



Properties of some edible carbohydrate polymer coatings for potential use in osmotic dehydration

Wayne Camirand,^{a*} John M. Krochta,^b Attila E. Pavlath,^a Dominic Wong^a & Margaret E. Cole^a

^aUnited States Department of Agriculture, Agricultural Research Service, Western Regional Research Center, Process Chemistry and Engineering, 800 Buchanan St., Albany, California, 94710, USA

^bDepartment of Food Science and Technology, University of California, Davis, California, 95616, USA

(Received 7 July 1990; revised version received 15 October 1990; accepted 31 October 1990)

A limited number of carbohydrate polymer coating compositions were evaluated for their potential usefulness for improving the osmotic dehydration of foods. A parameter P_r , or 'performance ratio', defined as (water out)/(solute in) (wt/wt), was found to be a valuable index for comparing literature results of quality judgements of coated and non-coated, osmotically dehydrated foods. Use of a model system with a diffusion cell to obtain P_r values partially eliminated the need for conducting large-scale experiments with each food, osmotic agent and coating. Three osmotic agents (69% sucrose, 96% glycerol, 51.6% dextrose) were tested in combination with ten calcium crosslinked polysaccharide ionotropic gel coatings and two coatings produced by evaporation of ethyl cellulose in ethanol. When glycerol was used as an osmotic agent, the highest P_r values were obtained with an ethyl cellulose coating. When a calcium ion crosslinked polymer coating was used, the greatest P_r values were obtained with 69% sucrose as the osmotic agent and aqueous 3% low-methoxyl pectin solution in the coating.

INTRODUCTION

In osmotic drying, food pieces are mixed with an osmotic medium, for example, sucrose or sugar syrup, and the mixture is allowed to stand. Diffusion of water from food pieces into the surrounding medium results in product dehydration. Properties of food tissues and the medium or osmotic agent can considerably influence drying behavior and the resulting product quality (Lerici *et al.*, 1985).

In such a system the ratio of solid food to osmotic agent (wt/wt), approximation to equilibrium, initial osmotic pressure differential between the food and the osmotic agent, and the relative diffusion rates of water and solute determine the extent of water loss and weight loss. The ratio of 'protopectin/soluble pectin' within fruit tissues may also determine suitability of the food for osmotic dehydration (Forni *et al.*, 1986).

Most past research on osmotic dehydration focused

on fruit where the osmotic dehydration step was used as an intermediate process prior to thermal drying, freezing and freeze-drying (Ponting *et al.*, 1966; Farkas & Lazar, 1969; Dixon & Jen, 1977; Hawkes & Flink, 1978); however, more recently a higher water product was processed with vacuum-packaging and pasteurization (Maltini & Torreggiani, 1986).

In 1968 we found that in some instances a more complete osmotic dehydration of food products was obtained by coating the food pieces with a water-permeable polymeric material prior to osmotic treatment (Camirand *et al.*, 1968; Camirand & Forrey, 1969). Near equilibrium the weight loss and the ratio (water loss (g)):(solute gained (g)) were greater with the coated food than with the same non-coated food.

Little additional research has been done on this process, hereafter referred to as the OSMEMB (osmotic membrane) process. Potential advantages of the OSMEMB process are greater water loss, final product preservation without supplemental drying (e.g. thermal or vacuum), reduced loss of desired constituents

*To whom correspondence should be addressed.

(e.g. colorants, flavor components and nutrients), less solute diffused into the food, improved product integrity, lower levels of oxygen and microbial contamination, greater aesthetic appeal (particularly when clear polysaccharide coatings are used) and a possible source of dietary fiber.

Intermediate moisture food (IMF) can be defined as any food with a water activity (A_w) between 0.65 and 0.90 (Guilbert, 1988). Researchers have suggested that IMF products can be used with edible films and coatings containing beneficial functional ingredients such as antimicrobials, antioxidants, carriers of pH gradients, flavors, colors and preservatives (Guilbert, 1988; Karel & Langer, 1988; Vojdani & Torres, 1989; Kester & Fennema, 1986). Furthermore, initially a food could be coated for use in the OSMEMB process and then coated with another mixture that contains slow-release agents and/or water-barrier coatings (Wong *et al.*, 1990; Krochta *et al.*, 1988).

Potential of carbohydrate polymers

Edible films and coatings have been prepared from a variety of polysaccharides, proteins, lipid materials and composites consisting of a blend of polysaccharides, proteins and/or lipids (Kester & Fennema, 1986). For the purposes of the OSMEMB process, edible coatings should have the following properties: good mechanical strength (gel strength), satisfactory sensory properties, easy and rapid film formation with simple techniques, high water diffusivity, low solute diffusivity and maintenance of the coatings in an intact state after formation on a wet surface (as on a raw food piece) without dissolving into the osmotic agent. If we look at edible films and coatings considered in the past, polysaccharides generally would fit these requirements better than protein or lipid films/coatings alone.

Solutions of sodium or potassium alginates and low-methoxyl pectinates or pectates (LMP) form strong ionic gels when crosslinked with calcium ions. Therefore, strong coatings or films can be easily formed by dipping an object or food piece in a sodium alginate or sodium-LMP solution and then by dipping it in a concentrated calcium salt solution such as calcium chloride. Several polysaccharides that do not form films without an evaporation step were incorporated into a film by forming an interpenetrating polymer network (IPN). This IPN is defined as a combination of two polymers in network form, at least one of which was synthesized and/or crosslinked in the immediate presence of the other (Sperling, 1988). The IPN coatings tested in our experiment were of the type characterized as semi-IPNs where one polymer is crosslinked and the other is linear. The only two well-known crosslinked polysaccharide ion gels that form strong films almost immediately are sodium alginate or sodium pectinate/pectate solutions formed by treat-

ment with calcium salt solutions at the interface. It has been found recently that these polysaccharide ionotropic gels with calcium chloride solutions can produce higher polymer concentrations at the surface than further into the gel (Skjåk-Bræk *et al.*, 1989). Coatings or films that are denser at the surface may improve the diffusion characteristics. These two polysaccharide ionotropic gels, alone or in polycarbohydrate mixtures, were tested in our study. Research reported here is limited to coatings of polysaccharide or polysaccharide mixtures formed below 30°C to help evaluate the potential in the OSMEMB process.

THEORETICAL CONSIDERATIONS

The performance ratio concept

Measurements on osmotically dehydrated food should include both the quantity of water removed from a food and the quantity of osmotic agent which passes through the surface or coating into the food piece. One useful parameter could be defined as the performance ratio (P_r) where $P_r = (\text{g water out})/(\text{g solute in})$.

P_r can be calculated operationally for a food during osmotic dehydration by first determining the initial solids (IS) of the food and initial gross weight (IGW). Then the coating treatments, if required, are added, and the food pieces are submerged in an osmotic agent for a given time. Finally the food pieces are removed, rinsed, drained and patted with a towel. At this point final weight (FW) and final solids (FS) of the food pieces can be determined as shown below.

$$\text{Solids gained} = \frac{\text{FS} - \text{IS}}{\text{IGW}} \times 100\%$$

$$\text{Weight loss} = \frac{\text{IGW} - \text{FW}}{\text{IGW}} \times 100\%$$

$$\text{Water loss} = 1 - \left[\frac{\text{FW} - \text{FS}}{\text{IGW} - \text{IS}} \right] \times 100\%$$

Thus,

$$\begin{aligned} P_r &= \frac{\text{water out}}{\text{solids in}} \\ &= \frac{\text{solids gained (\%)} + \text{weight loss (\%)}}{\text{solids gained (\%)}} \\ &= 1 + \frac{\text{IGW} - \text{FW}}{\text{FS} - \text{IS}} \end{aligned}$$

The P_r is useful for evaluating any osmotically dehydrated food, and some data from the literature have been reinterpreted and plotted as P_r versus

percent water loss (g water loss/100 g original water) with the points also representing a number of parameters (e.g. temperature, time, concentration) as indicated in Figs 1–4. Figures 1 & 2 were derived from the corresponding references, and quality and acceptability statements demonstrate that these fruit products also had the highest P_r s values. Figure 3 was derived

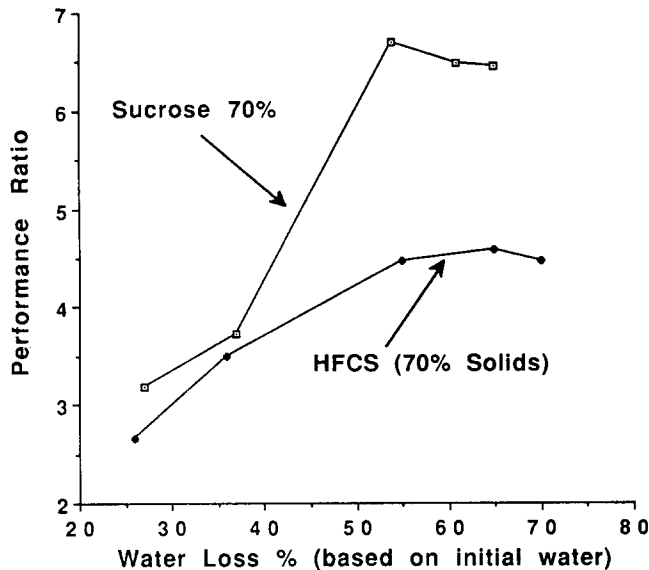


Fig. 1. Osmotic dehydration of water loss (%) from Red Delicious apples at 70°C, versus P_r (ratio: water loss (g) to solids gained (g)). In both curves, 70% sucrose and HFCS (70% solids) represent points at 0.50, 1.00, 3.00, 5.00 and 7.00 h (left to right) (fruit:syrup = 1:4, w/w) (Bolin *et al.*, 1983).

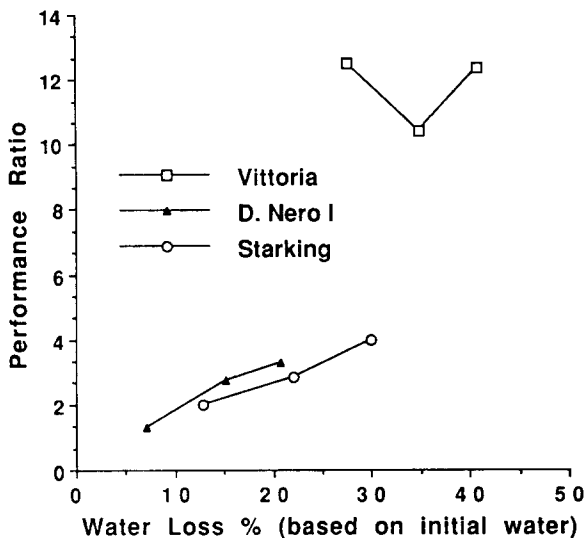


Fig. 2. Osmotic dehydration of water loss (%) from three cultivars of cherries ('Vittoria', 'Durone Nero I', 'Starking') at 25°C, versus P_r . In the three curves, the osmotic agent was a 50% (w/w) of 70° Brix sucrose and a 70° Brix corn syrup (24% glucose, 29% maltose, 12% polysaccharides, 35% water). Points along the curves were taken at 2, 4 and 6 h (left to right) (fruit:syrup = 1:5, w/w) (Torreggiani *et al.*, 1987 and Giangiacomo *et al.*, 1987).

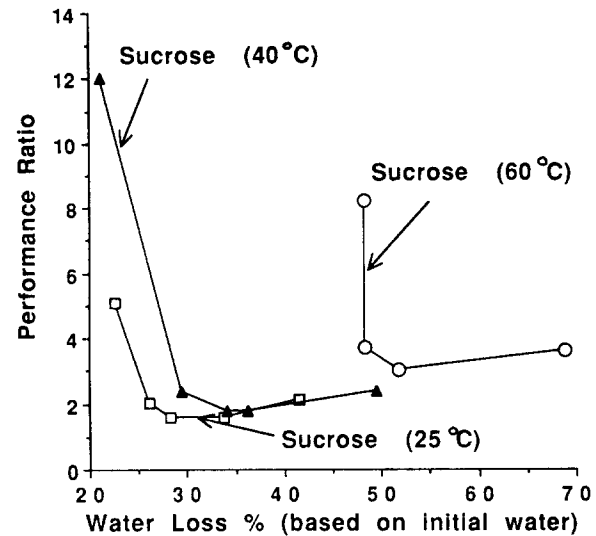


Fig. 3. Osmotic dehydration using Rabbit eye blueberries after 24 h of water loss (%) versus P_r (ratio: water loss (g) to solids gained (g)). Sucrose solutions were from curves at 25°C, 40°C, and 60°C. The 20°C and 40°C curves represent points at 20, 40, 50, 60 and 100% sucrose (left to right). The 60°C curve eliminates the 20% sucrose point. (fruit:syrup = 1:4.5) (Kim, 1987).

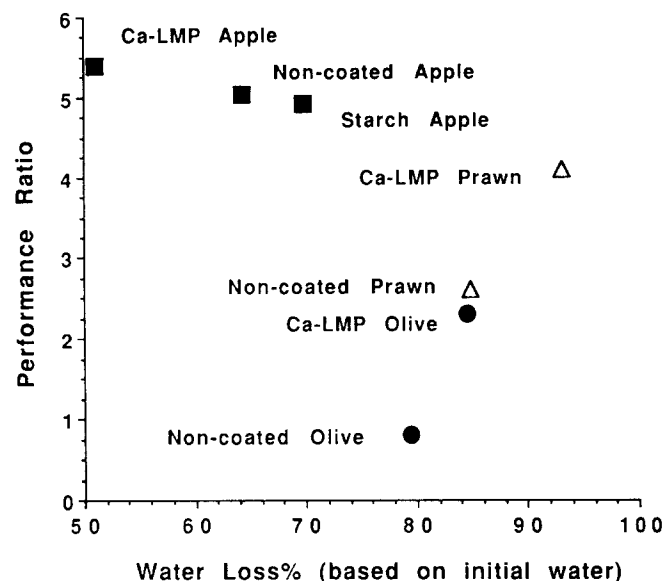


Fig. 4. Osmotic dehydration at 25°C of coated and non-coated apples, olives and prawns with water loss (%) based on initial water; P_r values are shown vertically. The osmotic agents were 69% sucrose syrup for apples and a 75° Brix syrup containing 50% (w/w) invert to sucrose for the olives and prawns. The apples (diced) were treated for 8 h, the olives 118 h and prawns 72 h. Ca-LMP coated prawns had a pleasing non-sticking sheen (food:syrup = 1:4, w/w) (Lewicki *et al.*, 1984; Camirand *et al.*, 1968).

from an extensive study of blueberries using various sucrose concentrations (after 24 hr) at three temperatures. The information presented here in a single plot helps comprehend the relationships and optimize various parameters. Figure 4 compares the results with coated

and non-coated foods, and demonstrates the remarkable improvement in P_r for coated olives and prawns.

To make comparisons of osmotically dehydrated foods for P_r versus water loss (%), Figs 1–4 need only be based on the initial foods prior to coating, if included; thus, we will maintain consistency by: (1) not computing the extra water in the coating as part of the food, which will be removed during the process, and (2) adding 'solids gained' to include edible coating solids which may also 'penetrate' into the food together with other osmotic agents added to the food. Comparing the OSMEMB process with 'standard' osmotic dehydration as stated above results in more conservative comparisons.

The P_r calculated for osmotic food pieces can depend on many variables, including size of the food piece, temperature, the osmotic agent, concentration, degree of mixing, ratio of food to osmotic agent (wt/wt) and duration of the experiment. If coatings are added, we must also consider interactions between the coating and the food, the presence of other coating treatments and changes in the properties of the coating during the process. Two other factors of importance to P_r are the diffusivities of water and osmotic solute in the coating and/or food tissue.

Mathematics of mass transport

We present below a mathematical model that can provide the framework for analysis of idealized mass transfer between a membrane-coated food piece and an osmotic drying medium or between two reservoirs separated by a mass-selective membrane.

In this model we assume Fick's law for diffusion through a membrane separating two well-mixed reservoirs. One of the reservoirs could be a food piece. There is an assumed pseudo-steady state in the membrane so that the concentration gradient is always equal to the differences in membrane surface concentrations divided by the membrane thickness. Then, if we make a material balance on any component 'i' in one of the reservoirs, for instance, the food piece, the result is

$$\frac{dM_{ij}}{dt} = -\frac{D_i K_i A}{h} (c_{i1} - c_{i2}) \quad (1)$$

where M_{ij} is the mass of 'i' in reservoir 'j', D_i is the diffusion coefficient of substance i through the membrane ($\text{cm}^2 \text{sec}^{-1}$); K_i is the distribution coefficient between the solution and the membrane, A is the interfacial area (cm^2); h is the thickness of the membrane (cm); and C_{i1} and C_{i2} are the upstream and downstream solution concentrations (g cm^{-3}) of component i at the membrane surface. The mass is equal to the product of C_{ij} and V_j , the volume of the reservoir j ; and the left-hand side of eqn (1) is equal to the mass of i transferred through the membrane.

In the general case for constant density, eqn (1) becomes

$$\frac{dM_{ij}}{dt} = -\frac{D_i K_i A}{h} (\rho_1 x_{i1} - \rho_2 x_{i2}) \quad (2)$$

where the x_{ij} are the mass fraction of component i in reservoir j , and ρ_j is the mass density where $V_j = x_{ij} \rho_j$. If density is constant and there are two components, water (w) and solute (s), then eqn (2) becomes

$$\frac{dM_{w1}}{dt} = -\frac{D_w K_w A \rho}{h} (x_{w1} - x_{w2}) \quad (3)$$

and

$$\frac{dM_{s1}}{dt} = -\frac{D_s K_s A \rho}{h} (x_{s1} - x_{s2}) \quad (4)$$

Since there are only two components then

$$x_{s1} + x_{w1} = 1 \quad (5)$$

Equations (3), (4), and (5) simplify to

$$\frac{dM_w}{-dM_s} = \frac{D_w K_w}{D_s K_s} \quad (6)$$

Equation (6) is the definition of P_r , the mass of water transferred through the membrane divided by the mass of solute transferred through the membrane. Therefore,

$$P_r = \frac{D_w K_w}{D_s K_s} \quad (7)$$

We conclude from this that, provided the assumptions are valid for the system analyzed, the value of P_r is independent of membrane thickness, time, interfacial area, and the geometry of the system.

The physical model system

In order to obtain the P_r on a large variety of coatings and osmotic agents, we proposed the use of a model system which could reduce the need for a large-scale food experiment. A bank of up to 24 simultaneous diffusion cells was operated. Figure 5 shows the details of a typical diffusion cell where

- a = porous thimble containing a solution of fixed volume;
- b = vial containing concentrated solution of osmotic agent;
- c = pipette (connected between tube and vial).

The definitions of terms are

- W_{bi} = initial weight of water in vial (b);
- W_{bf} = final weight of total water into thimble (a) from pipette (c);
- S_{bi} = initial weight of solute (drying agent) in vial (b);
- W_{ai} = initial weight of water in thimble (a);

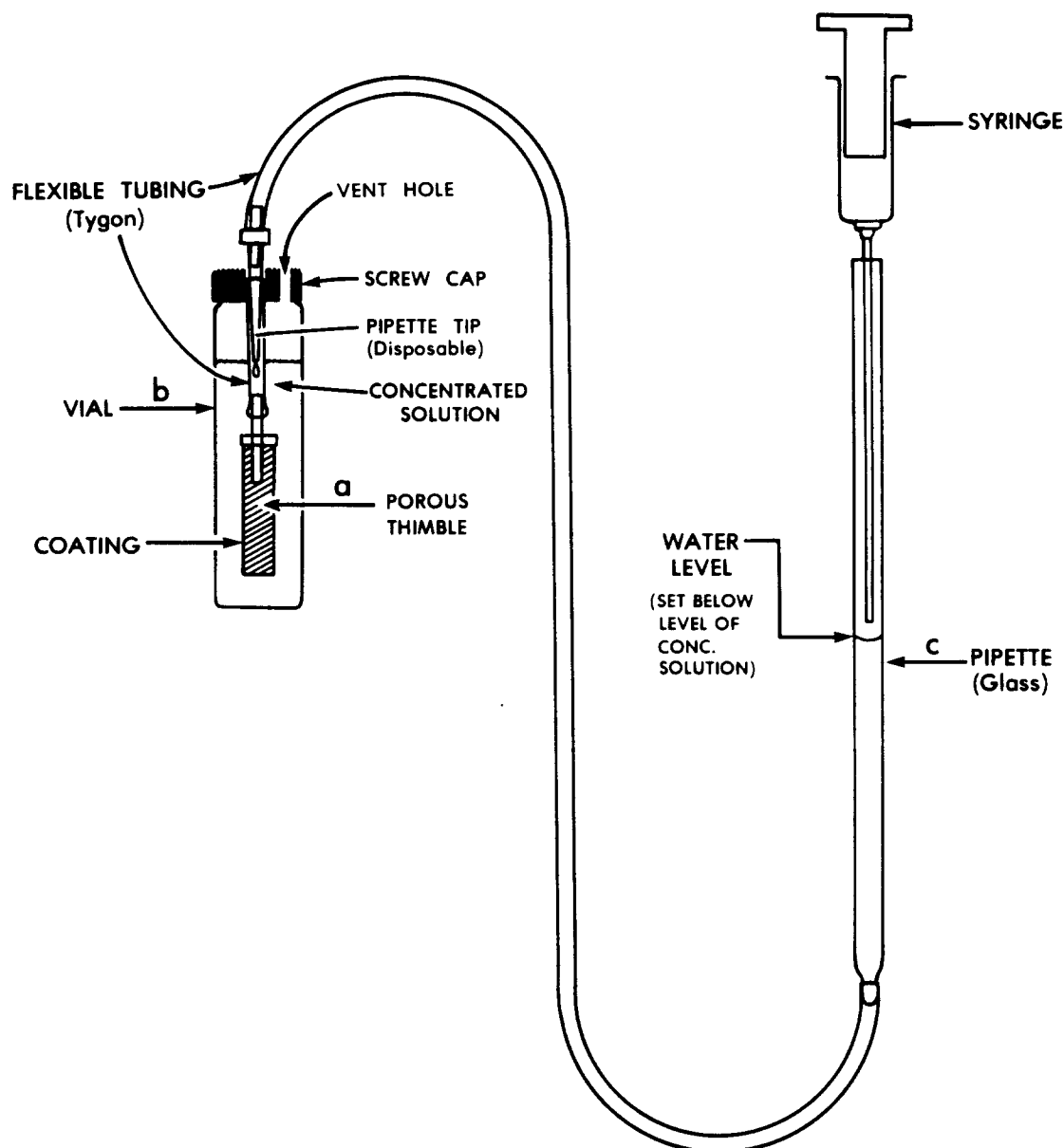


Fig. 5. Diffusion cell for determination of P_r (performance ratio) with polymeric coatings.

W_{af} = final weight of water in thimble (a) at equilibrium;

X = final weight of water in vial (b) at equilibrium;

Y = final weight of solute in thimble (a) at equilibrium;

Z = final weight of solute in vial (b) at equilibrium;

P_r = performance ratio = $(W_{bf} + Y)/Y$ where the mass balance requirements are

$$X = W_{bi} + W_{bf} + Y \quad (8)$$

The final weight of water in (b) must be equal to W_{bi} plus the weight of water that enters from (c) (pipette + tubing up to the plastic pipette tip) plus the water from the thimble that has been displaced by the solute from (b). From eqn (1) it is assumed that the weight of solute entering the thimble is the same as the weight of

displaced water entering (b). Density differences and volume contraction effects would have to be taken into account for this equation to be exact. This equation would then be extremely complicated. If we eliminated the Y term from the above equation, the equation remains true within about 5% of X since (b) usually contains over 20 g of drying solution and the thimble contains only about 1 g of water. Thus, the above assumption is considered justified.

$$W_{af} = W_{ai} - Y \quad (9)$$

This is another mass balance equation based on the same assumption discussed above.

$$Z = S_{bi} - Y \quad (10)$$

This simple material balance requires no assumptions.

Equilibrium requirements:

$$Z/X = Y/W_{af} \quad (11)$$

This equation simply states that the concentration of the osmotic agent must be the same on both sides of the coating at equilibrium.

This system of four equations and four unknowns (W_{af}, X, Y, Z) is easily solved.

MATERIALS AND METHODS

Initially the hydrostatic pressure in the system (Fig. 5) will be very small compared to the osmotic pressure. The osmotic pressure difference across the coating will reduce the level of water in the pipette (c). When the system approaches equilibrium the level in the pipette will remain steady. If the coating on the thimble leaks, the level in the pipette will begin to rise relatively rapidly. A coating fails when the level of water in the pipette falls at the time the diffusion cell is assembled.

The procedures for preparing cells are shown in steps 1–5 of Fig. 6 and are described below.

1. Record weight of empty thimble.
2. Weigh and record dry cap and thimble assembly.
3. Using a vacuum oven, vacuum-infuse all porous thimbles submerged in deionized water for at least 10 h. (Water refers to deionized water in the subsequent procedures.)
4. Place the thimble assembly in de-aerated water, draw up to the plastic pipette tip, weigh and record.
5. Remove water from the inside of the thimble assembly by pouring the water out (not water in pores), shake the water from the cap assembly at (a). The components are reattached at (b) and weighed. (Procedures 1–2 and 4–5 allow calculation of water in the 'pores'.)

As illustrated in steps 6–11 of Fig. 6 the coating procedure discussed below involves dipping the polymer solutions and 'fixing' or insolubilizing the coatings.

6. Dip the thimble assembly into polymer solution while an oscillating gentle vacuum/pressure action of a syringe is applied to the thimble assembly to force the viscous solution into the thimble pores. Dip the thimble assembly into a crosslinking agent (e.g. 5 M CaCl_2) for a given time and then rinse with water. Add further treatments if desired at this time (e.g. ethanol), or the polymer can simply be evaporated without further treatment in order to determine its effect on P_r values.
7. Use a syringe with a fine, flexible extension tube attached to a blunt needle to add 1.5–2.0 ml of dilution water to the thimble. Then remove most

of the polymer solution inside the thimble with the syringe. Weigh coated 'empty' assembly.

8. Use syringe attached to the small diameter tube to refill the thimble assembly with water up to the plastic pipette tip. Record the weight.
9. Fill thimble assembly with water up to the very top. Fill the rest of the system with water to the zero mark of the glass pipette (c).
10. Add osmotic agent (of known composition) to the vial. Record components and concentrations of solutes in previously prepared osmotic solution. Weigh and record quantity of osmotic solution and volume (calculated) in vial.
11. Assemble the full diffusion cell. Record zero time at which the pipette must be refilled. Place vial and assembly into a temperature bath. Measure water loss with time and replenish the pipette with water as necessary.

Osmotic agents and coatings

Osmotic agents used were sucrose solution (68–69%), dextrose solution (51.6%), and glycerol (96%). Granulated sucrose was obtained from C&H Sugar Co. (Concord, CA); anhydrous dextrose was from Mallinckrodt Chemical Works (St Louis, MO); and glycerol (Emery 912) was from Emery Industries (Los Angeles, CA). Polymeric coatings used in applications with diffusion cells were prepared with the following products or compounds: pure food corn starch (PFCS), sodium alginate (SA), calcium chloride, ethanol (95%) (E95), sodium polypectinate (SP), low-methoxyl pectin (LMP), ethyl cellulose (EC), maltodextrin (MD), potato starch (PS) and methyl cellulose (MC). Sources of these substances, formulations of polymeric coatings and procedures for their preparation are presented in Table 1.

Statistical analysis

The data points in Figs 1–4 were derived from their corresponding references; however, statistical data was not reported in these sources. With the diffusion cells, zero data were deleted prior to analyses of variance and Duncan's tests. Data were tested for homogeneity of variance between coatings. Where required, data were either transformed or weighting was used to compensate for the heterogeneity.

RESULTS AND DISCUSSION

Osmotic food products compared

In our search for general parameters useful for producing high-quality, osmotically dehydrated products, a number of studies were examined. P_r values

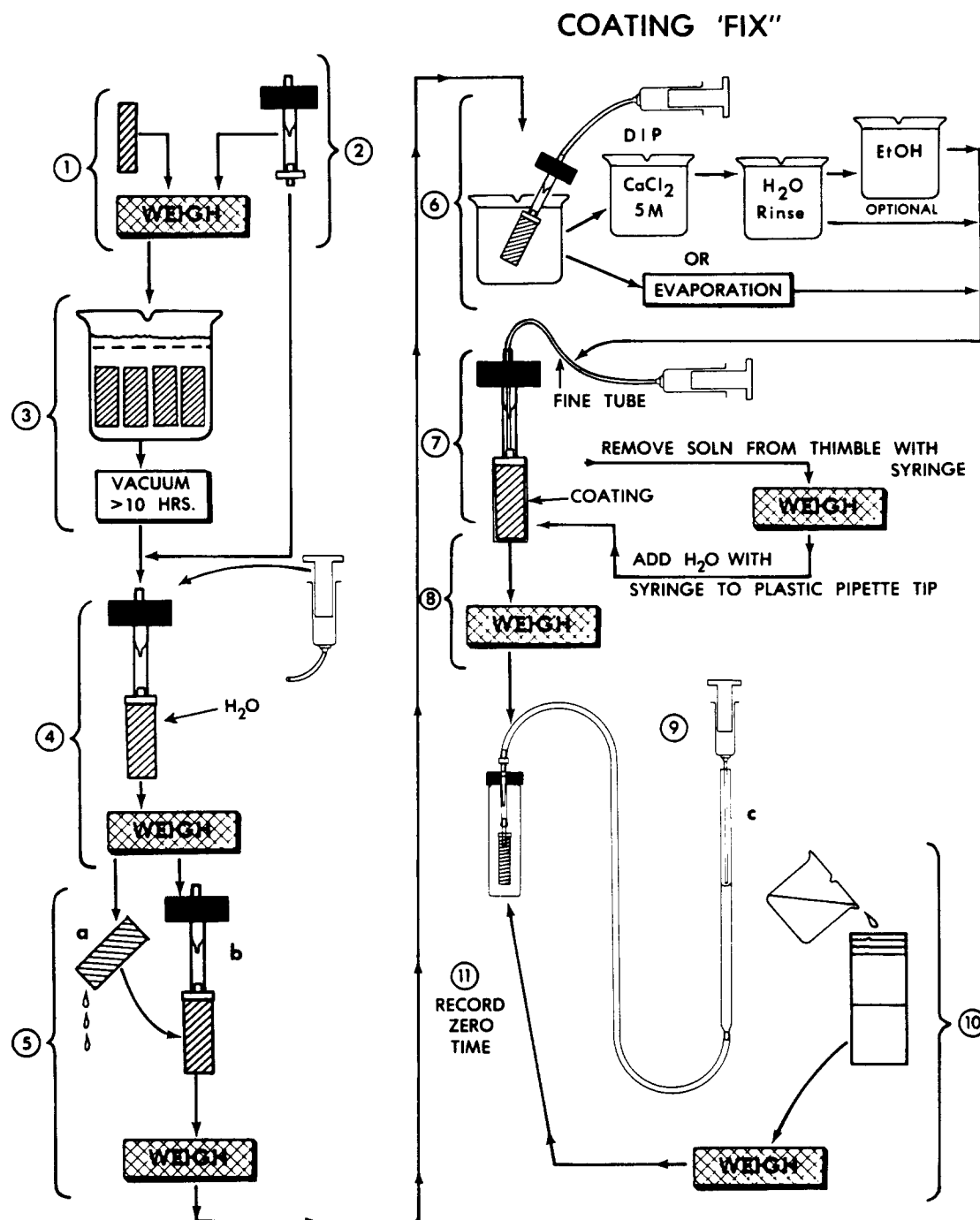


Fig. 6. Schematic procedure for preparing the diffusion cells.

(water loss/solids gained) (wt/wt) and percent water loss (based on the original water) were plotted as derived from data given in these reports (Figs. 1-4).

For Figs 1-3, P_r versus percent water loss was plotted for uncoated osmotically dehydrated fruit. Data were derived from various independent publications, using Red Delicious apples, three cherry cultivars, and Rabbiteye blueberries.

In comparing Red Delicious apples dehydrated in high fructose corn syrup (70% solids) to apples in a 70% sucrose, the product using sucrose which was claimed

to have the '... best quality' (Bolin *et al.*, 1983) also had the highest P_r according to our calculations (Fig. 1).

In comparing three cherry cultivars it was stated that, '... only the 'Vittoria' appearance was acceptable to the standards and the products obtained were of good quality.' (Torreggiani *et al.*, 1987). This cultivar also had the highest P_r values and percent water loss (Fig. 2).

Kim (1987) states that 'When comparing results using high sugar concentration (60%) with crystalline sucrose, the latter was a better osmotic agent than the

Table 1. Formulations and procedures for preparation of carbohydrate polymer coatings

Coating no.	Substances used in formulation ^a			procedure ^b
	A	B	C	
1	PFCS (4.2)	SA (1.4)	H ₂ O (150.0)	Mix A and B together and add to C at RT. Heat mixture to 87.7°C in a boiling water bath. Then cool to RT. (Allen <i>et al.</i> , 1963) Follow crosslinking procedure.
2	EC (10.0)	—	E95 (90)	Stir A into C at RT. Coat thimbles with mixture and evaporate for 10 h. Repeat dipping and evaporation.
3	SP (2.5)	—	H ₂ O (97.5)	Stir A slowly into C at RT. Follow crosslinking procedure.
4	LMP (3.75)	—	H ₂ O (146.2)	Stir A rapidly into C at 70°C. Cool to RT. Follow crosslinking procedure.
5	LMP (5.0)	MD (30.0)	H ₂ O (165.0)	Mix A and B together and add to C at RT. Follow crosslinking procedure.
6	LMP (3.75)	PFCS (4.25)	H ₂ O (142.0)	Mix A and B together and add to C at RT. Heat mixture to 87.7°C in a boiling water bath. Then cool to RT. Follow crosslinking procedure. Then dip in E95 for 30 s.
7	PFCS (4.2)	SA (1.4)	H ₂ O (150.0)	Follow same procedure as for Coating no. 1. Then dip in E95 for 30 s.
8	PS (2.0)	LMP (5.128)	H ₂ O (198.0)	Stir A into C at RT. Heat mixture to 65.0°C. Then cool to RT. Add B to mixture. Follow crosslinking procedure.
9	LMP (1.5)	MC (5.5)	H ₂ O (53.0. 40.0)	Stir A into 53 g C at 70.0°C and then cool mixture to RT. Stir B into 40 g C, heat mixture to > 70.0°C (Vojdani & Torres, 1989), and cool to 60.0°C. Add second mixture to first and stir while holding in an ice water bath until solution is clear. Follow crosslinking procedure.
10	LMP (3.75)	PFCS (4.25)	H ₂ O (142.0)	Follow same procedure as for Coating no. 1.
11	LMP (4.5)	—	H ₂ O (145.5)	Follow same procedure as for Coating no. 4.
12	EC (10.0)	—	E95 (90)	Follow same procedure as for Coating No. 2 except that the dipping and evaporation procedures are not repeated a second time.

^aNumber enclosed in parentheses following substance abbreviation indicates quantity of substance used in grams to prepare a formulation. PFCS = pure food corn starch (CPC International, Englewood Cliffs, NJ); SA = sodium alginate (GHB Manugel, Kelco International, London, UK); EC = ethyl cellulose (Type N-50) (Hercules Powder Co., Wilmington, DE); E95 = ethanol (95%) (Quintum Chemical Corp., Anaheim, CA); SP = sodium polypectinate (Sunkist Growers, Corona, CA); LMP = low methoxyl pectin (290 NA 95) (Sanofi Bio Ingredients, Germantown, WI); MD = maltodextrin (Maltrin M040) (ave. MW = 3600) (Grain Processing Co., Muscatine, IA); PS = potato starch (Eastman Kodak Co., Rochester, NY); MC = methyl cellulose (Methocel A15-LV) (Dow Chemical Co., Midland, MI).

^bRT = room temperature (25°C). Crosslinking procedure involved the following steps: (1) dipping previously prepared thimble assembly in polymeric coating for 60 s, (2) dipping assembly in 5 M CaCl₂ (Spectrum Chemical Mfg. Corp., Gardena, CA) for 30 s, and (3) rinsing in water for 10 s.

60% sugar solution since less sugar was gained with higher water loss at all temperatures (25°C, 40°C and 60°C) tested. This single plot is very useful for comprehending all of the data and determining the most useful parameters for different purposes (Fig. 3).

Comparing P_r versus percent water loss for coated and non-coated foods is shown in Fig. 4. When coated (Ca-LMP) and non-coated olives (at 118 h) and prawns (at 72 h) are compared, coated food had the greatest P_r values, the greatest percent water losses, and the best appearances (Camirand *et al.*, 1968). Diced apples were used to produce three treatments after 8 h of osmotic dehydration: (1) a non-coated product, (2) a product coated with a 1% starch, and (3) a coated product with 1% Na-LMP solution treated by submerging apples in a 2.4% CaCl₂ solution for 30 s. Of the three apple products, it appears that the starch coating resulted in

the greatest water loss. Rinsing and washing the syrup from coated, diced apples prevents gain of water and loss of flavor during the final treatment. Coatings were more efficient in removal of surface sugar during washing and retaining more of the initial soluble solids after the osmotic treatment (Lewicki *et al.*, 1984). Plotting P_r against water loss (%), however, would indicate that the coatings did not improve P_r values for the Lewicki experiment with only a slight improvement with the Ca-LMP. Potential reasons for Lewicki's results are: (1) the limited duration of these experiments (8 h); (2) the small size of the apple pieces (diced, about one cubic cm); thus, a considerable amount of polymer coating was added to the apple dice; and (3) uncoated apple pieces seem to have a relatively high P_r (4–5 at RT) which is comparable to the P_r for a 2.5% LMP, on a diffusion cell, at around 4 (see coatings 3 and 4, Tables

2 and 3). On the other hand the sucrose added to the coated diced apple was lower than the uncoated diced apple. It should be pointed out that Lewicki *et al.* (1984) included most of their published results using a short thermal drying step prior to osmotic dehydration.

Comparing graphs of P_r versus percent water loss (Figs 1–4) for a range of food experiments, including information on osmotic agents, temperature, times, and polymer coating (if any), may help predict both shelf life and quality. If $P_r(C)$ is the P_r (average) for a

given coating (using diffusion cells), and $P_r(F)$ is the value of P_r from a given non-coated food experiment, then if $P_r(C) \gg P_r(F)$ one will expect that $P_r(CF)$, the coated food, should result with $P_r(CF) > P_r(F)$ (using the same coat, food, temperature, osmotic area, etc.).

Model coating results

Applying 12 coatings containing polycarbohydrates and mixtures as edible coatings in the model system

Table 2. Solids composition of coating solutions and failure rates of coatings in osmotic agents

Coating no.	Solids composition of coating solutions ^a			Osmotic agents					
				Sucrose		Glycerol		Dextrose	
	Total solids (%)	LMP (%)	Other solids (%)	Frequency ^b	Failure rate	Frequency	Failure rate	Frequency	Failure rate
1	3.60	—	0.90 SA + 2.7 PFCS	3	1.00	3	1.00	—	—
2	10.0	—	10.0 EC	6	1.00	6	1.00	6	1.00
3	2.5	2.5 ^c	—	6	0.17	6	0.33	6	0.05
4	2.5	2.5	—	7	0.29	9	0.33	12	0.05
5	17.5	2.5	15.0 MD	6	0	6	0	6	0.33
6	5.33	2.5	2.83 PFCS	3	1.00	3	0	—	—
7	3.6	—	0.90 SA + 2.7 PFCS	3	0.67	3	0.67	—	—
8	3.47	2.5	0.975 PS	6	0	6	0	6	0.166
9	7.0	1.5	5.8 MC	3	0.33	3	0	—	—
10	5.33	2.5	2.83 PFCS	3	—	3	0	—	—
11	3.0	3.0	—	3	0.33	—	—	—	—
12	10.0	—	10.0 EC	3	1.00	6	0.33	—	—

^aDescriptions of substances used to prepare coating solutions are provided in Table 1.

^bFrequency refers to the number of experiments performed with individual cells.

^cSP was the type of LMP used in this coating.

Table 3. Mean P_r values and standard deviations for ten diffusion coatings in three osmotic agents

Coating no. ^a	Osmotic agent					
	Sucrose		Glycerol		Dextrose	
	Mean P_r value	Standard deviation	Mean P_r value	Standard deviation	Mean P_r value	Standard deviation
1	—	—	—	—	—	—
2	—	—	—	—	—	—
3	2.13 ^b	0.44	1.63 ^b	0.36	1.40 ^c	0.14
4	3.66 ^c	0.56	3.02 ^d	0.49	1.49 ^c	0.29
5	4.27 ^{cd}	0.83	2.82 ^d	0.43	1.56 ^c	0.28
6	—	—	3.47 ^{de}	1.92	—	—
7	4.66 ^{cde}	—	4.03	—	—	—
8	4.66 ^{cde}	0.87	4.37 ^c	0.31	2.04 ^f	0.20
9	7.31 ^{def}	1.26	3.70 ^{de}	1.69	—	—
10	8.06 ^{ef}	2.95	5.08 ^e	1.45	—	—
11	8.81 ^c	2.73	—	—	—	—
12	—	—	12.76 ^f	10.87	—	—

^aCoating nos 1 and 2 failed.

^{b–f}Within a column, means followed by the same superscript are not significantly different ($P > 0.05$) from one another by the Duncan multiple-range test.

Table 4. Mean values and standard deviations for equilibrium time of twelve diffusion coatings in three osmotic agents

Coating no. ^a	Osmotic agent					
	Sucrose		Glycerol		Dextrose	
	Mean equilibrium time (h)	Standard deviation	Mean equilibrium time (h)	Standard deviation	Mean equilibrium time (h)	Standard deviation
1	—	—	—	—	—	—
2	—	—	—	—	—	—
3	76.8 ^b	37.57	93.0 ^{bc}	44.97	24.0 ^b	0
4	66.4 ^b	28.34	51.0 ^{bc}	44.86	8.3 ^c	5.24
5	85.5 ^b	43.87	63.3 ^{bcd}	47.71	20.3 ^b	9.46
6	—	—	11.2 ^c	13.89	—	—
7	19.0 ^c	—	22.0 ^{cde}	—	—	—
8	47.3 ^b	14.53	46.0 ^{bcd}	13.37	14.2 ^{bc}	6.57
9	102.5 ^b	31.82	50.7 ^{bcd}	41.67	—	—
10	40.0 ^b	3.00	16.8 ^{cd}	14.37	—	—
11	40.0 ^b	14.14	—	—	—	—
12	—	—	205.0 ^b	151.9	—	—

^aCoating nos 1 and 2 failed.

^{b-c}Within a column, means followed by the same superscript are not significantly different ($P > 0.05$) from one another by the Duncan multiple-range test.

has allowed us to determine P_r values for a wet surface, its failure rate (0 = no failure, 1.0 = 100% failure), repetitions (frequency), and osmotic agents (69% sucrose, 96% glycerol, 51.6% dextrose). All coatings used crosslinked polysaccharide ion gels (e.g. sodium alginates or Na-LMP), mixtures of crosslinked polymers with linear (non-crosslinked) polymers (e.g. semi-IPNs), or evaporated coatings in the case of ethyl cellulose in ethanol.

Mean P_r values, equilibrium times, standard deviations, number of replications, solids in the coatings solutions, formulations and procedures, failure rates and Duncan groupings are shown in Tables 1–4. Mean P_r values for the coatings are summarized in Fig. 7. The highest mean P_r values were obtained with ethyl cellulose as a coating and glycerol as a dehydrating agent (coating #12). Experiments with EC using sucrose failed. Glycerol perhaps worked better as a plasticizer since the coating appeared to be more flexible. In all LMP (290 NA 95) coatings used, the best result was obtained with 69% sucrose as the osmotic agent and a 3% coating solution (Coating #11) whereas a 2.5% coating solution did not perform nearly as well. Coatings #5–6 and 8–10 formed from semi-IPN mixtures derived from a single Na-LMP solution at the same or lower solids (Coating #4). The semi-IPN coatings concept is thus a success because it allows use of non-crosslinked polymers in an edible coating for further examination of its properties.

In the future, by knowing the P_r and the original food's soluble solids, initial moisture, osmotic agent composition, ratio of food to osmotic agent (wt/wt) and

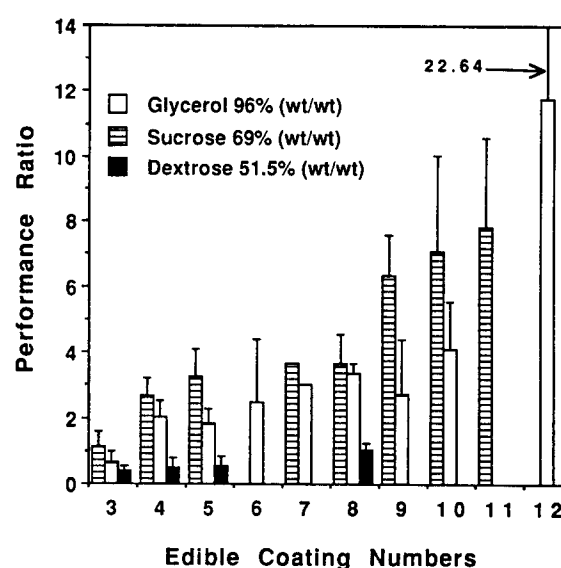


Fig. 7. Determination of the ratio of the water loss to solids gained in 10 coatings at 30°C comprised of 96% glycerol, 69% sucrose and 51.5% dextrose. (Refer to Tables 1, 2, 3 and 4 for compositions of coatings.)

approximate equilibrium time, we can better determine the weight loss, solids gained and water activity.

DEDICATION

This work is dedicated to the recent memory of Dr James D. Ponting, who pioneered research on osmotic dehydration of fruits.

ACKNOWLEDGEMENTS

We thank Dr George H. Robertson for his help with the mathematics of mass transport. We also thank Dr Bruce E. Mackey for assistance with the statistical analysis.

REFERENCES

- Allen, L., Nelson, A.I., Steinberg, M.P. & McGill, J.N. (1963). Edible corn-carbohydrate food coatings I. Development and physical testing of a starch-algin coating. II. Evaluation on fresh meat products. *Food Technol.*, **17**(11), 99-108.
- Bolin, H.R., Huxsoll, C.C., Jackson, R., & Ng, K.C. (1983). Effects of osmotic agents and concentration on fruit quality. *J. Food Sci.*, **48**, 202-5.
- Camirand, W.M. & Forrey, R.R. (1969). Osmotic dehydration of coated foods. U.S. Patent 3 425 848.
- Camirand, W.M., Forrey, R.R., Popper, K., Boyle, F.P. & Stanley, W.L. (1968). Dehydration of membrane-coated foods by osmosis. *J. Sci. Food Agric.*, **19**, 472-474.
- Dixon, G.M. & Jen, J.J. (1977). Changes of sugars and acids of osmovac-dried apple slices. *J. Sci. Food Agric.*, **42**(4), 1126-7.
- Farkas, D.F. & Lazar, M.E. (1969). Osmotic dehydration of apple pieces: Effects of temperature and syrup concentration on rates. *Food Technol.*, **23**, 688-90.
- Forni, E., Torreggiani, D., Battiston, P. & Polesollo, A. (1986). Research into changes of pectic substances in apricots and peaches processed by osmotic dehydration. *Carbohydr. Polym.*, **6**, pp. 379-93.
- Giangiacomo, R., Torreggiani, D. & Abbo, E. (1987). Osmotic dehydration of fruit: Part I. Sugars exchange between fruit and extracting syrups. *J. Food Proc. & Pres.*, **11**, 183-95.
- Guilbert, S. (1988). Use of superficial edible layer to protect intermediate moisture food: Application to the protection of tropical fruit dehydrated by osmosis. In *Food Preservation by Moisture Control*, ed. C.C. Seow, T.T. Teng & C.H. Quah. Elsevier Applied Science, London and New York, pp. 199-219.
- Hawkes, J. & Flink, J.M. (1978). Osmotic concentration of fruit slices prior to freeze dehydration. *J. Food Proc. Pres.*, **2**, 265-84.
- Karel, M. & Langer, R. (1988). Controlled Release of Food Additives. In *Flavor Encapsulation*, ACS symposium series 370, ed. S.J. Risch & G.A. Reineccius. American Chemical Society, Washington, DC, pp. 177-91.
- Kester, J.J. & Fennema, O.R. (1986). Edible films and coatings: A Review. *Food Technol.*, **40**(12), 47-59.
- Kim, M.H. (1987). Fluidized bed and osmotic processes for dehydration of Rabbiteye Blueberries. Dissertation, U.M.I., Ann Arbor, Michigan, pp. 1-151.
- Krochta, J.M., Hudson, J.S., Camirand, W.M. & Pavlath, A.E. (1988). Edible films for light-processed fruits and vegetables. Paper presented at International Winter Meeting of the American Society of Agricultural Engineers, Chicago, Illinois, (Paper No. 88-6523).
- Lerici, C.R., Pinnavaia, G., Dalla Rosa, M. & Bartolucci, L. (1985). Osmotic dehydration of fruit: Influence of osmotic agents on drying behavior and product quality. *J. Food Sci.*, **50**, 1217-26.
- Lewicki, P.P., Lenart, A. & Pakula, W. (1984). Influence of artificial semi-permeable membranes on the process of osmotic dehydration of apples. *Annals of Warsaw Agricultural University, SGGW-AR. Food Technol. and Nutr.*, **16**, 17-24.
- Maltini, E. & Torreggiani, D. (1986). Production of shelf-stable fruits by osmosis. In *Progress in Food Engineering*, presented at a European Symposium of the Food Working Party of the EFCE (European Federation of Chemical Engineering), Milan, Italy, pp. 471-6.
- Ponting, J.D., Watters, G.G., Forrey, R.R., Jackson, R. & Stanley, W.L. (1966). Osmotic dehydration of fruits. *Food Technol.*, **20**(10), 125.
- Skjåk-Bræk, G., Grasdalen, H. & Smidsrød, O. (1989). Inhomogeneous polysaccharides ionic gels. *Carbohydr. Polym.*, **10**, 31-54.
- Sperling, L.H. (1988). Interpenetrating polymer networks. *Chemtech.*, **18**(2), 104-9.
- Torreggiani, D., Forni, E. & Rizzolo, A. (1987). Osmotic dehydration of fruit. Part 2: Influence of the osmotic time on the stability of processed cherries. *J. Food Proc. & Preserv.*, **12**, 27-44.
- Vojdani, F. & Torres, J.A. (1989). Potassium sorbate permeability of polysaccharide films: Chitosan, methylcellulose and hydroxypropyl methylcellulose. *J. Food Proc. & Engng.*, **12**, 33-48.
- Wong, D.W.S., Gregorski, K.S. & Pavlath, A.E. (1990). Edible films from carbohydrates and fatty acids. Paper presented at 19th Meeting of the UJNR Protein Panel, August 6-7, 1990, San Diego, CA, pp. 8.