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Combination model for the spatial partition of surimi protein and hydroxylpropylmethylcellulose

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Abstract

The objectives of this study were to investigate the combination model of hydroxylpropylmethylcellulose (HPMC) added with surimi protein gel. HPMC forms and addition timing apparently affected the structural geometry of the combination gel matrix and resulted in the variation in rigidity, thermal stability and gel strength. Upon viewing of the scanning electron microscopy (SEM) photographs, the horse mackerel surimi displayed a large holes and smooth protein matrix, while, the HPMC added surimi displayed a porous or rough structure. According to thermal gelation and microstructure analysis, the filling effect existed in HPMC added surimi. HPMC existed as dispersed particles in HPBC (HPMC powder added before chopping) and HPDC (HPMC powder added during chopping) surimi. HPMC remained soluble in the interstitial fluid of the surimi gel matrix in HGBC (HPMC gel added before chopping) and HGDC (HPMC gel added during chopping) surimi. These four 'simple' filled gels turned to 'complex' filled gels during the process of thermal gelation caused by certain interaction of the gelled HPMC and protein gel matrix. HGAC (HPMC gel added after chopping) surimi formed an interpenetrating structure together with a gelling protein and a gelling HPMC.

Keywords: HPMC; Horse mackerel; Surimi; Combination gelation; Microstructure

1. Introduction

Polysaccharides have been extensively used in meat product manufacturing to provide the emulsifying, viscous and gelation properties in such products (Chin, Keeton, Longnecker, & Lamkey, 1998; Glicksman, 1991). Among them, starch is the most widely used 'filler' ingredient in surimi products. The gelatinized and expanded starch granules cause pressure to be exerted on the surimi protein matrix. This compression effect results in a firmer and slightly cohesive gel matrix even though the interaction between the starch and protein is weak (Iso et al., 1985; Kong, Ogawa, & Iso, 1999; Lee & Kim, 1986; Wu, Lanier, & Hamann, 1985; Yamazawa, 1990).

Consumer interest in reduced-fat and reduced-calorie foods has led to an expanded use of some polysaccharides gums as a food ingredient. In our previous study, four kinds of polysaccharides, including konjac, hydroxylpropylmethylcellulose (HPMC), curdlan and gellan gum, were added as gelation-aid materials for the preparation of horse mackerel surimi. A decrease in the on-set temperature of the first-heat gelation and resolution degree was noted for the produced surimi containing these polysaccharides (Chen, 2000).

In most case, the added ingredients will be entrapped within the surimi protein gel matrix and thus fill the gel, exerting their functional effects through influencing the formation of the continuous surimi gel matrix during heat-induced gelation or modifying the viscosity, mobility and other properties of the aqueous phase. Furthermore, the ingredients may possibly influence the texture and appearance of the surimi products by virtue of their particle size, distribution, rheological properties and relative volume

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fraction of the gel (Lee, Wu, & Okada, 1992). Lee and Chung (1990) mentioned that several factors will influence the physical properties of filled surimi gel including the integrity and properties of the primary gel matrix, the properties of the dispersed coingredients, and the interactions between the continuous matrix and dispersed coingredients.

Ziegler and Foegeding (1991) envisioned five possible models for the spatial partitioning of a gelling protein and a coingredient. In model A, the filler remains soluble in the interstitial fluid of the gel matrix. In model B, the filler exists as dispersed particles. In model C, the coingredient may associate with the gelling fraction without constituting a continuous linkage in the gel matrix. In model D, the coingredients may copolymerize with the primary gelling fraction to form a single, heterogeneous network. In model E, an interpenetrating network formed by the coingredient, not directly interacting with the continuous matrix formed by the primary gelling fraction, but structurally cooperative due to the entwining of the two gel networks. Model A and B are termed 'simple' filled gels, where the dispersed phase is unassociated with the gel matrix. While, model C and D are termed 'complex' filled gels and the coingredient associates directly via nonspecific interactions with the primary gelling component.

In combination gelation of HPMC added horse mackerel surimi protein, addition of HPMC lowered the breaking force, but increased the gel strength of surimi gel caused by the substantial rise in deformation (Chen, 2000). We have found the addition of HPMC did not markedly increase SS bonds and hydrophobic interactions in the combinative myofibrillar protein (MP) solution or surimi gel. Secondary bonds, such as H-bond, are the major contributor to the interaction between HPMC and MP within the combination gelation (Chen & Chen, 2001). Nevertheless, this phenomenon is still not well understood is worthy of further investigation with respect to the possible models for the spatial partitioning of the surimi protein gel and HPMC.

As the structure has a strong influence on the sense of texture and quality of surimi products, it is essential to understand the mechanisms that control the structural formation of such systems under processing conditions. The object of the present research was to realize the combination model of HPMC and surimi protein combinative gel, and to establish the desirable conditions for the production of HPMC containing horse mackerel surimi by investigating the thermal gelation properties and the microstructure of such two component systems.

2. Materials and methods

2.1. Materials

Frozen horse mackerel (*Trachurus japonicus*) was acquired from Cheer-Foods Enterprise Co. Ltd (Ilan, ROC) The fish block used for this study (10 kg), had been previously frozen for 2 months at −20 °C, and was thawed at 25 °C for 12 h. Ordinary muscles were collected from the thawed fish and then squeezed through a sieve with 3 mm dia holes. The HPMC (Celacol CC-001, Courtaulds Chemicals Celacol Co., Derby, UK) was acquired from Toong Yeuan Enterprise Co., Ltd (Taipei, ROC).

2.2. Preparation of samples

2.2.1. HPMC gel solution

The HPMC powder was weighed and hydrated with distilled water (w/w = 1/4) in a Stephan vertical vacuum cutter (Model UM 5 Universal; Stephan Machinery Co., Hameln, Germany) for 1 min at 1600 rpm, vacuum around 600 mmHg was performed at last 0.5 min, in order to a obtain final concentration of 20%.

2.2.2. Surimi

The washed mince was dewatered and adjusted to have a water content of 80%, and then chopped in a Stephan vertical vacuum cutter (Model UM 5 Universal) with a circulating chiller to keep its temperature at below 10 °C. The mince was first chopped for 1 min at 1600 rpm (i.e. no-salt-chopping). Salt equivalent to 2.5% weight of the mince was added and then chopped at 1600 rpm for another 2 min (i.e. salt-chopping) under vacuum of approximately 600 mmHg in order to become surimi. The HPMC powder or gel equivalent to 2.0% weight of the mince was added (i) before chopping (BC), (ii) during the chopping (between no-salt-chopping and salt-chopping process, DC), (iii) after chopping (the HPMC was added after salt-chopping and manually blended for 0.5 min, AC). The surimi was also kept at 40 °C for 30 min before the measurement of rigidity.

2.2.3. Kamaboko

The surimi paste was packed in a 3 cm dia polyvinylidene chloride casing, was allowed to set at 4 °C for 30 min, and then heated in a water bath at 85 °C for 30 min in order to become surimi sausage (defined as 'kamaboko'). The heated kamaboko was cooled immediately with running water. The cooled gel was then kept at 4 °C overnight before the measurement of gel strength.

2.3. Gelation properties determination

2.3.1. Rigidity

The rigidity of the surimi was measured in a thermal scanning rigidity monitor (TSRM) assembled as previously described (Chen & Lee, 1998). Briefly, the surimi was pasted evenly in a testing cell to 5 cm in height. The blade (adapter No. 24 of the SUN RHEOMETER CR-150, Sun Scientific Co., Ltd, Tokyo, Japan) was oscillated within 1 mm in the surimi at 2 mm/min at 2 °C intervals. Temperature of sample was raised at 1.5 °C/min over

the range of 15–90 °C. Three replicate scans were made and the rigidity of surimi was then calculated according to Hamann, Purkayastha, and Lanier (1990); Sue, Chen, and Kong (1995):

$$R = (F/A)/(D/T) = F \times T/(2L \times W \times D)$$

where R is the rigidity (kPa), F the pulling force (kgw) applied to specimen, A the contact area (m^2) between blade and specimen, D the moving distance (m) of the blade, T the thickness (m) of specimen, and L and W the length and width (m) of the blade in contact with the specimen.

2.3.2. Gel strength

Gel-forming ability of the heated surimi gel was determined on 3 cm dia, 3 cm length sections by a Rheometer (SUN RHEOMETER CR-150) as described in a previous paper (Chen, 1995). Briefly, using a 5 mm diameter spherical head plunger to press into one end of each sample with 60 mm/min deformation rate. When the test sample lost its resistance and ruptured, the load (as breaking force) and the depth of depression (as deformation) were recorded. Six replicates were determined and the gel strength (G) was calculated as: $B \times D$, where B represents breaking force (N) and D represents deformation (mm).

2.4. Thermal analysis

The HPMC and surimi were analyzed with differential scanning calorimetry (DSC) using a TA Instruments DSC 2010 (TA Instruments, New Castle, USA). The specimen (about 15 mg) was placed in a sealed aluminum pan and scanned at 10 °C/min over the range of 10–100 °C. Three replicate scans were made for each sample. The temperature at each peak maximum on a representative curve was recorded as the transition temperature (T_m).

2.5. Microstructure analysis

2.5.1. Scanning electron microscopy (SEM) of surimi

The surimi specimens were ruptured into small cubes following being frozen with liquid nitrogen and dried with a LABACONCO freeze-drier (LABCONCO corporation, Kansas city, USA). The dried specimens were mounted on aluminum studs and coated with gold palladium under vacuum conditions. The specimens were examined with a SEM (ABT-55I, International Scientific Instrument, Tokyo, Japan) to observe the structure of ruptured surface.

2.5.2. Phase contrast and optic photomicrograph of HPMC

HPMC powder or HPMC gel solution was observed and photographed with phase contrast microscope (Nikon OPTIPHOT-2, Tokyo, Japan).

2.6. Statistical analysis

Statistical analysis of the data was performed using the system developed by SAS Institute, Inc. (1985). When analysis of variance (ANOVA) revealed a significant effect ($P < 0.05$), data means were compared with the least significant difference (LSD) test.

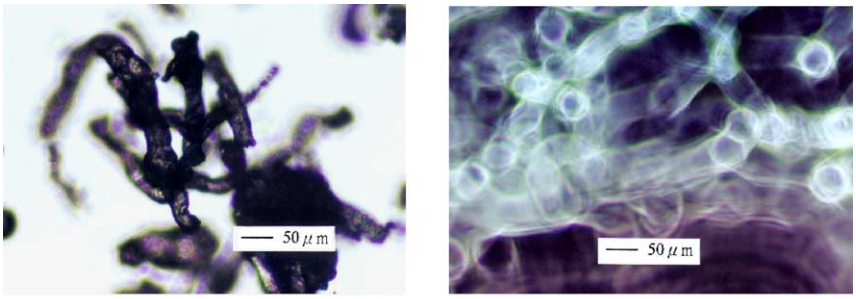
3. Results and discussion

3.1. Appearance of HPMC

Phase contrast photomicrograph showed HPMC powder has a threadlike appearance of 20–30 μm width (Fig. 1a). After imbibing water, the transparency of threadlike fibers in HPMC increased and such fibers distributed themselves in a continuous reticular arrangement. A lot of light circles around 50 μm, which were at the intersection positions of the polymer, could be observed on the photomicrograph (Fig. 1b). They rose from a jacketing of the long threadlike polymer molecules with layers of water molecules, which increased the bulk of the polymer aggregate.

3.2. Thermal gelation of HPMC added surimi

From DSC analysis, the position and size of single endothermic peak in the second scanning thermograms of HPMC gel solution exhibited patterns analogous to that in the first scanning. Their on-set temperature (T_o) and transition temperatures (T_m) of gelation were around 60 and 72 °C, respectively (Fig. 2). The thermo-reversible HPMC gel used in this study exhibited an on-set thermal



(a) HPMC powder (b) HPMC gel

Fig. 1. Phase contrast microphotographs of HPMC.

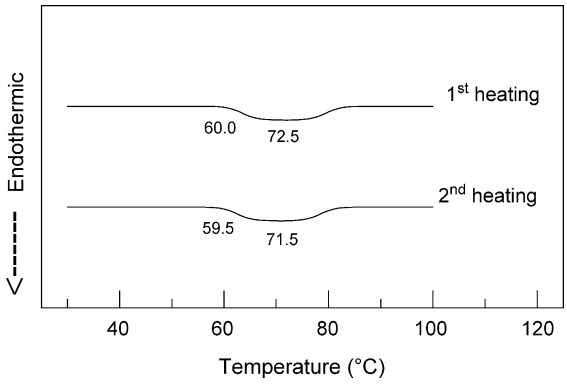


Fig. 2. DSC programs for rescanning test for a 20% HPMC gel solution.

gelation temperature of approximately 60 °C. The rescanning treatment did not significant alter the thermal gelation temperature.

During the rescanning test of HPMC added horse mackerel surimi, a four-stage processing of change in rigidity was observed in surimi as its temperature was increased in the first scanning (Fig. 3). These stages were classified as softening, the first heat gelation, resolution and the second heat gelation stages of horse mackerel surimi paste with the first, second and third break-points of the TSRM curves being defined as the on-set temperature of the first heat gelation (T_1), gel resolution (T_2) and the second heat gelation (T_3), respectively (Chen, 2000; Hamman et al., 1990). During the first scanning, the rigidity was the lowest for the surimi with HPMC powder added after chopping (HPAC) among these surimis with HPMC addition (Fig. 3). The HPAC surimi showed sharper increases and decreases at the first heat gelation stage and resolution stage, respectively, than those of HPBC (HPMC powder added before chopping) and HPDC (HPMC powder added during chopping) surimi. Furthermore, the rigidity value and thermogram change pattern of HPAC were more similar to that of the horse mackerel surimi without HPMC addition, reported by Chen and Chen (2001), than what those of HPBC and HPDC surimi were. The T_3 in the TSRM thermograms of horse mackerel surimi reported by Chen and Chen (2001) was 64 °C. For HPBC, T_3 shifted to 58 °C and the first heat gelation and resolution stage became less observable. This revealed that the addition of HPMC powder before or during chopping led the thermal gelation properties to apparently alter themselves in the surimi.

During the second scanning, the rigidity of HPMC powder added surimi maintained a higher value (Fig. 3). The rigidity decreased up to around 60 °C, and a higher temperature yielded HPDC and HPAC surimi with a higher rigidity (Fig. 3b and c). This transition in HPBC surimi was not obvious and the rigidity decreased as the second scanning temperature increased (Fig. 4a). Since HPMC is thermo-reversible with on-set gelation temperature around 60 °C (Fig. 2) and the surimi protein is thermo-irreversible, the increase of rigidity in the second scanning at temperature higher than 60 °C is due to the thermal gelation of HPMC.

For HPAC surimi, the first scanning thermogram was similar to the surimi without HPMC addition, and the second scanning thermogram exhibited the observable thermal gelation of HPMC. This reflected that the HPAC surimi structure might be comprised of an independent surimi gel network and a dispersed HPMC powder system, which are different from the combination model of HPBC or HPDC surimi. That is to say, the HPMC and surimi gelation properties having being changed for HPBC and HPDC surimi might be owing to the participation of HPMC in the network forming of heated surimi protein gel and certain interaction existing between surimi protein and HPMC. Meanwhile, the earlier the HPMC was added into chopping process the firmer the HPMC entrapped in surimi gel matrix became. This caused a more noticeable change in their thermal gelation.

For HPMC gel solution added surimi, the thermogram of HGAC (HPMC gel solution added after chopping), including the rigidity value (Fig. 4), was similar to that of HPAC surimi in this study and that of the horse mackerel surimi without HPMC addition as reported by Chen and Chen (2001). The rigidity of HGBC (HPMC gel solution added

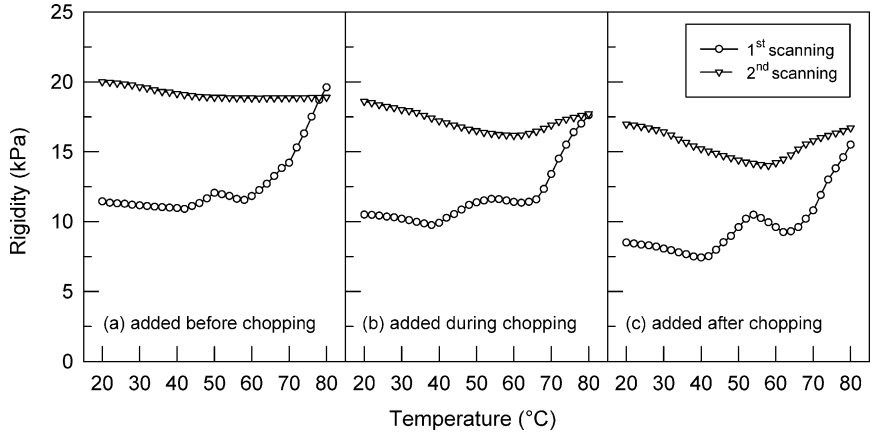


Fig. 3. TSRM programs of rescanning tests of surimi with 2% HPMC powder added.

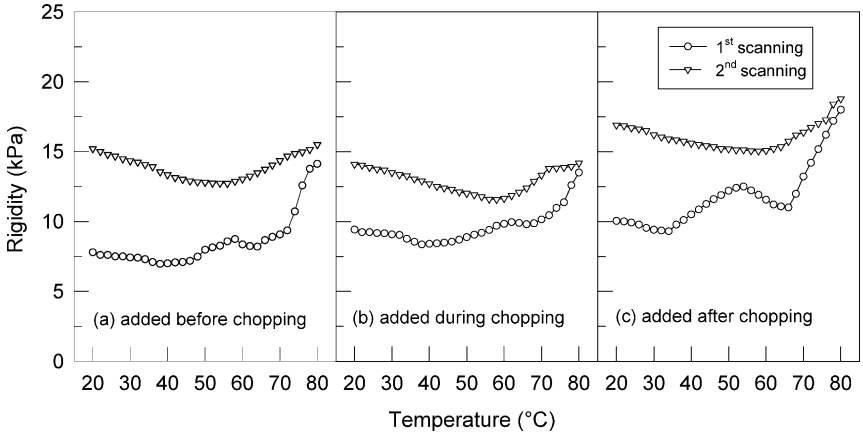


Fig. 4. TSRM programs of reheated test of surimi with 2% HPMC gel solution added.

before chopping) and HGDC (HPMC gel solution added during chopping) surimi in both of the first and the second scanning was lower than those in surimi without HPMC, and the four-stage processing of changes in rigidity became less observable. Since the bulk of HPMC polymer aggregate increased because of the absorption of water molecules within its long threadlike polymer molecules, this would enlarge the gap between linked protein molecules in its gel matrix in HGBC and HGDC surimi. As the scanning temperature rose, the attached water molecules were driven from the cellulose ether chains, the chains locked and the bulk volume shrank. The enlarged gap influenced the cross-link within the interaction of protein molecules to form a three-dimensional network and a loose-fitting surimi structure was developed. This phenomenon caused the lowering in rigidity. It was also observed that an apparent curve in the second scanning thermogram in all of the HPMC gel solution added surimi, which might have been caused by the reversible thermal gelation from the loosely- or un-entrapped HPMC gel.

In DSC analysis, thermograms of horse mackerel surimi showed two clearly discernible endothermic peaks at 51.2 and 66.7 °C (Fig. 5a). Assuming that the horse mackerel DSC profiles follows the same pattern as that of cod, two larger endothermic peaks are thought to be the thermal denaturation of myosin and actin, respectively (Chen, Lou, Chen, Chen, & Lee, 1996; Hasting, Rodjer, Park, Matthews, & Anderson, 1985). The shoulder peaks around 56.0–58.0 °C were found in the first scanning when HPMC was added into the surimi. These peaks were attributed to the rupture of HPMC gel as reported by Chen and Chen (2001). Only a small endothermic peak was remained in the second

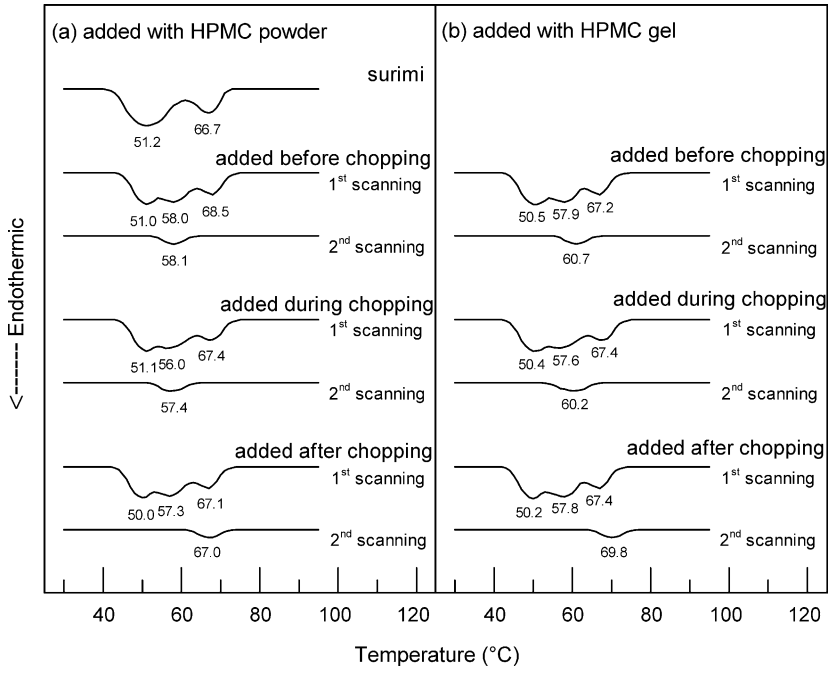


Fig. 5. DSC programs of reheated test in surimi with (a) HPMC powder and (b) HPMC gel solution added at various chopping stage.

scanning thermogram, but the T_m dispersed from 58.1 to 69.8 °C (Fig. 5). Since the on-set and peak-maximum thermal gelation temperatures (T_m) of HPMC gel used in this study were around 60 and 72 °C, respectively (Chen & Chen, 2001), the shift of T_m reflected that the HPMC gelation properties had been changed in the combinative gel system. Surimi with HPMC addition before or during chopping showed a lower T_m in the second scanning thermogram and was farther from the T_m of HPMC than that of surimi with HPMC addition after chopping. This might be due to the impacted HPMC transforming into a gel with a lower energy requirement. On the other hand, the T_m of second scanning in HPBC and HPDC surimi shifted to a lower temperature than those of HGBC and HGDC surimi. It was postulated that the powder form HPMC was filled and took up a smaller space in the protein gel matrix than what gel form HPMC did. The congested space moved HPMC cellulose ether chains closer to each other, and the interaction of threadlike polymer molecules made cellulose ether chain lock and transform into a gel more easily. Conversely, when the HPMC was added after chopping, though two oppositely charged polymer (protein and HPMC) formed a complex gel, they would eventually phase separate. Therefore, the T_m of HPAC and HGAC during second scanning approached the peak-maximum temperatures of the thermal gelation of HPMC gel.

Addition of HPMC decreased the breaking force but increased the deformation of horse mackerel kamaboko. However, the gel strength increased for HPMC added kamaboko except for HPAC and HGBC (Table 1). Since the breaking force was the force required to break a sample and HPMC gel offered a softer non-rubbery structure (Glicksman, 1969), it is not surprising that the lower breaking force was observed in HPMC added kamaboko. Among the HPMC added samples, HPAC showed the lowest breaking force and gel strength. The reason behind the lowering in gel-forming ability for HPAC might have been the blending process causing the HPMC powder to disrupt surimi protein gel matrix which had been developed after salt-chopping, and the competition in water molecules with dispersed HPMC powder further interfering with the development of a continuous matrix formed by the primary gelling protein. After all, the coexistence of two incomplete gels in HPAC kamaboko could not build a strong enough conformation to resist high levels of stress. On the other hand, though HGAC kamaboko might have no continuous combinative network, it owned two independent gels. The cooperation of protein gel and HPMC gel developed a certain pliable gel system and resulted in a high value of deformation. In addition, the various impacted intensity of HPMC on the protein gel matrix caused a breaking force diversity, which was observed in the surimi when a powder form or gel form HPMC was added.

Table 1
The effect of HPMC addition during the chopping period on the gel strength of kamaboko

	Breaking force (N)	Deformation (mm)	Gel strength (N×mm)
Surimi	3.630±0.037a	5.8±0.4d	21.054±0.412c
HPBC[a]	3.592±0.040a	7.0±0.3bc	25.142±0.382a
HPDC[a]	3.480±0.028b	6.4±0.4cd	22.272±0.304b
HPAC[a]	2.655±0.031e	6.6±0.2c	17.522±0.235e
HGBC[b]	2.995±0.050d	6.7±0.3c	20.066±0.392d
HGDC[b]	2.982±0.043d	7.3±0.2b	21.770±0.255b
HGAC[b]	3.237±0.039c	7.7±0.2a	24.924±0.245a

Values in a column for each sample with different alphabets (a–e) are significantly different ($P<0.05$).

[a] HPBC, HPDC and HPAC representing the HPMC powder was added before, during and after chopping, respectively.

[b] HGBC, HGDC and HGAC representing the HPMC gel solution was added before, during and after chopping, respectively.

3.3. Microstructures of HPMC added surimi

To verify the assumption in the combination model for the spatial partition of surimi protein and HPMC, the microstructure of surimi was observed with a SEM. The surimi displayed a smooth protein gel matrix with fewer but bigger cavities (Fig. 6a) than that which was added with HPMC (Fig. 6b–g). Some fiber was entrapped in HPBC surimi gel matrix with numerous small apertures, which formed a porous network structure (Fig. 6b). These small apertures were the remaining spaces from the absorbed water in HPMC following dehydration. Small apertures distributed in gel matrix were also found in the HPDC surimi except that the network was not as porous as the HPBC surimi. Pieces of gel matrix congregated and formed a rough and solid structure (Fig. 6c). Instead of a porous network and aperture-distributed gel matrix, a layered structure was observed in the HPAC surimi (Fig. 6d). It seems that the surimi gel matrix accompanied HPMC was a rearrangement following the stress direction during blending which caused the HPMC fibers to not be entrapped, but rather adhere to the gel matrix.

Lots of hemisphere hollows were observed in electron microphotographs when the HPMC gel solution was added before or during chopping (Fig. 6e and f). The dimension of such hollows was bigger than that of the apertures distributed in the HPBC and HPDC surimi. Furthermore, the gel matrix was denser in the HGAC surimi than in the HGBC and HGDC surimi (Fig. 6g). Since HPBC and HGAC kamaboko exhibited higher gel strength than the other HPMC added kamaboko, it was suggested that both of the uniform protein gel dispersion (HPBC) and the dense gel matrix (HGAC) could contribute to the better viscoelasticity of surimi products.

3.4. Possible filling model of HPMC added surimi

To illustrate the filling model of HPMC added surimi according to Ziegler and Foegeding (1991), the HPMC existed as dispersed particles in HPBC and HPDC surimi

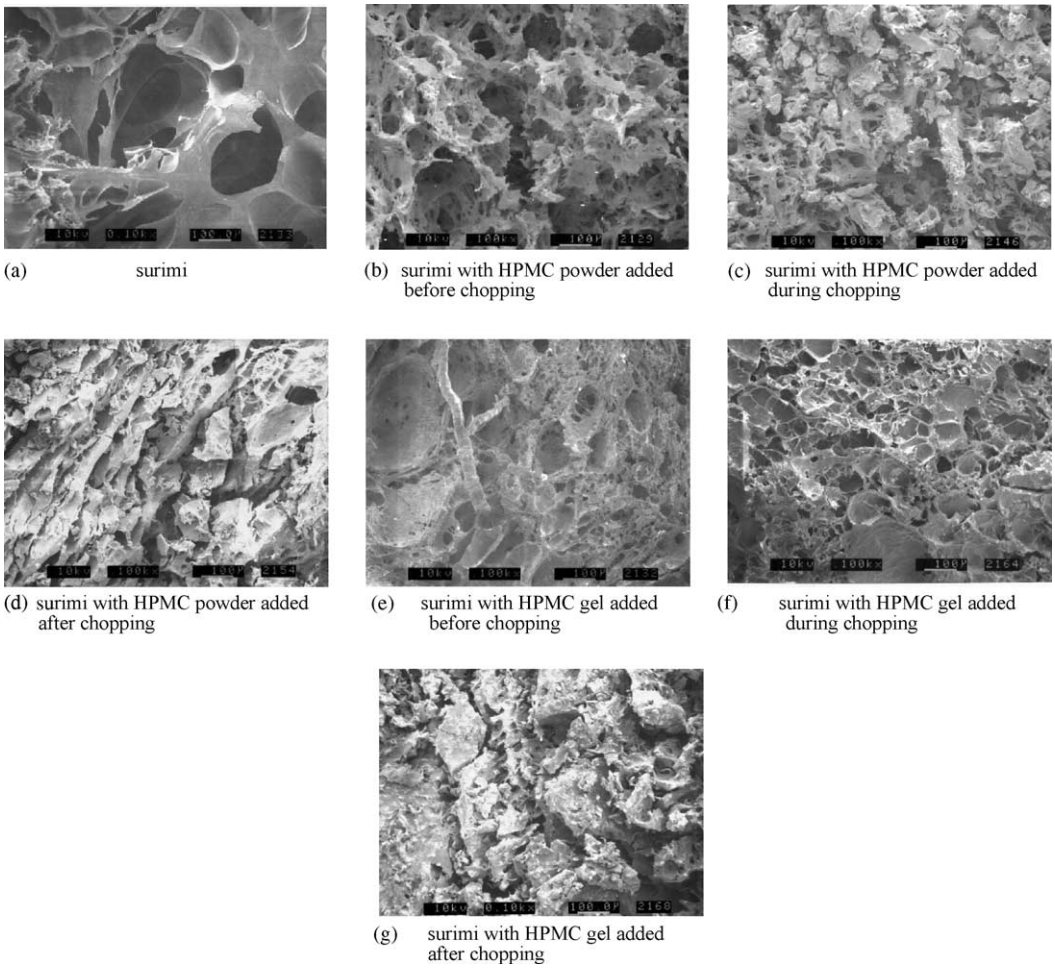


Fig. 6. Electron microphotographs of surimi with HPMC added.

thus belonging to model B. While, HPBC and HPDC surimi turned to model C during heating caused by the gelling of HPMC. Since HPMC remained soluble in the interstitial fluid of the surimi gel matrix, the possible filling model of HGBC and HGDC surimi during thermal gelation was transformed from Model A to C. In Model C, the gelled HPMC might associate with the surimi protein gelling fraction. For HPBC and HPDC surimi, such association not only did not affect the gelation properties of surimi proteins but also resulted in their high rigidity and gel strength. For HGBC and HGDC surimi, the swollen HPMC showed a negative effect regarding the structural geometry of the protein gel matrix and resulted in their low rigidity and breaking force.

The filling type of HPAC and HGAC surimi tended toward Model E. It was postulated that there was no direct interacting between the HPMC and surimi protein, but they rearranged alongside the stress during blending and were structurally cooperative due to the entwining of the gels. Therefore, a non-porous gel matrix was observed in both of the HPAC and HGAC surimi. On the other hand, though HGAC kamaboko did not have a continuous and perfect protein network, it owned two independent gel systems. This conformation, structurally cooperated by a gelling protein and a gelling HPMC, created a certain pliable but strong combinative gel which was reflected in its high deformation and gel strength.

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