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D.TIROUTCHELVAME[1]
V. SIVAKUMAR[2]
J. PRAKASH MARAN[3]

[1]Department of Food Processing and Engineering, Karunya University, Coimbatore, Tamil Nadu, India
[2]Department of Chemical Engineering, AC Tech Campus, Anna University, Chennai, Tamil Nadu, India
[3] Department of Food Technology, Kongu Engineering College, Perundurai, Erode, Tamil Nadu, India

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MASS TRANSFER KINETICS DURING OSMOTIC DEHYDRATION OF AMLA (*Emblica officinalis* L.) CUBES IN SUGAR SOLUTION

Article Highlights

- Osmotic dehydration of amla cubes were studied using RSM
- Mass transfer properties and quality characteristics of dehydrated amla cubes were studied
- Second order polynomial models were developed for responses
- Optimization of osmotic dehydration process was done by desired function methodology

Abstract

A four-factor three-level Box-Behnken response surface design was employed in this study to investigate and optimize the effect of process variables (osmotic solution concentration, fruit to liquid ratio, temperature and dehydration time) on mass transfer properties such as weight reduction, solute gain, water loss, rehydration ratio, shrinkage and overall acceptability of the osmotically dehydrated amla cubes. The cubes of uniform size (10 mm×10 mm×10 mm) were impregnated into sugar solution of different solution concentration (30-50 °Brix), temperature (30-50 °C), fruit to liquid ratio (1:5-1:15 w/V) and time (30--180 min). It was observed from the results that the process variables have significant effects on osmotic dehydration process. The optimum condition was found to be: sugar concentration of 50 °Brix, solution temperature of 30 °C, fruit to liquid ratio of 1:5 and immersion time of 133 min, respectively. The microstructural changes during osmotic dehydration were also investigated using scanning electron microscopy (SEM).

Keywords: amla, osmotic dehydration, response surface methodology, microstructure.

Fruits and vegetables are considered to be important components of life for the health of human beings by providing the required nutrients. Since fruits and vegetables are highly perishable in nature, they should be processed quickly after harvest in order to reduce the post-harvest losses during glut period. As a perishable commodity, they are available at much cheaper in terms of selling price during the peak season, and can also lead to more financial losses to the grower resulting in the spoilage in larger quantities. The preservation of fruits and vegetables can prevent a huge wastage and make them available during the off-season at remunerative prices [1].

The removal of water from solid food is a form of food preservation, inhibiting the growth of microorganisms, besides preventing a large part of biochemical reactions that occur due to the presence of moisture [2]. The dehydration process of fruits and vegetables provides many advantages, such as reduced weight, inexpensive packaging, dry shelf stability and negligible deterioration in quality due to enzymatic changes. However, this method has some disadvantages related to the preparation and handling of large volumes of osmotic solutions, high water consumption and losses of soluble nutritional compounds in the osmotic solution. Proper management and reuse of the spent osmotic solution need to be addressed in order to develop this process to be economically viable and cost effective.

Correspondence: V. Sivakumar, Department of Chemical Engineering, AC Tech Campus, Anna University, Chennai, Tamil Nadu, India.
E-mail: drvsivakumar@yahoo.com
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Osmotic dehydration is the process of partial removal of water from fruits and vegetables using hypertonic solutions. These solutions can contain one or more solutes. For fruits, sugar is the commonly used solute. Other solutes that are used as osmotic agents include glucose, fructose, maltodextrose and corn syrup [3]. Common salt (sodium chloride) is the most used solute for vegetables. In some cases, a combination of solutes can also be used. Osmotic dehydration is governed by the osmotic pressure difference between the food material (hypotonic medium) and concentrated osmotic solution (hypertonic medium) [4]. The cell membrane works as a semi-permeable tissue and allows water to pass through faster than solutes [5]. The mass transfer rate is higher at the beginning of the process because of the largest difference in the osmotic pressure between the osmotic solution and cell tissue of the material and less mass transfer resistance at the initial stage of the process [6]. The rate of water removal from the food material and changes in the chemical composition depend on the type of osmotic solute used, its concentration, temperature, time of impregnation in the solution, fruit to solution ratio, size and kind of the material and the type of apparatus used. During osmotic dehydration, two main mass fluxes (water loss and solid gain) take place as counter-current. The effectiveness of the osmotic treatment can be evaluated by the ratio of water loss to solid gain, considering that the water removal must be greater than the solid intake [7]. The low values of this ratio establish the best condition of osmotic dehydration [8].

Amla (*Emblica officinalis* L.), commonly called as Indian gooseberry is an important seasonal fruit crop of the Indian subcontinent [9]. The plant has the ability to grow in wastelands and cultivation area has increased in the recent years [10]. Owing to hardy nature, suitability to various wastelands, high productivity/unit area (15-20 t/ha), nutritive and therapeutic value, amla is becoming more and more commercially important now a days. Amla is highly nutritious and is an important dietary source of Vitamin C, minerals and amino acids. It contains 500-1500 mg of ascorbic acid per 100 g of pulp [11]. The medicinal properties of amla were studied as a preventive and therapeutic drug for cancer in humans [12]. Studies have also been performed for the evaluation of amla properties on in vivo tests [13].

Amla is highly perishable in nature and it can be stored under atmospheric conditions only for about 5-6 days after harvesting [14]. The storage and shelf life of the fruits can be increased by adopting appropriate processing methods and it could eliminate the post--harvest losses up to 30% [15]. The methods adopted for extending the shelf life of amla fruit include cold storage, sun drying and hot air drying or by value addition of the fruits by converting to murabba, pickle, juice, syrup, squash and dehydrated powder [16]. Among the various drying methods available, osmotic dehydration is one of the most simple and inexpensive alternate processes. It is an energy-saving and low capital investment process that offers a way to make available this highly perishable and valuable crop for the regions away from production zones and also during off-season. The cultivation of amla has increased, thus augmenting the post-harvest storage problems. Development of shelf stable products from amla is important for the reduction of post-harvest losses. Hence, the objective of the present research was to evaluate and optimize the influence of the process variables such as osmotic solution concentration, time of dehydration, temperature and fruit to liquid ratio on the osmotic dehydration characteristics (water loss, solid gain, weight reduction, rehydration ratio and shrinkage) of amla cubes in sugar solution using statistical experimental design. In addition, the overall acceptability of the product was also analyzed with the help of trained panelist using a nine point hedonic scale sensory evaluation method. Scanning electron microscopy (SEM) was also used to analyze the microstructural changes during osmotic dehydration of amla.

MATERIALS AND METHODS

Raw materials

Fresh amla fruits (uniform size, shape, and maturity) were procured from local market near Coimbatore, Tamil Nadu, India. Osmotic solutions with different concentration (30-50 °Brix) were prepared by dissolving appropriate amount of sugar in distilled water. The concentrations (°Brix) of the prepared osmotic solutions were checked by using a hand refractometer. The initial moisture content of the raw fruit and the final moisture content after impregnation in sugar solution were determined by following AOAC procedure [17]. The initial moisture content (wet basis) of fresh amla fruit was 80.5% and the final moisture content after osmotic treatment in sugar solution was found to be 61.5%.

Experimental procedure

The amla fruit was cut in to 1 cm^3 (10 mm×10 mm×10 mm) size cubes for each experiment. The amla fruits were washed in running tap water in order to remove any adhering foreign matters on the surface. The outer skin of the fruit is waxy in nature which will resist the mass transfer rate. Hence, the fruits were blanched for 5 min in hot water, which could remove the unacceptable odor and to increase the mass transfer by loosening the tissues of the fruit. The fruits were then removed from hot water and the excess moisture on the surface was removed by using muslin cloth. The seeds were removed by using seed remover and the pulp of the fruit was used for further experimental analysis.

The desired concentration of osmotic solution of sugar was prepared and the known weight of amla cubes were immersed in Erlenmeyer flasks containing osmotic solutions of different concentrations (30-50 °Brix) at different temperatures (30-50 °C), fruit to liquid ratio (1:5-1:15 *w*/*V*) and time (30-180 min). From the preliminary experimental results, the process variables and their ranges were selected. The osmotic dehydration process was carried out in a temperature-controlled chamber. The Erlenmeyer flasks were covered with a plastic wrap during the experiments in order to prevent evaporation of the osmotic solution. During osmotic treatment, at a particular interval of time, the cubes were removed from the osmotic solution and weighed after removing the solution adhering on the surface using filter paper (Whatman No. 1). The experiments were carried out in randomized order to minimize the variability in the observed responses due to extraneous factors. All the experiments were performed in triplicate and the mean value was used for the determination of water loss, weight reduction and solid gain.

Mathematical calculations

Determination of mass transfer properties

In osmotic dehydration, both water loss and solid gain take place simultaneously. The reduction in weight is attributed to the loss of water from the sample and increase in the weight of the sample due to solute gain from the osmotic solution. The evaluation of mass transfer between the solution and samples during osmotic dehydration process were estimated by using the parameters such as weight reduction (*WR*), solid gain (*SG*) and water loss (*WL*) and the parameters were calculated by using the following equations:

$$WR(\%) = 100\frac{W_0 - W_t}{W_0} \tag{1}$$

$$SG(\%) = 100\frac{S_t - S_0}{W_0} \tag{2}$$

$$WL(\%) = WR + SG \tag{3}$$

where W_0 is the initial weight of amla cubes (g), W_t the weight of amla cubes after osmotic dehydration for any time t (g), S_0 is the initial dry weight of amla (g), and S_t is dry weight of amla after osmotic dehydration for any time t (g).

Estimation of quality parameters

The quality parameters such as rehydration ratio (*RR*) and shrinkage (*SH*) of the osmotic dehydrated amla cubes were studied in the present work. The rehydration characteristics of the osmotically dehydrated amla cubes were determined by soaking a known amount of sample in 50 ml of water and kept at room temperature [18] until constant weight was attained. The rehydration ratio was computed by using the formula:

$$RR(\%) = \frac{\text{Weight of rehydrated amla cube (g)}}{\text{Weight of dehydrated amla cube (g)}} \times 100 \tag{4}$$

Shrinkage (*SH*) of the dehydrated sample was determined by using the following equation:

$$SH(\%) = 100\left(1 - \frac{V_i}{V_0}\right) \tag{5}$$

where V_i is the volume displaced by the dehydrated sample and V_0 is the volume displaced by the fresh sample [19].

Organoleptic evaluation

Overall acceptability (OA) of dehydrated amla cubes was evaluated by a trained panel of 10 members of all age groups (15-50). The samples were served in plastic cups, coded with three-digit randomly selected numbers to the panelists in a random order. The panelists were instructed to cleanse their palates using water between the samples. All the panelists evaluated the odor and taste of the samples using quantitative descriptive analysis (QDA) technique with a line scale of 0-9 [20] where 0 and 9 are assigned to negative and positive intensity of overall acceptability of dehydrated amla cubes. The overall acceptability was computed by the average scores of all the 10 panelists.

Response surface methodology modeling

Response surface methodology (RSM) is an empirical statistical modeling technique employed for multiple regression analysis using quantitative data obtained from properly designed experiments to solve multivariate equations simultaneously. A Box Behnken Design (BBD) with four factors at three levels was used to design the experiments and it is exhibited in

Table 1. The process parameters (independent variables) selected for the optimization were osmotic solution concentration (X_1), fruit to liquid ratio (X_2), osmotic temperature (X_3) and osmotic dehydration time (X_4). The number of experiments (N) required for the development of BBD is defined as $N = 2k(k-1) + C_o$ (where k is the number of factors and C_o is the number of central point) [21]. The design included 29 experiments and with 5 central points. Each independent variable was coded at three levels between +1, 0 and -1, whereas osmotic solution concentration: 30-50 °Brix, fruit to liquid ratio: 1:5-1:15 g/ml, osmotic temperature: 30-50 °C and time: 30-180 min. Coding of the variables was done according to the following equation:

$$x_i = \frac{X_i - X_{cp}}{\Delta X_i},\ i = 1,2,3\ldots,k \tag{6}$$

where x_i is the dimensionless value of an independent variable; X_i is the real value of an independent variable; X_{cp} is the real value of an independent variable at the center point; and ΔX_i is the step change of real value of the variable i corresponding to a variation of a unit for the dimensionless value of the variable i.

Performance of the process was evaluated by analyzing the responses (Y), which depend on the input factors x_1, x_2,…,x_k, and the relationship between the response and the input process parameters is described by:

$$Y = f\left(x_1, x_2, \ldots, x_k\right) + e \tag{7}$$

where f is the real response function the format of which is unknown and e is the error which describes the differentiation [22].

A second-order polynomial equation was used to fit the experimental data to identify the relevant model terms using statistical software (Design Expert 8.0.7.1). A quadratic model, which also includes the linear model, can be described as

$$\mathrm{Y} = \beta_0 + \sum_{j=1}^{k} \beta_j x_j + \sum_{j=1}^{k} \beta_{jj} x_j^2 + \sum_{i} \sum_{<j=2}^{k} \beta_{ij} x_i x_j + e_i \tag{8}$$

where x_i and x_j are variables (i and j range from 1 to k); β_0 is the model intercept coefficient; β_j, β_{jj} and β_{ij} are interaction coefficients of linear, quadratic and the second-order terms, respectively; k is the number of independent parameters ($k = 4$ in this study); and e_i is the error [23].

The statistical analysis was performed using Design Expert Statistical Software package 8.0.7.1 (Stat Ease Inc., Minneapolis, MN, USA). The experimental data was analyzed using multiple regressions and the significance of regression coefficients was evaluated by F-test. Modeling was started with a quadratic model including linear, squared and interaction terms and the model adequacies were checked in terms of the values of R^2, adjusted R^2 and prediction error sum of squares (*PRESS*). The significant terms in the model were found by Pareto analysis of variance (ANOVA) for each response at significance level of 95% and ANOVA tables were generated. The regression coefficients were used to make statistical calculations to generate response surface plots from the regression models.

Microstructure analysis

The fresh and osmotically treated (optimal condition) amla pieces were examined using scanning electron microscopy (SEM) in order to determine the effect of osmotic dehydration process on the microstructure of the tissue. Samples were cut into cubes with a sharp blade and mounted on aluminium SEM stubs for gold coating with a fine coat. The microstructure of the tissue was examined by a JEOL scanning electron microscope (JSM-6390) and the images were recorded at the magnification of 100×.

RESULTS AND DISCUSSION

Model fitting and statistical analysis

A total number of 29 experiments were performed with different combinations of process variables in order to study and optimize the combined effect of independent variables (osmotic solution concentration, fruit to liquid ratio, osmotic temperature and time) on the responses (*WR*, *SG*, *WL*, *RR*, *SH* and *OA*) and the results are shown in Table 2.

By applying multiple regression analysis on the experimental data, Design-Expert software generated the second-order polynomial equation, which can

Table 1. Coded and uncoded values of process variables and their levels during osmotic dehydration of amla cubes in sugar solution

Independent variables	Coded levels	−1	0	+1
Solution concentration, °Brix	X_1	30	40	50
FL ratio, g/ml	X_2	1:5	1:10	1:15
Temperature, °C	X_3	30	40	50
Time, min	X_4	30	105	180

Table 2. Box-Behnken experimental design matrix with observed values of responses for the osmotically treated amla cubes in sugar solution; WR: weight reduction, SG: solid gain, WL: water loss, RR: rehydration ratio, OA: overall acceptability, SH: shrinkage

Std. order	X_1	X_2	X_3	X_4	*WR* / %	*SG* / %	*WL* / %	*RR* / %	*OA*	*SH* / %
1	-1	-1	0	0	22.91±1.03	7.75±0.43	28.19±1.21	4.6±0.13	6.2±0.18	3.8±0.23
2	1	-1	0	0	32.55±1.18	17.35±0.65	37.84±1.30	14.3±0.27	7.9±0.21	11.9±0.48
3	-1	1	0	0	22.34±1.09	7.18±0.23	27.62±1.26	4.0±0.14	6.1±0.19	3.4±0.09
4	1	1	0	0	26.26±1.24	11.06±0.43	31.51±1.32	8.0±0.32	6.8±0.22	6.6±0.31
5	0	0	-1	-1	27.30±1.17	12.11±0.26	32.54±1.05	9.0±0.37	7.0±0.36	7.5±0.28
6	0	0	1	-1	31.46±1.13	16.26±0.68	36.71±1.28	13.2±0.28	7.7±0.43	11.0±0.45
7	0	0	-1	1	23.86±1.31	8.71±0.29	29.10±1.15	5.6±0.17	6.4±0.61	4.6±0.17
8	0	0	1	1	33.68±1.26	18.53±0.41	38.93±1.29	15.4±0.33	8.1±0.24	12.8±0.29
9	-1	0	0	-1	24.56±1.14	9.36±0.09	29.81±1.14	6.3±0.19	6.5±0.08	5.2±0.22
10	1	0	0	-1	32.95±1.21	17.75±0.62	38.23±1.35	14.7±0.52	8.0±0.25	12.2±0.26
11	-1	0	0	1	24.55±1.09	9.35±0.33	29.81±1.26	6.3±0.18	6.5±0.36	5.2±0.17
12	1	0	0	1	33.87±1.21	18.71±0.53	39.11±1.17	15.6±0.53	8.2±0.28	13.0±0.26
13	0	-1	-1	0	26.96±1.15	11.76±0.27	32.24±1.24	8.7±0.16	7.1±0.23	7.2±0.19
14	0	1	-1	0	17.14±1.04	1.94±0.06	22.38±1.18	1.2±0.11	5.2±0.23	1.0±0.06
15	0	-1	1	0	28.05±1.12	12.86±0.47	33.30±1.27	9.8±0.47	7.0±0.18	8.1±0.25
16	0	1	1	0	31.44±1.36	16.25±0.18	36.73±1.35	13.2±0. 53	7.7±0.17	11.0±0.39
17	-1	0	-1	0	19.48±1.07	4.29±0.08	24.76±1.13	1.2±0.12	5.6±0.37	1.0±0.05
18	1	0	-1	0	31.21±1.27	16.06±0.41	36.46±1.24	12.9±0.61	7.7±0.19	10.8±0.24
19	-1	0	1	0	31.44±1.32	16.24±0.29	36.73±1.28	13.2±0.53	7.7±0.31	11.0±0.27
20	1	0	1	0	37.11±1.35	21.91±0.63	42.35±1.35	18.8±0.61	8.7±0.22	15.7±0.39
21	0	-1	0	-1	26.83±1.17	11.63±0.25	32.11±1.23	8.5±0.24	6.9±0.27	7.1±0.21
22	0	1	0	-1	28.11±1.09	12.92±0.37	33.36±1.19	9.8±0.29	7.1±0.37	7.6±0.14
23	0	-1	0	1	27.46±1.05	12.27±0.31	32.71±1.16	9.2±0.27	7.0±0.62	7.6±0.18
24	0	1	0	1	23.90±1.13	8.71±0.26	29.19±1.14	5.6±0.18	6.4±0.65	4.7±0.21
25	0	0	0	0	39.29±1.28	24.10±0.72	44.53±1.25	21.0±0.67	9.1±0.53	17.5±0.29
26	0	0	0	0	39.29±1.25	24.11±0.63	44.53±1.27	21.0±0.62	9.1±0.45	17.5±0.22
27	0	0	0	0	39.29±1.21	24.14±0.68	44.53±1.23	21.0±0.58	9.1±0.32	17.5±0.36
28	0	0	0	0	39.29±1.19	24.10±0.47	44.54±1.19	21.0±0.64	9.1±0.31	17.5±0.32
29	0	0	0	0	39.29±1.16	24.10±0.61	44.54±1.28	21.0±0.39	9.1±0.56	17.5±0.41

express the relationship between process variables and the responses. The final equations obtained in terms of coded factors are as follows:

$$WR = 39.29 + 4.06X_1 - 1.301X_2 + 3.94X_3 + 1.43X_1X_2 - 1.52X_1X_3 + 3.30X_2X_3 - 1.21X_2X_4 + 1.42X_3X_4 - \\ -4.97X_1^2 - 8.13X_2^2 - 4.98X_3^2 - 5.05X_4^2 \tag{9}$$

$$SG = 24.11 + 4.06X_1 - 1.30X_2 + 3.93X_3 + 1.43X_1X_2 - 1.52X_1X_3 + 3.30X_2X_3 - 1.21X_2X_4 + 1.42X_3X_4 - \\ -4.97X_1^2 - 8.13X_2^2 - 4.98X_3^2 - 5.06X_4^2 \tag{10}$$

$$WL = 44.53 + 4.05X_1 - 1.30X_2 + 3.94X_3 + 1.44X_1X_2 - 1.52X_1X_3 + 3.32X_2X_3 - 1.19X_2X_4 + 1.41X_3X_4 - \\ -4.95X_1^2 - 8.11X_2^2 - 4.98X_3^2 - 5.05X_4^2 \tag{11}$$

$$SH = 17.50 + 3.38X_1 - 0.97X_2 + 3.12X_3 - 1.19X_1X_2 - 1.26X_1X_3 + 2.27X_2X_3 - 0.87X_2X_4 + 1.18X_3X_4 - \\ -4.20X_1^2 - 6.66X_2^2 - 3.97X_3^2 - 4.34X_4^2 \tag{12}$$

$$RR = 21.00 + 4.06X_1 - 1.11X_2 + 3.75X_3 - 1.43X_1X_2 - 1.52X_1X_3 + 2.73X_2X_3 - 1.21X_2X_4 + 1.42X_3X_4 - \\ -5.06X_1^2 - 7.94X_2^2 - 4.79X_3^2 - 5.15X_4^2 \tag{13}$$

$$OA = 9.12 + 0.71X_1 - 0.23X_2 + 0.66X_3 - 0.25X_1X_2 - 0.27X_1X_3 + 0.67X_2X_3 - 0.20X_2X_4 + 0.25X_3X_4 - \\ -0.87X_1^2 - 1.42X_2^2 - 0.87X_3^2 - 0.89X_4^2 \tag{14}$$

Pareto analysis of variance (ANOVA) was used to analyze the experimental data and the results are listed in Table 3. The higher model *F*-value (84.10 for

Table 3. Analysis of variance (ANOVA) for the observed responses; DF: degree of freedom, RC: regression coefficient

Source	*DF*	*WR* / %		*SG* / %		*WL* / %		*RR* / %		*OA* / %		*SH* / %	
		RC	*p* value	RC	*p* value	RC	*p* value	RC	*p* value	RC	*p* value	RC	*p* value
Model	14	39.29	< 0.0001	24.11	< 0.0001	44.53	< 0.0001	21	< 0.0001	9.12	< 0.0001	17.50	< 0.0001
X_1	1	4.06	< 0.0001	4.06	< 0.0001	4.05	< 0.0001	4.06	< 0.0001	0.71	< 0.0001	3.38	< 0.0001
X_2	1	-1.30	0.0004	-1.30	0.0004	-1.30	0.0004	-1.11	0.0013	-0.23	0.0004	-0.97	0.0005
X_3	1	3.94	< 0.0001	3.93	< 0.0001	3.94	< 0.0001	3.75	< 0.0001	0.66	< 0.0001	3.12	< 0.0001
X_4	1	-0.32	0.2660	-0.31	0.2823	-0.33	0.2609	-0.32	0.2639	-0.05	0.2972	-0.22	0.3189
X_{12}	1	-1.43	0.0101	-1.43	0.0105	-1.44	0.0097	-1.43	0.0098	-0.25	0.0114	-1.19	0.0061
X_{13}	1	-1.52	0.0071	-1.53	0.0071	-1.52	0.0070	-1.52	0.0069	-0.27	0.0081	-1.26	0.0042
X_{14}	1	0.23	0.6341	0.24	0.6241	0.22	0.6548	0.23	0.6326	0.04	0.6413	0.20	0.6058
X_{23}	1	3.30	< 0.0001	3.30	< 0.0001	3.32	< 0.0001	2.73	< 0.0001	0.67	< 0.0001	2.27	< 0.0001
X_{24}	1	-1.21	0.0248	-1.21	0.0252	-1.19	0.0267	-1.21	0.0242	-0.20	0.0326	-0.87	0.0346
X_{34}	1	1.42	0.0108	1.42	0.0110	1.42	0.0108	1.42	0.0105	0.25	0.0122	1.18	0.0065
X_1^2	1	-4.97	< 0.0001	-4.97	< 0.0001	-4.95	< 0.0001	-5.06	< 0.0001	-0.87	< 0.0001	-4.20	< 0.0001
X_2^2	1	-8.13	< 0.0001	-8.14	< 0.0001	-8.11	< 0.0001	-7.94	< 0.0001	-1.43	< 0.0001	-6.66	< 0.0001
X_3^2	1	-4.98	< 0.0001	-4.98	< 0.0001	-4.98	< 0.0001	-4.79	< 0.0001	-0.87	< 0.0001	-3.97	< 0.0001
X_4^2	1	-5.05	< 0.0001	-5.06	< 0.0001	-5.05	< 0.0001	-5.15	< 0.0001	-0.89	< 0.0001	-4.34	< 0.0001
R^2		0.988		0.988		0.988		0.988		0.988		0.990	
Adj-R^2		0.976		0.976		0.976		0.976		0.975		0.979	
Pre-R^2		0.932		0.932		0.932		0.930		0.929		0.940	
CV / %		3.24		6.66		2.75		8.33		2.32		7.73	
Adeq. Pre.		31.25		31.09		31.24		29.44		31.13		31.94	

WR, 83.27 for *SG*, 83.89 for *WL*, 80.78 for *RR*, 94.65 for *SH* and 80.55 for *OA*) and the associated lower *p*-values ($p < 0.0001$) demonstrated the significance of developed models and also indicated that most of the variation in the responses could be explained through the regression equations. The high value of R^2 (0.9882 for *WR*, 0.9881 for *SG*, 0.9882 for *WL*, 0.9878 for *RR*, 0.9875 for *SH* and 0.9867 for *OA*) and adj-R^2 (0.9765 for *WR*, 0.9763 for *SG*, 0.9764 for *WL*, 0.9755 for *RR*, 0.9791 for *SH* and 0.9755 for *OA*) clearly demonstrated that the form of the model chosen to represent the actual relationship between the response and independent variables is well correlated and accurate. Low values of coefficient of variance (3.24 for *WR*, 6.66 for *SG*, 2.75 for *WL*, 8.33 for *RR*, 7.73 for *SH* and 2.32 for *OA*) exhibited the high degree of precision and good reliability of the conducted experiments. In this study, the adequate precision (signal to noise ratio) was found to be > 29 for all the responses, which indicated the best fitness of the developed models.

Diagnostics of model adequacy

Generally, it is important to confirm that the fitted model gives a sufficient approximation to the actual values. Unless the model shows a satisfactory fit, proceeding with an investigation and optimization of the fitted response surface likely gives poor or misleading results. In addition to determination coefficient, the adequacy of the models was also evaluated by the residuals (difference between the observed and the predicted response value) and the influence plots for the experimental data obtained from this study. Diagnostic plots such as predicted versus actual (Figure 1) help us to evaluate the model suitability and find out the relationship between predicted and experimental values. The data points on this plot lie reasonably close to the straight line and indicated that an adequate agreement between real data and the data obtained from the models. Hence, trends observed in Figure 1 revealed that no obvious patterns were found and residuals appeared to be randomly scattered.

Effect of process variables

To understand the interaction between the independent variables and dependent variables, three dimensional (3D) response surface plots were plotted from the developed model. In this study, the model has more than two factors. Hence, the 3D plots were drawn by maintaining two factors at constant level (in turn at its central level), whereas the other two factors were varied in their range in order to understand their main and interactive effects on the dependent vari-

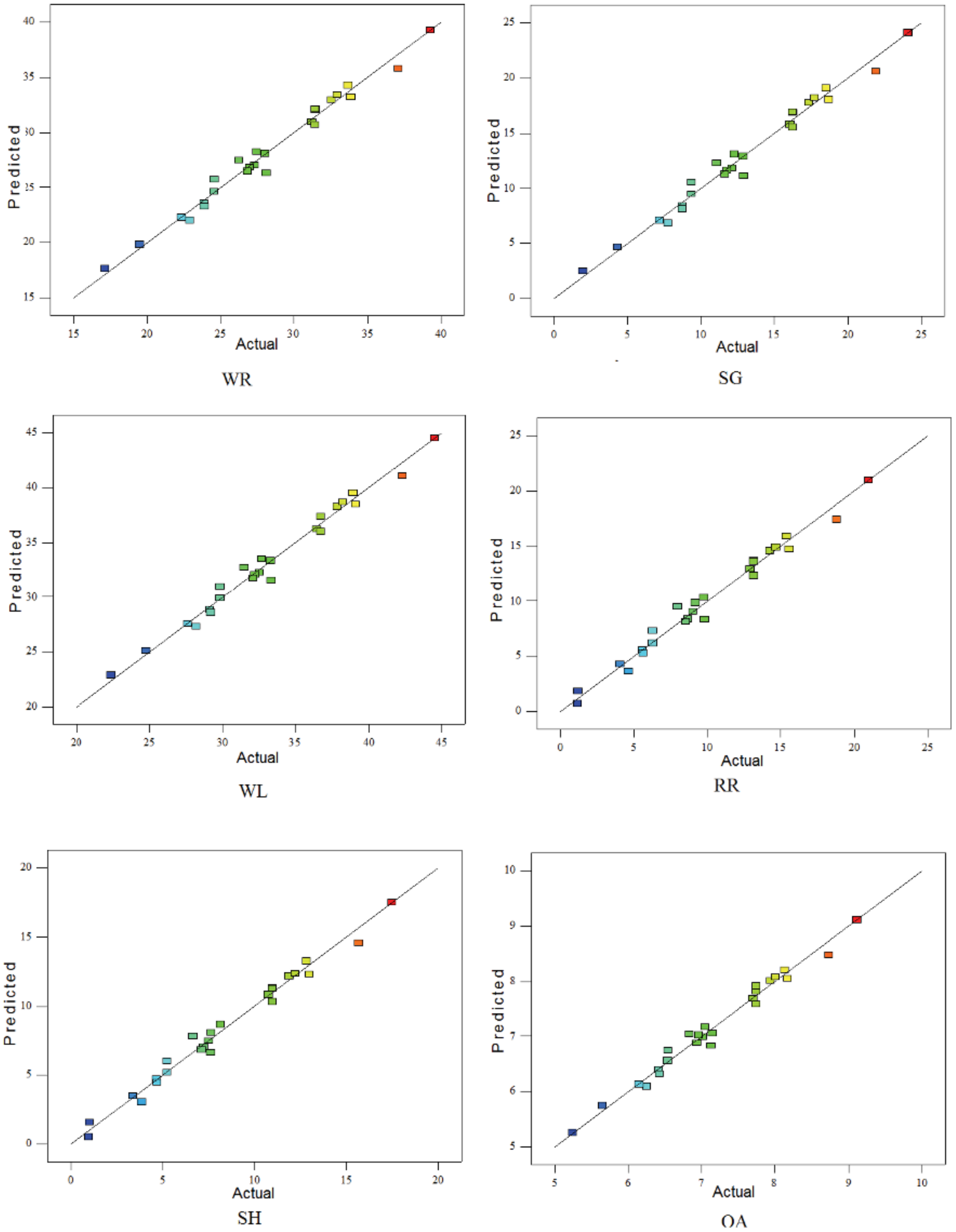


Figure 1. Model adequacy plots (experimental vs. predicted) for responses (WR: weight reduction, SG: solid gain, WL: water loss, RR: rehydration ratio, SH: shrinkage, OA: overall acceptability).

ables. The model was also used to locate the optimum conditions.

Mass transfer properties

At the beginning of the process, due to high osmotic driving force between the concentrated solution and the fresh sample, the rate of water removal and solid gain was relatively high. The osmosis effect increased with the increasing sugar concentration from 30-45 °Brix (Figure 2a-b). Increase in solution concentration up to 45 °Brix resulted in an increase in the osmotic pressure gradients and hence, higher

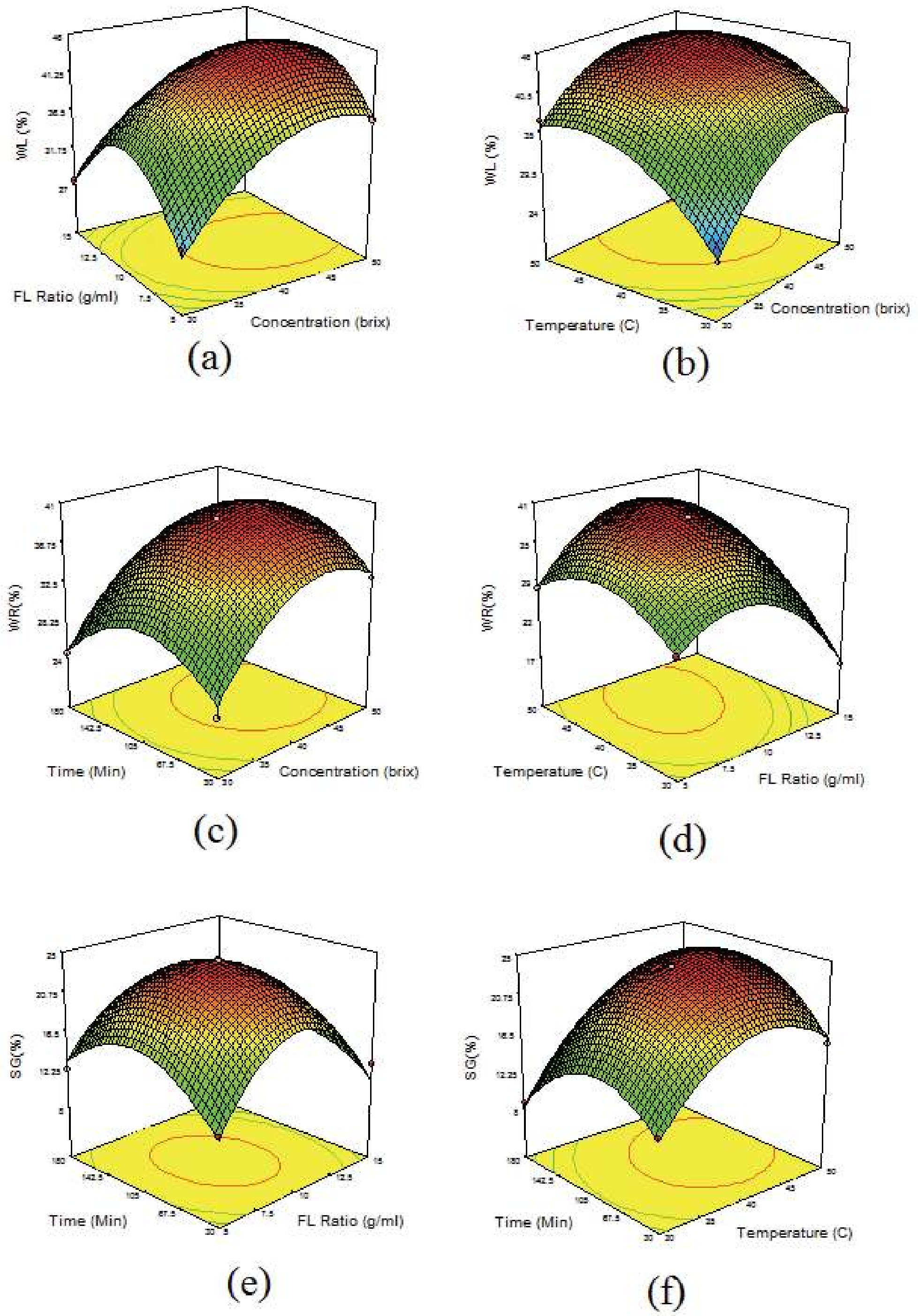


Figure 2. Response surface plots for different mass transfer parameters (WL: water loss, WR: weight reduction, SG: solid gain) during osmotic dehydration of amla cubes.

water loss (and solid uptake) values throughout the osmosis period were obtained. When the osmotic solution concentration was increased, water loss and solid gain took place in parallel mode; the rate of water loss is always higher than the solid gain. In osmotic dehydration, the concentration gradient between the intracellular fluid and osmotic solution create a difference of osmotic pressure, which leads to diffusion of water and solid molecules through the semi-permeable membrane of this fruit to achieve osmotic equilibrium. Thus, the increase in solute concentration led to increases in SG and WL. Further increase of sugar concentration reduced the water loss that might have led to sugar gain by the fruits, which was not desirable [24]. This is attributed to the diffusion of water from dilute medium to concentrated solution (hypertonic solution) through a semi-permeable membrane until the concentration equilibrium was reached. The driving force in this process is the water activity gradient caused due to the osmotic pressure. For solute concentrations of above 50 ºBrix and below 30 °Brix, there was impregnation and crystallization of sugar and poor moisture removal respectively. This strongly suggested that for optimal osmotic dehydration, the sugar concentrations should be in the range of 40-50 °Brix.

High temperatures combined with high concentration of osmotic solution were shown to facilitate osmotic dehydration. Increase in temperature (up to 45 °C) led to more water loss than solid gain, which caused an increase in the weight reduction. This phenomenon is attributed to the diffusion difference between water and solutes as related to their molar masses [25-28]. Further increase in temperature affects the semi-permeability of the cell walls and reduces the rate of osmosis. This may be due to reduction in viscosity of hypertonic solution and increase in diffusion coefficient of water increased at high temperature [29,30].

Osmotic treatment time is one of the most influential variables during osmotic dehydration of fruits, which is due to the fact that the *WL*, *WR* and *SG* were based on the time. From the results, it was observed that increasing time (0 to 135 min) resulted in an initial increase of the *WL*, *WR* (Figure 2c-d) and *SG* (Figure 2e-f), followed by a decrease. This can be explained by the ionization characteristics and low molecular weight of sugar that allow it to diffuse easily into the product and increase the driving force for dehydration. On the other hand, SG increased in the early stages, remained almost constant for a short period of time and finally showed a decreasing trend. This declining trend in SG at the end of the process could be attributed to the loss of some original solids in amla due to the osmotic driving force between the amla and the surrounding sugar solution [31]. But in this study, osmotic dehydration time did not statistically have a significant effect of the process ($p <$ < 0.05, Table 3).

Increasing the volume of osmotic media increases the mass transfer rate. More solution volume could increase the rate of water loss and solid gain but it has some adverse effects also. Increasing the volume of solution causes an increase in operating costs. At the same time, solid gain is increased and the overall mass transfer could lower the product quality by altering the taste of the product due to more migration of natural substances to the osmotic media. Therefore, the weight ratio of solution to sample should be optimized. From the results, it was observed that, increasing sample to solution ratio from 1:5 to 1:13 caused a major increase of WL, WR and SG which is due to an appreciable degree of dehydration achieved by the contact of the fruit pieces with sufficient level of osmotic syrup to prevent dilution and resulted in steady water-solute transfer. Beyond 1:13 ratio, mass transfer properties of amla during osmotic dehydration had decreased and increased the operational cost of the process. This result is compliance with other researchers' observations [32-34].

Quality characteristics

Rehydration ratio

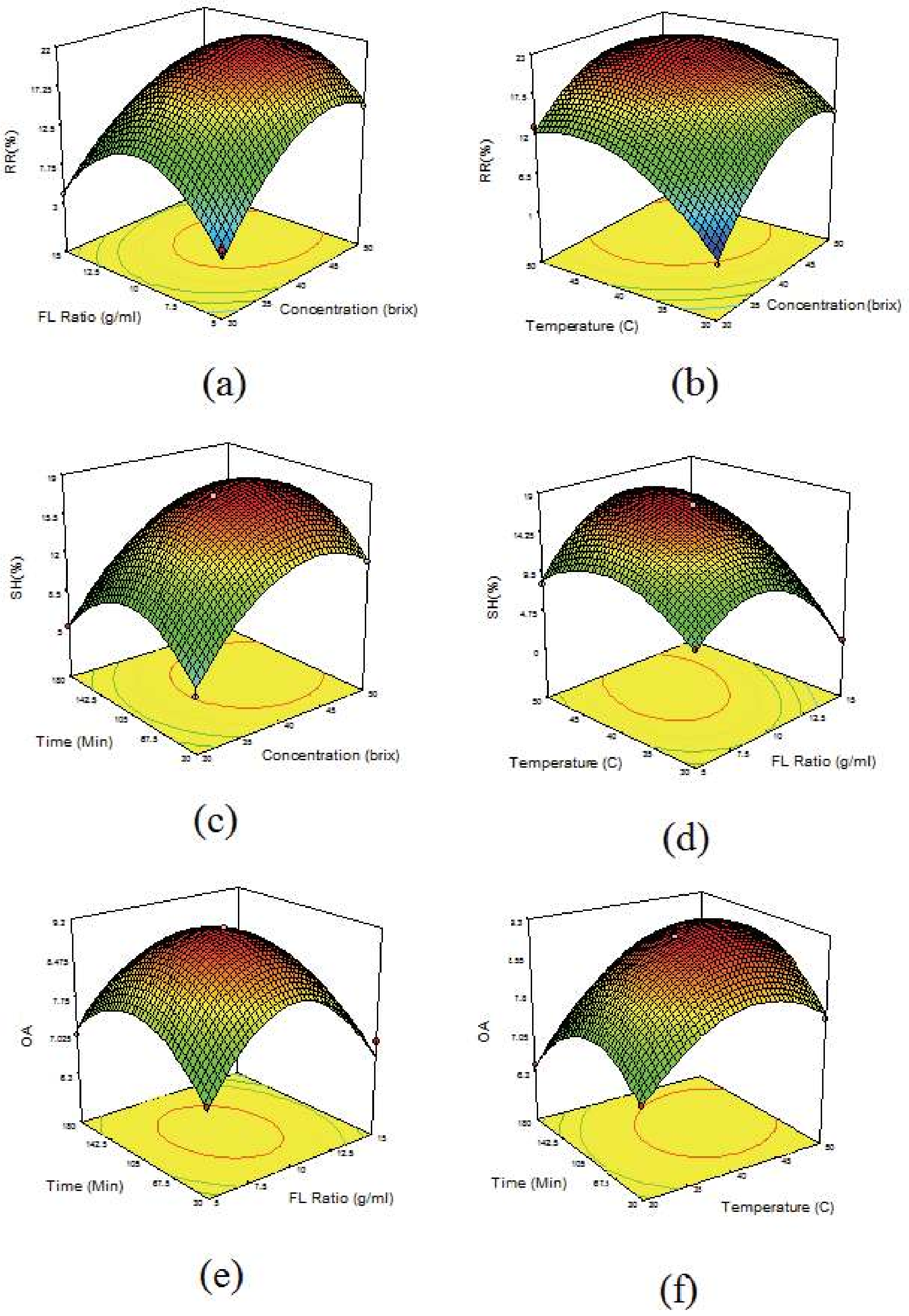
The effect of osmotic dehydration on rehydration ratio increased linearly with the increase of the process variables (Figure 3a and b) up to sugar concentration of 45 °Brix, process time of 120 min, solution temperature of 45 °C and FL ratio of 1:12. This is due to the fact that the rehydration ratio is inversely related to the solute gain during osmotic dehydration process, which has to be leached out during rehydration process.

Shrinkage

The magnitude of the effect of process variables is shown in Figure 3c and d, indicating a linear decrease in shrinkage with increase of osmotic solution temperature and concentration at 180 min of process duration. All the process parameters (except osmotic time and interaction of solution concentration and osmotic time) showed significant effect on the shrinkage of amla cubes statistically based on the p-value (Table 3). The increase in shrinkage with increase in temperature is that higher temperatures seem to promote faster water loss through swelling and plasticizing of cell membranes as well as the better water transfer characteristics on the product surface due to lower viscosity of the osmotic medium [35]. The rapid loss of water, especially in the beginning of OD, and the temperature of the osmotic solution account for a great proportion of shrinkage. Beyond that, decrease in shrinkage of the product with the advancement of process duration was observed, which may be due to the attainment of the saturated conditions. The solid gain, even in small amounts, can reinforce the strength of the solid material, creating more resistance to water removal and reduced shrinkage of the material.

Overall acceptability

Overall acceptability is useful to select the best quality of osmotically dehydrated product with maximum consumer perception. The sensory attributes



(a) (b) (c) (d) (e) (f)

Figure 3. Response surface plots for different quality parameters (RR: rehydration ratio, SH: shrinkage, OA: overall acceptability) during osmotic dehydration of amla cubes in sugar solution.

are affected by different process variables and the results are shown in Figure 3. All the process parameters (except osmotic time and interaction of solution concentration and osmotic time) showed significant effect on the overall acceptability of amla cubes statistically based on the *p*-value (Table 3). The maximum acceptance was noticed for the product osmotically dehydrated under process condition of temperature of 45 °C, concentration of 46 °Brix, FL ratio of 1:11.5 g/ml and time of 120 min. The overall acceptability is achieved due to the prevention of enzymatic and oxidative browning as the fruit pieces were surrounded by sugar and making it possible to retain good consumer observation. Similar results were observed in the osmotic dehydration of sweet anola flakes [36].

Optimization and validation of the optimized conditions

The second-order polynomial models obtained in this study were utilized for each response in order to determine the specified optimum conditions. These regression models are valid only in the selected experimental domain. Therefore, the operating region was determined considering some economical, industrial and product quality-related constraints. In this study, sucrose concentration, FL ratio and immersion temperature were selected in the range of 30-50 °Brix, 1:5-1:15 g/ml and 30-50 °C. Considering the costs involved to increase the process time and also immersion time (X_4) was not statistically significant for all responses in ANOVA table, the immersion time was fixed at 30 min. By applying the desired function methodology, the following optimized conditions were obtained: solution concentration of 43 °Brix, FL ratio of 1:10 g/ml, temperature of 30 °C and time of 30 min. At this optimum point, *WR*, *SG*, *WL*, *RR*, *SH* and *OA* found to be 27.5, 12.32, 32.68, 9.2, 7.3 and 7.16%, respectively.

Microstructure

The microstructural changes during osmotic dehydration are important in order to understand the changes that occurred in the compositional changes of fruits. Images of transversal cross-sections of treated and untreated samples are presented in Figure 4. Figure 4a shows the control sample of amla tissue, which did not receive any treatment other than the preparation for SEM. The bright regions in the micrograph are mainly the cytoplasmic membrane and the cell walls. The cells appeared torn and irregular in shape, with the presence of many empty regions (regions which were not occupied by cells, Figure 4a).

Figure 4b shows that osmotic dehydration process (at optimal condition) changes the tissue structure compared to the untreated sample. In fact, the cells appeared shrunk and distorted and their contour appeared irregular and wrinkling. This fact was probably due to the solubilization of polysaccharides (cellulose, hemicellulose and pectin) that compose the cells walls, the water loss and the pre-concentration of sucrose on the surface of the tissue during the process [26,37-39]. Moreover, water loss induces the plasmolysis of cells and solid gain gives consistency to the tissues. There are several experimental findings in the literature that are consistent with our claims regarding the occurrence of cell structure modification during osmotic processing [38-40].

CONCLUSION

Box-Behnken response surface design was successfully employed in this study to evaluate and identify the optimal osmotic condition in order to prepare osmotically dehydrated amla cubes using sugar solution as an osmotic agent. From the experimental results, second order polynomial models were developed for the responses (water loss, solid gain, weight reduction, rehydration ratio, shrinkage and overall acceptability). The results exhibited that osmotic solution concentration, FL ratio and temperature have significant effects on the osmotic dehydration process of amla. The optimal conditions were found to be: sugar concentration of 50 °Brix, solution temperature of 30 °C, fruit to liquid ratio of 1:5 g/ml, and immersion time of 133 min.

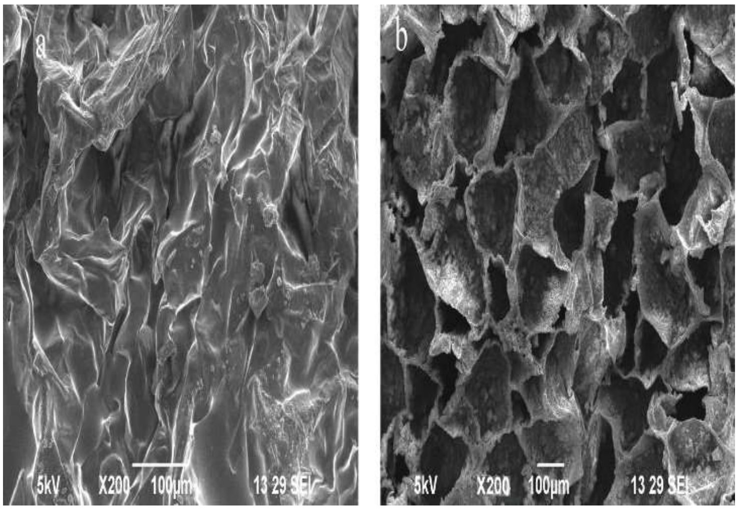


Figure 4. SEM images of amla cubes a) raw and b) osmotically treated sample at optimal condition (sugar concentration of 50 °Brix, solution temperature of 30 °C, fruit to liquid ratio of 1:5 g/ml and immersion time of 133 min).

Nomenclature

WR - Weight reduction
WL - Water loss
SG - Solid gain
RR - Rehydration ratio
SH - Shrinkage
OA - Overall acceptability
RSM - Response surface methodology
BBD – Box-Behnken design
°Brix – Degree Brix
ANOVA - Analysis of variance
SEM - Scanning electron microscopy
FL ratio - Fruit to liquid ratio

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D.TIROUTCHELVAME[1]
V. SIVAKUMAR[2]
J. PRAKASH MARAN[3]

[1]Department of Food Processing and Engineering, Karunya University, Coimbatore, Tamil Nadu, India
[2]Department of Chemical Engineering, AC Tech Campus, Anna University, Chennai, Tamil Nadu, India
[3] Department of Food Technology, Kongu Engineering College, Perundurai, Erode, Tamil Nadu, India

NAUČNI RAD

KINETIKA PRENOSA MASE U OSMOTSKOJ DEHIDRATACIJI KOCKI INDIJSKOG OGROZDA (*Emblica officinalis* L.) U RASTVORU ŠEĆERA

Box-Behnken dizajn sa četiri faktora na tri nivo je korišćen za optimizacije uticaja različitih procesnih faktora (osmotska koncentracija, odnos voća i tečnosti, temperature i vreme dehidratacije) na karakteristike prenosa mase kao što su smanjenje mase, količina rastvorka, gubitak vode, redehidratacioni odnos, smanjenje i ukupna asceptibilnost osmotski dehidratisanih kocki ogrozda. Kocke jednake veličine (10 mm×10 mm×10 mm) su potapane u šećerni rastvor različite koncentracije (30-50 °Brix) određeno vreme (30--180 min) na različitim temperaturama (30-50 °C) i pri različitim odnosima voće-tečnost (1:5-1:15 g/ml). Rezultati pokazuju da procesni faktori imaju značajni uticaj na osmotsku dehidrataciju. Definisani su sledeći optimalni uslovi: koncentracija šećera od 50 °Brix, temperature rastvora 30 °C, odnos voće-tečnost 1:5 g/ml i vreme potapanja od 133 min. Praćene su takođe mikrokristalne promene za vreme osmotske dehidratacije korišćenjem SEM metode.

Ključne reči: indijski ogrozd, osmotska dehidratacija, metodologija površine odziva, mikrostruktura.