Effect of acid and alkaline solubilization on the properties of surimi based film

Thummanoon Prodpran¹ and Soottawat Benjakul²

Abstract

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The effect of acid and alkaline solubilizing processes on the properties of the protein based film from threadfin bream surimi was investigated. Surimi films prepared from both processes had the similar light transmission, tensile strength (TS) and elongation at break (EAB) (P<0.05). However, film with alkaline process had slightly lower water vapor permeability (WVP), compared to that prepared by acid solubilizing process. The protein concentration in the film-forming solution directly affected the properties of the film. Increase in protein concentration resulted in an increase in TS, EAB as well as WVP. The film prepared by acid solubilizing process had an increase in yellowish color as evidenced by the continuous increase in b* and E* values during the storage at room temperature. The acid and alkali solubilizing processes caused the degradation of muscle protein in surimi, especially with increasing exposure time. Therefore, solubilizing process had the influence on the properties of the protein film from threadfin bream surimi.

Key words : surimi, film, threadfin bream, acid, alkali, solubilization

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Acid and alkaline solubilization on properties of surimi based film

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Surimi is stabilized myofibrillar proteins prepared from fish mince that is washed with water and blended with cryoprotectant (Park and Morrissey, 2000). Surimi possesses the functionality, especially gelling property, which make it useful as a food base in seafood analogue. Apart from gelation, film formation is another functionality of fish protein, which has been paid increasing attention (Shiku et al., 2003; Cuq et al., 1995; Paschoalick et al., 2003). Edible/biodegradable films from fish myofibrillar protein have been successfully produced (Shiku et al., 2003). The formation of edible packaging films from sardine meats and their properties was also investigated (Cuq et al., 1995). Proteins are thermoplastic heteropolymers containing both polar and non-polar amino acids, which are able to form numerous intermolecular linkages. Generally, globular proteins must be denatured by heat, acid, base and/or solvent to form more extended structures that are required for film formation (Krochta, 1997). Procedure of film solubilization affected the film formation and its properties. Shiku et al. (2003) reported that pH was shown to influence the mechanical and physical properties of myofibrillar protein film. Tensile strength of films was greater when prepared at the very acidic and alkaline conditions, compared with the neutral pH range. However, pH of film forming solution had no effect on water vapor permeability, light transmission, film solubility and enzymatic hydrolysis (Shiku et al., 2003). Recently, transparent and flexible edible/biodegradable films were made from frozen Alaska pollack surimi (Shiku et al., 2004). Due to the high content of hydrophilic components, protein film generally is a poor barrier to moisture (Kim and Ustunol, 2001). Additionally, thickness of hydrophobic film was associated with the increased water vapor permeability (McHugh et al., 1993).

Thailand is one of the largest surimi producers in Southeast Asia. Most of fish used for surimi production include threadfin bream (Nemipterus spp.), bigeye snapper (Priacanthus spp.), croaker (Pennisia and Johnius spp.) and lizardfish (Saurida spp.) (Benjakul et al., 2003). Apart from gelation, the appropriate development of protein film from surimi produced from tropical fish should be an alternative promising means to obtain the nutritional and biodegradable film.
Though the effect of pH on film properties of fish myofibrillar protein has been reported, no information concerning the different pHs and thickness on the properties of film produced from frozen surimi containing cryoprotectants has been reported. Therefore, the objective of this study was to study the effect of acidic and alkaline solubilization as well as thickness on properties of film produced from frozen surimi from threadfin bream.

Materials and Methods

1. Frozen surimi

Frozen surimi (grade A), produced from threadfin bream (*Nemipterus bleekeri*) was purchased from Man A Frozen Foods Co., Ltd., Muang, Songkhla. Surimi was kept at -20ºC until used.

2. Proximate analysis

Surimi was determined for moisture, ash, fat, protein and carbohydrate according to the method of AOAC (1999).

3. Preparation of film-forming solution

Frozen surimi was thawed using a running water (26-28ºC) until the core temperature reached 0ºC. The film-forming solution was prepared as described by Shiku *et al.* (2003) with a slight modification. The surimi was added with the distilled water and homogenized for 1 min at a speed of 3 using a homogenizer (IKA Labortechnik, Malaysia). The protein concentration of the film-forming solution was fixed at 1 and 2% (w/v) and glycerol was added at 50% (w/w) of protein. The mixtures were stirred gently for 30 min at room temperature. Subsequently, the pH of the film-forming solution was adjusted to 3 and 11 using 1 M HCl and 1 M NaOH, respectively. The solution was subjected to centrifugation at 3,000xg for 5 min at room temperature. The supernatant was transferred carefully using a transfer pipette. The solution was used for film casting.

4. Film casting and drying

The film-forming solution (4 g) was cast onto a rimmed silicone resin plate (50x50 mm) and dried overnight using an electric fan prior to drying in a ventilated oven at 25ºC and 50% relative humidity (RH) for 48h in an environmental chamber (WTB Binder, Tuttingen, Germany). The resulting films were manually peeled off and used for analyses.

5. Determination of film properties

5.1 Film thickness

Film thickness was measured using a micrometer (Gotech, Model GT-313-A, Gotech testing machines Inc, Taiwan). Five random positions of each film of ten films were used for thickness determination.

5.2 Light transmission and film transparency

The transmission of films was measured at the ultraviolet and visible range (200-800 nm) using the UV-1601 spectrophotometer (Shimadzu, Kyoto, Japan) as described by Shiku *et al.* (2004).

5.3 Color

Color of the film was determined as L*, a* and b* using CIE colorimeter (Hunter associates laboratory, Inc., VA, USA). E* was calculated as follows: (Paschoalick *et al.*, 2003)

\[ \Delta E^* = \sqrt{(\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2} \]

The films were applied on the surface of a white standard plate and the color parameters were measured.

5.4 Mechanical properties

The films were conditioned for 48h at 25ºC and 50%RH prior to testing. Tensile strength (TS) and elongation at break (EAB) were determined using the Universal Testing Machine (Lloyd Instruments, Hampshire, UK). Ten samples (2x5 cm) with initial grip length of 3 cm were used for testing. Cross-head speed was 0.5 mm/s.

5.5 Water vapor permeability (WVP)

WVP of films was determined using a modified ASTM method (American Society for Testing & Materials, 1989). The film was sealed on a glass permeation cup containing silica gel (0%RH) with silicone vacuum grease and a rubber
band. The cups were placed at 30°C in a desiccator containing the distilled water. The cups were weighed at 1 h intervals over an 8 h period. WVP of the films was calculated as follows (McHugh et al., 1993):

\[ WVP = \frac{wxA^i}{t^i(P_2 - P_1)^i} \]

where \( w \) is the weight gain of the cup (g), \( x \) is the film thickness (m), \( A \) is the area of exposed film (m\(^2\)), \( t \) is the time of gain (s) and \((P_2 - P_1)\) is the vapor pressure differential across the film (Pa). Four films were used for WVP testing and the measurement was run in duplicate.

5.6 SDS-polyacrylamide gel electrophoresis (SDS-PAGE)

SDS-PAGE was performed according to the method of Laemmli (1970) using a 10% running gel and 4% stacking gel. To prepare the protein sample, surimi (3 g) was solubilized in 27 ml of 1% SDS (85°C) as described by Benjakul et al. (2001). To solubilize the films, the samples were mixed with a solublizing solution containing 1% SDS and 8M urea. The mixtures were homogenized with a homogenizer for 1 min at a speed of 3 (~16,000 rpm). The homogenate was stirred continuously for 12 h at room temperature, followed by centrifugation at 3,000xg for 15 min. The supernatants obtained were also subjected to SDS-PAGE analysis in presence and absence of \( \beta \)-mercaptoethanol.

5.7 Protein determination

The protein content was determined using the biuret method (Robinson and Hodgen, 1940). Bovine serum albumin was used as the protein standard.

5.8 Scanning electron microscopy

Surface morphology was examined by scanning electron microscope (JEOL JSM-5800 LV, Tokyo, Japan). Film samples were mounted on a bronze stub and sputter-coated with gold (Sputter coater SPI-Module, PA, USA). The surface was observed at an acceleration voltage of 10 kV.

6. Changes in protein pattern of surimi protein under acidic and alkaline conditions

The surimi was solubilized using acid and alkaline at pH 3 and 11, respectively as mentioned previously. The solution was allowed to stand at room temperature and taken for analysis at 0, 2, 5, 7 and 10 h. At the time designated, the solution was neutralized using either 1N NaOH or 1 N HCl. Then the neutralized solution was mixed with 5% SDS at a ratio of 1:2 (v/v). The mixture was incubated at 85°C for 30 min and the undissolved debris was removed by centrifuging at 3,500xg for 20 min. The supernatants were subjected to SDS-PAGE (10% running gel and 4% stacking gel).

7. Changes in color of surimi film during storage

Surimi films prepared either by acid or alkaline solubilizing processes were placed in the polyethylene bag and stored at room temperature (28-30°C). The films were taken for \( b^* \) and \( E^* \) measurement periodically up to 30 days.

8. Statistical analysis

Analysis of variance (ANOVA) was per-

<table>
<thead>
<tr>
<th>Table 1. Proximate compositions of surimi from threadfin bream</th>
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</thead>
<tbody>
<tr>
<td>Composition</td>
</tr>
<tr>
<td>-------------</td>
</tr>
<tr>
<td>Moisture</td>
</tr>
<tr>
<td>Protein</td>
</tr>
<tr>
<td>Carbohydrate</td>
</tr>
<tr>
<td>Fat</td>
</tr>
<tr>
<td>Ash</td>
</tr>
</tbody>
</table>

^a Mean±SD from triplicate determinations.
formed and mean comparisons run by Duncan's multiple range test (Steel and Torrie, 1980). Analysis was performed using the SPSS package (SPSS 8.0 for Windows, SPSS Inc, Chicago, IL).

Results and Discussion

1. Surimi composition

Threadfin bream surimi consisted of 77.19% moisture content. Surimi contained a high protein content (16.05%) with the negligible fat and ash contents. The flesh of fish normally contains 11-24% crude protein, depending on the species of the animal, the type of muscle, etc. (Sikorski et al., 1990). Carbohydrate at a level of 6.15% was found in the surimi. Generally, cryoprotectants are added with surimi to prevent the denaturation of protein during extended frozen storage. The most commonly used cryoprotectant are sucrose and sorbitol, typically added in a blend of 4:4% (MacDonald et al., 2000).

2. Effect of protein concentration and pH on surimi film properties

2.1 Film thickness

Films cast from surimi solution with 2% protein at either pH 3 or 11 had a thickness ranging from 0.035 to 0.037 mm, whereas those with 1% protein possessed the thickness of 0.017-0.019 mm (Table 2). From the result, protein level in film solution was directly associated with the thickness of films. However, no marked differences in thickness were observed between films prepared by acid and alkaline solubilizing process. Solubilization of globular protein was necessary for film formation. Adjustment of pH to very acidic or alkaline pH ranges resulted in the repulsion of protein molecules. At pH values above and below pI, where a protein has a net negative or positive charge more, water interacts with protein charges. Charge repulsion contributes to the greater protein solubility. Unfolding at low or high pH values occurs owing to a decrease in electrostatic bonds (Vojdani, 1996). From the result, acid and alkaline solubilization was effective in solubilizing the muscle protein in surimi prior to film casting.

2.2 Light transmission and color

Light transmission at some selected wavelength of surimi films prepared by acid and alkaline solubilization is shown in Table 2. Films prepared from both processes had the low transmission in UV ranges (200-280 nm). The film prepared from 2% protein casting solution, which had a greater thickness, showed the lower light transmission at 280 nm. Therefore, the thicker film would prevent the UV light more effectively than the thinner film. The result was in accordance with Shiku et al. (2004) who reported that the film from Alaska pollack surimi exhibited the excellent barrier to UV light. For the visible ranges, particularly from 400 to 800 nm, the light transmission of 72.9-87.3% was found. No marked differences between light transmission were observed between films with different solubilizing processes. However, the film made from the higher protein content exhibited the slightly lower light transmission.

Table 2. Thickness and light transmission (%) of protein-based film from threadfin bream surimi

<table>
<thead>
<tr>
<th>Conc. (%)</th>
<th>Thickness (mm)</th>
<th>Wavelength (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>200</td>
</tr>
<tr>
<td>1/3</td>
<td>0.0198±0.003a**</td>
<td>0.1</td>
</tr>
<tr>
<td>1/11</td>
<td>0.0171±0.002a</td>
<td>0.1</td>
</tr>
<tr>
<td>2/3</td>
<td>0.0379±0.003b</td>
<td>0.1</td>
</tr>
<tr>
<td>2/11</td>
<td>0.0352±0.005b</td>
<td>0.1</td>
</tr>
</tbody>
</table>

* Mean±SD from ten determinations.
** The different superscripts in the same column indicate the significant differences (P<0.05).
From the result, the films from surimi had the low preventive effect on visible light transmission.

Color of films obtained from acid and alkaline solubilizing processes had the different color as evidenced by different \( b^* \) value (Table 3). The film from acid process was more yellowish than that from alkaline process. For \( L^* \) and \( a^* \) values, no marked differences were observed between samples. With the same pH used, film with different thickness had the similar color. However, for film with 1% protein content, \( b^* \) value of the film made by alkaline solubilizing process was lower than that of acid process (\( P<0.05 \)). Thus, it was postulated that acid used might hydrolyze the sucrose used as the cryoprotectant in surimi, resulting in the increased reducing sugar, glucose and fructose. As a consequence, those reducing sugars might react with amino acid in surimi via Maillard reaction, especially during drying and the brown or yellowish compounds were formed (Wong, 1989). For \( E^* \) value, total color difference, the lowest value was observed in the surimi film with 1% protein with alkaline solubilizing process.

### 2.3 Mechanical properties

TS and EAB of surimi film prepared using acid and alkaline solubilizing process are presented in Table 4. At the same protein concentration used, no differences in TS and EAB were observed between surimi film prepared by acid and alkaline solubilizing process (\( P>0.05 \)). The unfolded proteins obtained from either acid or alkaline solubilizing process underwent the aggregation through hydrogen, ionic, hydrophobic and covalent bonding. The degree of chain extension and the nature of sequence of amino acid

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**Table 3. Color parameters of the protein-based film from threadfin bream surimi**

<table>
<thead>
<tr>
<th>Conc.(%)/pH</th>
<th>( L^* )</th>
<th>( a^* )</th>
<th>( b^* )</th>
<th>( E^* )</th>
</tr>
</thead>
<tbody>
<tr>
<td>1/3</td>
<td>90.60±0.23(^{**})</td>
<td>-1.37±0.30(^{**})</td>
<td>2.59±1.21(^{**})</td>
<td>3.44±0.28(^{**})</td>
</tr>
<tr>
<td>1/11</td>
<td>91.77±0.73(^{**})</td>
<td>-1.28±0.12(^{**})</td>
<td>1.41±0.21(^{**})</td>
<td>2.20±0.38(^{**})</td>
</tr>
<tr>
<td>2/3</td>
<td>90.22±0.23(^{**})</td>
<td>-1.39±0.21(^{**})</td>
<td>2.25±0.27(^{**})</td>
<td>3.87±0.26(^{**})</td>
</tr>
<tr>
<td>2/11</td>
<td>90.54±0.54(^{**})</td>
<td>-1.49±0.15(^{**})</td>
<td>2.02±0.52(^{**})</td>
<td>3.51±0.59(^{**})</td>
</tr>
</tbody>
</table>

\(^{**}\) Mean±SD from three determinations.

** The different superscripts in the same column indicate the significant differences (\( P<0.05 \)).

**Table 4. Tensile strength (TS), elongation at break (EAB) and water vapor permeability (WVP) of protein-based film from threadfin bream surimi**

<table>
<thead>
<tr>
<th>Conc.(%)/pH</th>
<th>TS(^{**}) (MPa)</th>
<th>EAB(^{**}) (%)</th>
<th>WVP(^{**}) (x10(^{-11}) gm(^{1})s(^{-1})Pa(^{-1}))</th>
</tr>
</thead>
<tbody>
<tr>
<td>1/3</td>
<td>4.06±0.61(^{***})</td>
<td>46.84±12.55(^{a})</td>
<td>8.76±0.69(^{a})</td>
</tr>
<tr>
<td>1/11</td>
<td>4.69±0.91(^{ab})</td>
<td>54.33±12.19(^{a})</td>
<td>6.93±0.78(^{b})</td>
</tr>
<tr>
<td>2/3</td>
<td>5.02±0.78(^{b})</td>
<td>115.87±12.68(^{a})</td>
<td>17.12±0.72(^{c})</td>
</tr>
<tr>
<td>2/11</td>
<td>5.07±0.53(^{b})</td>
<td>125.98±18.23(^{b})</td>
<td>14.47±1.32(^{a})</td>
</tr>
</tbody>
</table>

\(^{**}\) Mean±SD from eight determinations

\(^{***}\) Mean±SD from four determinations

** The different superscripts in the same column indicate the significant differences (\( P<0.05 \)).
residues affect the mechanical properties of protein film (Krochta, 1997). In general, low molecular weight plasticizers are added to protein film in order to improve film flexibility by reducing protein-protein interaction (Krochta, 1997). The increase in glycerin resulted in the lowered puncture force of film from Nile tilapia muscle proteins with the increased puncture deformation (Paschoalick et al., 2003). From the result, film with greater protein content exhibited the higher TS and EAB (P<0.05). The higher amount of protein might aggregate intermolecularly to a great extent, compared with the lower amount, leading to the stronger interaction as evidenced by the increased TS. Since the weak bonds including hydrogen bonds, hydrophobic and ionic interactions were dominant in surimi film as reported by Shiku et al. (2004), it was most likely that proteins with the greater amount could undergo aggregation with those bonds, leading to the network with a greater numbers of those bonding. This resulted in the increased EAB of the film.

2.4 Water vapor permeability

Water vapor permeability of surimi film prepared using acid and alkaline solubilizing processes is shown in Table 4. WVP of surimi film either from acid or alkaline solubilizing process increased with increasing protein concentration in the film-forming solution (P<0.05). A higher amount of protein was probably associated with a higher amount of polar groups in surimi film, which could absorb more water from the surrounding atmosphere. Blue marlin myofibrillar proteins contained a large amount of ionized polar amino acids (approximately 33%) (Shiku et al., 2003). Transmission of water vapor through protein-based film is also facilitated by the presence of glycerol, a hydrophilic plasticizer (Cuq et al., 1995). Additionally, cryoprotectants including sucrose and sorbitol also provided the polar groups in the surimi film. Those polar groups provide for hydrogen bonding. As a result, the film can absorb the water from the surrounding air or from the food product (Kim and Ustunol, 2001). From the result, film with the greater surimi content contained more cryoprotectant amount, leading to the greater WVP. Cuq et al. (1995) who found that the WVP of myofibrillar protein-based biopackaging was much greater than those of typical polymeric packaging materials such as low density and high density polyethylene films. From the result, film prepared by acid solubilizing process had the greater WVP than that prepared by alkaline solubilizing process (P<0.05). Therefore, acid or alkaline used for pH adjustment might affect the WVP property of film by modifying the charge of protein molecule differently, resulting in the differences in WVP.

2.5 Protein pattern

Protein patterns of surimi and surimi film prepared by acid and alkaline solubilizing process are shown in Figure 1. Myosin heavy chain constituted as the major protein in surimi, followed by actin. The film contained the lower band intensity of myosin heavy chain and actin, compared to that of surimi, with the concomitant appearance of proteins with the lower molecular weight. The result suggested that the major proteins were degraded via acid and alkaline process. From the result, different protein patterns were found between both films. The degradation proteins with molecular weight of 150-160 kD were found in the film with acid process, while the proteins with the molecular weight ranging from 60 to 70 kD were found in the film with alkaline process. Therefore, the cleavage of myosin occurred at different sites, leading to the differences in the molecular distribution. Actin was also degraded into different degradation products by two different processes. Therefore, the acid or alkaline process might induce the degradation of muscle protein in surimi. This might contribute to the different characteristic of surimi films produced by both processes. The result was in agreement with Cuq et al. (1995) who found the degradation of myosin heavy chain in edible films based on sardine myofibrillar proteins, especially in the acidic pH ranges, due to the cathepsins. However, no degradation of myosin heavy chain was found in the film from Alaska pollack surimi (Shiku et al., 2004). Therefore, different enzymes in muscle of various fish were postulated, leading to the varying degree
Figure 1. Protein patterns of protein-based films from threadfin bream surimi prepared by acid and alkaline solubilizing processes. M: high-molecular-weight protein marker; S: surimi; acid: film prepared by acid solubilizing process; alkali: film with alkaline solubilizing process.

Figure 2. SEM micrographs of protein-based films from threadfin bream surimi prepared by acid and alkaline solubilizing processes with different protein contents. (magnification: 7,500x) A: 1% protein content, pH 3; B: 2% protein content, pH 3; C: 1% protein content, pH 11; D: 2% protein content, pH 11.
of hydrolysis in the film based on muscle protein from different fish types. For the film produced with the same process, there were no marked differences in protein patterns between the films analyzed under reducing and non-reducing conditions. However, some minor bands disappeared under reducing condition. Therefore, disulfide bond did not contribute as the major bonds in film formation of surimi produced by acid and alkaline solubilizing process. The result was in agreement with Shiku et al. (2004) who reported that no disulfide bond was found in the Alaska pollack surimi film.

2.6 SEM micrograph

The micrographs of surimi film prepared by acid and alkaline solubilizing processes are shown in Figure 2. The surface of surimi film prepared by acid process was smoother, while surface of surimi film from alkaline process exhibited more protruding structure. With the same solubilizing process, no marked differences in surface micrograph between the films with different protein contents were observed. The differences in protein compositions induced by both processes might contribute to the difference in aggregation patterns. As a result, the slight difference was noticeable between films from two solubilizing processes.

3. Changes in protein pattern as affected by acid and alkaline solubilizing processes

The protein patterns of acidic and alkali film forming solutions are depicted in Figure 3. At 0 h, the protein pattern of film forming solution

![Figure 3. Protein patterns of acidic and alkali film forming solutions of threadfin bream surimi with different exposure times. MHC: myosin heavy chain. The numbers designate the exposure time (h). A: 1% protein content, pH 3; B: 2% protein content, pH 3; C: 1% protein content, pH 11; D: 2% protein content, pH 11.](image-url)
was very similar to those of surimi. However, the myosin and actin were more degraded as the exposure time increased up to 10 h. At 10 h, the lowest myosin band intensity was observed with the coincidental appearance of low-molecular-weight products. This result revealed that the muscle proteins underwent the degradation during drying process. From the result, the degradation pattern of myosin and actin in film forming solutions was different between two processes. Chawla et al. (1996) reported the degradation of myosin heavy chain of acid treated threadfin bream mince, due to the presence of acid protease. However, with extended exposure time, the pattern of film forming solution was similar to that found in the film (Figure 1). Therefore, the degradation patterns induced by acid and alkaline process might be associated with the different properties and characteristics of surimi film.

4. The change in color of surimi film during the storage

The color of films prepared from acid and alkaline processes is shown in Figure 4. The increase in both $b^*$ and $E^*$ values was observed in film prepared by acid solubilizing process with increasing storage time ($P<0.05$), but no marked changes in those values were found in surimi film prepared by alkaline process ($P>0.05$). The greater changes were found in the film with higher protein contents. The increase in $b^*$ value was associated with the formation of yellow hue. The formation of yellow color might be due to the non-enzymatic browning reaction, which might be induced during the extended storage. The result was in accordance with Cuq et al. (1996) who found the increase in $b^*$ value of fish myofibrillar protein-based film formulated with saccharose as a plasticizer. From the result, the surimi films prepared by acid process were more susceptible to color changes, compared with those prepared by alkaline process. This was presumed that acid might induce the hydrolysis of sucrose added in surimi as cryoprotectant. Hydrolysis of glycosidic bonds joining monosaccharide units can be catalyzed by acid (BeMiller and Whister, 1996). As a result, the free reducing sugars were formed and underwent the browning reaction with amino groups in surimi. Additionally, the muscle proteins were also degraded by acid as shown in Figure 3. Thus, the browning could occur to a greater extent in the surimi film prepared by acid process. Similar changes in $E^*$ values were found, compared to the changes in $b^*$ value. $E^*$ is useful for expressing the total amount of color changes. From the result, the increase in $E^*$ was associated with the increase in $b^*$ value.

![Figure 4. Changes in $b^*$ and $E^*$ values of protein-based films from threadfin bream surimi during storage at room temperature.](image-url)
Conclusion

The properties of film from threadfin bream surimi were affected by the solubilizing processes, which were possibly associated with the different degradation patterns of proteins. The film with acid solubilizing process had the smoother surface but was susceptible to the browning reaction, compared to that prepared with alkaline process. However, similar mechanical properties were found in the two films.

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References


