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Mass Transfer Kinetic Study of Honey Based Apple Preserve through Osmotic Dehydration

R. Kamboj[1,*], M.B. Bera[2] and V. Nanda[2]

[1]Department of Food Technology, Ch. Devi Lal State Institute of Engineering and Technology, Panniwala Mota-125 077, India
[2]Department of Food Engineering and Technology, Sant Longowal Institute of Engineering and Technology, Longowal-148 106, India

*Corresponding author: E-mail: rajni_kamboj@rediffmail.com

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The present work was focused on study of mass transfer kinetics through osmotic dehydration of apple for solid gain and water loss which was performed for different process times ranging from 10 to 540 min, having three concentrations of honey to sucrose solution (v/w) (100:5, 100:10 and 100:15) at 45, 55 and 65 °C having solution to fruit ratio 3:1. Three empirical kinetic models *i.e.* Peleg, Power law and Azuara were used for analysis of experimental data. Among these, the Power law model was best fitted. Coefficient of correlation (R^2), mean relative deviation modulus (E) and root mean square error (RMSE) were used to determine the best suitable model. The present work depicted that the Power Law model suitably described the osmotic dehydration kinetics with the highest R^2 (> 0.982), the lowest E (< 6.028 %) and RMSE (< 2.398).

Keywords: Apple, Honey, Osmotic dehydration, Water loss, Solid gain.

INTRODUCTION

Preserves are prepared from various fruits and vegetable like amla, ber, apple, bael and carrot. Apple preserve is attributed as highly beneficial for heart patients as it rejuvenate heart and increases muscular strength. In ayurvedic tradition it is considered as heart tonic and digestive stimulant. Preserves are prepared from matured fruit cooked in concentrated sugar solution either whole or in pieces till it become soft and transparent [1]. Generally white sugar is used as an osmotic agent for the preparation of preserve which contain approximately 99.7 % sucrose. Extreme intake of sucrose may cause various health problems like coronary thrombosis and heart ailment [2]. Considering the drawbacks of consumption of excessive sugar, prospects of replacing white sugar with natural sweeteners have been investigated. Nowadays, honey is preferred in processing of fruits as substitute of sugar completely or partially. Durrani and Verma [3] developed honey based amla murabba. Honey is a natural potential energy food consisting sugars *i.e.* fructose and glucose (60-85 %) as the predominant monosaccharide while maltose and sucrose (7-10 %) as disaccharides. Honey is also having enzymes (invertase, diastase, catalase and glucose oxidase), amino acids, proteins, vitamins, phenolic acids, flavonoids, minerals (potassium, calcium, sodium, phosphorous, magnesium and iron) and various phytochemicals [4-9]. It is an excellent source of numerous bioactive phenolic compounds [10-12]. It is a strong inhibitor of the causing agents of peptic ulcers and gastritis [13,14].

Apple (*Malus pumila*) is the main temperate fruit that contains major food constituents like 13.4 % carbohydrates, 84.6 % water, 0.3 % minerals, 1 % fibre (soluble and insoluble fiber), energy upto 59 Kcal per 100 g, iron (0.66 mg per 100 g), calcium (10 mg per 100 g) and phosphorus (14 mg per 100 g) [15]. Phyto-nutrients present in apple play vital role in optimal health as these reduce the risk of cardiovascular disease, colon cancer, prostate cancer, asthma and lung cancer [16]. An enhanced nutritional synergy value is also expected by blending apple as a honey based preserve.

In order to increase shelf life of the fruit as well as its availability throughout the year, preservation techniques like drying and osmotic dehydration may successfully be employed. Water removal by high temperatures and long dehydration may cause decline in the nutritive value and other properties [17]. Osmotic dehydration is a preservation technique which involves partial removing water without change of phase and is used to produce high quality products [18-20]. In osmotic dehydration, salt is used as osmotic agents for vegetables whereas sugar for fruits [21]. The major outcome of this phenomenon is removal of water from the fruit due to concentration gradient. Optimum ratio of fruit to solution is important to consider water loss and solid gain in osmosis [22]. Fruit to solution ratio of 1:2 or 1:3 is most favourable for practical point of view [23]. Rate of mass transfer depends upon various factors such as size and geometry of the sample, temperature and concentration of solution and fruit to solution ratio [24,25]. Water loss and solid gain increased with increasing the solution temperature [26-28].

The objectives of this study is to investigate mass transfer kinetic of honey based apple preserve through osmotic dehydration and to examine the analytical capacity of Peleg, Power law and Azuara equations for the experimental data along with preparation of preserve having high nutritive value.

EXPERIMENTAL

Experiments were conducted to study mass transfer kinetic of honey based apple preserve through osmotic dehydration. Apples used for experiment were obtained from the market of Sirsa, Haryana. Apples were thoroughly washed with water to remove dirt and dust, peeled and cut into pieces of (10 mm × 10 mm × 10 mm). Blanching of samples was done in hot water to prevent browning. Osmosis of the sample was done and initial moisture content was determined.

Osmotic dehydration: Sucrose and honey were used as the osmotic agents. Already prepared three concentrations of honey to sucrose solution (v/w) (100:5, 100:10 and 100:15) were used at temperature of 45, 55 and 65 °C with solution to fruit ratio (STFR) 3:1 as per preliminary experiments and literature. Subsequently, apple cubes were submerged in the osmotic solution in beakers and placed in a water bath. After, specific intervals of 10, 20, 30, 45, 60, 90, 120, 180, 240, 300, 360, 420, 480 and 540 min samples were taken out from the osmotic solution, then excess solution at the surface was blotted with absorbent paper and weighed.

The initial and final moisture content (after osmosis) of samples were determined by using hot air oven method recommended by Ranganna [29] for fruits and vegetables:

$$\text{Moisture content (\%)} = \frac{(M_1 - M_2)}{M_2} \times 100 \quad (1)$$

where M_1 = Weight of sample (g), M_2 = Weight of dried sample (g).

Water loss (g/100 g fresh sample):

$$WL_t = \frac{(M_0 - M)}{W} \times 100 \quad (2)$$

where M_0 = Weight of initial moisture (g), M = Weight of final moisture (g) and W = Initial weight of sample (g).

Solid gain (g/100 g fresh sample):

$$SG_t = \frac{(S_t - S_0)}{W} \times 100 \quad (3)$$

where S_0 = Weight of initial solid (g), S_t = Weight of final solid (g) and W = Initial weight of sample (g).

Kinetic models for osmotic dehydration: To setup a relation among water loss (WL) and solute gain (SG) with immersion time during osmotic dehydration, the mass transfer kinetics were get modeled as per Peleg's model [30,31], Power's law model [32] and Azuara's model [33-35]. These models are specified below.

Peleg's model: Peleg model was applied to study the mass transfer kinetics through the equation:

$$X_W = X_{W_0} \pm \frac{t}{k_1 + k_2 t} \quad (4)$$

where 'X_W' represents the moisture content at time 't' and 'X_{W_0}' is used for initial moisture content both on dry basis; k_1 (h g/g) is given for Peleg rate constant and k_2 (g/g) for Peleg capacity constant. In this '±' becomes '+' if the process is absorption and '–' if the process is drying.

However to represent water loss or solid gain in Peleg model, the equation used is:

$$Y = \frac{t}{K_1 + K_2 t} \quad (5)$$

where K_1 is Peleg rate constants and K_2 is Peleg capacity constants for water loss and solid gain.

Power law model: The equation used for this model is:

$$\text{WL or SG} = k \times t^n \quad (6)$$

where 'k' and 'n' are the power law parameters at time 't'.

Azuara's model: In this model, the equation used for water loss (WL) is:

$$WL_t = \frac{\beta_1 t \times WL_\infty}{1 + \beta_1 t} \quad (7)$$

where 'WL_t' shows the water loss at time 't' in osmotic dehydration, 'WL_∞' shows the corresponding value at infinite time and β_1 (min^{-1}) is the constant related to the diffusion rate of water out of the apple.

The eqn. no. 7 is re-arranged as:

$$\frac{t}{WL_t} = \frac{t}{WL_\infty} + \frac{1}{\beta_1 WL_\infty} \quad (8)$$

The water loss at equilibrium (WL_∞) and the constant β_1 were estimated respectively from the slope and intercept of the plot (t/WL_t) *vs.* t using the eqn. 8.

In this model, the equation for solid gain is:

$$SG_t = \frac{\beta_2 t \times SG_\infty}{1 + \beta_2 t} \quad (9)$$

which is rearranged as:

$$\frac{t}{SG_t} = \frac{t}{SG_\infty} + \frac{1}{\beta_2 SG_\infty} \quad (10)$$

where SG_t is the solid gain at time t, SG_∞ is the corresponding value at infinite time (*i.e.* at equilibrium) and β_2 is the constant related to the incoming solute diffusion rate in the apple. Similarly to WL_∞ and β_1, SG_∞ and β_2 parameters are obtained from the straight line (t/SG_t) *vs.* t using eqn. 10.

Adequacy of fit for empirical models: The non-linear regression and statistical analysis were performed by using Statistica version 10.0 software (StatSoft, Inc., USA). The best predictive capacity of models was evaluated from the coefficient of correlation (R^2), percent mean relative deviation of modulus (E) and root mean square error (RMSE). These parameters can be calculated as given equation:

$$RMSE = \sqrt{\frac{\sum_{t=1}^{N} (\text{Experimental Value} - \text{Predicted Value})^2}{N}} \quad (11)$$

$$E\ (\%) = \frac{100}{N} \sum_{t=1}^{N} \left| \frac{\text{Experimental Value} - \text{Predicted Value}}{\text{Experimental Value}} \right| \quad (12)$$

A model with E value below 10 % is considered acceptable [36]. Therefore, the best model should follow the highest R^2, least RMSE and E values as criteria.

RESULTS AND DISCUSSION

Water loss and solid gain: Through this work, it is found that the water loss and solid gain of apple pieces increased with increase in immersion time. Fig. 1 shows that water loss and solid gain is higher at initial level. The similar results have been found for different osmotically dehydrated foods reported by various researchers [34,35,37-40]. These results can also be predicted that the huge osmotic driving force in between apple cubes and the surrounding medium is the basic reason for the speedy water loss during starting phase and it also indicates the closeness of the system to the end of the osmotic process (pseudo-equilibrium) [18]. Osmotic treatment at 45 °C and having honey to sucrose ratio (100:5), the solid gain and water loss were obtained as 11.52 (g/100 g of initial mass) and 43.83 (g/100 g of initial mass), respectively.

It was also found that because of greater osmotic pressure gradients, the amount of water loss and solid gain are higher with the increase in concentration of the solutions. The various researchers are also of the same opinion [41,42]. On average about 11.64, 13.19 and 14.25 (g/100 g of initial mass) of solid gain and 43.61, 45.21 and 46.43 (g/100 g of initial mass) of water loss was observed at 55 °C and honey to sucrose ratio (100:5, 100:10 and 100:15, respectively).

The mass transfer is accelerated due to high temperatures of osmotic media as depicted in the Fig. 1. Because of this acceleration of mass transfer due to high temperatures, the diffusion rate is also improved owing to swelling and plasticizing of cell membrane [43,44]. Present study revealed (Fig. 1), that during an increase in temperature, the water loss is enhanced but solid gain does not change significantly. These results are in agreement with various workers [45,46]. On an average about 14.38, 14.25 and 14.07 (g/100 g of initial mass) of solid gain and 47.48, 46.43 and 48.42 (g/100 g of initial mass) of water loss was observed in treatments at 45, 55 and 65 °C, respectively of honey to sucrose ratio (100:15).

Modelling of mass transfer kinetics: Peleg, Power law and Azuara's equations were used to evaluate osmotic dehydration kinetics of apple cubes. Tables 1 and 2 show the values of model parameters obtained from the non-linear regression analysis which are en suite to experimental data, along with the coefficient of correlation R^2, percent mean relative deviation of modulus (E) and root mean square error (RMSE) for Peleg, Power law and Azuara model. Peleg rate constants 'K_1' (0.435-0.594 and 1.858-3.152 for water loss and solid gain, respectively) decreased with rising in the temperature and honey to sucrose ratio. However, 'K_1' also decreased at 45 and 65 °C while it increased at 55 °C for solid gain. These findings are comparable to earlier work [34,40,47]. The Peleg capacity constants 'K_2' (0.014-0.016 and 0.042-0.059 for water loss and solid gain, respectively) describe the rate of water loss and solid gain at the equilibrium stage of osmotic dehydration process. 'K_2' decreases with increased in solution concentration and temperature for water loss and solid gain.

In power law model, parameter 'k' for water loss increased with increase in concentration of honey to sucrose at 45 and 55 °C. But at 65 °C 'k' value decreased with increase in honey to sucrose concentration (100:10 and 100:15). The value 'n' decreased with increase in concentration of honey to sucrose

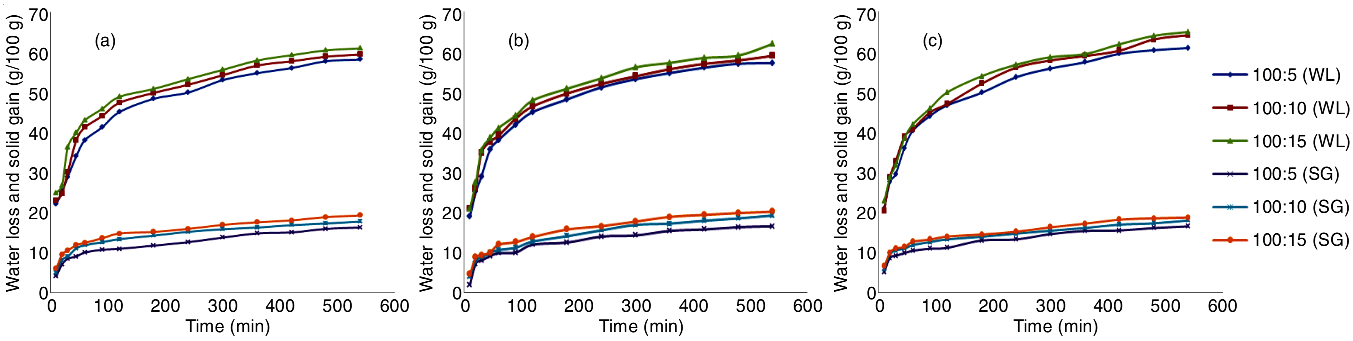

Fig. 1. Effect of osmotic solution concentration and time on water loss and solid gain during osmotic dehydration of apple at different temperatures (a) 45 °C, (b) 55 °C, (c) 65 °C at solution to fruit ratio (STFR) 3:1

TABLE-1
MODEL'S PARAMETERS AND GOODNESS OF FIT FOR WATER LOSS DURING OSMOTIC DEHYDRATION OF APPLE

Models		Peleg					Power law					Azuara				
Temp. (°C)	Honey: Sucrose	K_1	K_2	R^2	RMSE	E (%)	k	n	R^2	RMSE	E (%)	WL_∞	β_1	R^2	RMSE	E (%)
45	100:5	0.594	0.016	0.997	3.236	6.990	13.651	0.232	0.992	1.600	3.695	60.875	0.027	0.997	3.236	6.991
	100:10	0.528	0.016	0.998	2.751	5.432	15.315	0.211	0.983	2.398	5.518	62.111	0.030	0.998	2.751	5.432
	100:15	0.491	0.015	0.998	4.108	8.245	17.590	0.202	0.986	2.211	4.593	62.500	0.035	0.997	2.977	5.807
55	100:5	0.572	0.016	0.998	2.672	6.433	13.717	0.234	0.987	2.230	5.733	60.941	0.029	0.998	2.672	6.433
	100:10	0.517	0.016	0.998	2.869	6.714	15.655	0.215	0.989	2.192	5.330	60.975	0.031	0.998	2.869	6.714
	100:15	0.489	0.015	0.998	2.422	5.415	16.205	0.214	0.987	2.348	5.570	62.500	0.032	0.998	2.422	5.415
65	100:5	0.571	0.015	0.997	3.444	7.473	14.241	0.237	0.988	1.936	4.546	64.102	0.026	0.997	3.446	7.477
	100:10	0.510	0.015	0.996	3.013	7.143	14.813	0.236	0.988	1.974	4.504	66.667	0.027	0.996	3.013	7.144
	100:15	0.435	0.014	0.997	4.178	8.659	15.284	0.234	0.982	1.895	4.511	67.567	0.027	0.997	4.174	8.652

TABLE-2
MODEL'S PARAMETERS AND GOODNESS OF FIT FOR SOLID GAIN DURING OSMOTIC DEHYDRATION OF APPLE

Models		Peleg					Power law					Azuara				
Temp. (°C)	Honey: Sucrose	K_1	K_2	R^2	RMSE	E (%)	k	n	R^2	RMSE	E (%)	SG_∞	β_2	R^2	RMSE	E (%)
45	100:5	2.789	0.059	0.988	1.143	10.729	3.259	0.255	0.986	0.620	6.028	16.949	0.021	0.988	1.143	10.729
	100:10	1.960	0.054	0.996	0.808	6.812	4.242	0.231	0.978	0.729	5.838	18.518	0.027	0.996	0.808	6.812
	100:15	1.899	0.050	0.994	1.139	8.284	4.881	0.219	0.987	0.668	4.579	20.000	0.028	0.994	1.139	8.248
55	100:5	3.152	0.056	0.992	1.049	10.303	2.772	0.289	0.968	0.807	9.467	17.557	0.017	0.992	1.049	10.293
	100:10	2.703	0.049	0.992	1.153	9.436	3.089	0.285	0.997	0.621	5.865	20.408	0.018	0.992	1.152	9.432
	100:15	2.370	0.046	0.994	1.127	8.276	3.477	0.284	0.986	0.664	5.303	21.739	0.019	0.994	1.125	8.272
65	100:5	2.357	0.056	0.992	1.291	10.720	3.944	0.227	0.986	0.581	5.053	17.641	0.025	0.992	1.291	10.717
	100:10	1.925	0.044	0.993	1.309	9.854	4.825	0.207	0.987	0.611	4.663	18.518	0.028	0.993	1.308	9.847
	100:15	1.858	0.042	0.992	1.469	10.484	5.088	0.207	0.988	0.600	4.330	19.608	0.028	0.992	1.468	10.483

whereas increased with increase in temperature. These results are comparable with other researchers [48]. For solid gain 'k' increased with increase in concentration of honey to sucrose but at the same time parameter 'k' decreased at 45 and 55 °C, however, increased at 65 °C. Parameter 'n' decreased with increase in the concentration. This value increased at 45 and 55 °C, but decreased at 65 °C.

In Azuara's equation parameters β_1 and β_2 represent water loss and solid gain, respectively. In the present study it was found that β_1 increased with increase in the concentration and temperature whereas it seems to decrease at 65 °C, for water loss. It agrees with the fact that increases in the concentration of osmotic solution, concentration gradient also increased [49]. The predicted equilibrium values of water loss (Table-1) are 60.241, 60.975 and 62.500 g/100 g of sample for different concentrations at 55 °C. It was also observed that WL_∞ increased with increase in the temperature and concentration of the hypertonic solution. Results are consistent with other researchers [50-52]. The predicted values (Table-2) of equilibrium solid gain at 45, 55 and 65 °C and at concentration of honey to sucrose ratio 100:5 are 16.949, 17.557 and 17.641 (g/100 g of sample), respectively. This indicates that with increase in the temperature of osmotic solution, solid gain at equilibrium increased. It depicted that with increase in the concentration of the osmotic solution, solid gain increased. The values of β_2 increases with increase in concentration and temperature.

Process to validate the empirical models for osmotic dehydration of apple cubes: Among the study of different models for osmotic dehydration kinetics, Power law model was best fitted to the experimental data for water loss and solute gain. Coefficient of correlation (R^2), mean relative deviation modulus (E) and root mean square error (RMSE) were used for best fitting of the model. The present work revealed that the Power law model having highest R^2 (> 0.982), the lowest E (< 6.028 %) and RMSE (< 2.398) adequately described the osmotic dehydration kinetics. However, Azuara model was best fit for osmotic dehydration reported by various researchers [53,54], but in the present study this model did not fit to the experimental data.

Conclusion

The effect of solution concentration (honey to sucrose *i.e.* 100:5, 100:10 and 100:15) and temperature (45, 55 and 65 °C) with solution to fruit ratio (STFR) 3:1 on mass transfer kinetics of the apple were investigated in terms of water loss and solid gain. The results of the present study revealed that a trend of increase in the water loss and solid gain with increase in the concentration and temperature was observed. Among different applied models, Power law's model was best fitted to the experimental data for higher coefficient of correlation (R^2), minimum root mean square error (RMSE) and less than 10 % mean relative deviation modulus (E). Power law's parameters 'k' and 'n' were varied from 13.651 to 17.590 and 0.202 to 0.237, respectively for water loss and 2.772 to 5.088 and 0.207 to 0.289, respectively for solid gain.

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