}/M_{\text{SS}}$) and the initial weight fractions of water and NaCl in the tuna meat ($X_0^w$ and $X_0^{\text{NaCl}}$, respectively) and in the brine solution ($y_0^w$ and $y_0^{\text{NaCl}}$, respectively). It can be calculated with Eq. (7) according to mass balance in the system [9].

$$Z_{\text{NaCl}}^e = y_{\text{NaCl}}^e = \frac{M_{\text{SD}}^0 X_{\text{NaCl}}^0}{M_{\text{SS}}^0 (X_0^w + X_0^{\text{NaCl}})} + \left( y_0^w + y_0^{\text{NaCl}} \right) \times 100\% \quad (7)$$

Fig. 1. Total weight changes (a), water weight changes (b), and salt weight changes (c) in tuna meat as functions of brining time and NaCl concentration. Static brining group: brining process without ultrasound application; Ultrasound group: brining process with ultrasound application.
2.7. Salt effective diffusion coefficient (De) and other kinetic parameters

The effective diffusion coefficient of NaCl (De) during tuna sample brining can be calculated with the values of $Z^{NaCl}$ (salt weight fraction in the liquid phase of tuna meat) and $y^{NaCl}$ (salt weight fraction in the brine solution) by using the integrated solution of Fick’s equation for semi-infinite flat plate (Eq. (8)) [20]. The semi-infinite slab assumption was applied to simplify the problem to an one-dimensional problem. A constant $De$ was assumed in the brining process since the brine could be considered as a dilute solution [53]. Negligible environmental temperature variation and sample volume, homogeneous initial NaCl and moisture content within the sample and negligible external resistance were also assumed during brining.

\[ 1 - Y_{NaCl}^0 = 1 - \left( \frac{Z^{NaCl} - y^{NaCl}}{Z_0^{NaCl} - Z_0^{NaCl}} \right) = 2e \left( \frac{D_{e,1}}{\pi t^2} \right) + K \]  

where, $Y_{NaCl}^0$ described the driving force of mass transfer between the liquid phase of the tuna sample and the brine solution; $I$ was half of the thickness of the tuna meat block (m); $De$ was the effective diffusion coefficient of NaCl (m²/s); $Z_0^{NaCl}$, $Z^{NaCl}$ and $Z_e^{NaCl}$ denoted the salt content in the liquid phase of the tuna sample at brining time 0 and $t$ and at the equilibrium point, respectively (%); $Y_{NaCl}^0$ denoted the salt content (%) of the brine solution at brining time $t$; The independent term $K$ was influenced by the NaCl concentration and can be used to correct the deviation caused by kinetic mechanisms at the beginning of the brining process.

2.8. Salt distribution

Salt distribution uniformity is an important factor of brining effect evaluation. Samples in static brining were taken out at 1, 5, 10, and 16 h, respectively, while those in ultrasound brining were taken out at 30, 60, 90, and 120 min for salt distribution study. Each prepared sample was equally divided into 9 aliquots and the salt content of each aliquot was determined using a hand-held salinity meter (PAL-ES1, Atago Co., Ltd., Tokyo, Japan). The measurements were repeated three times for each sample.

2.9. Water holding capacity (WHC)

Water holding capacity was determined with a centrifugation method [29]. The tuna samples were taken out at various brining time periods, their surface water was absorbed with filter paper, and samples were then ground and churned manually. Around 5 g of the brined tuna meat was weighed and recorded as $W_1$, then put into a centrifuge tube, centrifuged (Sigma 1-16KL, Sigma Zentrifugen, Saxony, Germany) at 10,621 g for 10 min at 4 °C, and taken out and weighed as $W_2$. The measurement was repeated three times for each time period and the average value was taken. The water holding capacity was calculated with Eq. (9):

\[ WHC(\%) = \frac{W_2 - W_1}{W_1} \times 100(\%) \]  

2.10. Color

The surface color of brined samples was measured using a hand-held colorimeter (Minolta CR-400, Tokyo, Japan). A white standard plate was used for calibration. The probe was pressed vertically on the meat surface to obtain $L^*$ (Lightness), $a^*$ (redness), and $b^*$ (yellowness) values. The measurements were repeated six times for each sample.

2.11. Textural properties

The textural properties of brined tuna samples were determined using a texture analyzer (TA. XT Plus, Stable Micro System Corp, Godalming, UK). The brined tuna samples were cut into $3 \times 2 \times 1$ cm pieces, and then being compressed using a P/50 probe at a fixed moving rate of 1 mm/s until the probe reached 50% of the sample height, held for 5 s and then returned at 1 mm/s. The sample hardness, adhesiveness, springiness, and chewiness values were measured. Experiments were repeated six times.

2.12. Statistical analysis

All experiments were performed in triplicates. Results of salt and moisture content in tuna samples were statistically compared using analysis of variance (ANOVA). Linear regressions were performed for obtaining the kinetic parameters. Statistical analyses were conducted using SPSS software (SPSS, Inc., Chicago, IL, USA). The significance of difference for all statistical analyses was set at $P < 0.05$.

3. Results and discussion

3.1. Calorimetry

Calorimetry is usually employed to determine the efficiency of ultrasound energy transformation. The ultrasonic intensity was calculated as 19.26 W/cm² at ultrasound power of 840 W from Eq. (1). During experiment, part of the ultrasonic energy is converted into heat or reflected back to the transducer, while the other part of the energy can be absorbed by the sample and its surrounding medium [11].

3.2. Mass changes in total, water and salt weight

During brining, each tuna sample could be seen as a combination of tuna muscle, salt, and water. The weight changes of the water and salt in brined tuna and the total weight change of brined tuna sample during static and ultrasound brining at different salt concentration are shown in Fig. 1. Under both static and ultrasound brining, all samples exhibited similar trends of variation in the total weight, water, and salt content at all salt concentration conditions. All of the sample weights increased with brining time, and the magnitude of these significant changes were observed before a brining time of 6 h (static brining) and 1 h (ultrasound). As the brining process reaching the end, the trend of the total weight, water, and salt weight changes of the tuna decreased to reach an equilibrium due to the decrease in the concentration gradient between the samples and the salt solution. The total and water weight changes of tuna were influenced by salt concentration and showed an increasing trend and then decreased with the increase of salt concentration. Among them, the largest change of total and water weight were obtained in 5% NaCl brine, which was related to the highest process yield. These results were consistent with a previous study in that Nguyen et al. [38] found the highest yield of cod muscle sample at 6% salt concentration when immersed in different concentrations of brine (6, 15, 18 and 24% (w/w)). When the brine concentration reached 1 mol/L, the muscle absorbed a large amount of water with mass flow through the intercellular space under capillary force to achieve the maximum water retention capacity [17]. This is because the complex actin-myosin-NaCl interactions generates a complex mass transfer mechanism during muscle brining, The osmosis effect caused muscles either absorb water and swell or lose water and contract [46]. The changes of salt weight of tuna muscle increased with the increase of salt concentration, the smallest change of salt weight were obtained under 2.5% NaCl concentration among all. The salinity of tuna muscle reached 2.0% at the end of the brining process when the salt solution concentration was 5%, which meets the salinity requirement of lightly brined tuna product (~2%) [4,5]. At the same treatment time, the total, water, and NaCl weight changes of the samples subjected to ultrasound brining were higher than those in static brining treatment at all NaCl concentration levels. Similarly, Ozuna et al. [41] and Gómez-Salazar et al. [24] reported that the changes of the total, water and salt weight in meat products with ultrasound treatment was higher than that of samples without ultrasound.
treatment. The effect of ultrasound application on mass transfer can be explained by the cavitation and sponge effects. Ultrasound produces a cavitation effect, where asymmetric cavitation bubble bursts on the surface of the muscle and form micro-jets into the meat to allow NaCl entering [32]. Meanwhile, the sponge effect caused the air trapped in the intercellular space of tuna muscle to be expelled and the salt solution could permeate with a lower resistance [48]. For comparison, at a brine solution concentration of 5%, 1 h of ultrasound treatment and 2 h of static brining treatment result in the same salinity in the tuna sample. Therefore, ultrasound-assisted brining significantly reduced the brining time to 1/2 for tuna brining.

3.3. Mass transfer kinetics

To determine the influence of brining conditions and NaCl concentration on the mass transfer rate, the experimental transport kinetic model was developed assuming a pseudo-diffusional transportation and considering the weight changes were related to the square root of time ($t^{0.5}$). As shown in Fig. 2 and Table 1, the fitting correction factor ($R^2$) of the experimental kinetic model achieved percentages of explained variance for total, water, and NaCl weight changes, ranging from 82.3 to 96.1% (static brining) and from 93.3 to 98.8% (ultrasound), respectively. These results suggested that the weight of total, water, and salt changes of tuna followed first-order kinetics at different brining conditions. In terms of the total weight changes ($\Delta M^o$), the slope term ($k_3$) was
related to the kinetics of the diffusion mechanisms and therefore it is related to the product yield [3]. As shown in Table 1, regarding the total weight changes, the $k_2$ values increased and then decreased with the increase of NaCl concentration. The differences in $k_2$ values indicated that salt concentration affected the diffusion mass transfer kinetics, and therefore the changes in total weight were unequal. With regard to $k_1$ values, the same trend as for $k_2$ values was observed. In general, the independent term ($k_1$) could describe what happens at the beginning of the brining process, and it is influenced by hydrodynamic mechanisms (HDM) and pressure gradients [33]. The observed results for $k_1$ and $k_2$ values of total weight changes were similar to other reports [21,54].

In terms of water weight changes ($\Delta M^w$) and salt weight changes ($\Delta M^{\text{NaCl}}$), the values of $k_2$ were related to the effective diffusion rate of water and sodium chloride. When brine concentration increased, the $k_1$ and $k_2$ values for $\Delta M^w$ showed a trend firstly increasing and then decreasing. On the contrary, the $k_2$ values of $\Delta M^{\text{NaCl}}$ gradually increased with increasing NaCl concentration. Similar results have been reported on the $k_2$ values of cod loins and Chinese cabbage during brining [38,55]. This is because NaCl mass transfer was driven by the concentration gradient between the muscle and brine. Within the muscle, and the driving force was stronger in higher concentrations compared to lower concentrations, so the $k_2$ value of $\Delta M^{\text{NaCl}}$ was higher in higher concentrations of brine [38]. With ultrasound assistance, the $k_1$ values of the total weight, water weight and NaCl weight changes were lower and the $k_2$ values were higher than that of static brining. Zhao et al. [56] reported the effects of different ultrasound intensities on mass transfer kinetics of brining processes, the results showed that the $k_1$ values of the ultrasound-treated samples were lower than that in the static brining group, and the $k_2$ values were higher. As the ultrasonic intensity increased, the $k_1$ values decreased and the $k_2$ values gradually increased. This is because the cavitation effect forms microjets impinging on the muscle and accelerates the salt diffusion.

| NaCl concentration (%) | Static brining | Ultrasound brining |
|------------------------|---------------|--------------------|
|                        | $\Delta M^w$  | $\Delta M^{\text{NaCl}}$ |
|                        | $k_1$ | $k_2$ | $R^2$ | Adjusted time (h) | $k_1$ | $k_2$ | $R^2$ | Adjusted time (h) |
| 2.5                    | 2.55 | 6.79 | 0.9184 | 16 | -1.32 | 16.93 | 0.9627 | 2 |
| 5                      | 5.02 | 10.57 | 0.9467 | 16 | -0.79 | 25.43 | 0.9637 | 2 |
| 7.5                    | 4.66 | 9.26 | 0.9295 | 16 | -0.92 | 23.04 | 0.9719 | 2 |
| 10                     | 4.43 | 8.63 | 0.9013 | 16 | -1.44 | 20.10 | 0.9860 | 2 |

Fig. 3. Reduced driving force ($1 - Y^{\text{NaCl}}$) versus $t^{0.5}$ in tuna at different NaCl concentrations with or without ultrasound assistance. Static brining group (a): brining without ultrasound; Ultrasound group (b): brining with ultrasound.

Table 1
| NaCl concentration (%) | Static brining | Ultrasound brining |
|------------------------|---------------|--------------------|
|                        | $\Delta M^w$  | $\Delta M^{\text{NaCl}}$ |
|                        | $k_1$ | $k_2$ | $R^2$ | Adjusted time (h) | $k_1$ | $k_2$ | $R^2$ | Adjusted time (h) |
| 2.5                    | 2.55 | 6.79 | 0.9184 | 16 | -1.32 | 16.93 | 0.9627 | 2 |
| 5                      | 5.02 | 10.57 | 0.9467 | 16 | -0.79 | 25.43 | 0.9637 | 2 |
| 7.5                    | 4.66 | 9.26 | 0.9295 | 16 | -0.92 | 23.04 | 0.9719 | 2 |
| 10                     | 4.43 | 8.63 | 0.9013 | 16 | -1.44 | 20.10 | 0.9860 | 2 |

Table 2
| NaCl concentration (%) | Static brining | Ultrasound brining |
|------------------------|---------------|--------------------|
|                        | $\Delta M^w$  | $\Delta M^{\text{NaCl}}$ |
|                        | $k_1$ | $k_2$ | $R^2$ | Adjusted time (h) | $k_1$ | $k_2$ | $R^2$ | Adjusted time (h) |
| 2.5                    | 1.09 | 0.12 | 0.9706 | 8.21 | 0.01 | 0.9332 | 653.21 |
| 5                      | 2.17 | 0.15 | 0.9024 | 12.39 | 0.12 | 0.9521 | 470.97 |
| 7.5                    | 1.88 | 0.04 | 0.9749 | 9.44 | 0.11 | 0.9263 | 402.13 |
| 10                     | 1.76 | 0.09 | 0.9572 | 8.87 | 0.11 | 0.9680 | 403.98 |
Fig. 4. Salt distribution in tuna during brining as functions of brine concentration and time. (a): static brining (b): ultrasound assisted brining.
| Ultrasound brining | NaCl concentration (%) |
|-------------------|------------------------|
| 2.5               | 5                      | 7.5                | 10                   |
| 30 min            | 1.40 1.05 1.35         | 1.95 1.85 2.05     | 3.10 2.30 2.95       | 4.40 3.70 3.95 |
|                   | 0.95 0.85 1.00         | 1.55 1.10 1.85     | 2.10 1.95 2.60       | 3.50 2.35 3.20 |
|                   | 1.30 1.05 1.45         | 2.00 1.95 2.00     | 3.35 2.85 3.40       | 4.40 3.85 4.30 |
|                   | 1.16 ± 0.21            | 1.81 ± 0.29        | 2.73 ± 0.50          | 3.74 ± 0.63 |
| 60 min            | 1.55 1.20 1.45         | 2.20 2.00 2.10     | 4.10 3.35 3.90       | 5.45 5.00 5.25 |
|                   | 1.10 1.00 1.10         | 1.95 1.35 1.90     | 3.40 3.05 3.70       | 4.35 3.50 4.45 |
|                   | 1.40 1.20 1.50         | 2.20 1.90 2.20     | 3.95 4.05 4.00       | 5.25 4.90 5.25 |
|                   | 1.28 ± 0.19            | 1.98 ± 0.25        | 3.72 ± 0.35          | 4.82 ± 0.59 |
| 90 min            | 1.55 1.20 1.40         | 2.25 2.25 2.25     | 4.10 3.85 4.20       | 5.55 5.05 5.35 |
|                   | 1.10 1.00 1.15         | 2.30 1.55 2.00     | 4.30 3.25 3.60       | 4.50 3.55 4.60 |
|                   | 1.40 1.30 1.60         | 2.20 2.05 2.25     | 4.10 3.95 4.00       | 5.35 5.05 5.35 |
|                   | 1.30 ± 0.19            | 2.12 ± 0.22        | 3.95 ± 0.31          | 4.93 ± 0.49 |
| 120 min           | 1.60 1.20 1.50         | 2.35 2.20 2.30     | 4.05 3.80 4.30       | 5.65 5.15 5.40 |
|                   | 1.15 1.05 1.25         | 2.25 1.65 2.05     | 4.10 3.35 3.80       | 4.9 3.70 4.85  |
|                   | 1.40 1.35 1.65         | 2.35 2.05 2.25     | 4.20 4.15 4.30       | 4.75 5.20 5.35 |
|                   | 1.35 ± 0.19            | 2.16 ± 0.21        | 4.01 ± 0.29          | 4.99 ± 0.44 |

Fig. 4. (continued).
3.4. Salt diffusion coefficients (De) and other kinetic parameters

When the system reached equilibrium, the salt concentration in the liquid phase of the tuna sample was equal to the concentration of the brine solution. The equilibrium salt content of brined tuna in brines with four NaCl concentrations (2.5, 5, 7.5 and 10% w/w NaCl) could be derived from the above mentioned Eq. (7) as 2.2%, 4.2%, 6.2% and 8.2%, respectively. The effective diffusion coefficient (De) of salt with different NaCl concentrations were determined by using the $Z_{\text{NaCl}}$ (NaCl concentration in the aqueous phase of tuna sample) and $Y_{\text{NaCl}}$ (NaCl concentration in the brine solution) values according to Fick’s second law equation for a semi-infinite slab and short time (Fig. 3). The independent term (K), in Eq. (8), is a time-independent constant to adjust the deviation from the origin of the coordinates. The differences in De values at different NaCl concentrations might be due to differences in component activity gradients and osmosis pressure gradients [8]. As shown in Table 2, the highest De value of $2.17 \times 10^{-10}$ m$^2$/s was observed in 5% NaCl (w/w) solution among all salt concentrations in the static brining group. Moreover, the highest K value was also found in the 5% brine solution, indicating an initial weight increase due to the fast initial water gain based on the hydrodynamic mechanism [7]. As shown in previous studies, ultrasound accelerated the diffusion rate of salt in meat muscles [35]. Therefore, ultrasound application significantly enhanced the De values of tuna at different salt concentrations, especially at the lowest salt concentration (2.5% w/w NaCl). For instance, for the 5% NaCl brine solution, the De value in the static brining group was $2.17 \times 10^{-10}$ m$^2$/s while that in ultrasound assisted brined samples reached $12.39 \times 10^{-10}$ m$^2$/s. Visy et al. [53] reported an increase of 1011.1% in the total solid diffusion coefficients in brined pork meat with ultrasound. While a lower increase in De ranging from 147.1 to 812.2% was observed in Chinese cabbage brined in solutions with different salt concentrations [55]. This is because ultrasound waves generated bubbles inside the liquid, which formed asymmetric implosions near the solid surface and produced microjet in the direction of the solid surface. These microjets of NaCl solution entered the tuna samples and accelerated the diffusion rate [14], which can explain the significant increase in De values in the tuna samples. Thus, the acceleration of brining kinetics was caused by a series of effects.

3.6. Water holding capacity (WHC)

Water holding capacity (WHC) reflects the ability of fish muscle to retain water. The higher the WHC, the stronger the WHC of fish protein and the better the quality of meat. The WHC changes of brined tuna sample during static and ultrasound brining at different salt concentration are shown in Fig. 5. As can be seen from Fig. 5a, the WHC of tuna blocks in the static brining group shows an increasing trend in the early stage of brining, and the highest WHC of tuna blocks was observed when the salt concentration was 5%. Afterward, the WHC began to decrease with the increase of brining time, and the longer the brining time, the WHC decreases, and proteins denature, leading to charge changes and disruption of hydrogen bonds, resulting in a decrease in WHC. As can be seen from Fig. 5b, the WHC of tuna block in the ultrasound brining group shows an increasing trend during brining process and the WHC was highest at 5% salt concentration. Under the same brining concentration and period, the WHC of samples in each treatment group gradually increased. This proves that ultrasound application significantly enhanced the WHC of the samples at each treatment group gradually increased. This proves that ultrasound application significantly enhanced the WHC of the samples at each treatment group. Besides, ultrasound also induces oxidation of myosin and protein-water interactions during ultrasound treatment [2]. Besides, ultrasound also induces oxidation of myosin and protein-water interactions during ultrasound treatment [2]. The Ultrasound group: brining process with ultrasound application.

3.7. Color

The color changes of brined tuna sample during static and ultrasound brining at different salt concentration are shown in Fig. 6. It can be seen...
that the $L^*$ value gradually increased with the increase of brining time, and the $L^*$ value of 2.5% salt concentration was the highest, which may be related to its minimal water holding capacity. Bras and Costa [13] found that the change of $L^*$ value was partly due to the loss of water, and the decrease of surface moisture of fish led to the decrease of light scattering degree, which in turn led to the decrease of fish surface transparency, so the $L^*$ value increased. The $a^*$ value decreases gradually during the brining process. Myoglobin is the main compound that contributes the red color to meat. When meat is soaked in aqueous solution, myoglobin, as a water-soluble compound, will be diluted, so the $a^*$ value of the sample decreased after brining [31]. The $b^*$ value increased gradually during the brining process, and the lower the salt concentration, the higher the $b^*$ value, because the enzymatic activity is stronger at low salt concentration, which promotes high lipid oxidation. In addition, the $L^*$ and $b^*$ values increased, the $a^*$ values decreased after ultrasound brining compared to static brining, but there was no significant effect. Carrillo-Lopez et al. [16] studied the effects of ultrasound intensity (16, 28, 90 W/cm$^2$) on the physicochemical properties and

Fig. 6. Changes in color in tuna during brining as functions of brine concentration and time. (Static brining group: brining without ultrasound; Ultrasound group: brining with ultrasound application).
Fig. 7. Changes of textural properties in tuna during brining as functions of brine concentration and time. (Static brining group: brining without ultrasound; Ultrasound group: brining with ultrasound application).
microstructure of bovine Longissimus dorsi, and found that ultrasound treatment had no effect on whiteness, redness and yellowness. Contreras-Lopez et al. [18] evaluated the effects of high intensity ultrasound (37 kHz, 22 W/cm²) on the physicochemical and sensory properties of longissimus muscle of cured porcine M. longissimus lumbarum, and found that ultrasound brining had no significant effect on the L* value of meat. It may be because the ultrasound power used in this study is not significant enough to induce pigment (myoglobin) denaturation and oxidation. Sikes et al. [47] and Stadnik and Dolatowski [49] also found similar results.

3.8. Textural properties

Texture is an important index to evaluate the quality of aquatic products. The effects of salt concentration and ultrasound application on the tissue structural characteristics of tuna are shown in Fig. 7. As the brining time increased, the hardness and chewiness of tuna samples in both static brining and ultrasound group decreased significantly, while the adhesiveness and springiness increased. This might have been caused by the diffusion of salt and the loss of water during brining which have an impact on the secondary structure and quantity of actomyosin. Microorganisms and enzymes make the tissue structure of fish lose and produce a large number of sticky polymers [27]. As can be seen from Fig. 7, when the salt concentration reached 2.5%, the hardness and chewiness of the sample were the highest, because the water content of brined fish with 2.5% concentration was the lowest. Toyohara [51] found that brining caused a large amount of sarcoplasmic protein precipitation in fish meat, which led to texture change, and decrease of water content in fish meat after brining also led to an increase in hardness. In addition, compared with the static brining group, the application of ultrasound significantly lowered the hardness and chewiness of tuna and enhanced the adhesiveness and springiness, which was due to the fact that the ultrasound destroyed the tissue structure of muscle and produced micro-channels, which reduced its hardness and improved its adhesiveness [26].

4. Conclusion

In this study, the effects of tuna muscle brining in brines with different NaCl concentrations (2.5, 5, 7.5 and 10%) with and without ultrasound on the brining kinetics were evaluated. Results showed that NaCl concentration and ultrasound application both had significant effects on mass transfer and kinetic parameters during the tuna brining process. With the increase of salt concentration, the changes of total weight and water weight showed a trend of increasing and then decreasing while the changes of salt weight increased continuously. At the end of the brining process, the sample treated in 5% salt concentration brine had a moderate NaCl content, the highest moisture content, and the highest yield. The maximum effective diffusion coefficient (De) was obtained for 5% salt concentration in both of the static and the ultrasound brining group samples. Meanwhile, ultrasound greatly enhanced the De value in all treatments, with a more pronounced effect at low concentration conditions. Compared with traditional brining, ultrasound assistance shortened the brining time and produced a more uniform NaCl distribution, thus showed positive effects on brining processes. In addition, the hardness and chewiness of the samples decreased, while the adhesiveness and springiness increased after ultrasound assisted brining. Therefore, ultrasound is an effective method to accelerate the brining process of muscle food products. Future studies would be needed on enlarging the processing scale and further improving the brining uniformity. This study provides theoretical support for the application of ultrasound technology to improve brined muscle food production rate and quality.

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.

Acknowledgement

The authors acknowledge China National Science Foundation (31801613) for its financial support to this research.

References

[1] M. Alino, R. Grau, A. Fernández-Sánchez, A. Arnold, J. Barat, Influence of brine concentration on swelling pressure of pork meat throughout salting, Meat science 86 (2010) 600–606.
[2] A. Amiri, P. Shariﬁan, N. Soltanzadeh, Application of ultrasound treatment for improving the physicochemical, functional and rheological properties of myofibrillar proteins, International Journal of Biological Macromolecules 111 (2018) 139–147.
[3] A. Andres, S. Rodriguez-Barana, J.M. Barat, P. Fito, Note: Mass Transfer Kinetics During Cod Salting Operation, Food Science and Technology International 8 (2002) 309–314.
[4] E. Aykin-Dincer, M. Erbas, Drying kinetics, adsorption isotherms and quality characteristics of vacuum-dried beef slices with different salt contents, Meat Science 145 (2018) 114–120.
[5] E. Aykin-Dincer, M. Erbas, Quality characteristics of cold-dried beef slices, Meat Science 155 (2019) 36–43.
[6] J.M. Barat, A. Chiralt, P. Fito, Effect of Osmotic Solution Concentration, Temperature and Vacuum Impregnation Pretreatment on Osmotic Dehydration Kinetics of Apple Slices, Food Science and Technology International 7 (2001) 451–456.
[7] J.M. Barat, L. Gallart-Jornet, A. Andres, L. Alke, M. Carleborg, O.T. Skjerdal, Influence of cod freshness on the salting, drying and desalting stages, Journal of Food Engineering 73 (2006) 9–19.
[8] J.M. Barat, S. Rodriguez-Barana, A. Andres & P. Fito, Cod salting manufacturing analysis, Food Research International 36 (2003) 447–453.
[9] J.M. Barat, S. Rodriguez-Barana, A. Andres, J.B. Ibanez, Modeling of the Cod Desalting Operation, Journal of Food Science 69 (2004) 183–189.
[10] J.M. Barat, S. Rodriguez-Zurilloana, A. Andres, P. Fito, Influence of Increasing Brine Concentration in the Cod-Salting Process, Journal of Food Science 67 (2010) 1922–1925.
[11] J. Belian, T.J. Mason, Sonochemistry: from research laboratories to industrial plants, Ultrasonics 30 (1992) 203–212.
[12] N. Boudrioua, N. Djendoubi, S. Bellagha, N. Kechaou, Study of moisture and salt transfers during salting of sardine fillets, Journal of Food Engineering 94 (2009) 85–89.
[13] A. Brais, R. Costa, Influence of brine salting prior to pickle salting in the manufacturing of various salted-dried fish species, Journal of Food Engineering 100 (2010) 490–495.
[14] J.A. Carcel, J. Benedito, J. Bon, A. Mulet, High intensity ultrasound effects on meat brining, Meat Science 76 (2007) 611–619.
[15] J.A. Carcel, J. Benedito, C. Roselló, A. Mulet, Influence of ultrasound intensity on mass transfer in apple immersed in a sucrrose solution, Journal of Food Engineering 78 (2007) 472–475.
[16] L.M. Carrillo-Lopez, M. Huerria-Jimenez, I.A. Garcia-Galicica, A.D. Alarcon-Rojo, Bacterial control and structural and physicochemical modification of bovine Longissimus dorsi by ultrasound, Ultrasonics Sonochemistry 58 (2019), 106608.
[17] Q. Cheng, D.W. Sun, Factors affecting the water holding capacity of red meat products: a review of recent research advances, Critical Reviews in Food Science & Nutrition 48 (2008) 137–159.
[18] G. Contreras-Lopez, A. Camero-Hernandez, M. Huerria-Jimenez, A.D. Alarcon-Rojo, I. Garcia-Galicica, L.M. Carrillo-Lopez, High-intensity ultrasound applied on cured pork: Sensory and physicochemical characteristics, Food Science & Nutrition 8 (2020) 786–795.
[19] C. Corzo, N. Barcelo, J. Rodriguez, Modeling mass transfer during salting of catfish sheets, Journal of Aquatic Food Product Technology 24 (2015) 120–130.
[20] J. Crank, The mathematics of diffusion, WSEAS Transactions on Systems and Control 8 (1975) 625–626.
[21] L. Du, G. Zhou, X. Xu, C. Li, Study on kinetics of mass transfer in water-boiled salted duck during wet-curing, Journal of Food Engineering 100 (2010) 578–584.
[22] L. Gallart-Jornet, J. Barat, T. Rustad, U. Erikson, I. Escriche, P. Fito, Influence of brine concentration on Atlantic salmon fillet salting, Journal of Food Engineering 80 (2007) 267–275.
[23] L. Gallart-Jornet, J.M. Barat, T. Rustad, U. Erikson, I. Escriche, P. Fito, A comparative study of brine salting of Atlantic cod (Gadus morhua) and Atlantic salmon (Salmo salar), Journal of Food Engineering 79 (2007) 261–270.
[24] J.A. Gómez-Salazar, D.A. Ochoa-Montes, A. Cerón-García, C. Orzna, M.E. Sosa-Morales, Effect of Acid Marination Assisted by Power Ultrasound on the Quality of Rabbit Meat, Journal of Food Quality 2018 (2018) 1–6.
[25] L. González-González, L. Luna-Rodríguez, L.M. Carrillo-López, A.D. Alarcon-Rojo, I. García-Galicica, R. Reyes-Villagrana, Ultrasound as an Alternative to
Conventional Marination: Acceptability and Mass Transfer, Journal of Food Quality 2017 (2017) 1–8.

[26] L. Grill, F. Ringdorfer, B. Baumung, B. Fuerst-Wald, Evaluation of ultrasound scanning to predict carcass composition of Austrian meat sheep, Small Ruminant Research 123 (2015) 260–268.

[27] C.C. Hwang, C.M. Lin, H.F. Kung, Y.L. Huang, D.F. Hwang, Y.C. Su, Y.H. Tsai, Effect of salt concentrations and drying methods on the quality and formation of histamine in dried milkfish (Chanos chanos), Food Chemistry 135 (2012) 839–844.

[28] S.D. Jayasasiriya, P.J. Torley, B.R. D’Arcy, B.R. Bhandari, Effect of high power ultrasound and ageing on the physical properties of bovine Semitendinosus and Longissimus muscles, Meat Science 75 (2007) 628–639.

[29] Q. Jiang, R. Jia, N. Nakazawa, Y. Hu, K. Osako, E. Okazaki, Changes in protein properties and tissue histology of tuna meat as affected by salting and subsequent freezing, Food Chemistry 271 (2019) 550–560.

[30] D.-C. Kang, A.-R. Wang, G.-H. Zhou, W.-G. Zhang, S.-M. Xu, G.-P. Guo, Power ultrasonic on mass transport of beef: Effects of ultrasound intensity and NaCl concentration, Innovative Food Science & Emerging Technologies 35 (2016) 36–44.

[31] M.M. Ladeira, L.C. Santarosa, M.L. Chizzotti, E.M. Ramos, O.R. Machado Neto, D. M. Oliveira, J.R. Carvalho, L.S. Lopes, J.S. Ribeiro, Fatty acid profile, color and lipid oxidation of meat from young bulls fed ground soybean or rumen protected fat with or without monensin, Meat Science 96 (2014) 597–605.

[32] M.Y. Leal-Ramos, A.D. Alarcon-Rojo, T.J. Mason, L. Panivnyk, M. Alarjah, Ultrasound-enhanced mass transfer in Halal compared with non-Halal chicken, Journal of the Science of Food and Agriculture 91 (2011) 130–133.

[33] X. Leng, L. Zhang, M. Huang, X. Xu, G. Zhou, Mass transfer during high pressure brining of chicken breast, Journal of Food Engineering 118 (2013) 296–301.

[34] T. Mason, J. Lorimer, Applied Sonochemistry: Uses of Power Ultrasound in Chemistry, (2002).

[35] C.K. McDonnell, P. Allen, G. Duane, C. Morin, E. Casey, J.G. Lyng, One-directional modelling to assess the mechanistic actions of power ultrasound on NaCl diffusion in pork, Ultrasonics Sonochemistry 40 (2018) 206–212.

[36] C.K. McDonnell, J.G. Lyng, P. Allen, The use of power ultrasound for accelerating the curing of pork, Meat Science 98 (2014) 142–149.

[37] H.S. Muralidhara, D. Esminger, A. Putnam, Acoustic dewatering and drying (low and high frequency): State of the art review, Drying Technology 3 (1985) 529–566.

[38] M.Y. Nguyen, S. Arzon, K.A. Thorarinsdottir, G. Thorkelson, A. Gudmundsdottir, Influence of salt concentration on the salting kinetics of cod loin (Gadus morhua) during brine salting, Journal of Food Engineering 100 (2010) 225–231.

[39] G. Offer, J. Trinick, On the mechanism of water holding in meat: the swelling and shrinking of myofibrils, Meat science 8 (1983) 245–281.

[40] K.S. Ojha, D.F. Keenan, A. Bright, J.P. Kerry, B.K. Tiwari, Ultrasound assisted diffusion of sodium salt replacer and effect on physicochemical properties of pork meat, International Journal of Food Science & Technology 51 (2016) 37–45.

[41] C. Ozuna, A. Puig, J.V. Garcia-Perez, A. Mulet, J.A. Carcel, Influence of high intensity ultrasound application on mass transport, microstructure and textural properties of pork meat (Longissimus dorsi) brined at different NaCl concentrations, Journal of Food Engineering 119 (2013) 84–93.

[42] E. Pena-Gonzalez, A.D. Alarcon-Rojo, I. Garcia-Galicia, L. Carrillo-Lopez, M. Huerta-Jimenez, Ultrasound as a potential process to tenderize beef: Sensory and technological parameters, Ultrasonics Sonochemistry 53 (2019) 134–141.

[43] N.A. Peppas, L. Brannon-Peppas, Water diffusion and sorption in amorphous macromolecular systems and foods, Journal of Food Engineering 22 (1994) 189–210.

[44] J. Raso, P. Maas, R. Pagan, J.F. Sala, Influence of different factors on the output power transferred into medium by ultrasound, Ultrasonics Sonochemistry 5 (1999) 157.

[45] M. Rasunnen, E. Puolanne, Reducing sodium intake from meat products, Meat Science 70 (2005) 531–541.

[46] F.C. Schmidt, B.A.M. Carciofi, J.B. Laurindo, Application of diffusive and empirical models to hydration, dehydration and salt gain during osmotic treatment of chicken breast cuts, Journal of Food Engineering 91 (2009) 553–559.

[47] A.L. Sikes, R. Mawson, J. Stark, R. Warner, Quality properties of pre- and post-rigor beef muscle after interventions with high frequency ultrasound, Ultrasonics Sonochemistry 21 (2014) 2138–2143.

[48] I. Sir, C. Ven, C. Balla, G. Jonás, I. Zeke, L. Friedrich, Application of an ultrasonic assisted curing technique for improving the diffusion of sodium chloride in porcine meat, Journal of Food Engineering 91 (2009) 353–362.

[49] J. Stadmä, Z.J. Dolatowski, Influence of sonication on Warner-Batzlizer shear force, colour and myoglobin of beef (m. semimembranosus), European Food Research and Technology 233 (2011) 553–559.

[50] V.R.N. Telis, P.F. Romanelli, A.L. Gabas, J. Telis-Romero, Salting kinetics and salt diffusivities in farmed Pantanal caiman muscle, Pesquisa Agropecuaria Brasileira 38 (2003) 529–535.

[51] Toyohara, Texture Changes Associated with Insolubilization of Sarcolemma Proteins During Salt-vinegar Curing of Fish, Journal of Food Science 64 (1999).

[52] C. Vanseegaard, J. Risum, J. Adler-Nissen, 23Na-MRI quantification of sodium and water mobility in pork during brining cure, Meat Science 69 (2005) 663–672.

[53] A. Visy, G. Jonas, D. Szalos, Z. Horvath-Mezofz, K.I. Hadzs, A. Barko, L. Friedrich, Evaluation of ultrasound and microbubbles effect on pork meat during brining process, Ultrasonics Sonochemistry 75 (2021), 105589.

[54] Z. Wang, Z. He, H. Li, Mass transfer dynamics during brining of rabbit meat, World Rabbit Science 25 (2017).

[55] C.C. Zhao, J.B. Eun, Influence of ultrasound application and NaCl concentrations on brining kinetics and textural properties of Chinese cabbage, Ultrasonics Sonochemistry 49 (2018) 137–144.

[56] X. Zhao, Y. Sun, Y. Zhou, Y. Leng, Effect of ultrasonic-assisted brining on mass transfer of beef, Journal of Food Process Engineering 42 (2019), e13257.