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## หองสมุตกรมวทยาศาส**ตร์บร**์หาร

#### REVERSE OSMOSIS TRANSPORT AND MODULE ANALYSIS FOR GREEN TEA JUICE CONCENTRATION

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#### ABSTRACT

Reverse osmosis experiments were performed to concentrate the green tea juice, and the experimental data was analyzed by using a set of transport equations together with osmotic pressure and density data of the green tea juice. The transport parameters obtained numerically by the above analytical procedure were then used to calculate several dimensionless quantities that can characterize a reverse osmosis module. An example of module design calculation was attempted using the dimensionless parameters so obtained. It has been found that an increase in operating pressure reduces the module length significantly when the feed tea juice concentration is high. An increase of the mass transfer coefficient on the high pressure side of the membrane has the same effect.

#### **INTRODUCTION**

Membrane separation processes have been found useful in many food processings involving the concentration of liquid foods. Typical examples are the treatment of milk whey, the concentration of fruit juices, and the treatment of alcoholic beverages. The low energy requirement inherent in the membrane separation process has been the force to promote wider applications of this separation technology in various branches of food processing industries. The concentration of tea juice is one of those applications and several attempts have already been made (Bonneau and Demande 1981; Buhler and Olofsson 1981; Nogy 1985; Schreier and Mick 1984; Tamaki *et al.* 1986).

In our previous work we have also attempted to concentrate green tea juice using membranes prepared from different polymeric materials and of different pore sizes (Zhang *et al.* 1991). In particular, emphasis was placed on the removal of the caffeine component into the membrane permeate while concentrating other tea components in the retentate. Because of severe membrane fouling observed during the process a detailed study was carried out for the effect of the membrane polymer and operating conditions on membrane fouling. It was concluded from the above investigations that, among various membranes tested, the membranes cast from cellulose diacetate material and used without further shrinkage demonstrated the best result in terms of the permeation rate at a given level of separation of tea components and also in terms of the high rate of removal of caffeine into the permeate. Fouling was found less with the membrane produced from cellulose diacetate material compared with those prepared from some other synthetic polymers.

The experimental data obtained in the previous work are considered to be local values. In reality, the membrane separation is performed by a reverse osmosis module in which the local operating conditions change from position to position as the feed tea juice advances from the module entrance to the module exit. Considerations were given to this aspect in the work of Ohya and Sourirajan (Sourirajan 1970) for the design of the reverse osmosis module and several design parameters were identified.

The objective of this work is to determine these design parameters numerically through a number of experiments specially designed for this purpose. Furthermore, the design parameters so obtained are used in the calculation of the module performance and discussions are made on the effect of the operating conditions on the performance of the entire module.

#### THEORETICAL

#### **Reverse Osmosis Transport**

Fundamental equations have been established and proved valid for reverse osmosis membrane transport (Sourirajan 1970). For the reverse osmosis concentration of liquid food that consists of many components of different molecular weights, transport equations based on the weight of the solute have been developed (Matsuura *et al.* 1973). They are,

$$A_{(wt)} = \frac{PWP}{S \times 3600 \times P} \tag{1}$$

$$N_{B(wt)} = A_{(wt)} [P - \pi (X_{A2(wt)}) + \pi (X_{A3(wt)})]$$
(2)

$$= \left(\frac{D_{AM}}{K\delta}\right)_{(wt)} \left(\frac{1 - X_{A3(wt)}}{X_{A3(wt)}}\right) \left(\rho_2 X_{A2(wt)} - \rho_3 X_{A3(wt)}\right)$$
(3)

$$= k_{wt} \rho_1 (1 - X_{A3(wt)}) \ln(\frac{X_{A2(wt)} - X_{A3(wt)}}{X_{A1(wt)} - X_{A3(wt)}})$$
(4)

All symbols are defined in the end of the paper. The first equation indicates that pure water permeation rate, PWP, is proportional to the operating pressure and the proportionality constant is  $A_{(wt)}$ . The second equation indicates that the solvent flux,  $N_{B(wt)}$ , in the presence of the solute is proportional to the effective pressure that is equal to the operating pressure minus the osmotic pressure difference on both sides of the membrane. The third equation has been derived from a Fick's type equation where the solute flux is proportional to the solute concentration difference on both sides of the membrane. The fourth equation is for concentration polarization and enables to calculate the solute weight fraction in the boundary layer,  $X_{A2(wt)}$ , which is different from that in the bulk solution,  $X_{A1(wt)}$ . Under the steady condition a set of experimental data PWP, PR, and f enables the calculation of  $A_{(wt)}$ ,  $(D_{AM}/K\delta)_{(wt)}$ ,  $k_{(wt)}$  and  $X_{A2(wt)}$  by using Eq. (1)–(4). Conversely, the reverse osmosis performance data, PWP, PR, and f can be obtained by calculation once numerical values are given for  $A_{(wt)}$ ,  $(D_{AM}/K\delta)_{(wt)}$  and  $k_{(wt)}$ by using Eq. (1)–(4).

When Eq. (1)-(4) are combined we obtain (Sourirajan and Matsuura 1985),

$$(D_{AM}/K\delta)_{(wt)} = v_s \frac{1-f}{f} [\exp(\frac{v_s}{k_{(wt)}})]^{-1}$$
(5)

The above equation is convenient to calculate  $(D_{AM}/K\delta)_{(wt)}$  value from separation data, f, particularly when the permeation rate,  $v_s$ , and the mass transfer coefficient,  $k_{(wt)}$ , are already known. Conversely, f can be calculated by Eq. (5) for a known value of  $(D_{AM}/K\delta)_{(wt)}$ . As for mass transfer coefficient,  $k_{(wt)}$ , when the value is known for solute  $\alpha$  that for solute  $\beta$  can be obtained by

$$k_{\beta(wt)} = k_{\alpha(wt)} \left[ \frac{(D_{AB})_{\beta}}{(D_{AB})_{\alpha}} \right]^{2/3} \tag{6}$$

where  $k_{\alpha(wt)}$  and  $k_{\beta(wt)}$  and  $k_{(wt)}$  values for the solute  $\alpha$  and  $\beta$ , respectively, and  $(D_{AB})_{\alpha}$  and  $(D_{AM})_{\beta}$  and  $D_{AB}$  values for the solute  $_{\alpha}$  and  $_{\beta}$ , respectively. When

the diffusivity data  $D_{AB}$  is not known it can be obtained using the Wilke-Chang equation,

$$D_{AB} = 7.4 \times 10^{-8} \times \frac{(\chi M_B)^{1/2} T}{\mu V_A^{0.6}}$$
(7)

where  $\chi = 2.6$ , M<sub>B</sub> = 18.02, T = 298.2 and  $\mu = 0.893$  for aqueous solution at 25C. V<sub>A</sub> is the molar volume of the solute molecule at the boiling point and can be obtained by the summation of the contribution from all structural components involved in the solute molecule (Wilke and Chang 1955). A method to measure osmotic pressures at various concentrations has also been established (Matsuura *et al.* 1973) based on the fundamental transport equations.

#### **Reverse Osmosis System Analysis**

It has been pointed out that a reverse osmosis system can be specified in terms of three dimensionless parameters,  $\gamma$ ,  $\theta$ , and  $\lambda$  which are defined as

$$\gamma = \frac{\pi(X_{A1(wt)}^{\circ})}{P} = \frac{\text{osmotic pressure of the solution at the module entrance}}{\text{operating pressure}}$$
(8)

$$\theta = \frac{(D_{AM}/K\delta)_{(wt)}}{v_w^*} = \frac{solute\ transport\ parameter}{pure\ water\ permeation\ velocity}$$
(9)

$$\lambda = \frac{k_{(wt)}}{(D_{AM}/K\delta)_{(wt)}} = \frac{mass \ transfer \ coefficient \ on \ the \ high \ pressure \ side}{solute \ transport \ parameter}$$
(10)

where  $v_w^* = A_{(wt)}P$ . The performance of the reverse osmosis system so specified can be predicted by the system analysis of Ohya and Sourirajan (Soiurirajan 1970) under the following conditions.

- 1. Osmotic pressure of the solution is proportional to the weight concentration of the solute.
- 2. The weight concentration in the product is small compared with that of water.
- 3. Longitudinal diffusion of the solute is negligible in the process using a flow type cell apparatus.
- 4.  $(D_{AM}/K\delta)_{(wt)}$  and  $k_{(wt)}$  are independent of the feed concentration.
- 5. The density of the solution is constant, and it is close to that of pure water.
- 6. Membrane pore structure does not change during the process.
- 7. The operation pressure is constant throughout the system.

Under the above conditions all the system parameters, illustrated schematically in Fig. 1, have been interrelated with each other by eight equations describing a reverse osmosis system (Sourirajan and Matsuura 1985). The following dimensionless concentrations are involved in the above system equations:



FIG. 1. PERFORMANCE PARAMETERS FOR A REVERSE OSMOSIS MODULE [Reproduced from Sourirajan and Matsuura (1985).]

$$C_{1} = X_{A1(wt)}/X_{A1(wt)}^{\circ}, C_{1}^{\circ} = 1, C_{2} = X_{A2(wt)}/X_{A1(wt)}^{\circ}, C_{2}^{\circ} = X_{A2(wt)}^{\circ}/X_{A1(wt)}^{\circ},$$
$$C_{3} = X_{A3(wt)}/X_{A1(wt)}^{\circ}, C_{3}^{\circ} = X_{A3(wt)}^{\circ}/X_{A1(wt)}^{\circ}, \text{ and } \overline{C_{3}} = \overline{X}_{A3(wt)}/X_{A1(wt)}^{\circ}.$$

The dimensionless module length, X, defined as  $X = v_w^* x/\bar{u}^\circ h$ , and  $\Delta$ , the fraction recovery of the feed solution entering into the module to the membrane permeate, are also involved in the above equations. While there are eight equations, the number of unknown quantities involved in the equations was found to be twelve. They are  $\gamma$ ,  $\theta$ ,  $\lambda$ ,  $C_1$ ,  $C_2$ ,  $C_2^\circ$ ,  $C_3$ ,  $C_3^\circ$ ,  $\overline{C_3}$ , X,  $\Delta$ , and Z. (Z is a quantity given by one of the equations as a function of  $C_3$ .) Therefore, when four of the above quantities are given, others can be calculated. Since  $\gamma$ ,  $\theta$ , and  $\lambda$  are usually given as parameters to specify the system, only by giving another parameter, for example  $\Delta$ , we are able to calculate all the numerical quantities involved in the system. In other words, the module length, the feed concentration at the module exit, the concentration of the composite product, etc., can be calculated for a given fraction product recovery.

One of the requirements for the applicability of the above design equations is that the quantity  $(D_{AM}/K\delta)_{(wt)}$  and  $k_{(wt)}$  are independent of the feed concentration. It will be shown later, however, that  $(D_{AM}/K\delta)_{(wt)}$  and  $k_{(wt)}$  are functions of the feed tea juice concentration and the above equations are no longer applicable for the system where the concentration of the tea component increases as the feed tea juice flows in the module from the entrance to the exit. This problem can be solved when the entire length of the module is divided into many small segments

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and at each one of the segments the feed tea juice concentration is assumed to be constant (Tweddle *et al.* 1980). Therefore, the above design equations are supposed to be applicable at each module segment.

#### **EXPERIMENTAL**

#### Materials

Cellulose acetate E-398-3 powder supplied from Eastman Co. was used without further treatment. All chemicals used for the experiment were of reagent grade. Green tea powder was supplied from Zhejiang Province, P.R. China.

#### **Membrane Preparation**

Cellulose acetate membranes were prepared according to the method outlined in the literature (Sourirajan and Matsuura 1985). The details of the membrane preparation are as follows. A casting solution containing cellulose acetate polymer (17.0 wt %), acetone (69.2 wt %), magnesium perchlorate (1.45 wt %) and water (12.35 wt %) was kept at 5C. It was then cast on a smooth glass plate. The temperature of the casting atmosphere and the relative humidity of the casting atmosphere were 25C and 55%, respectively. The membrane thickness was 254  $\mu$ m. The cast film was left in the casting atmosphere for 30 s for the partial evaporation of the solvent before it was immersed into ice cold water where the gelation took place. The membrane so prepared was used for the tea juice concentration experiments without shrinkage, since it was known from our previous work (Zhang *et al.* 1991) that the above membrane exhibits very high permeation rate while maintaining a high level of separation for the major tea juice components.

#### Preparation of the Feed Tea Juice Solution

Green tea powder was weighed and dissolved in distilled water at 90C. Then, the solution was centrifuged at 6000 rpm to remove the sediment.

#### **Reverse Osmosis Experiment**

The static cell was used for the tea juice concentration. The details of the cell design are given in (Sourirajan 1970). The effective membrane area is 9.6 cm<sup>2</sup>. The details of the experimental method are also given in (Sourirajan 1970). All experiments are of short run type and carried out at the laboratory temperature under specified operating pressures. The product rate (PR) and pure water permeation rate (PWP) were corrected to 25C using the relative viscosity and density

data for water. The term "product" and "product rate" refer to membrane permeated solution when solute is present in the feed solution. The fraction solute separation obtained in each experiment was calculated from the equation,

$$f = \frac{solute \ ppm \ in \ feed - solute \ ppm \ in \ product}{solute \ ppm \ in \ feed}$$
(11)

In each experiment, (PWP) and (PR) in grams per hour per given area of film su rface  $(9.6 \text{ cm}^2)$  were determined at the operating conditions employed.

#### **Analysis of Tea Component**

Analysis of caffeine, polyphenols and amino acids was performed by spectrophotometry following the method described in *Tea Plant Physiology and Biochemistry Handbook*. According to the method, caffeine content was determined by ultraviolet spectrometry at 274 nm after adding lead acetate and sulfuric acid into the sample solution. The concentration of polyphenols was determined by ultraviolet spectrometry at 540 nm after adding ferrous tartrate into the sample solution and then diluting with buffer solution. The amino acids content was determined also by ultraviolet spectrometry at 570 nm after adding ninhydrin solution and buffer solution to the sample and heating at 100C. The Varian Cary 210 spectrophotometer was employed for the spectroscopic measurement. The total organic carbon (TOC ppm) content was determined by Beckman Total Carbon Analyzer, Model 915B. The total dissolved solid (TDS ppm) was determined by gravimetric method. The density of the green tea juice solution was determined by weighing known volumes of the green tea juice sample.

#### **RESULTS AND DISCUSSION**

#### Relationship Between $c_{A(wt)}$ , $\rho$ , $X_{A(wt)}$ , TOC and TDS

The quantities that are used in the transport equations for the tea juice concentration are weight concentration,  $c_{A(wt)}$  (kg of solute/m<sup>3</sup> of solution), density,  $\rho$  (kg of solution/m<sup>3</sup> of solution), and solute weight fraction  $X_{A(wt)}$  (dimensionless). Among those there exists the following relationship.

$$X_{A(wt)} \times 10^6 = TDS \tag{12}$$

$$c_{A(wt)} = \rho \times X_{A(wt)} \tag{13}$$

One way of determining  $c_{A(wt)}$  of a tea juice sample is to measure TDS and  $\rho$  and apply Eq. (12) and (13) to the results. This procedure is, however, very

cumbersome since it involves gravimetric method to determine TDS. The total organic carbon mesaurement requires much shorter time. A linear relationship was found between TOC and  $c_{A(wt)}$  as illustrated in Fig. 2. Therefore, except the measurement of the weight concentration of the feed tea juice freshly prepared, most of the concentration measurements were done by TOC method based on Fig. 2. A linear relationship was also found between  $\rho$  and  $c_{A(wt)}$  as illustrated in Fig. 3. Therefore,  $c_{A(wt)}$ ,  $\rho$  and  $X_{A(wt)}$  are all known for a given TOC value.  $c_{A(wt)}$  is called hereafter tea juice concentration.

#### **Osmotic Pressure**

Osmotic pressure data experimentally obtained by the method described in the theoretical section was plotted versus tea juice concentration as illustrated in Fig. 4. Although there is some scatter in the data, there is a straight line relationship between the above two quantities, suggesting the validity of the following equation.



$$\pi(X_{A2(wt)}) = B \times \rho X_{A2(wt)} \tag{14}$$

FIG. 2. TOC VS TEA JUICE CONCENTRATION, CA(wt)



FIG. 3. SOLUTION DENSITY, p, VS TEA JUICE CONCENTRATION, cA(wt)

It was replotted as osmotic pressure versus carbon weight fraction (TOC  $\times 10^{-6}$ ) in Fig. 5. The results in Fig. 5 were further compared with the osmotic pressure data of various fruit juices obtained in our earlier work (Matsuura and Sourira-jan 1978a). Surprisingly, osmotic pressure data of tea juice are exactly on the line correlating the osmotic pressure and the carbon weight fraction of fruit juices as illustrated in Fig. 6. Because the data involve fruit juices of many different origins, these results are worth receiving special attention.

#### **Transport of Tea Juice Reverse Osmosis**

Reverse osmosis experiments were conducted for various feed tea juice concentrations at different operating pressures and the results summarized in Table 1. These data were further analyzed by transport Eq. (1)–(4) together with the density and the osmotic pressure data given in Fig. 3 and 4. In the analysis the correlation between  $c_{A(wt)}$  and TOC given in Fig. 2 was assumed valid not only in the feed tea juice but also in the membrane permeate. Therefore, the separation based on TOC was regarded equal to that based on  $c_{A(wt)}$ . Strictly speaking, this assumption is not necessarily true, since the composition of the tea components in the feed is different from that in the permeate. However, since the separations of all tea juice components are nearly equal to 100%, the above assumption is a valid approximation. All the transport parameters reported hereafter are concerning the total weight of the tea juice components unless otherwise specified.

 $(D_{AM}/K\delta)_{(wt)}$  is plotted versus the feed tea juice concentration,  $c_{A1(wt)}$ , for different operating pressures in Fig. 7.  $(D_{AM}/K\delta)(wt)$  becomes less at a higher tea juice concentration and at a higher operating pressure. The correlation between  $log(D_{AM}/K\delta)_{(wt)}$  and the tea juice concentration for a given operating pressure can be approximated by a linear relationship and demonstrated by three straight lines in the figure. These three straight lines can be given as,

$$\log(D_{AM}/K\delta)_{(wt)} = -0.016c_{A1(wt)} - 7.540 \ at \ 1724 \ kPag \tag{15}$$

$$\log(D_{AM}/K\delta)_{(wt)} = -0.016c_{A1(wt)} - 7.810 \ at \ 3448 \ kPag \tag{16}$$

$$\log(D_{AM}/K\delta)_{(wt)} = -0.0156c_{A1(wt)} - 8.350 \ at \ 6895 \ kPag \tag{17}$$

A linear relationship between  $log(D_{AM}/K\delta)$  and concentration was also reported in our earlier paper (Matsuura and Sourirajan 1978b).



FIG. 4. OSMOTIC PRESSURE AS A FUNCTION OF TEA JUICE CONCENTRATION AT 25C



FIG. 5. OSMOTIC PRESSURE AS A FUNCTION OF CARBON WEIGHT FRACTION AT 25C



VARIOUS FRUIT JUICES

SOME REVERSE OSMOSIS DATA FOR			
TEA JUICE CONCENTRATION			
	Opera	ting pre	ssure, kPag
	1724	3448	6895
	РИ	$VP \times 10$	$^3$ , kg/h <sup>a</sup>
	37.24	72.00	141.50
CA1(wt)	P	$R \times 10^3$	, kg/h ª
10.94	22.71	41.96	64.29
28.73	11.90	9.67	8.98
47.11	4.22	3.06	5.23
	TC	C separ	ation, %
10.94	98.16	98.57	98.81
28.73	98.80	98.81	99.18
47.11	97.68	97.00	98.53
	Caff	eine sep	aration, %
10.94	87.97	92.30	94.30
28.73	92.48	92.47	94.90
47.11	92.64	93.22	97.42
	Polyp	henol se	paration, %
10.94	98.94	99.24	99.38
28.73	99.19	99.12	99.53
47.11	98.92	98.35	99.45
	Amino	acids s	eparation, %
10.94	99.72	99.78	99.85
28.73	99.88	99.66	99.96
47.11	99.71	99.58	99.91
<sup>a</sup> Effect	ive film	area, 9.	$6 \text{ cm}^2$

TABLE 1.
SOME REVERSE OSMOSIS DATA FOR
TEA JUICE CONCENTRATION

A similar relation was applied between  $k_{(wt)}$  and the tea juice concentration, although the effect of the operating pressure was not significant as illustrated in Fig. 8. The linear correlations are given as,

$$\log k_{(wt)} = -0.026c_{A1(wt)} - 5.020 \ at \ 1724 \ kPag \tag{18}$$

$$\log k_{(wt)} = -0.026c_{A1(wt)} - 5.106 \ at \ 3448 \ kPag \tag{19}$$

$$\log k_{(wt)} = -0.026c_{A1(wt)} - 5.179 \ at \ 6895 \ kPag \tag{20}$$



FIG. 7.  $log(D_{AM}/K\delta)_{(wt)}$  VS  $c_{A1(wt)}$  FOR TOTAL TEA JUICE COMPONENTS





As Table 1 shows the separation of caffeine is lower than that of other tea juice components. In other words, caffeine is partially removed into the membrane permeate. The degree of caffeine permeation through the membrane seems to be one of the important factors of the reverse osmosis design. Therefore, an attempt was made to calculate transport parameters for caffeine. For this purpose  $k_{(wt)}$  of caffeine solute was evaluated using Eq. (6), in which  $\alpha$  and  $\beta$  represent tea juice and caffeine, respectively.  $k_{(wt)}$  for tea juice is known from Fig. 8 for different feed tea juice concentrations and at different operating pressures. As for  $D_{AB}$  data, L-epigallocatechin gallate was assumed to represent the entire tea juice components since the latter compound constitutes the major component of the tea juice. Knowing its molecular structure  $D_{AB}$  of L-epigallocatechin gallate was calculated to be  $0.417 \times 10^{-9}$  m<sup>2</sup>/s by Eq. (7). The latter value was used as  $(D_{AB})_{\alpha}$  in Eq. (6). Similarly,  $D_{AB}$  for caffeine was calculated to be 0.656  $\times$  $10^{-9}$  m<sup>2</sup>/s and the latter value was used in Eq. (6) as  $(D_{AM})_{\beta}$ . Thus,  $k_{\beta(wt)}$  is obtainable from Eq. (6) for caffeine solute for different feed tea juice concentrations and at different operating pressures. The results are now called k<sub>caffeine(wt)</sub>. These  $k_{caffeine(wt)}$  are then used in Eq. (5) to calculate  $(D_{AM}/K\delta)_{caffeine(wt)}$ , and the results plotted versus feed tea juice concentration in Fig. 9. Linear relation-



FIG. 9.  $log(D_{AM}/K\delta)_{caffeine(wt)}$  VS  $c_{A1(wt)}$  FOR CAFFEINE

ships are found again between  $\log(D_{AM}/K\delta)_{caffeine(wt)}$  and feed tea juice concentration. These linear relationships can be given as,

$$\log(D_{AM}/K\delta)_{caffeine(wt)} = -0.026c_{A1(wt)} - 6.288 \ at \ 1724 \ kPag$$
(21)

$$\log(D_{AM}/K\delta)_{caffeine(wt)} = -0.026c_{A1(wt)} - 6.599 \ at \ 3448 \ kPag$$
(22)

$$\log(D_{AM}/K\delta)_{caffeine(wt)} = -0.026c_{A1(wt)} - 7.062 \ at \ 6895 \ kPag \tag{23}$$

Note that  $(D_{AM}/K\delta)_{(wt)}$  values are consistently higher for caffeine than for tea juice, indicating the higher permeation rate of caffeine solute. The validity of the parameters represented by Fig. 7, 8 and 9 was examined by backcalculating the product rate data using the parameters obtained from the figures in the transport equations and comparing the results with the experimental product rate values. Table 2 shows such a comparison. The agreement of the calculated and the experimental values is reasonable, testifying to the validity of the numerical values for the parameter and, particularly the linear relationship presented in Fig. 7, 8 and 9.

#### **Process Calculation**

Some design calculations were performed at two levels of the operating pressure, i.e., 3448 kPag and 6895 kPag, using system equations together with the numerical data obtained experimentally for  $(D_{AM}/K\delta)_{(wt)}$ ,  $k_{(wt)}$ , density and osmotic pressure. As design parameters  $\bar{u}^{\circ} = 9.01 \times 10^{-2}$  m/s,  $h = 0.25 \times 10^{-2}$  m and  $v_{w}^{*} =$ 

	TABLE 2.				
С	COMPARISON OF THE BACKCALCULATED AND				
	EXPERIMENTAL PRODUCT RATE DATA				
	cA1(wt) Pressure (PR)calcd (PR)exptl				
	$kg/m^3$	kPag	g/h ª	g/h ª	
	10.94	1724	25.69	20.13	
		3448	33.16	37.44	
		6895	34.09	37.78	
	28.73	1724	9.04	11.90	
		3448	9.94	8.98	
		6895	9.90	8.98	
	47.11	1724	2.59	4.22	
		3448	2.92	3.06	
		6895	2.99	5.23	
		170 01	0.0	2	

Effective film area 9.6 cm<sup>2</sup>

 $2.78 \times 10^{-5}$  m/s for 3448 kPag and  $5.56 \times 10^{-5}$  m/s for 6895 kPag, respectively, were used since the latter values are typical for the operation of some commercial module (Tweddle *et al.* 1980). As for the mass transfer coefficient, k<sub>(wt)</sub>, the value depends strongly on the hydrodynamic condition of the feed solution near the membrane and changes significantly when the design of the module and the feed flow rate are changed. The numerical values obtained by our experimental cells are not necessarily applicable for the module. However, the values given in Fig. 8 are in the same range as those obtained for the commercial module (Tweddle *et al.* 1980); therefore, they were used in the design calculation unless otherwise specified. The results of the calculation are given in Tables 3 and 4. These tables are extremely informative for the reverse osmosis concentration of tea juice. For example, looking into Table 3, when the feed concentration at the module entrance is 10 kg/m<sup>3</sup>, which is nearly equal to 1 wt%, 71% of water has to be

TABLE 3. RESULTS OF DESIGN CALCULATION FOR THE OPERATING PRESSURE OF 3448 kPag

	<b>L</b>	RESSORE O	1 J++0 KI a	5	
$c^{o}_{A1(wt)}$	$\Delta$	X	x	$C_{A1(wt)}^{b}$	CA3(wt)
$kg/m^3$			m	$kg/m^3$	$kg/m^3$
1.0	0.4614	0.5078	4.114	1.839	0.206
	0.7100	0.8082	6.548	3.387	0.251
	0.8437	0.9791	7.933	6.243	0.287
3.0	0.4614	0.5948	4.819	5.529	0.160
	0.7100	0.9630	7.803	10.20	0.199
	0.8437	1.2036	9.752	18.83	0.223
5.0	0.4614	0.6661	5.397	9.222	0.143
	0.7100	1.0968	8.887	17.07	0.175
	0.8437	1.3970	11.32	31.46	0.197
10.0	0.4614	0.8232	6.670	18.46	0.119
	0.7100	1.4047	11.38	34.12	0.149
	0.8437	1.8804	15.24	63.06	0.171
30.0	0.4614	1.4740	11.94	55.45	0.099
	0.7100	3.0580	24.78	102.4	0.142
	0.8437	18.057	146.3	181.9	0.623
50.0	0.4614	2.505	20.30	92.37	0.107
	0.7100	16.640	134.8	167.1	0.431
	0.8437	138.630	1123.0	255.1	2.403

<sup>e</sup> Results of the calculation with

 $\overline{u}^{\circ} = 9.01 \times 10^{-2} \text{ m/s}, h = 0.25 \times 10^{-2} \text{ m},$ 

and  $v_w^*=2.78\, imes\,10^{-5}$  m/s

<sup>b</sup> Weight concentration at the module exit

removed into the permeate to increase the tea juice concentration 3.4 times. The concentration in the permeate is  $0.149 \text{ kg/m}^3$  ( $\approx 0.015 \text{ wt}\%$ ). The module length required is 11.4 m. When the feed concentration at the module entrance is 50 kg/m<sup>3</sup>, which is nearly equal to 5 wt %, 71% of water has to be removed to increase the juice concentration 3.34 times. The concentration in the permeate isa bout 0.431 kg/m<sup>3</sup> ( $\approx 0.04 \text{ wt}\%$ ). The module length required is 134.8 m, which seems almost unrealistic. On the other hand, when the reverse osmosis concentration at the module entrance and achieving the same water recovery in the permeate, the tea juice concentration is increased 3.4 times. The permeate is uncentration will be 0.128 kg/m<sup>3</sup>. Thus, a smaller amount of tea essence is wasted into the permeate. More importantly, the module length required is only 33.97 m. It is particularly interesting that the module length becomes almost one quarter

PRESSURE OF 6895 kPag					
Contemporal Contem	$\Delta$	Х	x	$C_{A1(wt)}^{b}$	CA3(wt)
$kg/m^3$			m	$kg/m^3$	$kg/m^3$
1.0	0.4614	0.7165	2.903	1.815	0.486
	0.7100	1.1506	4.661	3.316	0.540
	0.8437	1.4184	5.746	6.085	0.580
3.0	0.4614	0.9009	3.650	5.502	0.264
	0.7100	1.4714	5.961	10.12	0.300
	0.8437	1.8491	7.491	18.67	0.322
5.0	0.4614	1.0298	3.726	9.196	0.204
	0.7100	1.7060	6.869	16.96	0.234
	0.8437	2.1801	12.69	31.32	0.250
10.0	0.4614	1.2958	5.250	18.44	0.145
	0.7100	2.2152	8.974	34.07	0.170
	0.8437	2.9512	11.96	62.98	0.186
30.0	0.4614	2.3063	9.343	55.45	0.095
	0.7100	4.4886	18.18	102.5	0.124
	0.8437	7.7125	31.24	189.4	0.156
50.0	0.4614	3.6087	14.62	92.45	0.089
	0.7100	8.3957	33.97	170.8	0.128
	0.8437	38.265	155.0	311.1	0.327

TABLE 4.
RESULTS OF DESIGN CALCULATION FOR THE OPERATING
PRESSURE OF 6805 kPag

<sup>a</sup> Results of the calculation with

 $\overline{u}^{\circ} = 9.01 \times 10^{-2} \text{ m/s}, h = 0.25 \times 10^{-2} \text{ m},$ 

and 
$$v_w^{\circ} = 5.56 \times 10^{-5} \text{ m/}$$

<sup>b</sup> Weight concentration at the module exit



by doubling the operating pressure, thus reducing the capital cost considerably. Operating cost however increases due to an increase in the operating pressure. All the above factors have to be taken into consideration for the optimal design of the system. Figure 10 illustrates the relation,  $\Delta$  versus X, when the parameter  $\lambda\theta$  are kept constant throughout the module at two different levels, i.e., 0.14 and 0.34. Note that the parameter  $\lambda\theta$  is equal to  $k_{(wt)}/v_{\mathbf{x}}$  and increases with an increase in the mass transfer coefficient  $k_{(wt)}$ . The figure clearly indicates that less X is required for a given  $\Delta$  when  $\lambda\theta$  is high. Therefore, a shorter module length is enough to obtain a given product recovery by increasing the mass transfer coefficient.

#### CONCLUSIONS

From the above investigations the following conclusions can be drawn.

(1) The osmotic pressure of the green tea juice is the same as those of fruit juices for a given carbon weight fraction of the solute.

(2) Both solute transport parameter and mass transfer coefficient of the green tea juice components decrease with an increase in the tea juice concentration and with an increase in the operating pressure.

(3) An increase in the operating pressure reduces the module length required to obtain a given fraction product recovery enormously, particularly when the green tea juice concentration is high.

(4) An increase in the mass transfer coefficient of the green tea juice component on the high pressure side of the membrane also reduces the module length.

#### NOMENCLATURE

A	=	Pure water permeability constant on weight basis, $kg/m^2\ s$
		Pa
В	=	Proportionality constant for osmotic pressure, Pa
D <sub>AB</sub>	=	Diffusivity of solute in water, m <sup>2</sup> /s
$(D_{AM}/K\delta)_{(wt)}$	=	Solute transport parameter on weight basis, m/s
f	=	Fraction solute separation
1/h	=	Membrane area per unit volume of channel space, 1/m
k <sub>(wt)</sub>	=	Mass transfer coefficient on weight basis, m/s
M <sub>B</sub>	=	Molecular weight, kg/kmol
N <sub>B(wt)</sub>	=	Solvent water flux through membrane on weight basis,
		kg/m <sup>2</sup> s
Р	=	Operating pressure, Pa
PWP	=	Pure water permeability through effective membrane area,
		kg/h
S	=	Effective membrane area, m <sup>2</sup>
ū°	=	The average velocity of feed solution at the module en-
		trance, m/s
x	=	A longitudinal distance from the module entrance, m
Х	=	v <sub>w</sub> x/ <u>u</u> °h
X <sub>A(wt)</sub>	=	Weight fraction of solute
V <sub>s</sub>	=	Permeation velocity of product solution, m/s
$V_{w}^{*}$	=	Pure water permeation velocity, m/s
		•

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#### ASSESSMENT OF A SEMI-EMPIRICAL FOUR PARAMETER GENERAL MODEL FOR SIGMOID MOISTURE SORPTION ISOTHERMS<sup>1</sup>

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#### ABSTRACT

Published sigmoid moisture sorption isotherms  $(0 < a_w < \sim 0.9)$  were fitted by the four parameter models  $m = k_1 a_w^{n_1} + k_2 a_w^{n_2}$  where m is the moisture contents (dry basis),  $a_w$  the water activity and the k and n values are constants  $(n_1 < 1 \text{ and } n_2 > 1)$ . Not surprisingly, the model had the same or better fit than the GAB model. In contrast with both the BET and GAB models, the proposed model is not based on the assumption that there exists a well-defined monolayer of absorbed water. At  $a_w < \sim 0.4$  and  $n_1 > \sim 0.35$ , however, the model produces a practically linear  $a_w/[m(1-a_w)]$  vs  $a_w$  plot, of the kind used to calculate the monolayer moisture with the BET model.

The proposed model can be a convenient means to catalog both sigmoid and nonsigmoid isotherms, and used to calculate the equilibrium water activity of dry mixtures with equations solving software.

#### INTRODUCTION

The basis of most of the kinetic moisture sorption models, especially for foods, is the BET model (Karel 1975). It was originally developed for physical adsorption of inert gases on solid surfaces. Its main feature is the monolayer concept, and that the sorption process is regulated by two mechanisms; one with a Langmuir kinetics, in which gas molecules are directly absorbed on selected sites at the

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<sup>&</sup>lt;sup>1</sup>Contribution of the Massachusetts Agricultural Experiment Station in Amherst.

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solid surface until the latter is covered by a layer of gas one molecule thick, and the other of condensation in subsequent layers (Van den Berg 1985). When applied to moisture sorption isotherms, the model is expressed by the equation

$$m = m_0 a_w / [(1 - a_w) (1 + (c_B - 1) a_w)]$$
(1)

where  $a_w$  is the water activity, m the moisture at a given  $a_w$ ,  $m_o$  the moisture contents of the monolayer and  $C_B$  a constant.

Eq. (1) can be rearranged to yield the linear relationship

$$a_w / [m(1-a_w)] = 1/m_o C_B + (C_B - 1) a_w / m_o C_B$$
 (2)

This relationship was found to be experimentally valid for water activities of up to 0.3–0.5, and traditionally the magnitude of the monolayer moisture contents ( $m_o$ ) has been calculated from the intercept and slope of the  $a_w/[m(1-a_w)]$  vs  $a_w$  plot in this water activity range (Karel 1975; Labuza 1984). The failure of the BET model at higher  $a_w$  levels had led to the development of the three parameter GAB model (Van den Berg 1985)

$$m = m_0 C_G K a_w / [1 - K a_w) (1 - K a_w + C_G K a_w)]$$
(3)

where  $C_G$  and K are constants. Its applicability range, as judged by the fit to experimental data, is up to about  $a_w = 0.9$ . Modified GAB models, with four or more constants, may have an improved fit but they do not significantly extend the applicable water activity range (Van den Berg 1985). The latter, it has recently been shown (Schuchmann *et al.* 1990), could be extended to water activities of up to 0.98–0.99 by replacing  $a_w$  in the GAB model (Eq. 3) by a transform such as  $-\ln(1-a_w)$ .

Although the BET and GAB models and the monolayer concept on which they are based are apparently useful in explaining various stability mechanisms, they are not always compatible with other aspects of the moisture sorption phenomenon. Examples are the discrepancy between the BET surface area measured with inert gases like N<sub>2</sub> or He, on the order of 1–4 m<sup>2</sup>/g and that determined by water vapor sorption, on the order of hundreds of square meters per gram, and the discrepancy between the observed and predicted heat of sorption and its moisture dependency.

There are numerous published mathematical models that describe water sorption on foods. According to Van den Berg and Bruins (1981) who list 77 isotherm equations, (some are variations of the same model), they can be divided into four groups. According to their classification these are: kinetic models based on a monolayer, kinetic models based on multilayer sorption and a condensed film, models imported from the polymer literature and ad hoc empirical models. The constants of the kinetic models, in contrast with those of the empirical model, are material physical properties. Therefore, their determination requires establishment of the appropriate sorption mechanism and verification of the model's parameter magnitude by an independent physical test. However, in most practical food applications of sorption isotherms, as in stability assessment, drying design or prediction of the equilibrium water activity of mixtures for example, verification of the kinetic model, which requires additional analyses and considerable effort is usually not done.

With the spread and ease of computer aided curve fitting techniques based on nonlinear regression, the traditional empirical models such as Henderson's, Smith, Oswin's and Kuhn's (Iglesias and Cheriffe 1982) have lost some of their advantages and main attraction — linearization and two constants only — in favor of the kinetic models of the BET/GAB family. However, when the constants of these are determined by nonlinear regression, their magnitude most importantly that of m<sub>o</sub>, the monolayer value, depends not only on the selected model and the experimental a<sub>w</sub> range, but also on any noise in the sorption data and the number of points taken for the regression. Therefore, it is theoretically very difficult to decide on the basis of the fit of the BET or GAB models equations alone whether they truly represent the molecular events that take place at the solid surface. Since confirmation of the validity of the monolayer value by independent physical tests can be a cumbersome task and probably unnecessary for most routine applications, it is worthwhile to consider an alternative model that does not require the existence of a certain moisture content level that corresponds to a monolayer in the molecular domain. Such a model, however, need not exclude the monolayer existence. It only implies that the latter cannot be determined unambiguously from the shape of the moisture sorption isotherm alone. The objective of this work is to propose such a model, test its applicability with published sorption data of foods, and examine its properties and potential uses.

#### THE MODEL

One of the simplest four parameter models that can describe a sigmoid m vs  $a_w$  curve is the double power expression:

$$\mathbf{m} = \mathbf{k}_1 \mathbf{a}_{\mathbf{w}}^{\mathbf{n}} \mathbf{1} + \mathbf{k}_2 \mathbf{a}_{\mathbf{w}}^{\mathbf{n}} \mathbf{2} \tag{4}$$

where  $k_1$ ,  $k_2$ ,  $n_1$  and  $n_2$  are constants ( $n_1 < 1$  and  $n_2 > 1$ ). When the k values are of comparable magnitude the first term ( $n_1 < 1$ ) is always the dominant at the lower range of water activity. At higher water activities either term or both can be dominant or of comparable magnitude. [To demonstrate this point, let us

examine a case where  $k_1 = k_2 = 1$ ,  $n_1 = 0.5$  and  $n_2 = 5$ . According to Eq. (4) the magnitude of the two terms will be 0.5 and 0.001 at  $a_w = 0.25$ , and 0.87 and 0.24, respectively, at  $a_w = 0.75$ ].

Eq. (4) is not derived from any theoretical considerations and is therefore in this respect, an empirical moisture sorption model, which compared with the BET or GAB model has an additional one or two constants. However, the number of constants was not increased to improve the model fit, and the mathematical structure of Eq. (4) was not arbitrarily selected. It was chosen in order to show that the shape of sigmoid sorption isotherms at low water activities is primarily determined by a process of a decreasing rate with respect to  $a_w$ ,  $(dm^2/da^2_w < 0)$ , similar to a Langmuir kinetic, (see below). The departure from the BET family of models is that this process does not require the existence of any asymptotic moisture contents as in the classic Langmuir kinetics and isotherm. At higher water activities there is a shift toward free condensation, manifested by the isotherm's monotonous and ever increasing slope. In fact, the mathematical format of both the BET and GAB models was initially derived on the basis of these very same two mechanisms, and they also can be written as a two term model in the form (Watt 1983; Van den Berg 1985):

$$m = Aa_w / (B+a_w) + Ca_w / (D-a_w)$$
(5)

where A, B, C and D are constants, and where the first term represents sorption with a decreasing rate with respect to  $a_w$  and the second with an ever increasing rate.

As already mentioned the difference between Eq. (4) and (5) is that in the former the first term has no asymptote. In Eq. (5) in contrast as  $a_w \rightarrow \infty$ , (no physical meaning)  $Aa_w/(B+a_w) \rightarrow A$ . The second term in Eq. (5) implies that as  $a_w \rightarrow$  $D \ m \rightarrow \infty$ , i.e., free condensation, while in Eq. (4) both m and dm/da<sub>w</sub> always have a finite magnitude. (This, however, is a moot point, since both types of models fail at  $a_w > \sim 0.9$ ).

The model expressed by Eq. (4) has no monolayer incorporated in it. However, plots of  $a_w/[m(1-a_w)]$  vs  $a_w$ , those used to determine the monolayer value with the BET model, are still expected to be practically linear over a water activity range of up to about 0.4. This is because at this range of  $a_w$  Eq. 4 can be approximated by

$$m \cong k_{1}a_{w}^{n_{1}} \qquad (n_{1} < 1) \qquad (6)$$

and

$$a_w / [m(1-a_w)] = a_w / [k_1 a_w^n (1-a_w)]$$
 (7)

As shown in Fig. 1, the plot of  $a_w/[a_w^{n_1}(1-a_w)]$  vs  $a_w$ ,  $(k_1 = 1)$  is indeed linear in this water activity range and for a large range of  $n_1$  values. The intercept is positive when  $n_1$  is bigger than about 0.3, the range actually found when the model is fitted to real food data (see below). In fact, the slight upward concavity of the simulated plot at the end of the range is moderated by the small contribution of the second term thus extending somewhat the  $a_w$  range at which the apparent linear relationship holds. Or, in other words, with

$$a_{w} / [m(1-a_{w})] = (a_{w} / k_{1}) / \{a_{w}^{n} [1 + (k_{1} / k_{2}) a_{w}^{n} 2^{-n} ] (1-a_{w}) \}$$
(8)

the added term,  $[1+(k_1/k_2) a_w^{n_2-n_1}]$ , in the denominator tends to lower the magnitude of  $a_w/[m(1-a_w)]$  as the magnitude of  $a_w$  increases.

Furthermore, experimental sorption data are prone to have some scatter, which can be simulated by generating a random noise. As demonstrated in Fig. 1 even a 2% scatter can make it impossible to decide, on the basis of the  $a_w/[m(1-a_w)]$ vs  $a_w$  plot, which of the two models, the BET or the double power [Eq. (4)] more faithfully describes the sorption phenomenon at this water activity range. (The simulated 2% noise was created by multiplying the calculated  $a_w/[m(1-a_w)]$  values by 0.98 + 0.04\*URN, URN being the random number generating operator of the SYSTAT package with 0 < N < 1.)

#### APPLICATION OF THE MODEL TO EXPERIMENTAL SORPTION DATA

Published equilibrium moisture sorption data of agar-agar, carrageenan, gelatin, low methoxyl pectin and wheat bran, from Labuza (1984), Bizot (1983) and Maroulis *et al.* (1988), in the range of water activity of up to about 0.85–0.95 were fitted with the proposed double power model and the GAB model [Eq. (3)]. This was done using the nonlinear regression program of the SYSTAT 5.1 package (SYSTAT, Inc., Evanston, IL) on a Macintosh SE 30 microcomputer. To confirm that the regression parameters were indeed unique the regression was repeated with various initial guessed values above and below those calculated.

Comparison of the fit of the GAB and double exponential models is shown in Fig. 2-11. As could be expected, the four parameter model [Eq. (4)] had a comparable or slightly better fit than the three parameter model [Eq. (3)] and this was also evident from the relative magnitudes of the mean square error in (Table 1). Like the GAB model the proposed model was only applicable up to an  $a_w$  level of about 0.9. Beyond this range ( $a_w > 0.95$ ) it also could only be applied if  $a_w$  was replace by a transform,  $-\ln(1-a_w)$  for example. Table 1 also shows that the magnitude of the power  $n_1$  in all the materials was on the order 0.3-0.8





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AGAR-AGAR

Open circle experimental data, from Labuza (1984), dashed line the fit of the GAB model [Eq. (3)], solid line the fit of the double power model [Eq. (4)].





Open circle experimental data, from Labuza (1984), dashed line the fit of the GAB model [Eq. (3)], solid line the fit of the double power model [Eq. (4)].





Open circle experimental data, from Labuzaa (1984), dashed line the fit of the GAB model [Eq. (3)], solid line the fit of the double power model [Eq. (4)].





Open circle experimental data, from Labuza (1984), dashed line the fit of the GAB model [Eq. (3)], solid line the fit of the double power model [Eq. (4)].


# FIG. 6. MOISTURE SORPTION ISOTHERM OF WHEAT BRAN

Open circle experimental data, from Labuza (1984), dashed line the fit of the GAB model [Eq. (3)], solid line the fit of the double power model [Eq. (4)].



# FIG. 7. MOISTURE SORPTION ISOTHERM OF RAISINS

Open circle experimental data, from Maroulis *et al.* (1988), dashed line the fit of the GAB model [Eq. (3)], solid line the fit of the double power model [Eq. (4)].







Open circle experimental data, from Bizot (1983), dashed line the fit of the GAB model [Eq. (3)], solid line the fit of the double power model [Eq. (4)].



dashed line the fit of the GAB model [Eq. (3)], solid line the fit of the double power model [Eq. (4)].



# FIG. 11. MOISTURE SORPTION ISOTHERM OF COFFEE

Open circle experimental data, from Bizot (1983), dashed line the fit of the GAB model [Eq. (3)], solid line the fit of the double power model [Eq. (4)].

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Material	GA	B mode	l (Eq.	3)	Doub	le pow	er law mo	odel (Ec	1.4)
	m <sub>o</sub> (%db)	k (-)	C <sub>G</sub> (-)	MSE	k1 (%db)	nı (-)	k <sub>2</sub> (%db)	n2 (-)	MSE <sup>4</sup> )
Agar-Agar <sup>1)</sup>	13.3	0.74	47.1	3.8	31.9	0.53	25.4	14.7	0.65
Carrageenan <sup>l</sup>	)13.4	0.84	19.3	3.9	34.3	0.61	119	12.0	0.74
Gelatin <sup>1)</sup>	11.4	0.85	22.8	3.3	28.8	0.58	105	12.1	0.90
Low methoxyl pectin	1) 9.5	0.94	3.9	3.7	14.0	0.39	90.8	6.0	0.25
Wheat bran <sup>1)</sup>	5.8	0.86	13.9	0.45	16.4	0.74	62.8	13.8	0.14
Raisins <sup>2)</sup>	18.2	0.88	0.87	3.8	11.0	0.34	97.2	4.9	0.63
Casein <sup>3)</sup>	8.7	0.65	7.0	0.06	9.2	0.36	12.7	2.3	0.03
Potato <sup>3)</sup> starch	9.9	0.75	8.3	0.05	20.9	0.70	16.3	5.5	0.05
Dextrin <sup>3)</sup>	11.5	0.46	34.9	0.08	17.9	0.38	4.4	8.1	0.18
Coffee <sup>3)</sup>	4.4	0.94	1.3	0.05	12.8	1.5	42.4	10.1	0.11

TABLE 1. EQUILIBRIUM MOISTURE SORPTION ISOTHERM CONSTANTS OF VARIOUS FOOD MATERIALS AT AMBIENT TEMPERATURE

1) Data from Labuza (1984).

2) Data from Maroulis et al.(1988).

3) Data from Bizot (1983).

4) MSE is the mean square error.

while that of  $n_2$ , between 6 and 15. The magnitude of the corresponding k values also varied considerably between the materials but remained on the order of 14–35 and 25–120% moisture on dry basis for  $k_1$  and  $k_2$ , respectively. Since most nonlinear regression procedures require an initial guess of the parameter's magnitude, the n and k values in the table can serve as a convenient starting point when the proposed model is applied to a new set of sorption data.

As shown in Fig. 1, when  $n_1$  is smaller than about 0.4 the conventional BET plot,  $a_w/[m(1-a_w)]$  vs  $a_w$ , from which traditionally the monolayer is determined has an intercept very close to zero or possibly even negative. This entails that if  $n_1 \sim 0.3$  for example, unambiguous determination of the BET monolayer value becomes very difficult, if not altogether impossible. The raisins data,  $n_1 = 0.34$ , clearly demonstrate this point (Fig. 12). The same applies to situations where  $n_1 > 1$ , in which case the BET plot has a downward concavity and cannot be



used to determine the monolayer value. This is demonstrated in the coffee results,  $n_1 = 1.46$ , shown in Fig. 13. In both the raisin's and coffee's cases the GAB model implied the existence of a substantial monolayer on the order of 18 and 4 gH<sub>2</sub>O/g solid, respectively, which again shows that the issue of whether there exists a monolayer value cannot be decided on the basis of the moisture isotherm curve shape and the statistical fit of any particular mathematical model.



### **Other Potential Applications of the Model**

The proposed model, Eq. (4), having four constants, is expected to fit tabulated data of a variety of foods very well. Because of its mathematical structure, its determination by nonlinear regression is fairly straightforward and, in contrast with other types of models, convergence is almost always guaranteed in the first trial. (When m is expessed as % on dry basis a standard initial guess of values similar to those reported in Table 1 will almost invariably work). Since the model's applicability does not depend on the validity of any specific kinetic assumption, and its water activity range is fairly large, it can serve as a standard model for cataloging the sorption data of a large group of food and other biological materials. The availability of such a standard model could improve on the currently available catalog (Iglesias and Chirife 1982) where the sorption data are presented in terms of six different equations each valid in a different water activity range.

In this capacity the model need not be limited to sigmoid moisture sorption isotherms (see Fig. 11 and the coffee constants in Table 1). By allowing  $n_1 > 1$  Eq. (4), will effectively describe isotherms that are distinctly concave upward, and with  $n_1 = 1$  isotherms that have a linear or practically linear region at low water activities.

Irrespective of whether  $n_1 < or n_1 > 1$ , the proposed model can also be employed, conveniently, to calculate or estimate the equilibrium water activity, a<sup>\*</sup>, of mixtures, using an equation solving software (Peleg and Norman 1992). Knowing the n and k values of the ingredients for the pertinent temperature, all that is needed is to write down in the appropriate format the expression:

$$a_{\mathbf{w}}^{\star} = \operatorname{root} \left[ m_{\mathrm{T}} \cdot \Sigma \mathbf{x}_{\mathrm{i}} \left( \mathbf{k}_{\mathrm{li}} a_{\mathbf{w}}^{n} \mathbf{li} + \mathbf{k}_{2\mathrm{i}} a_{\mathbf{w}}^{n} \mathbf{2i} \right) \right]$$
(9)

where  $m_T$  is the mixture's total moisture contents on a dry solid basis and the  $x_i$  values, the weight fractions of the ingredients on a dry basis. The value of  $a_x^*$  will then instantaneously appear on the screen as demonstrated in Fig. 14. [Calculation of the equilibrium water activities of a mixture of the first five materials having different levels of moisture was done using the MathCAD software, Mathsoft, Inc., Cambridge, MA using the procedure described by Peleg and Normand (1992)].

## CONCLUSIONS

According to scientific tradition, simpler models or models with fewer adjustable parameters are always preferable to more elaborate models. If the BET and GAB models parameters were indeed objective material properties, and could be determined by an independent test, then there would be no reason to doubt their

$$f(m, a) := root \begin{cases} m \cdot .2 \cdot \begin{bmatrix} 31.9 \cdot a & + 25.4 \cdot a \\ + 25.4 \cdot a & + 25.4 \cdot a \end{bmatrix} \dots \\ + \begin{bmatrix} ..2 \cdot \begin{bmatrix} 34.3 \cdot a & + 119 \cdot a \\ + 119 \cdot a & \end{bmatrix} \dots \\ + \begin{bmatrix} ..2 \cdot \begin{bmatrix} 28.8 \cdot a & + 105 \cdot a \\ + 105 \cdot a & + 105 \cdot a \end{bmatrix} \dots \\ + \begin{bmatrix} ..2 \cdot \begin{bmatrix} 14.0 \cdot a & + 90.8 \cdot a \\ + 90.8 \cdot a & \end{bmatrix} \\ + \begin{bmatrix} ..2 \cdot \begin{bmatrix} 14.0 \cdot a & + 62.8 \cdot a \\ + 62.8 \cdot a & \end{bmatrix} \end{bmatrix} \dots , a \\ + \begin{bmatrix} ..2 \cdot \begin{bmatrix} 16.4 \cdot a & + 62.8 \cdot a \\ + 62.8 \cdot a & \end{bmatrix} \end{bmatrix}$$

FIG. 14. CALCULATION OF THE EQUILIBRIUM WATER ACTIVITY OF 1:1:1:1:1 MIXTURES (DRY BASIS) OF THE FIRST FIVE MATERIALS IN TABLE 1 USING THE DOUBLE POWER MODEL [EQ. (9) WITH  $x_1 = x_2 = x_3 = x_4 = x_5 = 0.2$ ] AND THE MATHCAD<sup>R</sup> PROGRAM The equation has been edited to fit a one page format.

m = the total moisture contents (dry basis) a = the equilibrium water activity The first number inside the parentheses is the total moisture and the second a guessed value of the equilibrium water activity, in our case a standard (arbitrary) a = 0.25.

usefulness and admire their elegance and compactness. Unfortunately, these criteria are not satisfied when these models are applied to moisture sorption on foods. And since the magnitude of their parameters depends on the water activity range and number of points, for example, they are basically equivalent to an empirical model of the form (Schuchmann *et al.* 1990);

$$m = C_1 a_w / [(1 + C_2 a_w) (C_3 - a_w)]$$
(10)

with  $C_1$ ,  $C_2$  and  $C_3$  constants. (In the BET model  $C_3 = 1$  by definition).

As long as no physical significance is assigned to the constants of Eq. (10) it is unquestionably the most effective model to describe sigmoid moisture sorption curves. However, when the constants are used to calculate monolayer values, internal inconsistencies and discrepancies with other models and physical phenomena usually arise. In such cases it is, therefore, safer to use a model that does not require, though does not exclude, the existence of a monolayer. An example of such a model is given in Eq. (4). At the expense of an extra constant, admittedly a drawback, it captures the fact that sigmoid moisture isotherms are the result of two sorption mechanisms, and at the same time enables classification of other isotherm types, regulated by different kinetics, with the same

mathematical format. In either application, determination of this model's constants by nonlinear regression does not require elaborate guessing, and they can very conveniently be used to calculate the sorption characteristics of mixtures using equation solving software.

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# SURFACE HEAT TRANSFER COEFFICIENTS ASSOCIATED WITH HEATING OF FOOD PARTICLES IN CMC SOLUTIONS

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## ABSTRACT

The fluid-to-particle heat transfer coefficients ( $h_{\rm fp}$ ) associated with the heating of cylindrical specimens of potato and carrot were evaluated in carboxymethylcellulose (CMC) solutions (0–1%) at 50–80C. Average  $h_{\rm fp}$  values ranged from 80 to 450 W/m<sup>2</sup>C for potatoes and 100 to 550 W/m<sup>2</sup>C for carrots. Temperature and concentration of CMC influenced its power law parameters and hence,  $h_{\rm fp}$ values while fluid velocity (0.2 to 0.7 × 10<sup>-3</sup> m/s), sample size (0.016–0.023 m diameter, 0.02–0.04 m length) and flow direction (upward and downward) had marginal effects. Natural convection dominated the flow regime, giving good correlations between Nusselt and Rayleigh numbers.

# **INTRODUCTION**

Data on thermal properties and boundary values are required in the design of thermal processing equipment and process schedule, especially for predicting the time-temperature response of a food undergoing heat processing. Successful temperature prediction for particulate foods in viscous fluids requires data on the convective heat transfer coefficient ( $h_{fp}$ ) at the fluid-particle interface. In most conventional canning applications employing steam, the surface heat transfer coefficient is assumed to be infinite (Ball and Olson 1957), although lower values have been recognized for steam/air heating media (Pflug and Borrero 1967;

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Ramaswamy *et al.* 1983; Tung *et al.* 1984). For continuous sterilization of food particulates, de Ruyter and Brunet (1973) and Manson and Cullen (1974) assumed the heat transfer coefficient to be infinite. But, the existence of finite convective heat transfer coefficient at the fluid-particle interface has been demonstrated by several researchers (Chang and Toledo 1989; Sastry 1986; Deniston *et al.* 1987; Chandarana and Gavin 1989; Chandarana *et al.* 1990).

The majority of carrier fluids used in the food industry are viscous and non-Newtonian. Higher viscosities associated with such fluids prevent turbulence under practical situations, and laminar regimes generally prevail. The heat transfer mechanism associated with non-Newtonian fluids is more complicated as a result of the combination of natural and forced convection (Alhamdan and Sastry 1990). Irrespective of the mechanism, heat transfer rates from these fluids have been recognized to be lower than from water.

Zuritz et al. (1990) obtained h<sub>fp</sub> values between 548 and 1175 W/m<sup>2</sup>C for a mushroom-shaped aluminum particle immersed in carboxymethycellulose (CMC) with mean apparent viscosity between 2.08 to 17.70 Pas. They reported that  $h_{fp}$ values increased with particle size and fluid flow rate and decreased with apparent viscosity at a temperature of 71C. Under similar conditions, Alhamdan and Sastry (1990) found hfp values of 75-310 W/m2C for natural convection at 20-80C, while for water the  $h_{fp}$  values were 652-850 W/m<sup>2</sup>C. The authors also observed that  $h_{fp}$  values increased with the initial temperature difference and decreased with the apparent fluid viscosity. Chandarana et al. (1989) found h<sub>fp</sub> values between 8.1 and 35.9 W/m<sup>2</sup>C for a 25.4 mm silicone rubber cube held stationary at 135C in starch, decreasing with consistency coefficient between 3.0  $\times 10^{-3}$  and 3.17 Pas<sup>n</sup>. In water, the h<sub>fp</sub> value was 51.1 W/m<sup>2</sup>C. For water and starch (2-3%) at 129.4C, Chandarana *et al.* (1990) found h<sub>fp</sub> values of 65.7–107.1  $W/m^2C$  and 55.6–86.5  $W/m^2C$ , respectively. By recording the surface temperature of spherical potatoes in rotating cans containing water, Deniston et al. (1987) reported average  $h_{fp}$  of 160+30 W/m<sup>2</sup>C. Chang and Toledo (1989) found  $h_{fp}$ values for potato cubes in water to be 239 and 303 W/m<sup>2</sup>C at 0 and 0.86 cm/s, while in stationary 35% sucrose solution it was 146 W/m<sup>2</sup>C. Chang and Toledo (1990) found the average  $h_{fp}$  at ~135C to range from 600–1533 W/m<sup>2</sup>C and 356-735 W/m<sup>2</sup>C at 0 and 1.58 cm/s fluid velocity in a packed bed of carrot cubes (1 and 2 cm), assuming a carrot thermal diffusivity value of  $1.94 \times 10^{-7}$  m<sup>2</sup>/s. Lamberg and Hallstrom (1986) simulated temperature profiles and heat transfer coefficients during blanching of carrots, and found good correlation between the theoretical and experimental data when the heat transfer coefficient was 750  $W/m^2C$ .

The objectives of this study were to: (1) determine the magnitude of the fluidto-particle heat transfer coefficient for heating finite cylindrical samples of carrots and potatoes in CMC solutions, (2) study the effect of CMC concentration on  $h_{fp}$ , and (3) study the effect of fluid flow rate, direction of fluid flow, and particle size (diameter and length) on  $h_{fp}$ . Cylindrical test samples and related equations were employed in these studies in order to broaden the scope and validity of heat transfer concepts since the majority of previous studies employed spherical or cube-shaped particles.

# THEORETICAL CONSIDERATIONS

A finite cylinder is formed as the intersection of an infinite slab and an infinite cylinder (Heldman and Singh 1981). Therefore, the solution with respect to finite cylindrical objects under surface convection will involve heat transfer equations governing an infinite slab and an infinite cylinder. The solution for the temperature history of an infinite cylinder and infinite plate with uniform initial temperature when plunged into a constant temperature environment are detailed below (Luikov 1968):

Infinite cylinder:

$$U_{c} = 2 \operatorname{Bi}_{c} \sum_{n=1}^{\infty} [J_{0}(\gamma_{n}r/a) / ((\operatorname{Bi}_{c}^{2} + \gamma_{n}^{2}) J_{0}(\gamma_{n}))] \cdot \exp(-\gamma_{n}^{2} Fo_{c})$$
(1)

where  $\gamma_n$  is the nth positive root of

$$\gamma J_1(\gamma) = Bi_c J_0(\gamma) \tag{2}$$

Infinite slab:

$$U_{p} = \sum_{n=1}^{\infty} [2 \sin \beta_{n} \cos (\beta_{n} X/L)/(\beta_{n} + \sin \beta_{n} \cos \beta_{n})] \cdot \exp(-\beta_{n}^{2}Fo_{p})$$
(3)

where  $\beta_n$  is the nth positive root of

$$\beta \tan \beta = Bi_p \tag{4}$$

After a lapse of certain time (Fo > 0.2), the terms in the infinite series of Eq. (1) and (3) converge rapidly (to the first term) due to the presence of the exponential term (Heisler 1947). Therefore, in most cases, the series solutions can be approximated by the first term and represented as follows for an infinite cylinder:

$$U_{c} = R_{c} \exp(-S_{c}F_{o})$$
(5)

and an infinite slab:

$$U_{p} = R_{p} \exp(-S_{p}F_{0})$$
(6)

where  $R_c$ ,  $R_p$ ,  $S_c$  and  $S_p$  are characteristic functions of the Biot number defined as follows:

$$R_{c} = 2 \operatorname{Bi}_{c} J_{o}(\gamma_{n} r/a) / [(\operatorname{Bi}_{c}^{2} + \gamma_{1}^{2}) J_{o}(\gamma_{1})]$$
(7)

$$S_c = \gamma_1^2 \tag{8}$$

$$\mathbf{R}_{\mathbf{p}} = [2\sin\beta_1\cos(\beta_n X/L)]/[\beta_1 + \sin\beta_1\cos\beta_1]$$
(9)

$$S_p = \beta_1^2 \tag{10}$$

Solutions for the characteristic functions generally involve graphical methods or computer work. Simplified forms of the characteristic functions have been published (Ramaswamy *et al.* 1982), valid for Fo > 0.2, and 0.02 < Bi < 200. For a finite cylinder, Eq. (5) and (6) are combined to give

$$U_{fc} = U_p \cdot U_c = R_p R_c \cdot exp - [S_p Fo_p + S_c Fo_c]$$
(11)

Or,

$$U_{fc} = R_p R_c \cdot exp - [(S_p t/L^2 + S_c t/a^2) \alpha]$$
(12)

Ball and Olson (1957), introduced an equation for calculating the thermal process time as follows:

$$t = f_{h} \left[ \log j + \log (T_{a} - T_{i}) / (T_{a} - T) \right]$$
(13)

This represents an equation of the straight line portion of the curve on a semilogarithmic plot of temperature ratio versus time. Equation (13) can be rearranged for a finite cylinder as:

$$[(T_a-T)/(T_a-T_i)] = U_{fc} = j \exp [2.303t/f_h]$$
(14)

Comparing Eq. (12) and (14), it is evident that  $j = R_c R_p$  and that the exponential terms can be equated and written as:

$$f_{h} = 2.303/[(S_{p}/L^{2} + S_{c}/a^{2})\alpha]$$
(15)

It is important to note that a finite cylinder has two finite dimensions, and will therefore have one set of Bi and Fo related to an infinite slab and another set

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related to an infinite cylinder. In other to solve for the convective heat transfer coefficient, Eq. (15) has to be solved by an iterative technique.

# **MATERIALS AND METHODS**

#### **Raw Material and CMC Solution Preparation**

Carrots and potatoes were purchased from a local market and stored in a refrigerator at 5C. Cylindrical specimens were punched out using a cork borer of known diameter, and the required length was cut using a knife.

Two concentrations (0.5 and 1.0% w/w) of commercial CMC (Sigma; St. Louis, MO) were used. CMC solutions were prepared by adding a known amount of CMC powder to water and thoroughly mixing to break undispersed lumps. The solution was left for 24 h to ensure total dispersion of the powder. Final handmixing was done to obtain a homogeneous solution. The solution was heated to the desired temperature in a steam kettle and transferred to a constant temperature water bath.

#### **Rheological Properties**

Rheological properties of CMC were determined before and after each experimental run with a Haake rotational viscometer Model Rotovisco RV3 (Haake Mess-Technik GmbHU. Co., Kerlsruhe, Germany). In order to maintain the same rheological properties it was found necessary to add some fresh CMC solution to the bath at the end of each run. Needed rheological data were obtained from previous studies (Abdelrahim *et al.* 1991) in the form of coefficients for the power law model (Table 1):

$$\sigma = m \left( \dot{\gamma} \right)^n \tag{16}$$

#### **Thermal Properties**

The thermal conductivity and specific heat of CMC solutions were estimated at bath temperatures using the following equations (Heldman and Singh 1981):

$$k_f = [326.575 + 1.0412T - 0.00337T^2] [0.796 + 0.009346(\%W)] \times 10^{-3}$$
 (17)

$$C_{nf} = 1.675 + 0.025 \,(\%W)$$
 (18)

The specific heat of carrots (moisture content, 84%; negligible fat content) was determined by the relationship in British units (Charm 1978):

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Concentration	Temperature	Consistency <sup>b</sup> coefficient Pas <sup>n</sup>	Flow behavior index <sup>b</sup>
0.5	80	0.02	1.00
1.0	50	2.88	0.52
	60	2.23	0.55
	70	1.84	0.57
	80	1.03	0.7

			TA	BLE 1.					
POWER LAW	<b>PARAMETERS</b> <sup>a</sup>	FOR	CMC	RHEOLO	GY A	T V	VARIOUS	TEMPER	ATURES
		AND	CON	CENTRA	TION				

<sup>a</sup>From Abdelrahim et al. (1991)

<sup>b</sup>Average for up and down cycles.

$$C_{ps} = 0.5 X_F + 0.3 X_s + 1.0 X_m$$
(19)

The thermal conductivity was back calculated from reported thermal diffusivity (Kostaropoulos et al. 1975) and measured density ( $\rho = m/V$ ): k =  $\alpha \rho C_{ps}$ . Other property values obtained from literature have been summarized in Table 2.

laterial	Temp C	Density kg/m <sup>3</sup>	Specific heat kJ/kgC	Thermal conductivity W/mC	Thermal diffusivity x 10 <sup>-7</sup> m <sup>2</sup> /s
arrot	-	1035	3.738 <sup>a</sup>	0.657b	1.70 <sup>c</sup>
otatod	-	1070	3.270	0.556	1.59
atere	82	970	4.195	0.673	1.65
% CMC	80	1003 <sup>f</sup>	4.15g	0.668 <sup>h</sup>	
.5% CMC	80	1003 <sup>f</sup>	4.16 <sup>g</sup>	0.670 <sup>h</sup>	
.5% CMC	80	1003 <sup>f</sup>	4.16 <sup>g</sup>	0.670 <sup>h</sup>	

TABLE 2. PHYSICAL PROPERTIES OF TEST MATERIALS

a From Charm (1978)

b Calculated from  $k = \alpha \rho C_{ps}$ Kostaropoulos et al. (1975)

С

d Yamada (1970)

e Holman (1976)

f Determined with pycnometer at 20 C.

gh From Heldman and Singh (1981).

### **Experimental Setup and Procedure**

The experimental arrangement (Fig. 1) consisted of a water bath equipped with a heater and a pump, a Dash-8 data acquisition system (MetraByte Corporation, Taunton, MA), a test chamber (8.6 cm diameter and 20 cm long), and a personal computer. The bath and test chamber were insulated to reduce heat losses, and the bath was filled to about three quarters full with test fluid. The average linear velocity of the fluid was obtained from the measured volumetric through flow rate and the cross-sectional area of the chamber. Test fluid was introduced from either the bottom (upward flow) or from the top (downward flow). A perforated plate was used to distribute the medium, which was returned to the water bath and recirculated. Two needle-type copper-constantan thermocouples (Ecklund-Harrison Technologies Inc., Cape Coral, FL) calibrated against an ASTM mercury-in-glass thermometer were used to gather time-temperature data. One was attached to the particle with its tip at the geometric center, while the other was suspended in the fluid. A distance was marked on the thermocouple to co-



FIG. 1. A SCHEMATIC DIAGRAM OF THE TEST APPARATUS AND SETUP.

incide with the geometric center of the test cylinder in the central direction, and the thermocouple was carefully inserted up to the mark. A combination of a special industrial adhesive (Loctite Canada Inc., Mississauga, ON) and epoxy resin was used to hold the test sample to the probe and prevent possible flow of fluid along the thermocouple probe. Temperatures were continuously recorded in a spreadsheet format at 5-s intervals.

#### **Experimental Factors**

Experiments were carried out to test the effect of various parameters on the convective heat transfer coefficient. The influence of CMC concentrations was tested at three levels (0, 0.5, and 1.0% w/w) using 0.0223 m diameter, 0.04 m long cylinders of potatoes and carrots. Effect of sample size (diameters 0.016, 0.019, and 0.023 m with length kept at 0.04 m; sample lengths of 0.02, 0.03, 0.04 m with diameter at 0.023 m) was studied with both potatoes and carrots in 1.0% CMC solution at 80C and a flow rate of 4.2 ml/s. Effect of flow rate (2.3 and 4.2 ml/s) was evaluated at 80C, while the effect of temperature (50–80C) was evaluated at 0, and 2.3 ml/s using 0.04 mm length and 0.023 m diameter samples. The combined effects of flow rate and temperature were also studied at selected levels (1.4 ml/s at 50C, 2.6 ml/s 60C, 3.3 ml/s at 70C and 4.2 ml/s at 80C). All the factors mentioned above were evaluated with the CMC solution flowing upward. The effect of the direction of fluid flow of  $h_{fp}$  was evaluated at 50 and 70C with flow rates of 1.4 and 2.3 ml/s, respectively, for carrot in 1.0% CMC solution. A minimum of five runs was performed for each experimental variation.

#### **Data Analysis**

The logarithm of the temperature ratio  $(T_a - T)/(T_a - T_i)$  was plotted against linear time as shown in Fig. 2, and the heating rate index  $f_h$  was evaluated by linear regression of the straight-line portion of the curve. An iterative computer program (Ramaswamy *et al.* 1983) based on Eq. (15) was used for the purpose of computing the convective heat transfer coefficient ( $h_{fp}$ ). An estimate of  $h_{fp}$ was first made using the lumped capacity method ( $h_{fp} = 2.303 Dk_s/(\alpha f_h)$ ] assuming the Biot number to be less than 0.1 (Kreith 1973). Individual Biot numbers (Bi<sub>c</sub> and Bi<sub>p</sub>) and characteristic functions (S<sub>c</sub> and S<sub>p</sub>) for the two finite dimensions were computed (Ramaswamy *et al.* 1982) from this  $h_{fp}$  value. A heating rate index was calculated using Eq. 15 and compared with the experimental  $f_h$ . The  $h_{fp}$  value was changed in a stepwise fashion until the difference between the experimental and calculated values of  $f_h$  was equal to or less than 0.01% of the experimental value. This method is based on backcalculating the  $h_{fp}$  value from

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FIG. 2. A TYPICAL SEMI-LOGARITHMIC PLOT OF THE HEAT PENETRATION CURVE FOR CARROT

f<sub>h</sub>, which is independent of the thermocouple location and hence is not sensitive to errors in the placement of the thermocouple.

Dimensionless parameters were evaluated for further data analysis. The forced convection heat transfer coefficient for immersed bodies was predicted by the relationship (Heldman and Singh 1981):

$$Nu = h_{fn} D/k_f = A_1 (Re)^b (Pr)^{1/3}$$
(20)

For natural convection, the general relationship used was:

$$Nu = h_{fp} D/k_f = A_2 (Pr.Gr)^c$$
(21)

where  $A_1$ ,  $A_2$ , b and c are system constants with Re, Pr, and Gr representing Reynold, Prandtl, and Grashof numbers. For non-Newtonian fluids, Re, Pr, and Gr are replaced by their generalized forms GRe, GPr, and GGr (Skelland 1967; Zuritz et al. 1990):

$$GRe = [\{D^{n}V^{2-n}\rho\}/\{8^{n-1} m (3n+1/4n)^{n}\}]$$
(22)

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$$GPr = [\{C_{nfm} (3n+1/n)^n 2^{n-3}\} / \{k_f (D/V)^{n-1}\}]$$
(23)

GGr=[g 
$$\beta \rho^2(T_a - T_i) D^3] / [m {(3n+1)/n}^n 2^{n-1} / {4V^{1-n} D^{n-1}}]^2$$
 (24)

Dimensionless parameters were calculated using the length of the cylinder as the characteristic length. This is justified by an analogy between the particle's configuration in the test chamber and fluid flow in vertical pipes. In order to evaluate the effects of sample length and diameter on  $h_{\rm fp}$ , however, the volume to area ratios were adopted.

# **RESULTS AND DISCUSSION**

Tables 3–8 summarize average  $h_{fp}$  values for potato and carrot cylinders. Correlations between dimensionless numbers (Nusselt, Grashof and Prandtl) are plotted in Fig. 3 for potatoes and carrots, respectively. Depending on the experimental variation, the average  $h_{fp}$  ranged between 76 to 456 W/m<sup>2</sup>C and 96 to 556 W/m<sup>2</sup>C for potatoes and carrots, respectively. In general, carrots had higher associated  $h_{fp}$  values than potatoes under similar experimental conditions, possibly due to structural and textural differences. Our results compare well with published values (Alhamdan and Sastry 1990; Deniston *et al.* 1987; Chang and Toledo 1989; Zuritz *et al.* 1990). Chandarana *et al.* (1989) obtained  $h_{fp}$  values lower than found

Food	Concentration	h <sub>fp</sub>	Standard deviation
	%	W/m <sup>2</sup> C	W/m <sup>2</sup> C
Potato	0	456 <sup>a</sup>	35
	0.5	199b	22
	1.0	143 <sup>c</sup>	6
Carrot	0	556d	26
	0.5	233 <sup>e</sup>	26
	1.0	172 <sup>f</sup>	5

 TABLE 3.

 HEAT TRANSFER COEFFICIENTS FOR POTATO AND CARROT CYLINDERS IMMERSED

 IN CMC SOLUTIONS (0-1%)

a,b,c,d,e,f

Mean h<sub>fp</sub> values sharing the same superscript are not significantly different (p>0.05)

Particle length 40mm, diameter 23mm; medium at 80 C and a linear velocity of  $0.7 \times 10^{-3}$  m/s.  $0.7 \times 10^{-3}$  m/s.



FIG. 3. NUSSELT NUMBER AS FUNCTION OF REYLEIGH NUMBER (GGR  $\times$  GPr) FOR CARROT AND POTATO CYLINDERS HEATED IN CMC SOLUTIONS

in this study, while Lamberg and Hallstrom (1986) found higher values. Results for carrot and potatoes in water (0% CMC) also compared favorably with the minimum  $h_{fp}$  values reported by Alhamdan *et al.* (1990). The differences in results as compared to those published were attributed to differences in experimental conditions and methodology adopted for  $h_{fp}$  evaluation.

The concentration of CMC had a significant (p < 0.01) effect on  $h_{fp}$ , with  $h_{fp}$  values decreasing with increasing CMC concentration (Table 3), possibly due to the increased apparent viscosity at higher concentrations (Table 1). An increase in CMC concentration from 0 to 1.0% decreased  $h_{fp}$  values by about 70%, which is in agreement with the findings of Alhamdan and Sastry (1990). Tables 4 and 5 illustrate a small increase in  $h_{fp}$  values with a decrease in the associated volume to surface area ratio of test samples. This suggests possible effects of sample size (length and diameter) on the convective heat transfer coefficient. However, except with 16 mm-diameter, 40 mm-length potatoes (Table 4), the observed changes were not statistically significant (p > 0.05). Notwithstanding the above observation, it was suspected that the influence of sample dimensions on  $h_{fp}$  might have been masked by the relatively high apparent viscosity of the fluid. Zuritz *et al.* (1990) reported the contrary: an increase in  $h_{fp}$  with particle size. They reasoned that as the particle size increased, available cross section for fluid flow decreased.

Food	Diameter (mm)	Volume/area x 10 <sup>-3</sup> m	h <sub>fp</sub> ₩/m <sup>2</sup> C	Standard deviation W/m <sup>2</sup> C
Potato	23	4.5	143 <sup>a</sup>	6
	19	3.8	143 <sup>a</sup>	8
	16	3.3	175 <sup>b</sup>	12
Carrot	23	4.5	172 <sup>b</sup>	5
	19	3.8	173 <sup>b</sup>	15
	16	3.3	182 <sup>b</sup>	5

#### TABLE 4. HEAT TRANSFER COEFFICIENTS FOR POTATO AND CARROT CYLINDERS IN 1.0% CMC AS INFLUENCED BY PARTICLE DIAMETER

a,b Mean h<sub>fp</sub> values sharing the same superscript are not significantly different (p> 0.05)

Particle length 40 mm; CMC at 80 C and a linear velocity of  $0.7 \times 10^{-3}$  m/s

Food	Length	Volume/area	hfp	Standard deviation
	(mm)	x 10 <sup>-3</sup> (m)	(W/m <sup>2</sup> C)	(W/m <sup>2</sup> C)
Potato	40	4.5	143 <sup>a</sup>	6
	30	4.2	156 <sup>a</sup>	13
	20	3.7	157 <sup>a</sup>	9
Carrot	40	4.5	172 <sup>b</sup>	5
	30	4.2	188 <sup>b</sup>	. 13
	20	3.7	195 <sup>b</sup>	28

TABLE 5. HEAT TRANSFER COEFFICIENT FOR POTATO AND CARROT CYLINDERS IN 1% CMC AS INFLUENCED BY LENGTH

a,b Mean h<sub>fp</sub> values sharing the same superscript are not significantly different (p> 0.05).

Particle diameter 23mm; CMC at 80 C and a linear velocity of 0.4 x 10<sup>-3</sup> m/s

This resulted in an apparent increase in fluid velocity and enhanced localized turbulence, both contributing to higher  $h_{fp}$ . In the present study, however, the chamber diameter was 86 mm, corresponding to approximately four times the maximum sample diameter. Furthermore, the flow rates used were very low, minimizing the chances for localized forced convection.

Table 6 shows the effect of temperature on  $h_{fp}$  under stationary fluid conditions. With the exception of potato with  $h_{fp}$  values in the range of 118 and 120

80 80	120 <sup>a</sup> 118a	3
80	118a	4
(0)		0
60	93b	4
50	76 <sup>c</sup>	4
80	145d	4
80	133 <sup>e</sup>	3
60	115 <sup>a</sup>	5
50	96 <sup>b</sup>	8
_	50 80 80 60 50	50     76 <sup>c</sup> 80     145 <sup>d</sup> 80     133 <sup>e</sup> 60     115 <sup>a</sup> 50     96 <sup>b</sup>

		TA	BLE 6.				
HEAT TRANSFER	COEFFICIENT	IN 1.09	6 CMC A	S INFLUENCED	BY TE	MPERATU	JRE
	UNDER	STATIO	NARY C	ONDITIONS			

a,b,c,d,e

Mean  $h_{fp}$  values sharing the same superscript are not significantly different (p> 0.05).

Particle length 40 mm, diameter 23 mm, fluid 1.0% CMC.

W/m<sup>2</sup>C at 70 and 80C, there was a significant (p < 0.01) decrease of h<sub>fp</sub> values with temperature. The test fluid flowing in the downward direction had generally higher h<sub>fp</sub> values than in the upward direction (Table 7); however, the difference between downward and upward flow on h<sub>fp</sub> was not significant (p > 0.05). Table 8 shows the effects of temperature and flow rate on the convective heat transfer coefficient. There was a significant decrease in h<sub>fp</sub> for both carrots and potatoes when the flow rate was lowered from  $0.7 \times 10^{-3}$  m/s. Temperature had a similar decreasing effect on hfp values, except for potatoes between 80 and 70C.

Mixed convection (forced-on-free convection) usually occurs when there is a small amount of forced fluid along with significant temperature difference between the convecting body and surrounding fluid. As the Reynolds number increases, mixed convection gives way to a pure forced convection regime (ASHRAE 1981; Johnson *et al.* 1988). To analyze the effects on convective heat transfer coefficients of fluid velocity and temperature of viscous fluids, the generalized dimensionless parameters defined by Eq. (22), (23), and (24) were evaluated. Furthermore, in order to establish which regime reigned with regard to natural, forced or mixed convection, Nusselt numbers based on Eq. (20) and (21) were evaluated. Data used for the latter were those obtained from simultaneous variation of temperature and flow rate.

Our data did not follow the trend indicated by the generalized relationship relating Nusselt (Nu) to Reynold (Re) and Prandtl (Pr) numbers under forced convection (Eq. 20), probably due to the prevailing low flow rates (Table 8).

Flow rate ml /s	Temperature C	h <sub>fp</sub> W/m <sup>2</sup> C	Standard deviation W/m <sup>2</sup> C	
2.3	70	151 <sup>a</sup>	8	
2.3	70	147 <sup>a</sup>	9	
1.4	50	103 <sup>b</sup>	9	
1.4	50	97b	6	
	Flow rate ml /s 2.3 2.3 1.4 1.4	Flow rate ml /s         Temperature C           2.3         70           2.3         70           1.4         50           1.4         50	Flow rate ml /s         Temperature C         hfp W/m <sup>2</sup> C           2.3         70         151 <sup>a</sup> 2.3         70         147 <sup>a</sup> 1.4         50         103 <sup>b</sup> 1.4         50         97 <sup>b</sup>	Flow rate ml /s         Temperature C $h_{fp}$ W/m <sup>2</sup> C         Standard deviation W/m <sup>2</sup> C           2.3         70         151 <sup>a</sup> 8           2.3         70         147 <sup>a</sup> 9           1.4         50         103 <sup>b</sup> 9           1.4         50         97 <sup>b</sup> 6

		TAB	LE 7.			
HEAT TRANSFER (	COEFFICIENTS	FOR	CARROT	CYLINDERS	AS	INFLUENCED
	BY DIRECT	TON	OF FLUID	) FLOW		

a,b Mean  $h_{fp}$  values sharing the same superscript are not significantly different (p> 0.05).

Particle diameter 23mm, length 40mm in 1.0% CMC solution.

Food	Velocity x 10 <sup>-3</sup> (m/s)	Temperature C	h <sub>fp</sub> W/m <sup>2</sup> C	GRe x10 <sup>-3</sup>	GPr	GGr x10 <sup>4</sup>	Nu
Potato	0.7	80	143 <sup>a</sup>	14.7	1.23	10.96	8.56
	0.4	80	128 <sup>b</sup>	6.80	1.50	6.79	7.67
	0.4	70	122 <sup>b</sup>	2.70	3.79	0.66	7.30
	0.4	60	109 <sup>c</sup>	2.10	4.93	0.31	6.53
	0.4	50	85 <sup>d</sup>	1.49	7.02	0.10	5.10
Carrot	0.7	80	172 <sup>e</sup>	14.7	1.23	11.1	10.3
	0.4	80	155 <sup>f</sup>	6.80	1.50	7.79	9.30
	0.4	70	146 <sup>f</sup>	2.70	3.79	0.57	8.74
	0.4	60	131 <sup>b</sup>	2.10	4.93	0.37	7.84
	0.4	50	103 <sup>c</sup>	1.49	7.02	0.11	6.17

TABLE 8. AVERAGE HEAT TRANSFER COEFFICIENTS AT VARIOUS LINEAR VELOCITIES AND TEMPERATURES, AND CORRESPONDING DIMENSIONLESS PARAMETERS

a,b,c,d,e,f

Mean values for carrots or potatoes sharing the same letter are not significantly different (p> 0.01).

Particle length 40 mm, diameter 23 mm, fluid 1.0% CMC.

(p> 0.01).

This indicated the possibility of existence of natural convection (Eq. 21) as opposed to forced convection. According to Ozisik (1985), forced convection will characterize the flow when the ratio of  $Gr/Re^2$  is < 1.0, while natural convection prevails when the ratio is > 1.0. In between these two values, the flow is considered to be in mixed mode. In the present study, because of the low flow rates employed, the ratio generally exceeded 30,000, indicating a strong possibility that natural convection prevails. According to ASHRAE (1981) data, laminar region of natural convection is characterized by Rayleigh number (Pr.Gr) in the range  $10^4-10^8$  while turbulent flow will have a range  $10^8-10^{12}$ . Our data gave the product GPr.GGr in the  $10^3-10^4$  range, indicating the possibility of laminar flow natural convection. Dimensionless correlations were developed to relate Nu to Rayleigh number (GPr.GGr). The logarithm of Nu was plotted against the logarithm of GGr and GPr combinations as shown in Fig. 3 for potatoes and carrots, respectively. Regression analysis yielded the following equations:

Carrot:	Nu = 2.45 (GGr.GPr) $0.108$	$r^2 = 0.80$	(25)
Potato:	Nu = 2.02 (GGr.GPr) $^{0.113}$	$r^2 = 0.88$	(26)
Limits:	$3 \times 10^3 < GGr.GPr < 6 \times 10^4$		

The regression output gave standard errors for Nu of 0.025 and 0.019 for carrot and potato, respectively. Figure 3 shows good correlations between the experimental and predicted values, and demonstrates increasing Rayleigh (GGr.GPr) number with Nu. This clearly depicts the contribution of temperature to heat transfer at low flow rates.

# CONCLUSIONS

Heat transfer coefficients associated with food samples heated in low velocity viscous fluid flow ranged from 80 to 450 W/m<sup>2</sup>C for potato cylinders and 100 to 550 W/m<sup>2</sup>C for carrot cylinders. Fluid concentration and temperature had large impact on the convective heat transfer coefficient, with higher fluid concentrations and lower temperatures decreasing the  $h_{fp}$ . Sample size (0.016–0.023) m diameter, 0.02–0.04 m length), flow rate (0.2 to 0.7 × 10<sup>-3</sup> m/s) and flow direction (upward and downward) had only small effects on  $h_{fp}$ . Good correlations were obtained between Nusselt and Rayleigh numbers, indicating strong natural convection.

The study indicates the dominance of natural convection while heating food particles in viscous fluids. From a safety viewpoint, it might be conservative to assume natural convection as the mechanism of heat transfer associated with the processing of real foods at low relative flow rates. However, the sensitivity and response of fluid viscosity to higher temperatures may shift the above trends from natural to a mixed mode or forced convective heat transfer.

# ACKNOWLEDGMENTS

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# NOMENCLATURE

- a Significant dimension, such as the radius of a cylinder or halfthickness of slab (m)
- C<sub>pf</sub> Specific heat (J/kgC)
- C<sub>ps</sub> Specific heat of conduction heated material (J/kgC)
- D Characteristic length of sample or volume to area ratio (m)
- f<sub>h</sub> Negative reciprocal slope of the straight line portion of the heating curve on semi-log coordinates (heating rate index)
- h<sub>fp</sub> Convective heat transfer coefficient (W/m<sup>2</sup>C)
- g Acceleration due to gravity, m/s<sup>2</sup>
- J Bessel function of order zero or one
- j Lag factor of heating  $(T_{pi}-T_a)(T_1-T_a)$
- k Thermal conductivity (W/mC)
- L Thickness or half thickness of a slab depending on it being heated or cooled from one side or both (m)
- m Consistency coefficient (Pas<sup>n</sup>) or mass (kg)
- n Flow behavior index
- R A function of Biot number
- r Position along radius of cylinder ( $0 \le r \le a$ ) (m)
- S A function of Biot number
- T Temperature (C)
- U Dimensionless temperature ratio  $((T_a T)/(T_a T_1))$
- V Velocity (m/s) or volume (m<sup>3</sup>)
- W % Water
- X Distance from the coldest plane of a slab ( $0 \le X \le L$ ) (m)
- $X_F$  Mass fraction of fat (%)
- X<sub>s</sub> Mass fraction of solids (%)

# Nomenclature (continued)

Xm	Mass	fraction	of	moisture	(%)
----	------	----------	----	----------	-----

- $\alpha$  Thermal diffusivity (k<sub>s</sub>/ $\rho$ C<sub>ps</sub>)
- $\beta$  Root of the characteristic Eq. (4) or volumetric thermal expression coefficient of fluid (C<sup>-1</sup>)
- $\gamma$  Root of the characteristic Eq. (2)
- $\dot{\gamma}$  Shear rate (s<sup>-1</sup>)
- $\rho$  Density (kg/m<sup>3</sup>)
- $\sigma$  Shear stress (Pa)

# Subscripts

- a Ambient
- c Infinite cylinder
- f Fluid
- fc Finite cylinder
- i Initial
- n An integer
- o Center of cylinder or coldest plane of slab
- p Infinite plate or slab
- pi Pseudo-initial
- s Solid
- x Distance from the coldest point of slab
- 1 First root of characteristic function

# **Dimensionless Parameters**

- Bi Biot number  $(h_{fp}.D/k_s)$
- Fo Fourier number  $(\alpha t/a^2)$
- GGr Generalized Grashof number
- Nu Nusselt number  $(h_{fp}.D/k_f)$
- GPr Generalized Prandtl number
- GRe Generalized Reynold number

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# FUZZY LOGIC CONTROL FOR A CONTINUOUS CROSSFLOW GRAIN DRYER

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## ABSTRACT

This paper presents the assembly and the performance of a fuzzy logic control system in regulating a laboratory continuous crossflow grain dryer. With fuzzy control, similar outlet moisture contents and lower outlet breakage susceptibility of corn were obtained as compared to the manually controlled dryer operation. The results verified that drying-induced corn breakage could be reduced through the application of fuzzy logic control. Like the corn drying process, many food processes are difficult to control automatically with conventional control techniques due to their complex nature. Fuzzy logic control provides an attractive solution for control of those processes.

# **INTRODUCTION**

Fuzzy logic control uses human operators' experience as well as measured process variables in control decision making. The development of such a controller must consider the specific process configuration. In this study, we assembled a fuzzy logic control system for a laboratory grain dryer. The measured process variables were the drying-air temperatures, the ambient air temperature, the grain mass temperatures and moisture contents at different locations in the dryer, and the rotational speed of the dryer discharge rotor. The control variables were the drying-air temperature and the rotational speed of the discharge rotor.

The primary purposes of this paper are to explain the assembly of a fuzzy logic control system for a grain dryer and to present the results of testing the control

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system with a laboratory continuous crossflow grain dryer. Specific objectives were: (1) design fuzzy algorithms for different process states. (2) develop the software system for the controller, and (3) evaluate controller performance with a laboratory dryer.

# FUZZY LOGIC CONTROL SYSTEM

The fuzzy logic control system consists of four principal components: a process identifier, a knowledge base, a fuzzy computation unit, and an input defuzzifier (Fig. 1). The process identifier measures process variables (P) and disturbances (D), converts the measured variables into a fuzzy set (F), which contains the fuzzified state variables and the predicted process outputs. This fuzzy set (F) is then compared to the set range set (R) and presents a fuzzy state set (S), which contains both state variables and errors between predicted outputs and set ranges.

The knowledge base consists of several sub-bases. Each sub-base contains a governing fuzzy control rule and several supporting knowledge matrices for a specific process state. The fuzzy computational unit searches feasible control actions by mapping the fuzzy state set (S) into a fuzzy action set (A) according to the fuzzy algorithm for a specific process state and the setting level. The input defuzzifier maps the fuzzy action set (A) onto a crisp action set (U), and provides one definite control command for implementation.



FIG. 1. SCHEMATIC OF FUZZY LOGIC CONTROL SYSTEM

# **DESCRIPTION OF GRAIN DRYING PROCESSES**

The study was conducted with a laboratory-scale continuous crossflow dryer (Fig. 2). As shown in the schematic, the dryer consisted of a drying zone and a cooling zone. The drying zone was divided into two sections. The width of the dryer column was 26.7 cm. Wet corn was fed into the dryer from the top, flowed down through the perforated column, and was discharged at the bottom.



FIG. 2. LABORATORY SCALE CONTINUOUS CROSSFLOW GRAIN DRYER

The resident time of corn in the dryer was regulated by the variable speed discharge rotor.

The dryer used heated-air as the drying medium. Three electric heaters of 1500, 3400 and 3750 watts were installed in the air-chamber. Three switches were used to operate these heaters separately, so that different drying-air temperatures were achieved by either using (a) 1500 watt, (b) 3400 watt, (c) both 1500 and 3400 watt, (d) both 3400 and 3750 watt heaters, or (e) all three heaters. An electric blower (Model 2C820) drew the ambient air through the hot corn in the cooling zone and blew the heated-air through the wet corn in the drying zone. The airflows were perpendicular to the corn flow.

The drying-air velocity was > 1.5 m/s on the air exhaust-side of the drying zone. The drying-air temperature range was from 45 to 105C, and the rotor speed range was between 1.0 and 7.0 rpm. The corn volume discharged from the drying was  $3.0 \times 10^{-4}$  m<sup>3</sup>/rev. The drying capacity ranged from 13.0 to 90.0 kg/h in terms of the initial moisture content of the wet corn.

Three conductivity-type sensor assemblies (Shivvers Inc., Corydon, IA) measured both moisture and temperature of the corn mass with accuracies of  $\pm 1\%$  for moisture and  $\pm 1C$  for temperature. The sensors were located above the drying zone (Fig. 2, MS1), at the middle of the drying zone (MS2), and near the end of the cooling zone (MS3).

Five copper-constantan (type T) thermocouples, with accuracy of  $\pm 1$ C, were installed inside the heated-air plenums (Fig. 2, TC1 and TC2), inside the grain column (TC3 and TC4), and outside the dryer (TC5). The drying-air temperature was measured inside the heated-air plenum by TC1 and TC2. Corn mass temperatures were measured at the air exhaust-side, near the end of both the drying zone and the cooling zone by TC3 and TC4. The ambient-air temperature was measured by thermocouple TC5 at the air inlet-side of the grain column near the end of the cooling zone.

#### **DESIGN OF FUZZY ALGORITHMS**

The algorithms used in the fuzzy logic controller were classified as fuzzy algorithms (Zadeh 1973). A fuzzy algorithm provides an approximate description of control strategy and is represented in the form of an *if-then* rule (Zhang and Litchfield 1992b):

IF	the predicted process output at time $n = \text{linguistic level } l_{y,n}$ ;		
AND	the process state at time $n = linguistic level l_{s,n}$ ;		
AND	the disturbance of the process at time $n = linguistic level l_{d,n}$ ;		
AND	the dryer operation setting at time $n = linguistic level l_{u,n}$ ;		
THEN	the dryer operation setting at time $n+1 = linguistic level l_{u,n+1}$ .		

or more formally:

IF 
$$L(Y(n)) = l_{y,n}$$
 AND  $L(S(n)) = l_{s,n}$  AND  $L(D(n)) = l_{d,n}$  AND  $L(U(n)) = l_{u,n}$   
THEN  $L(U(n+1)) = l_{u,n+1}$ 

where L(Y(n)), L(S(n)), L(D(n)), and L(U(n)) are the linguistic variables of current process outputs process states, disturbances, and dryer operation settings. L(U(n+1)) is the linguistic variable of the new (or the selected) operation settings.

The likelihoods of the new operation settings were determined by fuzzy memberships of process conditions and current control inputs. Fuzzy memberships of a process condition were derived from historic data of dryer operation and experience of skillful human operators. Fuzzy memberships of control inputs were derived from the experience of the dryer operation (Zhang and Litchfield 1991b).

Control objectives of this fuzzy logic controller were to (1) dry corn to a desired final moisture level, and (2) decrease high levels of drying-induced breakage susceptibility. Based on predicted levels of process outputs and dryer operation conditions, the process was divided into states of acceptable, defect (including three defect sub-states), and disturbed (Zhang and Litchfield 1992b). Five fuzzy algorithms (Table 1) were designed for these process states.

For instance, in the large moisture defect state, the fuzzy algorithm would find an approximate range of dryer operation settings suitable to the state based on the current operation settings.

Process state	Applied premises	Operation setting	Fuzzy algorithm
acceptable	$L(Y(n)) = l_{y,n}$	$L(U(n+1)) = l_{u,n}$	$min\{\mu(l_{y,n})\}$
large MC defect	$L(Y(n)) = l_{y,n}$ $L(S(n)) = l_{s,n}$ $L(U(n)) = l_{u,n}$	$L(U(n+1)) = l_{u,n+1}$	$max\{\mu(l_{y,n}) \land \mu(l_{s,n} \land \mu(l_{u,n} \rightarrow l_{u,n+1})\}$
small MC defect	$L(Y(n)) = l_{y,n}$ $L(U(n)) = l_{u,n}$	$L(U(n+1)) = l_{\Delta u,n+1}$	$max\{\mu(l_{y,n}) \wedge \mu(l_{u,n}+l_{\Delta u,n+1})\}$
high BS defect	$L(Y(n)) = l_{y,n}$ $L(U(n)) = l_{u,n}$	$L(U(n+1)) = l_{u,n+1}$	$max\{\mu(l_{y,n}) \land \mu(l_{u,n} \rightarrow l_{u,n+1})\}$
disturbed	$L(S(n)) = l_{s,n}$ $L(D(n)) = l_{d,n}$ $L(U(n)) = l_{u,n}$	$L(U(n+1)) = l_{u,n+1}$	$max\{\mu(l_{u,n}) \land \mu(l_{d,n}) \land \mu(l_{u,n} \rightarrow l_{u,n+1})\}$

TABLE 1. FUZZY ALGORITHMS FOR DIFFERENT PROCESS STATES

IF 
$$L(Y(n)) = l_{y,n}$$
 AND  $L(S(n)) = l_{s,n}$  AND  $L(U(n)) = l_{u,n}$   
THEN  $L(U(n+1)) = l_{u,n+1}$ 

Mathematically, the fuzzy algorithm can be represented as

$$l_{u,n+1}: \quad \mu(l_{u,n+1}) = \left\{ \max_{i=1}^{k} (\mu(l_{u,n+1,i})) \right\}$$
(1)

and

$$\mu(l_{u,n+1,i}) = \mu(l_{s,n}) \wedge \mu(l_{heater,n}) \wedge \mu(l_{rotor,n})$$
(2)

where  $l_{u,n+1}$  is the selected operation setting (combination of the heater setting and the rotor speed) for time n+1,  $\mu(l_{u,n+1})$  is the likelihood of this selection,  $\mu(l_{u,n+1,i})$  is the likelihood of feasible operation setting i for time n+1,  $\mu(l_{s,n})$ is the fuzzy membership based on the process condition at time n,  $\mu(l_{heater,n})$  and  $\mu(l_{rotor,n})$  are the fuzzy memberships based on heater and rotor operating settings at time n, respectively, and the symbol  $\wedge$  represents the fuzzy operation of determining the minimum value.

### **DEVELOPMENT OF SOFTWARE SYSTEM**

Figure 3 is the flow chart for this control system, which consists of five major segments: a data acquisition segment, a fuzzy mapping segment, a fuzzy prediction segment, a process identification and knowledge inference segment, and a defuzzification segment.

The data acquisition segment first acquired state variables through three I/O interfaces: one for rotor speed reading, one for moisture readings, and one for temperature readings. To reduce the measurement error, the speed and moisture readings were averaged over five measurements with 1.5 s intervals, and the temperature readings were averaged over 50 measurements with 0.5 s intervals. The current heater switch settings were input from the keyboard.

To perform the fuzzy reasoning, the measured state variables needed to be mapped into linguistic variables. The fuzzy mapping segment converted all realvalue variables into corresponding linguistic variables. A standard triangle fuzzy membership function (Kaufmann and Gupta 1988) was used to perform these mappings.


FIG. 3. FLOWCHART FOR THE FUZZY LOGIC CONTROLLER

The fuzzy prediction segment estimated the final moisture and breakage levels through fuzzy max-min composition operations between the weighing factors and the fuzzy memberships of the state variables (Zhang and Litchfield 1991b).

The next segment was the knowledge base and the inference engine, which consisted of a knowledge base to handle dryer operation experience, a process identifier to classify process states, and an inference engine to perform fuzzy reasoning. This segment first classified the process into one of five states in terms of linguistic state variables and estimated process outputs. Workable solutions were chosen based on the process state and current dryer control inputs. Approximate reasonings were based on the corresponding fuzzy algorithm for the identified process state.

The last segment was for defuzzification. An  $\alpha$ -cut method (Dubois and Prade 1980) was used to defuzzify the selected dryer operation settings. This defuzzifier picked the highest fuzzy membership as the  $\alpha$  value.

#### **EXPERIMENTAL INVESTIGATION**

Figure 4 is the schematic diagram of hardware used with the fuzzy control system. The system consisted of (1) a 286 personal computer, (2) sensors (moisture sensors from Shivver Inc., type T thermocouple, and Model PU2E speed pickup



FIG. 4. SCHEMATIC DIAGRAM OF THE FUZZY LOGIC CONTROLLER HARDWARE FOR A LABORATORY SCALE CONTINUOUS CROSSFLOW GRAIN DRYER

sensor) and A/D converters (PIO, DAS8, and LABMASTER I/O boards), and (3) dryer control devices (electric heaters and discharge rotor). The control inputs were determined by the controller based on current state variables and dryer operation settings. The measured state variables, predicted process outputs, dryer operation settings, and selected new settings were listed on the screen. The dryer control devices were adjusted manually according to instructions shown on the screen.

Two experimental investigations were conducted to test the fuzzy logic controller. The first investigation used three varieties (FR27 × FRM017, FR618 × GH123, and FR1141 × LH123) of frozen corn harvested in September 1990 and stored in a freezer at -40C until July 1992. The frozen corn was removed from the freezer 20 h prior to the experiment to raise corn mass temperature close to indoor-air temperature (25C). Some moldy corn was observed in higher moisture samples. The second investigation used three varieties (FR27 × FRM017, FR618 × FR600, and FR1087 × LH123) of newly harvested corn (fresh corn) at various moisture contents and dried in September 1991.

During the experiments, the control system acquired state variables, predicted process outputs, and made control decisions at 5-min intervals. 400 g corn samples were collected under the discharge rotor after each control cycle and from the wet corn container before each loading of the wet corn. All the samples were measured for moisture content and breakage susceptibility according to the procedures introduced by Paulsen and Hill (1985).

# **RESULTS AND DISCUSSION**

# Moisture and Breakage Predictions for Frozen Corn

Figures 5 and 6 show predicted levels and measured values of corn moisture content and breakage susceptibility of the investigation of using frozen corn. Of 133 predictions in the 11-h investigation, 82 moisture predictions and 84 breakage predictions matched measured values. The rates of match were 61.7% and 63.2% for moisture and breakage, respectively. Most of the other predictions were very close to the corresponding prediction levels. For example, 102 of the moisture predictions (76.7\%) were either within target ranges or 0.5\% above and below the target ranges. Similarly, 95 of the breakage predictions (71.4\%) were either within target ranges or 1.0\% above and below the target ranges or 2.5\% above and below.



FIG. 5. PREDICTED LEVELS AND MEASURED VALUES OF MOISTURE CONTENT FROM THE INVESTIGATION USING FROZEN CORN



FIG. 6. PREDICTED LEVELS AND MEASURED VALUES OF BREAKAGE SUSCEPTIBILITY FROM THE INVESTIGATION USING FROZEN CORN

#### Moisture and Breakage Predictions for Fresh Corn

For the experiment using fresh corn, Fig. 7 and 8 show predicted levels and measured values of corn moisture content and breakage susceptibility. Of 133 predictions, 98 moisture predictions and 102 breakage predictions matched measured values. The rates of match were 73.7% for moisture and 76.7% for breakage. Including those close predictions, there were 118 (88.7%) within the target range or 0.5% above and below the target, and 132 (99.2%) within the target range or 1.0% above and below for moisture. Of the breakage predictions, there were 112 (84.2%) within the target range or 1.0% above and below the target range or 2.5% above and below.

Note that two of the varieties used in the evaluation test were different from those used in the data acquisition test. However, there was one variety each of soft, medium, and hard endosperm corn used in both tests. So fuzzy memberships derived from some representative varieties could be used to predict process outputs of other varieties of corn.

The breakage prediction accuracy was lower for drying frozen corn (63.2%) compared with that for drying fresh corn (76.7%). The moldy corn was the likely cause of this decrease in accuracy, since the fuzzy memberships were derived



FIG. 7. PREDICTED LEVELS AND MEASURED VALUES OF MOISTURE CONTENT FROM THE INVESTIGATION USING NEWLY HARVESTED CORN



FIG. 8. PREDICTED LEVELS AND MEASURED VALUES OF CORN BREAKAGE SUSCEPTIBILITY FROM THE INVESTIGATION USING NEWLY HARVESTED CORN

from experimental data for fresh corn drying. Frozen storage and molding of corn caused a change in drying characteristics.

# **Fuzzy Logic Control Performance for Frozen Corn**

Control objectives were to obtain (1) outlet moisture content between 15.0 and 16.0% and (2) outlet breakage susceptibility as low as possible. Inlet moisture contents ranged from 19.4 to 34.0% for the frozen corn. The dryer was operated at drying-air temperature settings from 75 to 105C and discharge rotor speed settings from 1.3 to 6.7 rpm.

The results for frozen corn drying are shown in Fig. 9. The outlet moisture content fluctuated dramatically. Heavy molding was observed from the inlet corn with moisture content over 25%. Furthermore, about one-third of the inlet corn had a moisture content over 28.0%, which surpassed the appropriate response range of the fuzzy logic controller. Both the high moisture and the mold contributed to poor controller performance.

The average outlet moisture content for the frozen corn experiment was 16.6% with a standard deviation of 2.39% for the entire testing period. The distribution



FIG. 9. RESULTS OBTAINED FROM THE FUZZY LOGIC CONTROL INVESTIGATION USING FROZEN CORN

of samples at various levels of outlet moisture were: 55 within the objective level (15.0-16.0%), 20 within the low level (13.0-15.0%), four within the very-low level (<13.0%), 22 within the medium-high level (16.0-18.0%), 16 within the high level (18.0-20.0%), and 16 within the very-high level (>20.0%). So 41.4% of the samples were within the objective level, and 31.6% of the samples were within levels adjacent to the objective level. Eliminating samples taken from the start-up period (the first 15 samples), the success rate was raised to 46.6%, the adjacent rate raised to 36.4%, and the average outlet moisture content was 16.0% with a standard deviation of 1.71%.

The average outlet breakage susceptibility for the frozen corn experiment was 16.4% with a standard deviation of 7.84%. The distribution of samples at various levels of outlet moisture content were: 118 samples within levels of medium-low or below (<25.0%), 15 in levels medium or above (>25.0%). So 88.7% of the samples were within the objective levels. Eliminating the samples taken from the start-up periods, the amended success rate of breakage control was 87.3%, and the amended average outlet breakage susceptibility was 18.2% with a standard deviation of 6.39%.

#### **Fuzzy Logic Control Performance for Fresh Corn**

In the fresh corn test, the inlet moisture content ranged from 18.8 to 27.3%, the drying temperature ranged from 45 to 105C, and the discharge rotor speed ranged from 1.5 to 5.5 rpm.

Figure 10 shows the results from the experiment using fresh corn. The outlet moisture content was more stable than that from the experiment using frozen corn. The average outlet moisture content was 15.6% with a standard deviation of 0.88%. The distribution of samples at various levels of outlet moisture content were: 92 within the objective level (15.0-16.0%), 14 within the low level (13.0-15.0%), zero within the very-low level (< 13.0%), 24 within the medium-high level (16.0-18.0%), one within the high level (18.0-20.0%), and one within the very-high level (> 20.0%). So 69.2% of the samples were within the objective level, and 28.6% were in the levels adjacent to the objective level. Eliminating the influence of the start-up period, the amended success rate was raised to 71.9% and the amended adjacent rate was 28.1%, all the samples were within the objective or its adjacent levels. The amended average outlet moisture content was 15.5% with a standard deviation of 0.53%.

The average breakage susceptibility was 17.7% with a standard deviation of 7.69%. The distribution of samples in various levels of breakage were: 102 samples in levels of medium-low or below (<25.0%), and 31 in levels of medium or above (>25.0%). So 76.7% of the samples were in the objective breakage levels. Eliminating the influence of the start-up period, the amended rate of suc-



FIG. 10. RESULTS OBTAINED FROM THE FUZZY LOGIC CONTROL INVESTIGATION USING NEWLY HARVESTED CORN

cess was 73.7%, and the amended average breakage susceptibility was 18.0% with a standard deviation of 7.56%. A short (about 10 min) power supply failure to the electrical heater occurred at the 4 h time point in the test, but the power failure did not significantly affect the test results.

# **Comparisons to Manual Control**

Figure 11 shows the results for fresh corn drying with manual control of the laboratory dryer (the start-up periods were eliminated). The objective level of the outlet moisture content was the same as those of the fuzzy logic control tests. The inlet moisture contents ranged from 18.2 to 21.7%. The control of the outlet moisture level was achieved by changing the drying-air temperature setting from 80 to 105C and the discharge rotor speed from 2.4 to 5.6 rpm.

The average outlet moisture content was 15.5% with a standard deviation of 0.53%. The average outlet breakage susceptibility was 34.7% with a standard deviation of 16.96%. Control actions were based mainly on large changes in the inlet moisture content and deviations in the outlet moisture content. The manual control kept the outlet moisture content within or close to the objective level.

High breakage levels occurred after 6.5 h, while the outlet moisture was mostly within the objective level (Fig. 11). It is noteworthy that the drying conditions were similar between the periods before 2.5 h and after 6 h. The major reason for the higher breakage after 6.5 h might be due to a change in corn variety, since a hard endosperm corn (FR1141× H123) was dried during this period. Therefore, with manual control it was difficult to prevent large variations of breakage based only in the inlet and/or the outlet moisture content.

Comparing the results from the manual control to those from fuzzy control, fuzzy control demonstrated a great advantage in reducing drying-induced breakage. The average outlet breakage was 34.7% with the peak value close to 60% with the manual control, and the comparable values were 18.2% and 18.0% with peak values less than 35% with fuzzy control for frozen and fresh corn tests. The outlet moisture was very similar for the tests with fresh corn. Compared with manual control, fuzzy logic control was comparable in moisture control and superior in breakage control because it directly considers breakage.

#### **Comparisons to Conventional Automatic Controls**

Conventional automatic control of continuous crossflow dryers is usually designed to minimize the overdrying/underdrying of the grain and to optimize the dryer capacity. Some conventional control systems have been successfully developed for achieving the above objectives. The outlet moisture content can be controlled close to the set range with standard deviations from 0.5 to 1.0%



FIG. 11. RESULTS OBTAINED FROM A MANUAL CONTROL EXPERIMENT USING NEW-LY HARVESTED CORN

(Moreira and Bakker-Arkema 1990). This study demonstrated that fuzzy logic control could achieve comparable performance (standard deviation of 0.8% for fresh corn drying).

It is difficult for conventional control systems to include another quality objective, such as breakage susceptibility, because we lack appropriate models for quality estimation. This study demonstrated that fuzzy logic control is superior to conventional controls because it can include control objectives for factors that cannot be modeled.

The results of this study show that fuzzy logic control has potential for improving dryer control, and side-by-side comparisons of fuzzy logic and PID controllers are justified in order to quantify performance differences.

# CONCLUSIONS

A fuzzy logic controller was tested in a laboratory grain dryer. Breakage susceptibility was successfully maintained at a low level while the discharged moisture content was maintained at its desired level. The results verified the hypothesis that drying-induced breakage susceptibility could be reduced by fuzzy logic control.

The use of a computationally intensive algorithm like fuzzy logic for control may be justified by the capability of the technique to handle multiple control of outlet moisture content which may be less precise with fuzzy logic control than with some other control techniques like adaptive control, proportional-integral control, optimal control, and even manual control. Yet this application of fuzzy control is still quite new, and improvements can be made in the fuzzy control of moisture content. Also, fuzzy control has demonstrated a capability to achieve control of objectives with uncertain influences, such as the control of breakage susceptibility. Therefore, it is expected that fuzzy logic control will be important in the control of many complex grain and food processes.

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