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Rapid Heating of Alaska Pollock and Chicken Breast Myofibrillar Protein Gels as Affecting Water-Holding Properties

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ABSTRACT: The gelation response of salted muscle minces to rapid versus slow heating rates is thought to differ between homeotherm and poikilotherm species. This study investigated water-holding (WH) properties of pastes prepared from refined myofibrils, at equal pH, of chicken breast versus Alaska pollock both during [cook loss (CL)] and following [expressible water (EW)] their cooking by rapid [microwave (MW)] versus slow [water bath (WB)] heating and whether such properties were related to gel matrix structure parameters and water mobility. Results did not confirm the industrial experience that pastes of meat from homeotherms benefit from slower cooking. Gels of equally high WH ability (low CL or EW) were made by rapid heating when the holding time did not exceed 5 min prior to cooling, which was sufficient for completion of gelation. Reduced CL and EW correlated with larger and smaller amplitudes of T_{21} and T_{22} water pools, respectively, measured by time-domain nuclear magnetic resonance (TD-NMR).

KEYWORDS: *Water holding, heating rate, water mobility, gels, pore size*

INTRODUCTION

Rapid heating technologies, such as electromagnetic radiation (i.e., microwave heating and radio frequency heating), have scarcely been implemented in the production of comminuted meat gels, such as frankfurters and deli meats, which are instead heated slowly at a rate of about 1 °C/min in "smokehouse" equipment. Several reports suggest that slow-heating processes, such as this, result in optimum texture development and water and fat stability of such products.[1−4] However, Riemann et al.[5] reported that rapidly (microwave) heated (20 and 98 °C/min) fish (surimi) and turkey meat pastes, if held for a few minutes at the end point temperature of 70 °C, displayed similar fracture and cook loss values as their counterparts heated at 1 °C/min to the same temperature.

Many surimi seafoods are typically rapidly heated to 90 °C or higher and yet still possess firm gel texture and exceptional water-holding (WH) properties. Surimi is a refined, largely myofibrillar protein manufactured from meats of poikilotherm (cold-blooded) fish species, such as Alaska pollock and Pacific whiting. It is not obvious whether the difference in WH between such surimi seafood gels and those made from homeotherm (warm-blooded) land animal species, such as frankfurters, is due to species differences in protein composition (myosin isoforms),[6,7] muscle fiber differences,[8,9] the presence of the sarcoplasmic fraction in homeotherm meat gels (removed by surimi processing),[10] and/or the presence of cryoprotectants typically added to surimi.[11] A better understanding of the causes for such a difference in species gelling performance could lead to improving the WH properties of rapidly heated comminuted meat gels.

Similar benefits would accrue from better understanding the mechanisms of WH properties and the nature of water in muscle food gels. Capillarity is the prevailing hypothesis for explaining how water is held in gels.[12,13] The three-dimensional network of the gel system has been compared to narrow capillary tubes that imbibe water and hold it against gravity. The water inside these tubes possesses a capillary pressure according to the Young−Laplace equation

$$\Delta P = \frac{2\gamma \cos\theta}{r}$$

where ΔP is the capillary pressure, γ is the liquid−gas surface tension, θ is the contact angle at the liquid−air interface, and r is the radius of the tube.

Several workers have used low-field time-domain nuclear magnetic resonance (TD-NMR) spectrometry to monitor changes in foods associated with the mobility of water as affected by the food microstructure,[14−16] and lower T_2 relaxation times have been associated with better WH properties and smaller pore sizes.[17]

The objectives of this research were therefore to (1) compare the WH properties of myofibrillar protein gels derived from representative cold- and warm-blooded animals (Alaska pollock and chicken), heated by slow ramp versus rapid (square-wave) heating and (2) investigate the WH mechanism(s) associated with differences in water mobility and matrix structure among these gels.

MATERIALS AND METHODS

Myofibrillar Protein (Surimi) Paste Preparation. Commercial Alaska pollock (fish) surimi (refined myofibrils), obtained from Trident Seafoods (Seattle, WA), was compared to a washed mince prepared, by a similar process as is used for making fish surimi, from fresh chicken breasts obtained from Pilgrim's Pride (Sanford, NC). Visible connective tissue and fat were removed prior to comminution at 2500 rpm and 5−10 °C in a Stephan mixer−cutter with three parts

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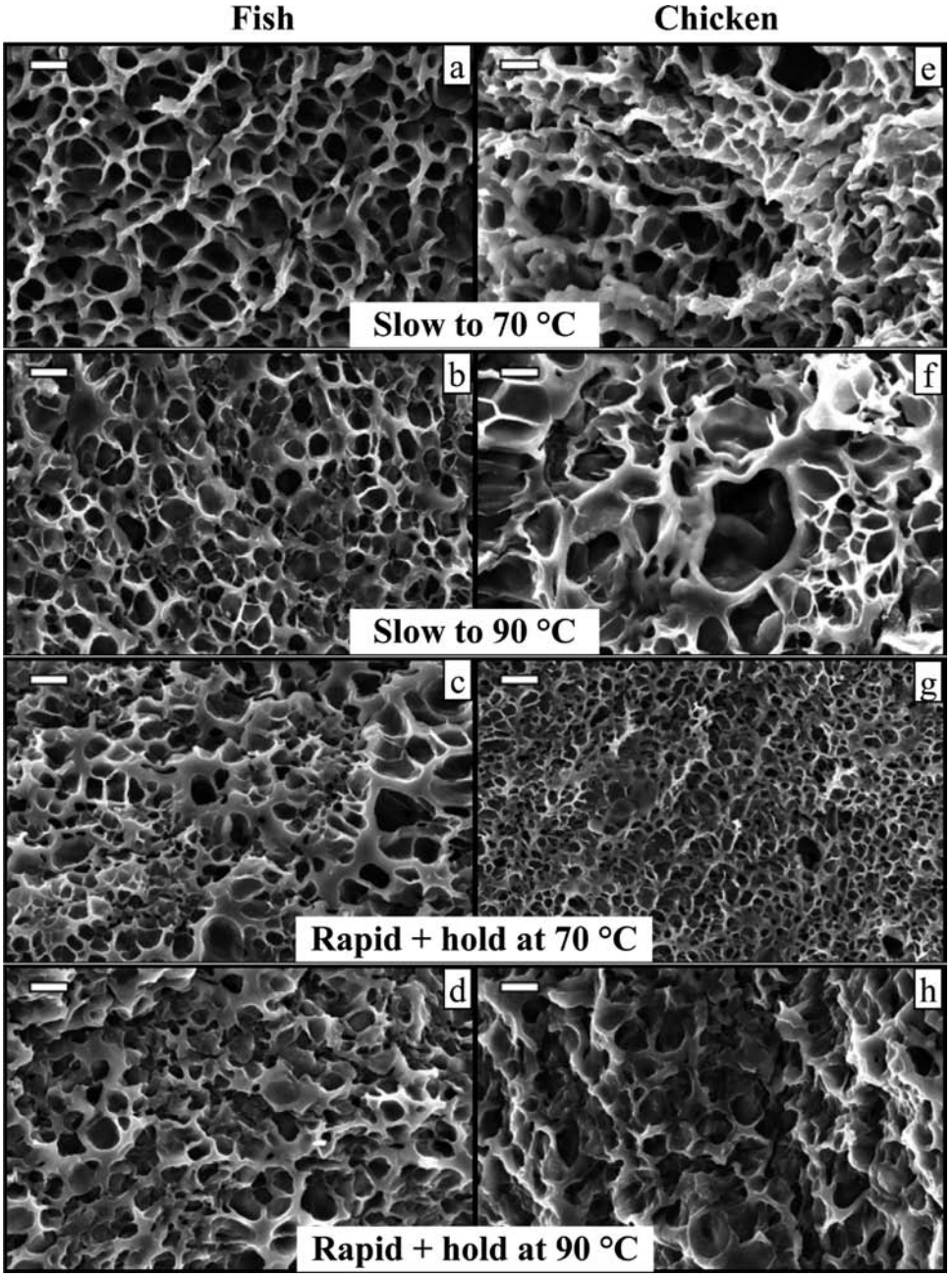

Figure 1. Scanning electron micrographs (1000×) of (a–d; left column) fish and (e–h; right column) chicken surimi gels slowly ramp heated from 5 °C to (a and e) 70 °C or (b and f) 90 °C or rapidly heated from 5 °C to (c and g) 70 °C or (d and h) 90 °C and held for 5 min. Scale bars are 10 μm long.

water. The resulting slurry was passed through a screw-fed strainer (Bibun Machine, Hiroshima, Japan) having a 2.5 mm diameter mesh. Organza cloth was then used to separate excess water from the myofibril extract, and the washing process was repeated twice more. NaCl (0.5%, w/v) was added to the last wash to facilitate subsequent water removal.[18] This refined myofibril preparation was then adjusted to contain the same protein (12%, w/w) and cryoprotectant (4% sucrose, 4% sorbitol, and 0.3% sodium tripolyphosphate, w/w) concentrations as the commercial fish surimi. Straining of the chicken surimi facilitated lowering its collagen content to a level similar to that of the fish surimi (<1%, as analyzed by hydroxyproline determination, AOAC).

Pastes were prepared from each species by combining surimi and water plus other ingredients sufficient to achieve final concentrations of 78% (w/w) water, 2% (w/w) NaCl, and 2% (w/w) porcine plasma (to inhibit possible endogenous protease activity) and then comminuting at 5–10 °C for 10–12 min at 2500 rpm in a Stephan cutter–mixer under vacuum. The pH of chicken surimi paste (initial pH 6.3) was adjusted with $NaHCO_3$ to that of the pollock surimi paste (pH 6.75). Pastes were vacuum-packaged with a Multivac 8941 (Multivac, Inc., Kansas City, MO) to remove as much air as possible. A corner of the evacuated bag containing the paste was cut before placing in a manually operated sausage stuffer for extrusion of the paste into Teflon (for microwave heating) or stainless-steel (for water bath heating) tubes, 1.9 cm inner diameter and 17.8 cm long. Tubes were sealed at both ends with threaded end caps before heating.

Heat Processing of Gels. The filled tubes were heated one of two ways: (a) slowly ramp heated (designated SRH for slow ramp heating) in a programmable (heating rate) water bath at 0.5 °C/min to simulate conventional smokehouse process heating rates to an end point temperature of 70 or 90 °C or (b) rapidly microwave heated at 100 °C/min (at 2450 MHz and 300 W) in an Industrial Microwave

Systems (Morrisville, NC) focused (equal energy distribution) chamber to an end point temperature of 70 or 90 °C and isothermally held at this temperature in a water bath for 5, 10, or 15 min (designated RHH for rapid heating plus isothermal hold). Target end point temperatures were validated as actual temperatures using a Reflex fiber optic thermocouple system (Neoptix, Inc., Fairport, NY). After heating, all gels were immediately placed in plastic bags, which were pressed to remove headspace air prior to sealing and then cooled in an ice water bath. Gels were removed from their tubes after they were cooled. At least three replicates of each gel treatment were provided for each method described below.

Gel Cook Loss (CL) and Expressible Water (EW). Loss of moisture during cooking of gels (CL) by microwave or water bath heating was measured in triplicate by subtracting the post-cooked weight of gels from the pre-cooked weight of the paste, expressed as a percentage of pre-cooked paste weight. Specimens were blotted with paper tissues before weighing and ice-bath cooling. The EW of each gel was measured in triplicate using the microcentrifuge-based method by Kocher and Foegeding.[19] The center of each gel was cut into 10 × 4.8 mm cylinder specimens using a cork borer, and these specimens were placed into the microcentrifuge filtration unit, which was comprised of a 2.0 mL microcentrifuge tube (Beckman Instruments, Inc., Palo Alto, CA) that collected moisture released through the bottom mesh of an inner tube containing the specimen. Specimens were centrifuged at 10000*g* for 10 min.[20] EW was calculated by subtracting the water expressed during centrifugation from the specimen weight, expressed as a percentage of the pre-centrifuged specimen weight.

TD-NMR. TD-NMR T_2 relaxation measurements were performed using a pulsed ^{1}H TD-NMR spectrometer (Minispec mq-20, Bruker Corp., Billerica, MA) with a magnetic field strength of 0.47 T, corresponding to a proton resonance frequency of 20 MHz. The instrument was equipped with a 10 mm probe. All hydrogen nuclei were excited by the Carr−Purcell−Meiboom−Gill (CPMG) radio frequency pulse sequence. These nuclei relaxed to their original state once the pulse is stopped, inducing voltage to the instrument receiver coil and observed as the NMR signal. All T_2 measurements were performed with a 40 μs time delay between the 90° and 180° pulses, a total of four scans, and a recycle delay of 15 s.

Core samples of gels were placed in NMR tubes that were capped and equilibrated at 5 °C in a dry air bath for 30−60 min. All experiments were performed at this temperature. Three aliquots of each gel type were analyzed to account for batch variation. The intensities and relaxation times of water pools were determined by analyzing the NMR relaxation data using the unbiased inverse Laplace transform with the CONTIN algorithm.[21]

Scanning Electron Microscopy (SEM) and Image Analysis. The center of each gel was cut out and shaped into a rectangular prism ca. 0.2 × 0.2 × 0.5 mm, and these specimens were frozen in liquid nitrogen, such that the possibility of ice crystal growth was minimized by immersing such small samples into the freezing medium. These samples were quickly taken out and fractured with a chilled stainless-steel scalpel knife and transferred to a 4.5 L benchtop Labconco (Kansas City, MO) freeze-dryer. Samples were freeze-dried at −100 °C or lower and 1000 mmHg for 24 h. These were then mounted on specimen stubs with colloidal silver, sputter-coated with gold−palladium, and imaged with a scanning electron microscope.

Pore diameters were measured using ImageJ image analysis freeware.[22] To minimize human bias while selecting pores for measurements, a 10 × 10 μm grid was overlaid on top of each micrograph and only the pores situated at grid intersections were selected and measured. Pores were identified as circular interstitial spaces between protein strands, and care was taken to not measure pores that appeared to be slanted along the *z* axis. At least two replicates from each gel treatment were taken. This procedure was repeated in three or four micrographs taken from separate fields for each gel; a total of 7−42 pores were identified in each micrograph, summating to a total of about 50−150 pores per gel.

Statistical Analysis of Data. Statistical analyses were performed using SigmaPlot, version 11.0 (Systat Software, Inc., Erkratch, Germany). Correlations were computed using the Pearson product moment correlation. Differences between interaction means were analyzed by individual, pre-planned *t* tests. Simple linear regression was also computed using SigmaPlot.

RESULTS AND DISCUSSION

Microstructure. Mean pore diameters ranged from 3 to 10 μm. Previous reports that have suggested relationships between pore size and WH properties of food protein gels have not analyzed pore size quantitatively.[20,23−25]

Mean pore diameters of either RHH or SRH chicken gels heated to 70 °C were smaller than those heated to 90 °C (both $p < 0.001$; Figures 1 and 2). Hermansson and Lucisano[26]

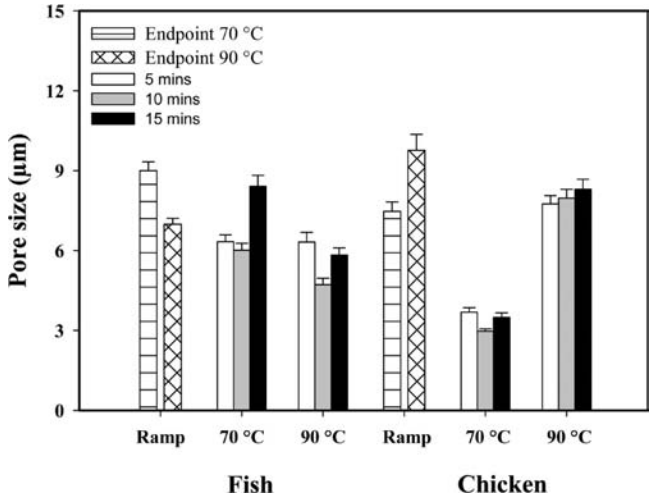

Figure 2. Average pore diameters of fish and chicken surimi gels (after cooling to 25 °C) either rapidly heated to and held isothermally at 70 or 90 °C for 5, 10, or 15 min or ramp heated (0.5 °C/min, starting at 5−10 °C) just to 70 or 90 °C. Bars represent standard errors.

similarly reported increased coarseness of blood plasma gel structure when heated to 92 °C, as compared to only 77 °C. Contrary to this, smaller or similar diameter pores were produced in fish gels at the higher heating temperature, for both heating regimes (panels e−h of Figure 1 and Figure 2). Park et al.[27] reported increases in apparent pore diameter during heating of fish surimi gels to an end point temperature of 90 °C; however, their assessment of pore size was qualitative and based on light micrographs. For both species, the SRH heating regime tended to produce gels with somewhat larger pores than those of RHH gels (each $p < 0.001$).

No significant trends in the mean pore size of either chicken or fish RHH gels were noted because of holding time at either temperature (both $p > 0.05$).

Barbut et al.[28] reported that raw meat batters already display an organized gel structure prior to cooking and that this structure is reinforced (thickened) by protein cross-linking induced during heating, resulting in apparent decreases in the pore diameter as the temperature is increased. It should be noted however that their heating protocol did not exceed 70 °C nor were rapid heating treatments compared nor were WH properties measured. If their observations were accurately interpreted, however, it is difficult to envision a mechanism whereby pore size might increase during cooking from 70 to 90 °C. Upon reaching 70 °C or within a very short time thereafter, further denaturation of myofibrillar proteins would seem unlikely. It is therefore not surprising that further heating

time at the end point temperature of RHH gels did not result in changes to pore diameter. There remains, however, no good explanation for why the pore size of RHH chicken gels at 90 °C were higher than those cooked to 70 °C.

WH Properties: CL. Overall, longer holding times at 70 and 90 °C after rapid heating (RHH) correlated with increased CL from gels of both species ($p < 0.12$; $r = 0.409$), as was similarly reported by Riemann et al.[5] for comminuted turkey breast meat but not surimi gels. RHH fish gels exhibited lower CL (both $p < 0.05$; Figure 3) than SRH gels, regardless of the temperature.

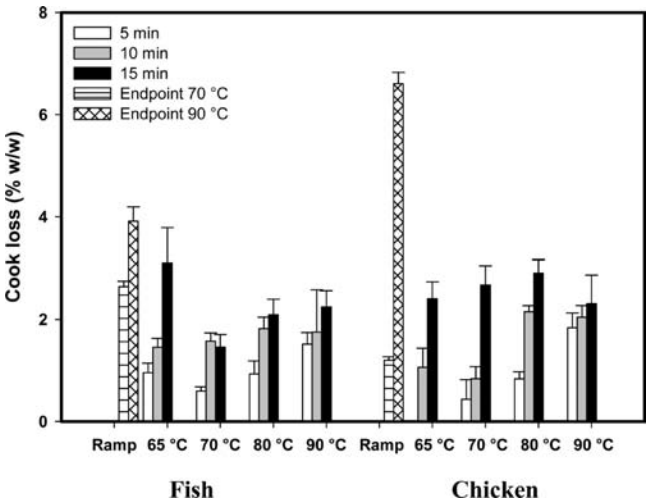

Figure 3. CL from fish and chicken surimi gels (after cooling to 25 °C) either rapidly heated to and held isothermally at the indicated temperatures for 5, 10, or 15 min or ramp heated (0.5 °C/min, starting at 5–10 °C) just to 70 or 90 °C. Bars represent standard errors.

The CL was also lower for RHH chicken gels at 90 °C ($p < 0.001$), but CL at 70 °C was especially low for SRH gels at this temperature ($p > 0.05$). It should be noted however that RHH gels at 5 min holding time essentially reached their final gel properties; further holding does not appreciably change the pore size either (Figures 1 and 2). Thus, there would be no incentive to exceed a 5 min holding time if rapid heating were used in commercial practice, and these gels, for both species, experienced no more CL than SRH gels overall.

No significant temperature (70 versus 90 °C) effects on CL were observed for RHH chicken or fish gels when data from all holding times (5, 10, and 15 min) were considered altogether. CL and temperature were however positively correlated for the 5 min holding times for both species (both $p < 0.05$; chicken $r = 0.524$; fish $r = 0.442$). Because the pore size did not increase with the holding time of RHH gels, this greater CL with increasing holding time may be merely a manifestation of the tortuous path within the gel that water must take to exit the gel, as explained by Darcy's law.

CL also increased at the higher end point temperature (90 °C) for SRH gels from both chicken and fish, most remarkably for chicken. Stangierski and Kijowski[29] reported a similar trend for chicken myofibril gels cooked to end points of 50–90 °C. Chicken gels were clearly more sensitive to the end point temperature when cooked by the slower conventional method, confirming industrial experience that end point temperatures above 70 °C may lead to excessive yield losses.

Interestingly, CL of RHH gels from both species was within a similar range, and CL of SRH fish gels was also within this same general range. However, CL of SRH chicken gels heated to 70 °C was markedly lower than that for SRH fish gels, and CL of chicken gels was much higher than SRH fish gels heated to 90 °C.

CL poorly correlated with the mean pore diameter when analyzed over all gels (Figure 4; $p = 0.12$; $r = 0.717$). This

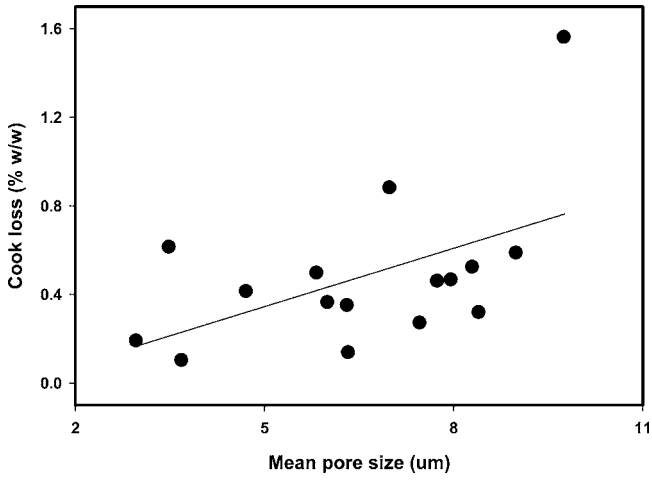

Figure 4. Effect of the mean pore size on CL of both chicken and fish gels ($p = 0.12$; $R^2 = 0.717$). Data include both rapidly and slowly heated gels.

relationship between pore size and CL was not statistically significant when analyzed by species or heating rate. While this result may seem not to support a capillary explanation for WH, as previously mentioned, we know that in certain cases (RHH gels held for longer times at end point temperatures) CL was clearly not correlated to change in gel pore diameter and the increasing CL could be explained by another mechanism (Darcy's law). Also, it should be noted that, in the present study, CL only varied approximately 1–7% and pore size only varied from 3 to 10 μm; perhaps the range of variation in pore size and CL was insufficient to provide evidence supporting capillarity as a main WH mechanism in meat gels. Certainly future work should not only seek to compare gels varying more widely in CL and pore size but additionally consider other parameters included in the capillarity hypothesis for explaining WH properties of gels, such as the surface tension of the solution and the contact angle at the polymer–water interface.[12,13]

WH Properties: EW. No clear significant relationships between EW and mean pore diameter of gels were evident either. Probably the variable cook losses sustained by the gels (not all cooked gels had the same moisture content prior to EW testing) additionally confounded this relationship.

EW was remarkably higher for RHH chicken gels at 90 °C and held for 15 min (Figure 5), perhaps reflecting the very high CL that was noted previously for SRH chicken gels to 90 °C (Figure 3). The EW of fish gels was, by contrast to chicken gels, relatively lower and insensitive to effects of the heating rate, end point temperature, or holding time.

Species Comparison. The present data suggest that the WH properties of chicken breast myofibrillar protein gels are more sensitive to end point cooking temperature effects than that of Alaska pollock. However, most fish muscle proteins are relatively more thermally labile, as compared to those from homeotherms,[30] owing to their lower body temperature.[31–34] A

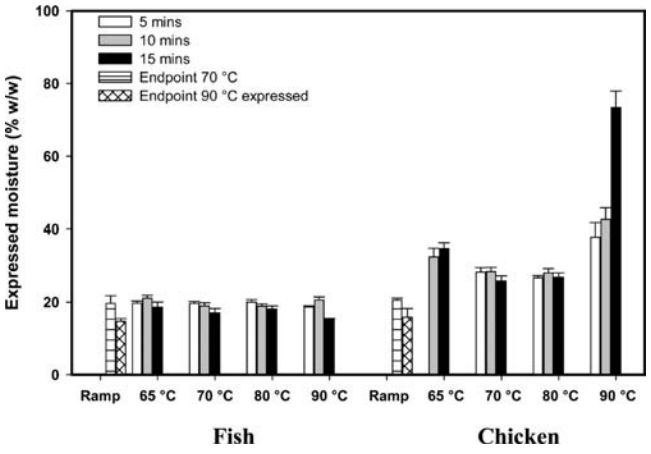

Figure 5. EW from fish and chicken surimi gels (after cooling to 25 °C) either rapidly heated to and held isothermally at the indicated temperatures for 5, 10, or 15 min or ramp heated (0.5 °C/min, starting at 5−10 °C) just to 70 or 90 °C. Bars represent standard errors.

higher cooking temperature seemed to result in chicken gels of larger mean pore diameter and correspondingly higher CL and EW (SRH and RHH gels, respectively). In contrast, higher cooking temperatures resulted in fish gels exhibiting pores of smaller diameter and less differences in CL and EW. Because conditions of the protein concentration, ionic strength, and pH were held constant for pastes from both species used in this study, these differences in gelling behavior may arise from species differences in myosin isoforms.[6] Muscle fiber differences in functionality, such as if chicken thigh meat were compared to breast,[9,35] may also be involved in comparing these two meats. Variations in the gel microstructure among these treatments were not as wide ranging in fish gels as in chicken gels, perhaps because the gelation events occurred sooner and/or at a lower temperature range.[6]

TD-NMR Indications of Water Mobility. When analyzed by distributed continuous curve fitting, each gel exhibited three water pools, T_{2B}, T_{21}, and T_{22}, corresponding to relaxation times of about 10, 100, and 300 ms, respectively (Figure 6). Different water populations within a food, delineated by temporally distinct peaks in TD-NMR data analysis, are commonly referred to as distinct "water pools". EW overall correlated only with shorter T_{22} relaxation times ($p < 0.05$; $r = 0.614$). However, this trend did not hold in a comparison to the T_{22} relaxation times during isothermal holding (comparing Table 1 and Figure 5); thus, it is unclear whether this correlation is meaningful. As previously mentioned, the EW of all gels spanned a relatively narrow range, with the exception of RHH chicken gels at 90 °C, and their differences were confounded by different initial (cooked gel) moisture contents prior to testing, reducing the likelihood for meaningful correlations. However, within each heating regime and temperature (e.g., SRH or RHH at 70 and 90 °C), there was a consistent relationship between EW and both relaxation time and water pool distribution; gels that expressed more water (Figure 5) had relatively smaller T_{21} water pools and larger T_{22} water pools (Table 1). Similarly, Bertram et al.[36,37] and Han et al.[20] reported that the amount of water expressed as gravity-induced drip over time correlated with a higher proportion of the T_{22} water pools prior to this gravimetric testing of water loss.

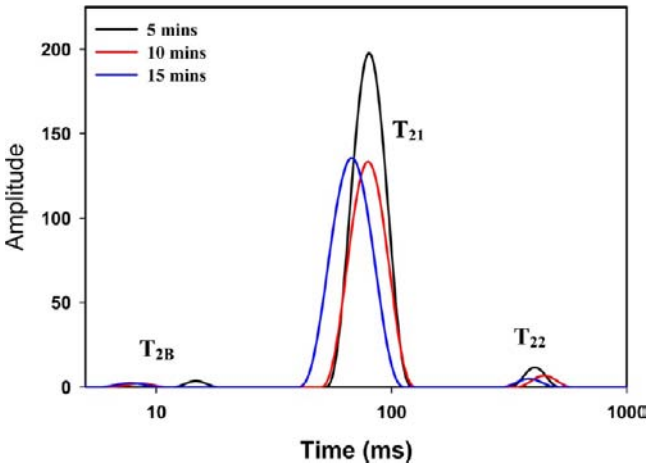

Figure 6. T_2 distribution as computed by the CONTIN algorithm of chicken gels rapidly heated to 70 °C and isothermally held for the indicated times.

Table 1. T_2 Relaxation Times and Water Pool Populations of Chicken and Fish Gels as Computed by the CONTIN Algorithm

	T_{2B} (%)	T_{21} (%)	T_{22} (%)	T_{2B} (ms)	T_{21} (ms)	T_{22} (ms)
			Chicken			
70 °C ramp	0.09	87	12	5	75	450
90 °C ramp	0.13	96	4	10	68	400
70 °C 5 min	0.20	85	15	15	80	410
70 °C 10 min	0.13	85	15	8	79	440
70 °C 15 min	0.14	90	10	8	67	380
90 °C 5 min	0.21	92	8	13	70	370
90 °C 10 min	0.16	93	6	8	72	400
90 °C 15 min	0.15	86	14	10	74	310
			Fish			
70 °C ramp	0.19	79	20	0.11	77	600
90 °C ramp	0.18	90	10	0.13	74	480
70 °C 15 min	0.1	77	23	0.12	71	590
90 °C 15 min	0.25	86	14	0.11	74	400

Gels that evidenced higher CL slightly correlated with increased and decreased values for the T_{21} ($r = 0.56$) and T_{22} ($r = -0.55$) water pools, respectively (both $p = 0.06$) (Table 2). Chicken gels isothermally held at 70 °C are provided in Figure 5 for example. CL correlated with a reduction in T_{21} relaxation times ($p < 0.05$; $r = 0.59$); however, no significant effects were observed with respect to T_{22} relaxation times.

Continuous distributed exponential curve fitting produces a nonbiased spectrum of relaxation components; by this approach, three separate water pools are generally reported for meat gels.[38] Bertram et al.[36] demonstrated that continuous distribution curve fitting explained much more variation in EW of pork meat, as compared to biexponential curve fitting (72 versus 36−56%, respectively). Bertram et al.[38] proposed the following origins of these water pools: the first peak, T_{2B} (~1−10 ms), represents water immobilized (bound) at the protein surface, T_{21} (~50−100 ms) represents a second layer of water around highly organized protein structures (e.g., water within protein tertiary and/or quaternary structures), and T_{22} (~300−400 ms) represents a third layer of water, which is more free in mobility and resides between protein fibers. Similarly in our

Table 2. Correlation Coefficients and Levels of Significance for the Correlations between Either EW or CL and T_2 Time Constants and Corresponding Integrals (Areas), Found by Either Biexponential Fitting or Continuous Distribution Analysis

T_2 parameter	variable	correlation coefficient	level of significance
T_{2B} (ms)	EW (%)	0.52	NS
T_{21} (ms)	EW (%)	0.03	NS
T_{22} (ms)	EW (%)	−0.61	$p < 0.05$
area T_{2B}	EW (%)	−0.01	NS
area T_{21}	EW (%)	0.12	NS
area T_{22}	EW (%)	−0.12	NS
T_{2B} (ms)	CL (%)	−0.07	NS
T_{21} (ms)	CL (%)	−0.59	$p < 0.05$
T_{22} (ms)	CL (%)	−0.09	NS
area T_{2B}	CL (%)	−0.03	NS
area T_{21}	CL (%)	0.56	$p = 0.06$
area T_{22}	CL (%)	−0.55	$p = 0.06$

data, T_{2B} water remained approximately constant across all gel treatments, while differences in the amounts of T_{21} and T_{22} water pools were associated with changes in WH properties EW and CL. The T_{22} water pool appeared to be lost during cooking losses, and subsequently, gels with proportionately larger T_{21} water pools held water more tightly during centrifugation.

Hills et al.[39,40] proposed that proton NMR relaxation times may be interpreted in terms of chemical exchange and molecular diffusion rather than the amounts and types of different types of water structures. This theory would suggest that the T_{2B} peak arises from non-exchanging protons from mobile parts of the biopolymer matrix, the T_{21} peak represents fast proton exchange between such biopolymer protons and water protons, and the T_{22} peak corresponds with a small amount of more mobile water in a few much larger pores in the biopolymer matrix or perhaps residual lipids. Our results support such an interpretation of the T_{2B} peak because it was near constant across all gel treatments, as well as T_{21} because larger amounts of it were associated with better WH properties. The assignment of the T_{2B} peak is less obvious because it most likely does not represent residual lipids because the materials tested in this study were surimis, which possessed very low or essentially negligible lipid contents, nor does is it likely to represent mobile water in a few large pores because the amounts of it did not show a significant correlation with EW.

The relaxation of all water pools in gels is reported to be more rapid than that of unconfined water, and therefore, it would seem logical to conclude that all water in gels is more structured/immobilized than free water. However, Lillford et al.[41] noted that the root-mean-square diffusion distance of water is about 100 μm [$\sigma = (6Dt)^{1/2}$; where the pure water diffusion constant, D, is 10^{-5} cm^{-2} s^{-1} and the diffusion relaxation time is about 2 s], yet pore diameters in protein gels are typically on the order of just 10 μm. This means that, before a "free" water molecule completely relaxes, it should deflect off the pore walls several times, thus encountering the largely immobilized water layer next to pore surfaces. Therefore, Lillford et al.[41] asserted that the varying "water pools" detectable in TD-NMR T_2 spectra rather than indicating water populations varying in mobility (degree of water structuring) merely reflect differences in the mean pore diameter. By this reasoning, a continuous distribution of relaxation times rather than temporally distinct water populations should be expected for meat gels because pore sizes vary considerably within any gel. Bertram et al.[37] reported a relationship between T_{21} relaxation times and sarcomere lengths in intact muscle; however, our results do not support this supposition because pore diameters did not correlate with the sizes of the different T_2 water pools or any T_2 times.

There is a separate body of literature that suggests water is highly structured in cells and gels. Wiggins,[42,43] for example, in studying solute partitioning by micro-osmosis, suggested that two types of water actually may exist in gels: high-density water (HDW), which resides at the hydrophilic surface/water interface, owing to tensile forces exerted by the hydrophilic surface that effectively increases the density of the water, and a low-density water (LDW), which is found at the center of capillaries. Pollack's group offered evidence that this more crystalline water originates at hydrophilic surfaces and extends far toward the center of gel capillaries.[44−46] Evidence of theirs and others[47] also suggests that this "structured" water has a higher density, higher viscosity, and lower mobility than bulk water and excludes solutes, owing to the nature of hydrogen-bond-coordinated packing between lattices. It is yet unclear whether or how such water structuring as described above may be associated with proton relaxometry experiments in protein gels, although this could be a constructive paradigm to further explain WH.

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Notes

The authors declare no competing financial interest.

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