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Original Article

Optimization of carrier agents using mixture design for tamarind powder production

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Abstract

This research investigates the collective use of maltodextrin (MD), octenyl succinic anhydride (OSA), and gum arabic (GA) to produce tamarind powders using the double drum dryer method. In this study, seven carrier mixture formulations were specified using a simplex lattice mixture design and the physicochemical characteristics of the tamarind products associated with the MD-OSA-GA combinations were assessed. Graphical optimization was utilized to identify the optimal MD-OSA-GA concentrations that would produce a tamarind powder with the essential physicochemical characteristics that would closely resemble the fresh tamarind pulp including total solids (TS), viscosity (VS), pH value (pH), and total acidity (TA). The optimal mixture concentrations were 61.52 g, 28.48 g, and 10.00 g/100 g for MD, OSA, and GA, respectively, with the tamarind powder possessing 39.88% TS, 0.27 VS, 3.04 pH, and 9.31% TA.

Keywords: tamarind powder, carrier agents, drum drying, mixture design, optimization

1. Introduction

The preparation of fresh tamarind juice or paste presents challenges to modern consumers who are time-strapped and prioritize convenience over cost. Thus, attempts have been made to transform the difficult-to-handle tamarind paste to dried tamarind powder using various drying technologies. The double-drum dryer method is commonly used in the production of heat-sensitive powdered food products, e.g., low-moisture baby foods and fruit powders (Nastaj, 2000). According to Henriquez, Cordova, Almonacid, and Saavedra (2014), the advantages of the drum dryer technique include shorter production time, less storage space, simpler process, and greater end-user convenience.

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Stickiness occurs in the drum drying of fruit juices, including tamarind, due to the low molecular weight of sugars and acids in the juices. According to Bhandari and Howes (2005), the sugars and acids tended to stick to the drum surface under high drying temperatures and were difficult to remove which resulted in impaired product quality, low yields, and damage to the dryer (Bhandari, Datta, & Howes, 1997).

To mitigate the stickiness, high molecular weight carrier agents, e.g., maltodextrin (MD), were introduced into the juices. MD is inexpensive and exhibits low viscosity at high solids concentrations (Carneiro, Tonon, Grosso, & Hubinger, 2013). Due to MD's low emulsifying capability, it is mixed with other carriers, e.g., gum arabic (GA) (Fernandes *et al.*, 2008) or octenyl succinic anhydride (OSA) (Bule, Singhal, & Kennedy, 2010), in the drum dryer to enhance the quality and quantity of juice powders.

There are publications on the collective use of MD, OSA, and GA in the production of fruit juice powders using the spray dryer method (Kunapornsujarit & Intipunya, 2013).

However, research on their collective use in the production of tamarind powder using the drum dryer is non-existent. This research investigates the collective use of MD, OSA, and GA in the production of tamarind powders under the double-drum dryer method. A simplex lattice mixture design was used for variable MD-OSA-GA mixture formulations. An assessment was performed on the physicochemical characteristics of the tamarind products, i.e. mixed tamarind, tamarind powder, and dissolved tamarind, associated with the various mixture formulations. Regression analysis was carried out to determine the statistical relationships between the carrier agents and various physicochemical properties. Moreover, graphical optimization was used to identify the optimal mixture of the carrier agents that would produce the final tamarind powder whose physicochemical characteristics, particularly TS, VS, pH, and TA, closely resembled the fresh tamarind pulp.

2. Materials and Methods

2.1 Raw materials

In this research, the tamarind flesh was from giant sour tamarinds (*Tamarindus indica* L.) from a plantation in the district of Non Sung of Thailand's northeastern province of Nakhon Ratchasima. The total acidity and moisture content of the tamarind flesh were 19.0-22.0% and 25.0-28.0%, respectively.

The experimental carrier agents included MD, OSA, and GA. Specifically, MD with a 10-12 dextrose equivalent and 5.0-6.0% moisture content was from Nutrition SC Co., Ltd. in Thailand's central province of Nakhonpathom, while OSA with a 4.0-8.0% moisture content was from Questex Co., Ltd. in Sumutprakarn Province. GA with a moisture content of 11.0-12.0% was acquired from Chemipan Co., Ltd., Bangkok.

2.2 Preparation of the fresh tamarind juice

The tamarind flesh was deseeded and mixed with 80 ºC hot water in a ratio of 1:5 (w/w, tamarind flesh: hot water). The mixture was kneaded into tamarind paste and filtered with two layers of cheesecloth for tamarind juice. The total soluble solids (TSS) of the tamarind juice were maintained at 11 ºBrix at a ratio of 1:5 (w/w). Table 1 tabulates the physicochemical properties of the experimental tamarind juice prior to the introduction of the carrier agents, i.e. MD, OSA, and GA.

Table 1. Physicochemical properties of the tamarind juice before mixing with the carrier agents.

Physicochemical properties	Mean±SD
Total soluble solid (ºBrix)	11.00±0.01
Total solid content (%)	11.20±0.02
Moisture content (%)	88.79±0.02
Total acidity (%)	20.73±0.05
pH value (-)	2.79±0.01
Viscosity (-)	0.03±0.01
Color of L (-)	33.37±0.01
Color of a (-)	6.99±0.02
Color of b (-)	9.59±0.01

2.3 Drum dryer configuration

A double drum dryer (New Way Manufacturing) was utilized to heat-dry the mixed tamarind, i.e. tamarind juice mixed with the carrier agents. The operating condition was at 140 ºC, 0.50 rpm, and 0.15 mm drum clearance (Prangpru, Jaito, Vanmontree, & Treeamnuk, 2015). The dried tamarind flakes were removed with the doctor blades and pulverized at a high speed for 50 s using a blender (HR2061, Philips) prior to sifting through an 80-mesh sieve and retained in sealed laminated aluminum foil bags for further analysis.

2.4 Experimental design

A simplex lattice mixture design with three factors, i.e. MD, OSA, GA, was used to determine the optimal carrier-mixture formulation for the tamarind powder and maintain the essential physicochemical characteristics to closely resemble the fresh tamarind pulp. In this research, the minimum thresholds of 50 g, 10 g, and 10g/100g for MD, OSA, and GA, respectively, were implemented for the ease of removal of the tamarind flakes by the doctor blades (Prangpru *et al.*, 2015). Furthermore, the MD, OSA, and GA concentrations were respectively varied between 50-80 g/100 g, 10-40 g/100 g, and 10-40 g/100 g with the MD-OSA-GA mixtures summing to 100g that gave rise to seven MD-OSA-GA mixture formulations (Figure 1).

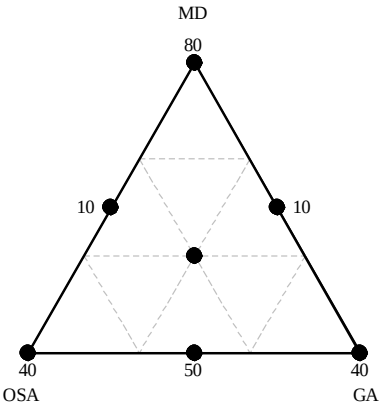

Figure 1. The experimental three-factor simplex lattice mixture design, where MD, OSA, and GA respectively denote maltodextrin, octenyl succinic anhydride, and gum arabic.

Table 2 tabulates the concentrations of the experimental carrier agents (MD, OSA, GA) and the 11 °Brix tamarind juice associated with the seven mixture formulations. In the preparation, MD, OSA, and GA according to the MD-OSA-GA mixture formulations were dissolved in 200 g of the tamarind juice and heat-dried under the 140 ºC, 0.50 rpm, and 0.15 mm drum clearance conditions. Specifically, the feed of each mixture formulation was 300 g which consisted of 100 g of the MD-OSA-GA mixture and 200 g of the 11 °Brix tamarind juice.

The physicochemical response variables of the tamarind products, i.e. mixed tamarind, tamarind powder, and dissolved tamarind, associated with the seven MD-OSA-GA formulations were also assessed and compared. The physicochemical response variables included the drying yield (DY),

Table 2. Concentrations of the carrier agents and tamarind juice of the experimental mixture formulations.

Ingredients	Mixture formulation (g)						
	1	2	3	4	5	6	7
MD	80	65	65	50	50	50	60
OSA	10	25	10	40	25	10	20
GA	10	10	25	10	25	40	20
Tamarind juice	200	200	200	200	200	200	200

water activity (Aw), moisture content (MC), total solids (TS), viscosity (VS), color value (ΔE), pH values (pH), total acidity (TA), bulk density (BD), and solubility (SO). Specifically, the seven MD-OSA-GA formulations were individually experimented in triplicate, giving rise to 21 experimental runs for each response variable.

In this research, the prediction models corresponding to the response variables (DY, Aw, MC, TS, VS, ΔE, pH, TA, BD, SO) were the polynomial regression without a constant term (i.e. intercept equals zero) (Ozturk *et al.*, 2014).

$$Y = \beta_1 X_1 + \beta_2 X_2 + \beta_3 X_3 + \beta_{12} X_1 X_2 + \beta_{13} X_1 X_3 + \beta_{23} X_2 X_3$$

where Y is the estimated response; β_1, β_2, β_3, β_{12}, β_{13}, and β_{23} are the regression coefficients of the linear and nonlinear (interaction) terms. X_1, X_2, and X_3 denote MD, OSA, and GA, respectively. X_1X_2, X_1X_3, and X_2X_3 denote the interaction terms of MD-OSA, MD-GA, and OSA-GA, respectively.

2.5 Optimization and validation of the model

Optimization was carried out using the response surface methodology (RSM) with the three-factor simplex lattice mixture design: X_1, X_2, and X_3 for MD, OSA, and GA, respectively. In light of the seven MD-OSA-GA formulations (Table 2) and each formulation experimented in triplicate, i.e. 21 experimental runs for each response variable, Minitab V. 16 was used for the statistical analysis of variance (ANOVA) and the squared correlation coefficients (R^2) were calculated and at 95% confidence level. In addition, the model fitness was determined through the lack-of-fit (LOF) test (P>0.05) which indicated how well the model fit the data (Khodadoust, Sadeghi, Pebdani, Mohammadi, & Salehi, 2017).

Furthermore, according to Parejiya, Patel, Mehta, Shelat, and Barot (2013), the experimental (validation) results were subsequently compared with the RSM-predicted outcomes and the relative error percentages were calculated by

$$\text{Relative Error} = \left| \frac{\text{Predicted value} - \text{Experimental value}}{\text{Predicted value}} \right| \times 100\%$$

2.6 Assessment of physicochemical properties

The assessment of the physicochemical properties was carried out on the tamarind juice, the mixed tamarind, i.e. tamarind juice mixed with carrier agents, the tamarind powder, i.e. tamarind powder produced by drum-drying, and the dissolved tamarind, i.e. tamarind powder dissolved with water in a ratio of 1:5 w/w (tamarind powder:water). All of the physicochemical measurements were carried out in triplicate.

2.6.1 Drying yield (DY)

The drying yield (DY) was determined using the dry solids weight ratio of the tamarind powder and mixed tamarind (Fazaeli, Emam-Djomeh, Ashtari, & Omid, 2012). The drying yield was estimated by equation (1).

$$Drying\ yield = \frac{M_a}{M_b} \times 100\% \quad (1)$$

where M_a and M_b are the weights (g) of the dry solids of tamarind powder and mixed tamarind, respectively.

2.6.2 Water activity (Aw)

The water activity of the tamarind powder was determined using a water activity meter (AquaLab, CX-3TE, USA) (Pua *et al.*, 2010).

2.6.3 Moisture content (MC) and total solids (TS)

The MC of the tamarind powder and TS of the mixed tamarind were determined using a convection oven (Shrivastav & Kumbhar, 2009), whereby the tamarind products were oven-dried at 105 ºC for 24 h. The MC and TS of the tamarind products were determined by equations (2) and (3).

$$Moisture\ content = \left(\frac{W_2 - W_3}{W_2 - W_1} \right) \times 100\% \quad (2)$$

$$Total\ solids = \left(1 - \frac{W_2 - W_3}{W_2 - W_1} \right) \times 100\% \quad (3)$$

where W_1 is the initial weight of a moisture can (g), W_2 is the weight of the moisture can with pre-heated tamarind products (g), and W_3 is the weight of the moisture can with post-heated tamarind products (g).

2.6.4 Viscosity (VS)

The viscosity of the mixed tamarind was determined using a modular compact rheometer (Anton Paar, MCR52, Austria) equipped with a spindle conic end concentric cylinder geometry with a rotor (CC24) and stainless steel cup (CC26) (Martinez-Flores, Garnica-Romo, Bermudez-Aguirre, Pokh rel, & Barbosa-Canovas, 2015). The analysis was carried out at 25±0.1 ºC with 20 mL of the mixed tamarind and the temperature was regulated using a Viscotherm VT 10 controlled by a Peltier system. The flow curves were in the 1-100s^{-1} shear rate range and taken every 1.01s^{-1}. The viscosity was calculated from the flow curve data of shear rates (s^{-1}) relative to shear stress (Pa) using the power law expression ($\tau = k\gamma^n$), and the results expressed as the flow behavior index (n).

2.6.5 Color value (ΔE)

The color of the tamarind products was determined using a colorimeter (Hunter Lab, ColorQuest XE, USA) for the total color change between the mixed tamarind and the dissolved tamarind. The color was expressed in terms of L (lightness), a (redness) and b (yellowness) (Shittu & Lawal, 2007). The change in the color was calculated by equation (4).

$$\Delta E = \sqrt{(L_0 - L_p)^2 + (a_0 - a_p)^2 + (b_0 - b_p)^2} \quad (4)$$

where L_0, a_0, and b_0 are the color values of the mixed tamarind and L_p, a_p, and b_p are the color values of the dissolved tamarind.

2.6.6 pH values (pH)

The pH of the dissolved tamarind was determined using a digital pH meter (Mettler Toledo, SevenEasy, Switzerland) (Zorba & Kurt, 2006).

2.6.7 Total acidity (TA)

TA was determined using a 250 mL Erlenmeyer flask containing 5 mL of the tamarind juice and dissolved tamarind with 10 mL of distilled water. Three drops of 1% phenolphthalein were added to the mixture as the indicator. The mixture was titrated with 0.1 N of NaOH until the endpoint, at which the solution color became light pink (Marikar & Wijerathnam, 2010). The total acidity was estimated by equation (5).

$$Total\ acidity = \frac{V \times N \times Eq.wt}{U \times 1000} \times 100\% \quad (5)$$

where V is the volume of NaOH in the titration until the endpoint (mL), N is the normality of NaOH, $Eq.wt$ is the equivalent weight of tartaric acid (=75), and U is the volume of the sample in the titration (mL).

2.6.8 Bulk density (BD)

To determine the BD of the tamarind powder, a cylinder of known volume was utilized whereby the cylinder was drop-filled with the tamarind powder (80 mesh) at a distance of 0.1 m from the cylinder. The drop-filling continued until the tamarind powder reached the cylinder rim (Goula & Adamopoulos, 2008). The BD was estimated by equation (6).

$$Bulk\ density = \frac{m}{v} \quad (6)$$

where m is the mass of tamarind powder (g) and v is the cylinder volume (mL).

2.6.9 Solubility (SO)

The tamarind powder solubility was determined by suspending 1 g of tamarind powder in 10 mL water at 30 ºC in a centrifuge tube. The suspension was stirred intermittently for 30 min prior to centrifugation at 3,000 rpm for 10 min. The supernatant was then transferred to a moisture can and oven-dried at 105 ºC for 24 h (Cano-Chauca, Stringheta, Ramos, & Cal-Vidal, 2005). The solubility of the tamarind powder was estimated by equation (7).

$$Solubility = \frac{M_s}{M_p} \times 100\% \quad (7)$$

where M_s is the dry solids weight of the supernatant (g) and M_p is the tamarind powder weight (g).

3. Results and Discussion

3.1 Model fitting

Table 3 tabulates the effects of variable MD-OSA-GA mixture combinations (the seven mixture formulations) on the physicochemical characteristics (DY, Aw, MC, TS, VS, ΔE, pH, TA, BD, SO) of the experimental tamarind products. The results were the inputs for regression analysis. In the analysis, the independent variables (MD, OSA, GA) and response variables (the physicochemical properties) were tested for the goodness of fit using both linear and interaction terms.

Table 4 tabulates the P-values of the response variables, i.e. physicochemical properties, in the linear and interaction terms and the LOF. The analysis results revealed that TS, VS, pH, and TA were statistically significant ($P \leq 0.05$) and exhibited very high R^2 ($R^2 > 0.9$), i.e. 0.92 for TS, 0.96 for VS, 0.99 for pH, and 0.98 for TA, and the corresponding LOF P-values greater than 0.05, i.e. 0.817, 0.255, 0.789, and 0.713, respectively. Meanwhile, DY, Aw, MC, ΔE, BD, and SO exhibited low R^2 and LOF P-values despite being statistically significant.

In this research, the optimal MD-OSA-GA mixture combination was that which yielded tamarind powder with TS, VS, pH, and TA values closely resembling the fresh tamarind pulp. The ideal TS, VS, pH, and TA values are 40.0% (Phomkong, Ekpong, & Onsaard, 2008), 0.27 (Kechinski, Schumacher, Marczak, Tessaro, & Cardozo, 2011), 3.00 (Phomkong *et al.*, 2008), and 9.33% (Muzaffar & Kumar, 2016), respectively.

In Table 5, the regression coefficients (β) associated with the linear and interaction terms revealed that the carrier agents (X_1, X_2, X_3) significantly and positively influenced the TS, VS, pH, and TA of the tamarind powder and the blended carrier agents (X_1X_2, X_1X_3, X_2X_3) were either significantly and positively or negatively correlated with the four attributes ($P \leq 0.05$).

Table 3. Physicochemical properties of the tamarind products associated with the experimental MD-OSA-GA mixture formulations.

Physicochemical properties (Response variables)	Mixture formulations						
	1	2	3	4	5	6	7
DY (%)	87.35±0.22	85.33±0.21	78.62±0.20	78.65±0.13	82.10±0.22	77.67±0.14	74.97±0.03
Aw (-)	0.21±0.01	0.22±0.01	0.19±0.01	0.23±0.01	0.23±0.03	0.18±0.02	0.20±0.01
MC (%)	3.07±0.04	2.75±0.06	2.77±0.05	2.79±0.01	2.77±0.01	2.43±0.04	3.01±0.01
TS (%)	40.15±0.09	39.92±0.09	39.59±0.11	39.81±0.06	39.78±0.11	40.46±0.08	39.62±0.01
VS (-)	0.19±0.01	0.26±0.01	0.27±0.01	0.33±0.01	0.36±0.01	0.35±0.01	0.29±0.01
ΔE (-)	9.12±0.02	7.50±0.02	10.26±0.02	8.21±0.01	4.65±0.01	4.56±0.01	4.22±0.02
pH (-)	3.33±0.01	3.07±0.01	3.13±0.01	2.99±0.01	3.02±0.01	3.17±0.01	3.04±0.01
TA (%)	8.43±0.05	9.21±0.05	9.09±0.01	9.48±0.05	9.39±0.05	9.03±0.05	9.30±0.05
BD (g/mL)	0.77±0.01	0.64±0.01	0.77±0.01	0.70±0.01	0.70±0.01	0.76±0.01	0.76±0.01
SO (%)	81.14±1.27	81.47±0.31	78.73±0.65	81.39±0.86	78.92±0.71	78.63±0.96	80.07±0.31

Table 4. P-values of the response variables in the linear and interaction terms and the lack-of-fit (LOF) P-values.

Source	df	P-value									
		DY (%)	Aw (-)	MC (%)	TS (%)	VS (-)	ΔE (-)	pH (-)	TA (%)	BD (g/mL)	SO (%)
Regression	5	0.001	0.006	0.000	0.000	0.000	0.000	0.000	0.000	0.001	0.000
Linear	2	0.080	0.991	0.001	0.000	0.000	0.001	0.000	0.000	0.021	0.404
Interaction	3	0.013	0.198	0.003	0.000	0.086	0.000	0.000	0.000	0.012	0.123
X_1X_2	1	0.926	0.660	0.129	0.263	0.469	0.009	0.000	0.000	0.003	0.446
X_1X_3	1	0.002	0.356	0.099	0.000	0.621	0.009	0.000	0.000	0.163	0.089
X_2X_3	1	0.407	0.066	0.001	0.000	0.019	0.002	0.000	0.001	0.765	0.111
Lack-of-fit	1	0.000	0.269	0.000	0.817	0.255	0.000	0.798	0.713	0.000	0.327

Table 5. Regression coefficients of the response variables under the linear and interaction terms.

Response variable	Linear terms			Interaction terms			R^2
	X_1	X_2	X_3	X_1X_2	X_1X_3	X_2X_3	
DY (%)	1.09	0.48	1.83	-7.09×10^{-4}	-2.83×10^{-2}*	6.42×10^{-3}	0.62
Aw (-)	2.48×10^{-3}	2.75×10^{-3}	2.33×10^{-3}	-2.05×10^{-5}	-4.34×10^{-5}	9.06×10^{-5}	0.50
MC (%)	3.44×10^{-2}*	3.66×10^{-2}*	-2.14×10^{-2}*	-4.37×10^{-4}	4.79×10^{-4}	1.09×10^{-3}*	0.78
TS (%)	0.42*	0.40*	0.57*	-2.76×10^{-4}	-3.21×10^{-3}*	-1.60×10^{-3}*	0.92
VS (-)	1.19×10^{-3}*	5.81×10^{-3}*	6.08×10^{-3}*	-2.24×10^{-5}	-1.52×10^{-5}	7.91×10^{-5}*	0.96
ΔE (-)	9.89×10^{-2}*	0.71*	-0.43*	-1.02×10^{-2}*	1.02×10^{-2}*	-1.27×10^{-2}*	0.75
pH (-)	3.93×10^{-2}*	4.17×10^{-2}*	5.34×10^{-2}*	-4.01×10^{-4}*	-5.20×10^{-4}*	-2.83×10^{-4}*	0.99
TA (%)	6.59×10^{-2}*	6.64×10^{-2}*	2.81×10^{-2}*	1.11×10^{-3}*	1.58×10^{-3}*	5.80×10^{-4}*	0.98
BD (g/mL)	8.79×10^{-3}*	1.99×10^{-2}*	6.07×10^{-4}*	-3.02×10^{-4}*	1.24×10^{-4}	-2.58×10^{-5}	0.64
SO (%)	0.83	0.76	0.97	1.82×10^{-3}	-4.23×10^{-3}	-3.94×10^{-3}	0.67

X_1, X_2, X_3 denote MD, OSA,GA, respectively;X_1X_2, X_1X_3, X_2X_3 denote the interaction terms for MD-OSA, MD-GA, and OSA-GA, respectively.
* The mean difference is significant at $P\leq0.05$.

Moreover, the regression models associated with the various physicochemical characteristics, i.e. response variables, were tested for reliability and validity (Homkhiew, Rawangwong, & Boonchouytan, 2015). As an example, Figure 2 demonstrates the reliability and validity tests and the predictive ability of the TS model. Specifically, Figure 2a illustrates the probability of the model residuals and the results indicate that the data is normally distributed. Figure 2b depicts the model residuals associated with the 21 experimental runs, i.e. seven MD-OSA-GA formulations experimented in triplicate, and the graph reveals no correlation between the two which indicated that the data were independent. Meanwhile, Figure 2c shows the model residuals relative to the predicted TS and the results indicate the variances are constant. Figure 2d plots the observed TS values (actual) against the predicted values and the findings revealed a high predictive power of the TS model. In sum, Figures 2a-d confirmed the validity and reliability of the TS prediction model (Homkhiew & Ratanawilai, 2014). More importantly, this was true also for the other three response variables of VS, pH, and TA.

3.2 Physicochemical properties of the tamarind products

Figure 3a illustrates the TS variations relative to the variable concentrations of MD, OSA, and GA. The illustration revealed that the TS of the mixed tamarind, i.e. tamarind juice mixed with carrier agents, and the carrier agents were positively correlated. According to Bhandari *et al.* (1997),

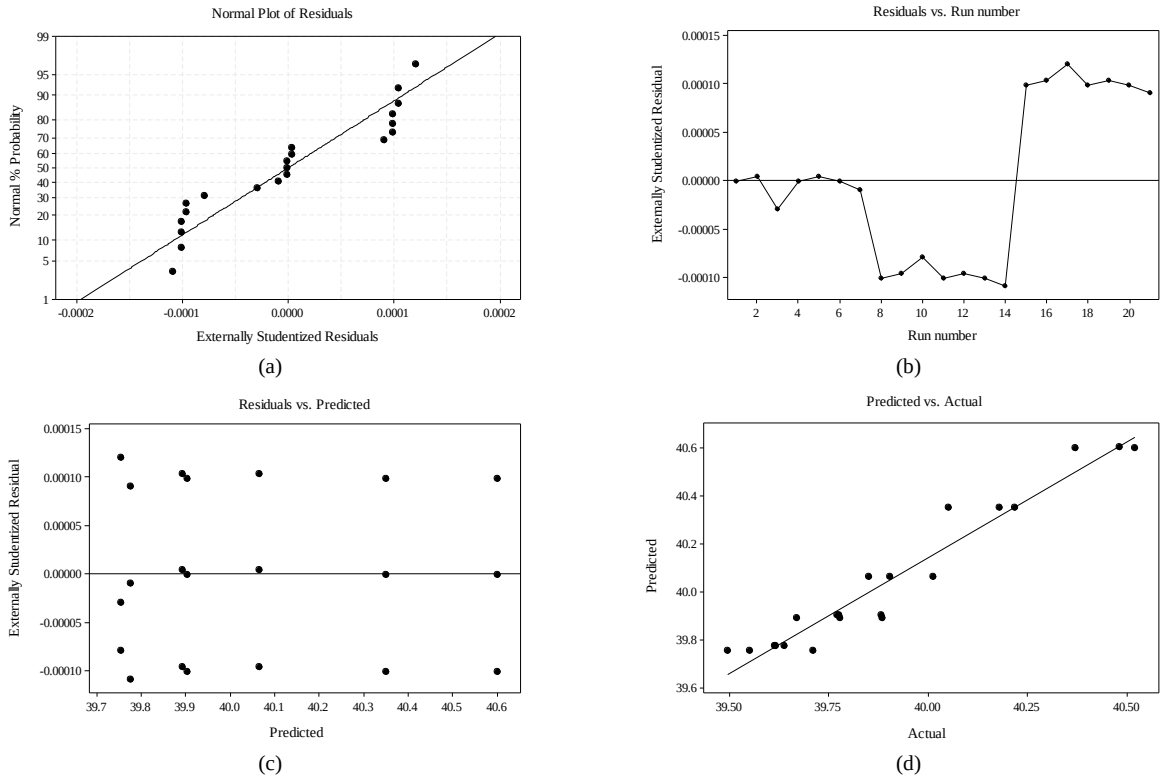

Figure 2. Reliability and validity tests of the regression model for, as an example, total solids (TS): (a) the probability of residuals, (b) the model residuals associated with the 21 experimental runs, (c) the residuals relative to the predicted TS, (d) the observed vs. the predicted TS values.

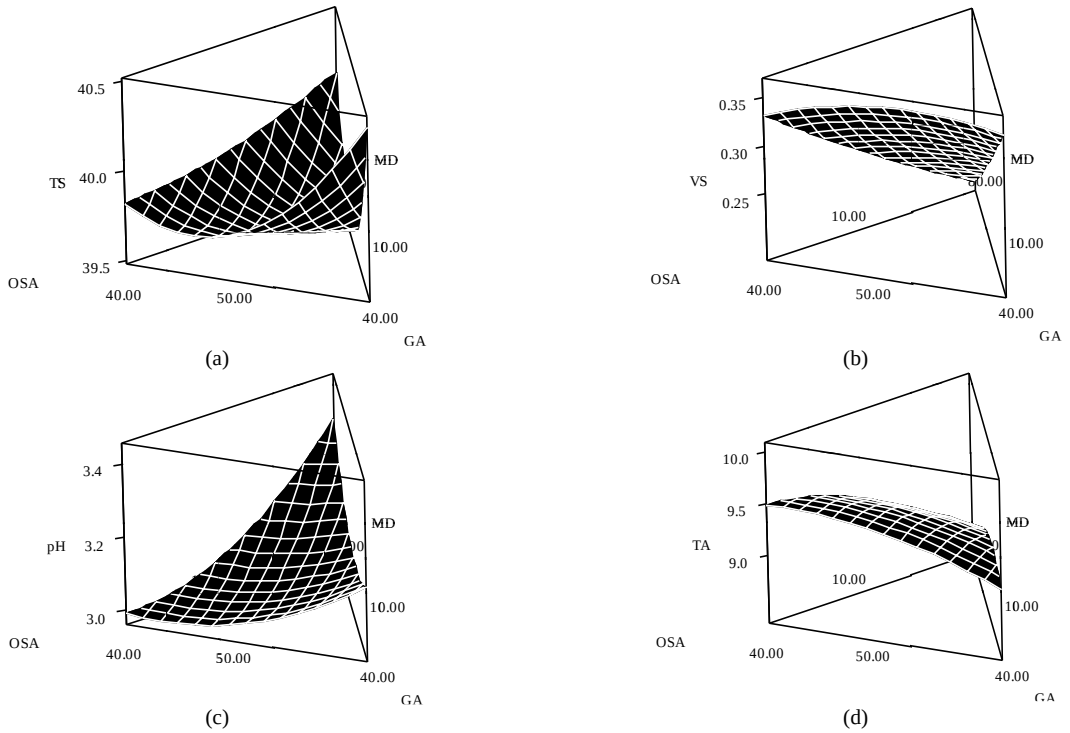

Figure 3. Response surface plots of the physicochemical characteristics of the drum-dried tamarind powders relative to the MD-OSA-GA mixture concentrations: (a) total solids (TS), (b) viscosity (VS), (c) pH values (pH), and (d) total acidity (TA).

Silva, Sobral, and Kieckbusch (2006), Carneiro *et al.* (2013), and Fernandes et al. (2008), the introduction of carriers into the fruit juice mitigated the stickiness during the drying process. The findings could be attributed to the carrier-induced incremental total solids and the subsequent increased molecular weight and glass transition temperatures. In this research, the TS of the mixed tamarind (tamarind juice mixed with the carrier agents) was positively significant under the linear terms (X_1, X_2, X_3) and negatively significant under the interaction terms for the MD-GA (X_1X_3) and OSA-GA (X_2X_3) combinations ($P \leq 0.05$), where X_1, X_2, and X_3 denoted MD, OSA, and GA, respectively (Table 5).

In Figure 3b, the VS of the mixed tamarind was positively correlated with the OSA and GA concentrations and negatively associated with MD. The viscosity of the mixed tamarind was expressed as the flow behavior index (n), which was in the range of 0.19-0.36. Given n<1, the mixed tamarind was a non-Newtonian fluid which is indicative of pseudo-plasticity. The pseudoplastic behavior is a characteristic of tomato ketchup (Koocheki, Ghandi, Razavi, Mortazavi, & Vasiljevic, 2009). In fact, the viscosity of the mixed tamarind, i.e. tamarind juice mixed with carrier agents, was positively significant under the linear terms (X_1, X_2, X_3). The correlation was positively significant under the interaction terms for the OSA-GA (X_2X_3) combinations ($P \leq 0.05$) (Table 5).

Figures 3c-3d illustrate the variations in pH and TA relative to the variable MD, OSA, and GA concentrations. The findings revealed that the MD and GA concentrations were positively correlated with pH of the mixed tamarind and negatively correlated with TA. Meanwhile, the increased OSA contributed to a lower pH and higher TA, which is desirable. Due to the low boiling points of the volatile organic compounds (VOCs) responsible for aroma and flavor (typically lower than that of water) according to Pua, Hamid, Rusul, & Rahman (2007), the VOCs are often lost during drying which increases the pH value and lowers the total acidity. OSA is thus normally used in the formulation for its flavor-encapsulating function. In fact, the pH of the mixed tamarind was positively significant under the linear terms (X_1, X_2, X_3) and negatively significant under the interaction terms for the MD-OSA (X_1X_2), MD-GA (X_1X_3), and OSA-GA (X_2X_3) combinations ($P \leq 0.05$). Meanwhile, TA was positively significant under the linear terms (X_1, X_2, X_3) and the interaction terms for the MD-OSA (X_1X_2), MD-GA (X_1X_3), and OSA-GA (X_2X_3) combinations ($P \leq 0.05$) (Table 5).

3.3 Optimization and validation of the mixed tamarind formulation

An ideal mixed tamarind formulation, i.e. tamarind juice mixed with the carrier agents, is a mixture that would yield a tamarind powder whose physicochemical characteristics closely resemble the fresh tamarind pulp, i.e. low ΔE, MC, Aw, pH, and BD and high TS, VS, TA, and SO. To obtain the optimal mixed tamarind formulation, graphical optimization is normally used whereby the surface plots with specific boundaries of various physicochemical attributes are superimposed and the overlapped region in which the physicochemical properties of the final product closely resemble the fresh tamarind pulp (Arteaga, Li-Chan, Arteaga, & Nakai, 1994).

In this research, given the very high R^2 of TS (0.92), VS (0.96), pH (0.99), and TA (0.98) ($P \leq 0.05$) and their corresponding high LOF P-values, i.e. 0.817, 0.255, 0.789, 0.713, respectively, ($P > 0.05$), the graphical optimization was thus carried out in terms of the TS, VS, pH, and TA attributes. Figure 4 illustrates the graphical optimization to determine the optimal mixture of MD, OSA, and GA given the drum drying process that yields the tamarind powder whose physico-chemical properties of TS, VS, pH, and TA closely resemble the fresh tamarind pulp. The results represented by the white area in Figure 4 indicate that the optimal concentrations of MD, OSA, and GA were 61.52-77.79 g, 11.96-28.48 g, and 10.00-33.18g, respectively, the sum of which must always equals 100 g.

In general, MD, OSA, and GA contributed to the improved physicochemical characteristics of the tamarind powder. However, MD increased the pH and lowered the TA which affected the quality of the tamarind powder and gave rise to the adoption of the lower limit of MD (61.52 g/100g). Meanwhile, due to the efficient flavor-encapsulating property of OSA, its upper limit of 28.48 g/100g was implemented to preserve the pH value and TA. The lower limit of GA (10.00 g/100g) was adopted due to its relatively high cost. The RSM-predicted optimal mixture of 61.52 g, 28.48 g, and 10.00 g for MD, OSA, and GA, respectively, would produce the tamarind powder with 39.88% TS, 0.27 VS, 3.04 pH, and 9.31% TA.

Furthermore, to validate the predictive ability of the proposed TS, VS, pH, and TA models, three actual experiments were carried out using the optimal MD-OSA-GA combination of 61.52 g, 28.48 g and 10.00 g/100g for MD, OSA, and GA, respectively, and the measurements were averaged. The experimental results were subsequently compared with the RSM-predicted outcomes and the relative error percentages were calculated. In Table 6, the differences in the TS, VS, pH, and TA relative errors were 3.34%, 3.70%, 1.32%, and 1.39%, respectively. Since these percentages were below 5%, a high predictive power of the models was indicated.

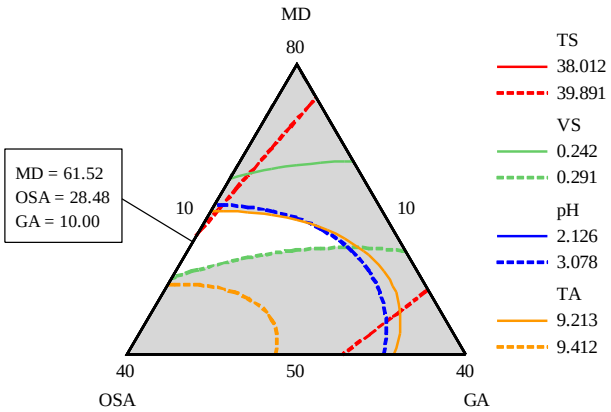

Figure 4. The optimal region of the carrier-mixture concentrations with four physicochemical attributes (TS, VS, pH, TA) of the final tamarind powder closely resembling the fresh tamarind pulp, where MD, OSA, GA denote maltodextrin, octenyl succinic anhydride, and gum arabic, respectively.

4. Conclusions

This research investigated the collective use of MD, OSA, and GA of varying concentrations using a double-drum dryer to produce tamarind powders. Seven carrier-mixture formulations were specified using a simplex lattice mixture design. Assessments of the physicochemical characteristics (DY, Aw, MC, TS, VS, ΔE, pH, TA, BD, and SO) were performed on the tamarind products with various MD-OSA-GA combinations. Regression analysis was carried out to determine the statistical relationships between the carrier agents and the physicochemical properties. The analysis results revealed that TS, VS, pH, and TA were statistically significant ($P \leq 0.05$) and exhibited very high R^2 and LOF P-values. Meanwhile, DY, Aw, MC, ΔE, BD, and SO exhibited low R^2 and LOF P-values, despite being statistically significant. Moreover, graphical optimization was used to identify the optimal mixture of the carrier agents that would produce a tamarind powder whose physicochemical characteristics closely resembled the fresh tamarind pulp. Given the very high R^2 and the LOF P-values ($p>0.05$) of TS, VS, pH, and TA, graphical optimization was carried out in terms of the TS, VS, pH, and TA attributes. The RSM-predicted optimal mixture concentrations were 61.52 g, 28.48 g, and 10.00 g/100g for MD, OSA, and GA, respectively, with the resulting tamarind powder exhibiting 39.88% TS, 0.27 VS, 3.04 pH, and 9.31% TA.

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