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# Moisture migration in idealized bilayer systems: relationships among water-associated properties, structure, and texture

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## Abstract

The effects of composition and thermal treatment on the water sorption and diffusional properties of idealized protein gels arranged in bilayer configurations were determined; these water binding/migration properties were related to the mechanical characteristics of the gels. Samples were prepared from whey protein concentrate (WPC), they consisted of water:WPC ratios of 1.5 to 5.67, and were thermally set for 20–60 min. Moisture migration rates from samples interfaced with filters were determined, as were moisture sorption capacities of samples immersed in water. The physical properties of the gels were assessed by uniaxial compression and microscopy. Results showed that gel strength and consequent extent of protein interaction—as affected by thermal treatment—controlled the ability of the gel structure to absorb water. Sorption was exponentially correlated with gel modulus and linearly correlated with a function of protein content, heating time, and immersion time. Rates of diffusion from interfaced gels were dependent solely on water content. It was concluded that the degree of protein interaction, whether influenced by concentration or thermal treatment, affected network extensibility and thus the capacity of the gels to act as receptors of moisture. Results have implications for the functionality of shelf-stable sandwiches and other multicomponent foods. © 1998 Elsevier Science Ltd. All rights reserved.

## 1. Introduction

Shelf-stable bilayer or multicomponent foods are becoming increasingly popular to both industry and the military. New army ration items under development include several sandwich-type products in which individual ingredients (i.e. bread/crust or filling) might be formulated to have different levels of water and/or water activity or in which the filling itself might consist of more than one component. Such nonuniformity will provide a driving force for migration and redistribution of moisture and other mobile ingredients, which can compromise the quality and stability of these products. Water binding is, furthermore, influenced by thermal treatment (i.e. baking time) through effects on macromolecular association.

In this study, the effects of process and formulation parameters on the water binding, water sorption, physical, and microstructural properties of a model proteinaceous system, consisting of whey protein concentrate

(WPC) gels interfaced with higher-moisture and lower-moisture constituents, were investigated. Solutions of whey protein, which is composed largely of beta-lactoglobulin and alpha-lactalbumin, form heat-set gels as a consequence of denaturation and aggregation of the protein molecules (Langley & Green, 1989). The extent and nature of intermolecular associations affect the microstructure and, consequently, the texture of the gels. These associations involve disulfide bonds (Katsuta & Kinsella, 1990; Watanabe & Klostermeyer, 1976; Schmidt et al., 1979; Kitabatake et al., 1985; Mangino et al., 1987; Shimada & Cheftel, 1989), ionic interactions (Schmidt et al., 1979; Johns & Ennis, 1981; deWit, 1981; Sone et al., 1983; Varunsatian et al., 1983; Mulvihill & Kinsella, 1988; Kohnhorst & Mangino, 1985), and hydrogen bonding and hydrophobic interactions (Kohnhorst & Mangino, 1985; Voutsinas et al., 1983; Oakenfull, 1987; Kinsella & Whitehead, 1989; Katsuka et al., 1990).

The physical and functional properties of WPC (and other) gels are influenced by many factors. Chief among

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these are protein level, in that a greater concentration of protein generally provides for a more tightly connected and stronger gel network (Katsuta et al., 1990; Tang et al., 1993; Ross-Murphy, 1991), and conditions of preparation that influence macromolecular structure and interaction. In addition to thermal treatment, which governs extent of denaturation and association through variation of heating temperature (Katsuta & Kinsella, 1990; Tang et al., 1993; Donovan & Mulvihill, 1987), heating time (Tang et al., 1993) or heating rate (Stading et al., 1993), pH also strongly determines macromolecular conformation and ability to bind water (Taylor et al., 1994; Langton & Hermansson, 1993).

Reported relationships between water-associated properties and physical properties or process parameters in milk-protein gel systems have not been generally consistent [e.g. no relationship between water-holding capacity and firmness in WPC gels (Karleskind et al., 1995); both decreased syneresis and increased hardness in calcium caseinate gels due to increased solids concentration (Konstance, 1993); decreased water binding in cheese due to increased thermal treatment (Marchesseau & Cuq, 1995); and differences in the ways structural changes can affect water binding and texture (Hermansson & Lucisano, 1982)]. More important, most studies of water holding properties of food gels have involved moisture being released from, rather than absorbed into, the gel. Such investigations have typically employed non-passive movement, such as that driven by centrifugation (Karleskind et al., 1995; Kocher & Foegeding, 1993) or expression (Konstance, 1993; Woodward & Cotterill, 1987; Chung & Lee, 1991), rather than interfacial transport of water. However, moisture release by interfacial diffusion was used to assess relative water-binding properties of processed eggs by Barrett et al. (1997); furthermore, NMR-assessed diffusion within concentrated starch gels was measured by Ohtsuka et al. (1994).

In this study, gel structure was varied through systematic adjustment of heating time and protein content. Relative tendencies of the gels to gain water through sorption and to lose water through diffusion were assessed. Moisture sorption properties were determined through immersion experiments in which water/protein ratios were measured over several intervals; a similar technique has been employed to assess the swelling behavior of polymer gels (Inomata et al., 1994; Yoshida et al., 1996). Tendency to release moisture through passive diffusion was evaluated by tests in which gel specimens were interfaced with filter paper; forced moisture-release properties were measured by centrifugation. The mechanical properties of the gels were evaluated by uniaxial compression tests, which provided measurements of modulus and elasticity. Selected samples were also evaluated by light and scanning electron microscopy. Diffusion and sorption properties were related to the texture and microstructure of the samples.

## 2. Materials and methods

### 2.1. Sample preparation

WPC gels (whey protein concentrate Alacen 878, 76.5% protein, New Zealand Milk Products, Santa Rosa, CA) with water:WPC ratios (by weight) of 1.5, 1.86, 2.33, 3, 4, and 5.67 (respectively: 40, 35, 30, 25, 20, and 15% solids content) were prepared by mixing the components in a stomacher and heating 200 mL of the slurry (measured pH was 6.5) in covered 10×15 cm plastic trays for 20, 40, 60, and 80 min in an 80°C water bath. Highly concentrated gels (%WPC > 30%) were maintained at room temperature for 1–2 h prior to heating, in order to allow maximal hydration of protein. Gel height in the trays was approximately 1.5 cm.

### 2.2. Passive diffusion tests

Water release by passive diffusion was measured in a manner similar to that of Barrett et al. (1997). Sections (3×3×1.5 cm) of each gel (sample weight, 13–14 g) were interfaced with weighed 1 cm diameter circles (approximately 0.2 g each) of Whatman 54 filter paper (the source of moisture for diffusion was far in excess of that actually transferred during the experiment and was thus considered invariant). The assemblies were wrapped tightly in plastic and placed filter-side down.

Water released by the gel and absorbed per weight of filter paper (dry basis filter moisture content) was calculated at six evenly-spaced timed intervals—which varied between 5 and 10 min, depending on gel moisture content. Diffusion tests were conducted at room temperature (22°C) and in triplicate. Data obtained at 30 min contact time for each sample were averaged across all heating times so that the influence of gel protein concentration on moisture migration properties could be determined. In order to assess the effect of gel heating time on moisture uptake by the filter paper, the data for each gel were fitted to the model:

$$FMC = a + b(\text{contact time}) + c(\text{gel heating time}) \quad (1)$$

where  $FMC$  = dry-basis filter moisture content, and  $a$ ,  $b$ , and  $c$  are fitted coefficients. Student  $t$ -ratio and probability values obtained from linear regression using a Minitab statistical program were used to determine the significance of heating time on moisture migration rate;  $t$  values greater than 2 and less than  $-2$  were taken to indicate parameter significance, as given by Box et al. (1978).

### 2.3. Centrifugation tests

Gels with water:WPC ratios of 3, 4, and 5.67 (respectively: 25, 20, and 15% solids content) were

cored and sectioned into 2 mm diameter×5 mm high cylinders. Weighed samples were placed in 1.5 mL microcentrifuge tubes containing a permeable mesh through which expressed water could pass, similar to the procedure of Kocher and Foegeding (1993). Samples were centrifuged in an Eppendorf Microcentrifuge 5414 (15 600 *g*) for 15 min and the samples reweighed. Percent expressible moisture was calculated as  $100 \times \text{expressed water} / \text{total water in the sample}$ .

#### 2.4. Immersion tests

Gels with water:WPC ratios of 1.5, 2.33, and 4 (respectively: 40, 30, and 20% solids content) were cored and trimmed into 1.5 cm diameter×1.5 cm height cylindrical sections. Five sections of each gel were weighed and immersed in an excess of distilled water maintained at room temperature (22°C). The samples were removed, blotted dry, and weighed at 1, 2, 5, 7, 9, 12, and 15-day intervals.

#### 2.5. Microscopy

Scanning electron and phase-contrast light micrographs were taken of 1.5 water:WPC ratio gels (40% solids concentration) heated for 20, 40, and 60 min. Samples were freeze dried and sputter-coated with approximately 100 nm gold palladium prior to analysis on a Zeiss CSM 950 scanning electron microscope. Light microscopy specimens were sliced into thin, transparent sections using a razor, treated by application of acetic orcein, and observed using phase contrast optics, a procedure similar to that employed by Cohen and Segars (1991). Polaroid film types 52 and 55 P/N were used for scanning electron and light microscopy, respectively.

#### 2.6. Compression tests

Gel texture was evaluated by uniaxial compression tests. Gels with water:WPC ratios of 1.5, 2.33, and 4 (respectively: 40, 30, and 20% solids content) were cored into 1 cm diameter sections. Samples were trimmed to 1 cm height and compressed on a Texture Technologies Texture Press at a rate of 0.2 mm/s to 50% deformation. Force-deformation data were automatically acquired by a Zenith 286 computer and subsequently converted to stress-strain units. The modulus determination method used to quantify gel strength accommodated the increase in sample diameter during compression according to the procedure described by Nussinovitch et al. (1990):

$$E = \text{corrected stress/corrected strain} = \frac{F(t)(L_0 - \Delta L(t)) / A_0 L_0}{[\ln(L_0 / (L_0 - \Delta L(t)))]} \quad (2)$$

where  $E$  is deformability modulus,  $F$  is force,  $\Delta L$  is deformation,  $L_0$  is initial sample height,  $A_0$  is initial sample cross sectional area, and  $t$  is time. Mathematical transformations were accomplished using Minitab statistical software, and samples were compressed in triplicate.

Gel elasticity, or ability to rebound after deformation, was evaluated by percent recoverable work measurements, according to the procedure described by Nussinovitch et al. (1990). Samples were sectioned and compressed as described for modulus determinations, then decompressed using the same crosshead speed. Percentage recoverable work (%*RW*) was calculated as:

$$\%RW = 100 \times \frac{\text{area under decompression curve}}{\text{area under compression curve}} \quad (3)$$

Compression and decompression curves were automatically integrated by the TXT2. Samples were compressed in triplicate.

### 3. Results and discussion

Gel properties and functionality varied extensively with protein concentration and thermal treatment. Mechanical effects, as expected, included increased compressive resistance with both increased protein level and heating time; increased network interaction due to prolonged heating was confirmed by microscopy. Water release properties were primarily affected by protein content, and were not significantly influenced by increased association due to thermal treatment. Sorption, however, was markedly reduced by increased heating time as well as by increased protein concentration.

#### 3.1. Moisture release properties

##### 3.1.1. Diffusion

Fig. 1 shows representative results (for the 3 water:WPC ratio gel) for measurements of passive diffusion into filter paper. The filter paper continuously absorbed moisture throughout the duration of the test, and the combined data reveal no discernible effect of heating time on moisture migration rate. (It should be noted that the water content of the gels did not vary with heating time.)  $t$ -Ratio values obtained from regression of sorption versus heating time and contact time (Table 1) indicate similar results: in four instances (for 1.5, 3, 4, and 5.67 water:WPC ratio gels),  $t$ -ratios between  $-2$  and  $2$  indicate no significant effect of heating time on moisture migration rate.  $t$ -Ratios are moderately significant in two instances, but show a positive influence of heating time in one case (the 2.33 water:WPC ratio gel) and a negative influence in the

other (the 1.86 water:WPC ratio gel). Moreover, *t*-ratios are randomly distributed with a mean near zero.

While heating time had no clear effect on diffusion from the gels, solids content had a considerable effect on moisture release. Fig. 2, in which data for all heating

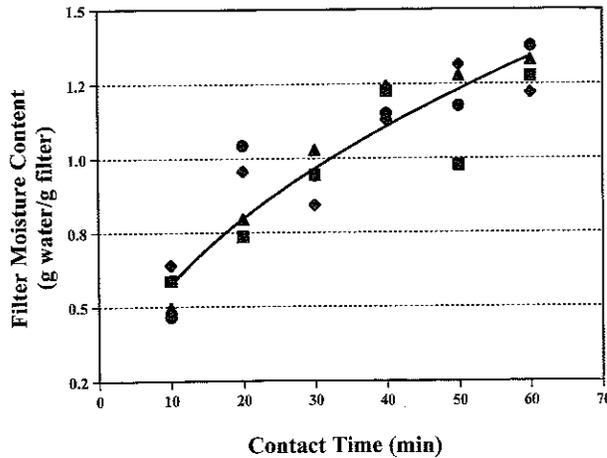


Fig. 1. Migration of moisture from a 3 water:WPC ratio gel into filter paper, expressed as filter moisture content versus contact time. ■, 20 min gel heating time; ◆, 40 min gel heating time; ●, 60 min gel heating time; ▲, 80 min gel heating time. Shows increasing filter moisture content with contact time but no consistent effect of heating

Table 1  
Effect of heating time on moisture migration rate

H <sub>2</sub> O:WPC ratio	<i>t</i> -ratio	<i>p</i>	Statistical effect
1.50	-1.04	0.308	None
1.86	-2.92	0.008	Negative influence
2.33	3.02	0.007	Positive influence
3.00	0.97	0.342	None
4.00	0.65	0.523	None
5.67	-0.66	0.519	None

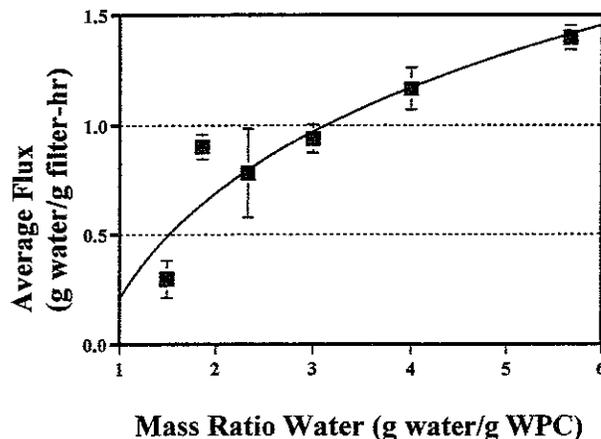


Fig. 2. Average moisture flux into filter paper versus mass ratio of water in the gels, measured after 30 min contact time. Flux measurements are averages for all gel heating times.

times are combined, shows a progressive increase in water diffusion rate into the filter paper, expressed as flux [g H<sub>2</sub>O/g filter-h (based on data at 30 min contact)], with increased water content. The curvature in the dependence is possibly due to partial saturation of filters interfaced with higher moisture gels, which reduced the driving force for diffusion. However, Ohtsuka et al. (1994) reported inhibited diffusion of water in concentrated starch gels—due to a greater number of structural barriers in, and thus decreased permeability of, the polymer network. These authors differentiated between very dilute gels, in which moisture is generally

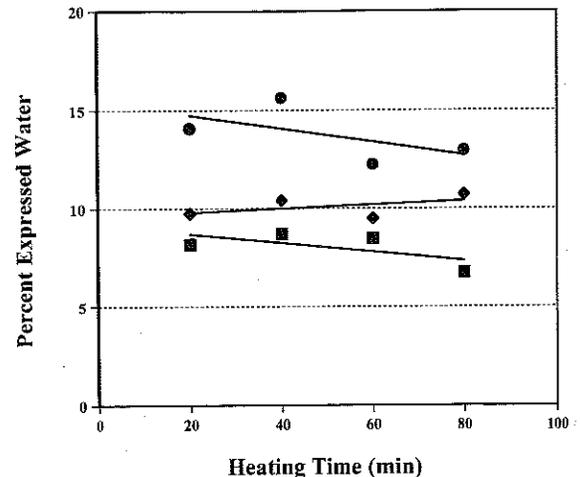


Fig. 3. Percentage expressed water by centrifugation versus gel heating time. ■, 3 water:WPC ratio; ◆, 4 water:WPC ratio; ●, 5.67 water:WPC ratio. Shows increasing percent expressed water with increasing moisture content.

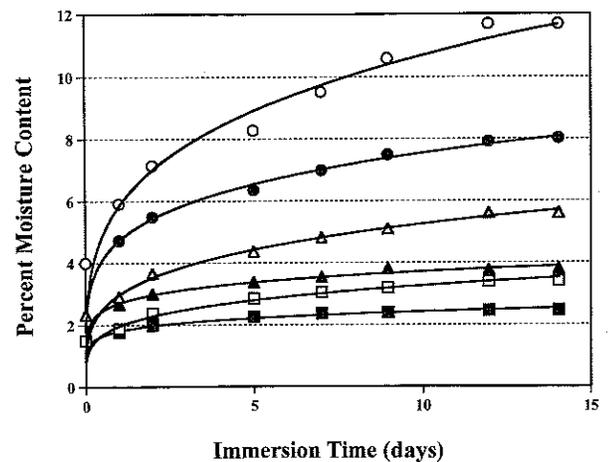


Fig. 4. Sorption into WPC gels immersed in water, expressed as percentage moisture content versus immersion time. ■, □, 1.5 water:WPC ratio gel; ▲, △, 2.33 water:WPC ratio gel; ●, ○, 4 water:WPC ratio gel. Open symbols, 20 min heating time; closed symbols, 80 min heating time. Shows increasing gel water content with immersion time, but a negative influence of heating time on moisture sorption.

'free' and unrestricted by the matrix (Callaghan et al. 1983), and concentrated systems, in which the macromolecular structure inhibits water mobility; they furthermore suggest that a higher concentration of dissolved fragments in gel voids will slow diffusion by increasing viscosity.

Based on data viewed separately at each WPC concentration in this study, changes in protein conformation and association due to thermal treatment did not

sufficiently alter structure to affect moisture transfer by diffusion.

### 3.1.2. Expression

Results from centrifugation experiments (given in terms of percentage water expressed) were similar to those obtained from diffusion tests (Fig. 3). Percentage expressed moisture at each heating time decreased with increasing protein content, but heating time did not

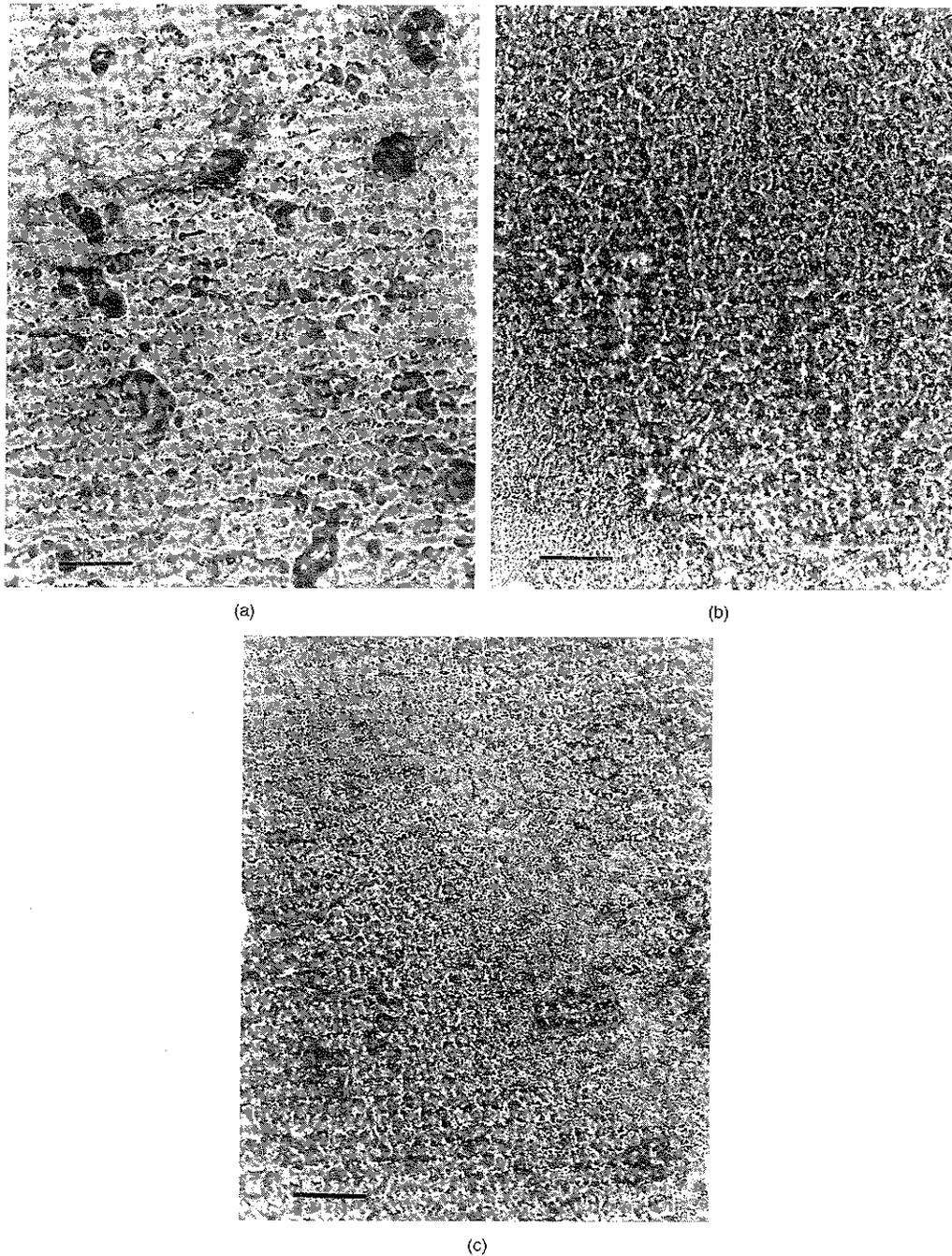


Fig. 5. Light micrographs of 1.5 water:WPC gels. (a) 20 min heating time; (b) 40 min heating time; (c) 60 min heating time. Shows increased network tightening with heating time. Bar = 30  $\mu$ m.

significantly reduce the amount of water released by centrifugation. A slight trend towards decreasing expression with increasing heating time was observed for the 15 and 25% solids content gels; however, while regression of percentage water expressed versus heating time for these systems showed slightly negative slopes,  $r^2$  values and  $t$ -ratios for each relationships were less than 0.15 and 1.9, respectively. Diffusion and centrifugation tests were analogous in that each measured how readily water could be removed by pressure (respectively, capillary and mechanical mechanisms), but differed in that only expression by centrifugation involved deformation of the gel.

### 3.2. Moisture sorption and structure

#### 3.2.1. Sorption

Conversely, heating time had a significant impact on absorption of water into immersed whey protein gel samples. Fig. 4 shows that water absorbed over an extended immersion time by 1.5, 4, and 5.67 water:WPC gels (respectively: 40, 30, and 20% solids content)

decreased progressively with heating time. Heating clearly enhances protein interactions, thereby tightening the structure and limiting extensibility and thus ability to swell with absorbed water. An analogous effect was observed for increasing protein content: higher solids content gels—hence, those with more highly associated structures—absorbed relatively less water. Inhibited sorption into more concentrated gels occurred despite the increased ionic driving force for mass transfer into these samples, due to soluble minerals native to whey protein.

#### 3.2.2. Microstructure and strength

Increased structural association was confirmed by both light and scanning electron micrographs of 1.5 water:WPC ratio (40% solids content) gels (Figs. 5 and 6, respectively). Samples heated for 20, 40, and 60 min clearly show decreased pore size and increased network tightening with increased thermal treatment. [That qualitatively similar conformational changes were observed at two different magnification levels, or 'length scales', suggests a fractal nature to the gel structure (Barrett & Peleg, 1995)].

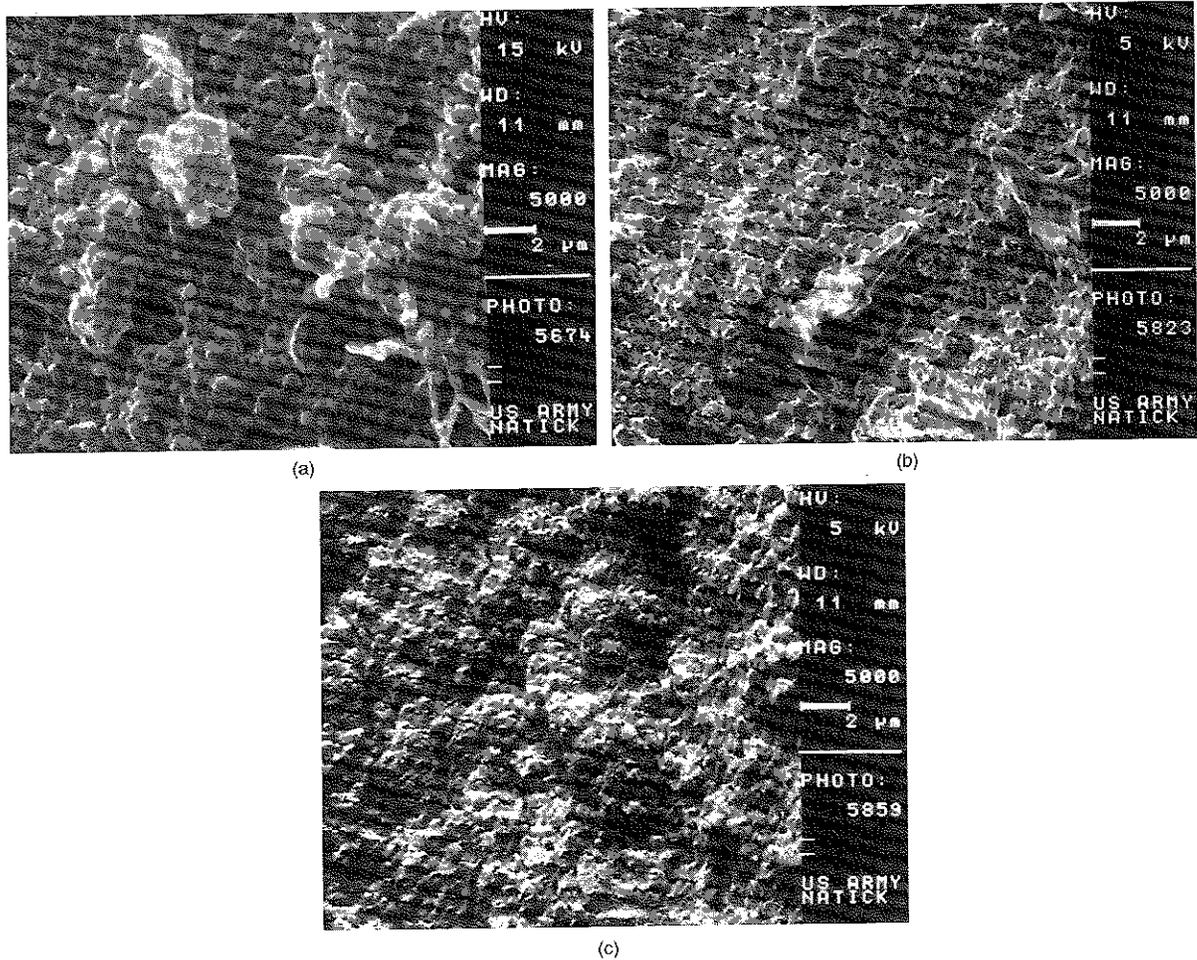


Fig. 6. Scanning electron micrographs of 1.5 water:WPC gels. (a) 20 min heating time; (b) 40 min heating time; (c) 60 min heating time. Shows increased network tightening with heating time.

Accordingly, gel modulus increased linearly with heating time (Fig. 7); modulus, as was expected, also increased with protein concentration. These observations for structural and mechanical effects of prolonged heating are in keeping with those of Schmidt et al. (1984) and of Langley and Green (1989) who reported, respectively, increased aggregation and increased strength of whey protein gels with severity of thermal treatment.

### 3.2.3. Sorption–texture and sorption–process parameter relationships

That the extent of sorption into the gels was highly dependent on the extent of association and connectivity is reflected in the discerned exponential relationship between resulting water/WPC ratio  $[M/P]$ , after each immersion interval, and initial modulus. For each immersion interval, data for all samples (all gel concentrations and heating times) were fitted to the function:

$$M/P = C \exp[-b(\text{initial modulus})] \quad (4)$$

where  $C$  and  $b$  are constants. Regression coefficients are approximately 90% (Table 2). Both  $C$  and  $b$  progres-

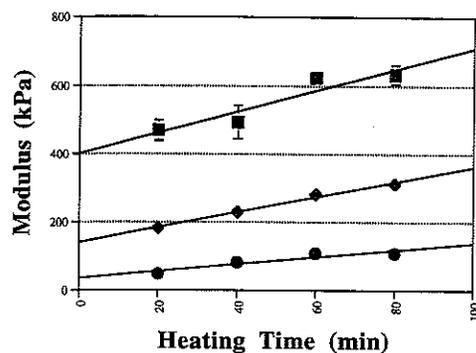


Fig. 7. Modulus versus gel heating time. ■, 1.5 water:WPC ratio gel; ◆, 2.33 water:WPC ratio gel; ●, 4.0 water:WPC ratio gel. Shows progressive increase in modulus with heating time for each gel concentration.

Table 2  
Dependence of water content<sup>a</sup> on initial modulus–regression parameters<sup>b</sup>

Immersion time (days)	$C$	$b (\times 10^3)$	$r^2$
0	1.44	1.87	0.89
1	1.68	2.00	0.88
2	1.87	2.07	0.90
5	2.03	2.12	0.91
7	2.16	2.26	0.90
9	2.24	2.37	0.89
12	2.32	2.45	0.89
14	2.33	2.47	0.89

<sup>a</sup> g water/g WPC.

<sup>b</sup> Water content in terms of water/WPC ratio =  $C \exp(-b(\text{initial modulus}))$ .

sively increase in magnitude with immersion time—illustrating both increased overall sorption and increased exponential dependence of sorption on initial modulus with increasing contact with water (Table 2). For pooled data, sorption is linearly dependent on a function of initial moisture/protein ratio ( $[M/P]_0$ ), heating time in minutes ( $H$ ), and immersion time in days ( $t$ ) by the following relationship:

$$M/P = 0.119 + 0.437[M/P]_0 - 0.000387H + 0.0458t \quad (5)$$

with a regression coefficient of 94%, and  $t$ -ratios of 34, –7, and 17 for the three parameters, respectively.

### 3.2.4. Elasticity

Gel elasticity increased proportionally with water:WPC ratio, as evidenced by increases in percentage of recoverable work (Fig. 8). It is possible that such an effect results from increased ‘swelling’ of water-containing pores in the gel as moisture content is increased. Percent recoverable work did not vary in any consistent manner with heating time, so results for all four heating times are combined in Fig. 8. Elasticity was apparently not a function of other mechanical properties, such as gel strength, that are governed by association and connectivity; results for recoverable work tests are similar to those for diffusion tests in that only total water in the matrix contributed to elasticity—suggesting that physical entrapment of water, rather than macromolecular association in the gel structure, was the dominant influence.

## 4. Conclusions

The overall influences of composition and heating time on the structure and physical properties of whey protein gels are complex and related to the extent of

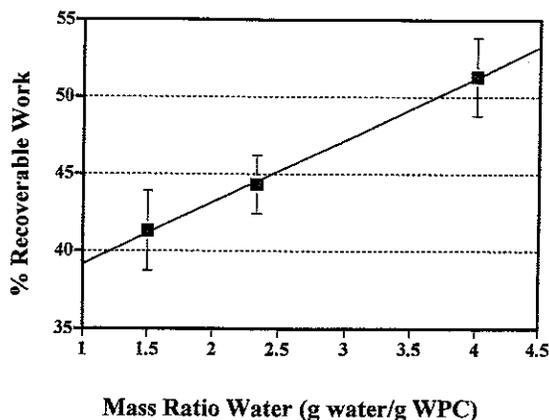


Fig. 8. Percentage recoverable work versus gel water content, with data combined for all gel heating times. Shows increasing elasticity with water content.

protein association and connectivity. Increased heating time significantly influences the mechanical, sorption, and microstructural characteristics of the gels, but not the water-release properties. Increased protein-protein association increases mechanical strength and decreases extensibility, thus decreasing the ability of the gel to expand or swell with absorbed moisture. Binding of water, and thus its release through diffusion, varies with protein concentration, but is not significantly affected by heating time. Moisture flux shows a non-linear relationship with water content, which may be reflective of inhibited diffusion in concentrated gel systems. These results have implications for the stability of gel-structured foods used in bilayer systems (for example, cheeses and processed meats)—in that macromolecular association will influence the ability of a gel network to bind and transport moisture.

## References

- Barrett, A. H., Cardello, A. V., Prakash, A., Mair, L., Leshner, L. L., & Taub, I. A. (1997). *Journal of Food Processing and Preservation*, 21, 225–244.
- Barrett, A. H., & Peleg, M. (1995). *Lebensm. -Wiss. u. -Tech.*, 28, 553–563.
- Box, G. E., Hunter, W. G., & Hunter, J. S. (1978). *Statistics for experimenters*. John Wiley & Sons.
- Callaghan, P. T., Jolley, K. W., Lelievre, J., & King, R. B. K. (1983). *Journal of Colloid Interface Science*, 92, 332–337.
- Chung, K. H., & Lee, C. M. (1991). *Journal of Food Science*, 56, 1263–1266.
- Cohen, S. H., & Segars, R. A. (1991). *Scanning Microscopy*, 5, 363–378.
- Donovan, M., & Mulvihill D. M. (1987). *Irish Journal of Food Science and Technology*, 11, 87–100.
- Hermansson, A. M., & Lucisano, M. (1982). *Journal of Food Science*, 47, 1955–1959, 1964.
- Inomata, H., Nagahama, K., & Saito, S. (1994). *Macromolecules*, 27, 6459–6464.
- Johns, J. E. M., & Ennis, B. M. (1981). *New Zealand Journal of Dairy Science*, 16, 79–83.
- Karleskind, D., Laye, I., Mei, F. J., & Morr, C. V. (1995). *Journal of Food Science*, 60, 731–737, 741.
- Katsuta, K., & Kinsella, J. E. (1990). *Journal of Food Science*, 55, 1296–1299.
- Katsuta, K., Rector, D. J., & Kinsella, J. E. (1990). *Journal of Food Science*, 55, 516–521.
- Kinsella, J. E., & Whitehead, D. M. (1989). *Advances in Food and Nutrition Research*, 33, 343–350.
- Kitabatake, N., Cuq, J. L., & Cheftel, J. C. (1985). *Journal of Agricultural and Food Chemistry*, 33, 125–129.
- Kocher, P. N., & Foegeding, E. A. (1993). *Journal of Food Science*, 58, 1040–1046.
- Kohnhorst, A. L., & Mangino, M. E. (1985). *Journal of Food Science*, 50, 1403–1405.
- Konstance, R. P. (1993). *Journal of Dairy Science*, 76, 3317–3326.
- Langley, K. R., & Green, M. L. (1989). *Journal of Dairy Research*, 56, 275–284.
- Langton, M., & Hermansson, A. M. (1993). *Food Hydrocolloids*, 5, 523–539.
- Mangino, M. E., Kim, J. H., Dunkerly, J. A., & Zadow, J. G. (1987). *Food Hydrocolloids*, 1, 277–282.
- Marchesseau, S., & Cuq, J. L. (1995). *Journal of Dairy Research*, 62, 479–489.
- Mulvihill, D. M., & Kinsella, J. E. (1988). *Journal of Food Science*, 53, 231–234.
- Nussinovitch, A. A., Normand, M. D., & Peleg, M. (1990). *Journal of Texture Studies*, 21, 37–43.
- Oakenfull, D. (1987). *Critical Reviews in Food Science and Nutrition*, 26, 1–12.
- Ohtsuka, A., Watanabe, T., & Suzuki, T. (1994). *Carbohydrate Polymer*, 25, 95–100.
- Ross-Murphy, S. B. (1991). In D. DeRossi, K. Kajiwara, Y. Osada, & A. Yamauchi (Eds.), *Polymer gels: fundamentals and biomedical applications*, (pp. 21–39). New York: Plenum Press.
- Schmidt, R. H., Illingworth, B. L., Deng, J. C., & Cornell, J. A. (1979). *Journal of Agricultural and Food Chemistry*, 27, 529–534.
- Schmidt, R. H., Packard, V. S., & Morris, H. A. (1984). *Journal of Dairy Science*, 67, 2723–2733.
- Shimada, K., & Cheftel, J. C. (1989). *Journal of Agricultural Food Chemistry*, 37, 161–168.
- Sone, T., Dosako, S., & Kimura, T. (1983). In *Instrumental analysis of foods*, Vol. 2. (pp. 209–223). Orlando, FL: Academic Press.
- Stading, M., Langton, M., & Hermansson, A. M. (1993). *Food Hydrocolloids*, 7, 195–212.
- Tang, Q., McCarthy O. J., & Munro, P. A. (1993). *Journal of Dairy Research*, 60, 543–555.
- Taylor, S. M., Gladden, L. F., & Fryer, P. J. (1994). *Journal of Dairy Research*, 61, 71–81.
- Varunsatian, S., Watanabe, K., Hayakawa, S., & Nakamura, R. (1983). *Journal of Food Science*, 48, 42–45.
- Voutsinas, L. P., Nakai, S., & Harwalkar, V. R. (1983). *Canadian Institute of Food Science Technology Journal*, 16, 185–190.
- Watanabe, K., & Klostermeyer, H. (1976). *Journal of Dairy Research*, 43, 411–417.
- de Wit, J. N. (1981). *Netherlands Milk & Dairy Journal*, 35, 47–51.
- Woodward, S. A., & Cotterill, O. J. (1987). *Journal of Food Science*, 52, 68–74.
- Yoshida, M., Safranj, A., & Omichi, H. (1996). *Macromolecules*, 29, 2321–2323.