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STUDY ON COLOUR COORDINATES AND MAILLARD PRECURSORS OF RE-FABRICATED RICE

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ABSTRACT

The effect of feed moisture, screw speed and barrel temperature on colour coordinates of re-fabricated rice grains were investigated in a co-rotating twin-screw extruder using response surface methodology. The rice flour obtained from broken rice (< 1/8th of actual kernel size) of cv. 'PR-116' was used in the study. The screw speed was set at five levels between 49 and 150 rpm, barrel temperature between 59 and 110°C and feed moisture between 31 and 45%. All colour coordinates of re-fabricated grains evaluated - L* (brightness), a* (redness), b* (yellowness), c* (chroma), ΔE (total colour difference) and hue angle were significantly affected by the three process variables. Barrel temperature was the most significant variable with quadratic effect on a*and ΔE. Response surface regression models were established to correlate the colour coordinates of re-fabricated rice grains to the process variables. The optimized extrusion conditions for preparation of re-fabricated grains were 38%, moisture content, 130 rpm screw speed and 89°C barrel temperature. A significant change was observed in maillard precursors i.e. sucrose, glucose, fructose and maltose levels of re-fabricated rice obtained at optimum extrusion conditions in comparison to natural rice (cv. 'PR-116').

Key words: Barrel temperature, colour coordinates, extrusion, re-fabricated rice, response surface methodology, screw speed

INTRODUCTION

Extrusion technology is a continuous process involving simultaneous mixing, shearing and cooking, in which the raw-material is subjected to various changes. Notable amongst these changes are starch and protein hydration, homogenization, starch gelatinization, fat liquefaction, protein denaturation, destruction of antinutritional factors, enzyme inactivation, plasticizing and expansion of the processed material. With little or no modification of basic equipment and with appropriate process control, it is possible to produce a great variety of marketable food products. Earlier studies were focused mainly on producing specific food products using single-screw extruders (Owusu-Ansah *et al*., 1983). Research on twin-screw extrusion of starches and cereal flours has shown that extrusion variables and ingredients markedly alter the quality characteristics of the finished product (Ryu *et al*., 1993; Manisha *et al*., 1998). Various expanded snacks, modified starch, ready-to-eat cereal foods have been produced and the relationship between process variables and product quality characteristics studied (Altan *et al*., 2009; Baljit *et al*., 2014). The extrusion appears to be a simple technological process for the development of re-fabricated rice, but the control of finished product characteristics is rather complicated. This is because of the complex nature of raw-material and large number of variables involved in the process (El-Dash *et al.*, 1983). Therefore, the relationship between process variables and quality characteristics of re-fabricated rice needs to be studied in detail.

The physical appearance of re-fabricated rice is important for popularity, and is a vital index governing the preference. Colour, a basic quality requirement, can be a useful trait for the acceptability of re-fabricated rice among the people in developing countries who seldom care for hidden quality attributes. Maillard reactions leading to the change in colour occur during high temperature treatment and storage of food products (Villamiel *et al.,* 2006). Martin and Van Boekel (2005) studied the kinetics of Maillard reaction pathways in glucose-glycine model systems and observed increased fructose formation with increase in temperature. The isomerization of maltose to maltulose is very common in cereals, when subjected to heat processing (Garcia *et al.,* 2004). The present study was thus aimed to investigate the effect of extrusion process variables on colour coordinates of re-fabricated rice and to optimize processing conditions for the production of re-fabricated rice with a close resemblance to natural rice. Further, regression models were established to predict the colour coordinates of re-fabricated rice as a function of process variables. The impact of optimized extrusion conditions on Maillard precursors was also assessed.

MATERIALS AND METHODS

Small rice brokens ($\leq 1/8^{th}$ of actual kernel size) of variety 'PR116' were ground in a lab mill model 3303 (Perten, Sweden) to fineness that passes through 200 µm sieve. The moisture content was determined by oven drying method (AACC, 2000) and appropriate amount of water was added to adjust the required moisture content of flour as per the experimental design. Extruded rice kernels were collected on stainless steel mesh trays. Trays were loaded into a tray dryer (S-Narang, Scientific Works, New Delhi, India) with a small fan and 220 volt Thermolyne© heating unit circulating air from the bottom of cabinet to the top. The re-fabricated rice grains were then pre-dried for 2-2.5 hr at 70°C. The partially dried re-fabricated grains were stacked in trays and placed in conditioning chamber for 8 hr for final drying at 60-70°C to reach a water content of 10-12 g 100 g^{-1}. Dried samples were stored in air tight plastic containers at room temperature and used for analysis.

Extruder and processing conditions

Extrusion was performed on a co-rotating, intermeshing twin-screw extruder Model BC 21 (Clextral, Firminy, France). The barrel diameter and its length to diameter ratio (L/D) were 2.5 mm and 16:1, respectively. The extruder had 4 barrel zones. Temperature of the 1^{st} , 2^{nd} and 3^{rd} zone were maintained at 20, 30 and 40°C, respectively, throughout the study period; while the temperature in last zone (compression and die section) was varied according to the experimental design. The screw configuration used in the entire experiment is shown in Fig. 1 and Table 1. The extruder was equipped with a torque indicator which showed percent of torque in proportion to the current drawn by the drive motor. Raw material was metered into the extruder with a single screw volumetric feeder. The extruder was thoroughly calibrated with respect to the combinations of feed rate and screw speed to be used. The feed rate was varied for optimum filling of extruder barrel corresponding to the screw speed. The moisture content of feed was varied by injecting water into extruder with water pump. A specially designed cutter with four bladed knives and a die (8 × 1.5 mm dimension), made of stainless steel were used for shaping of the re-fabricated rice grains.

Experimental design

A central composite rotatable design (CCRD) (Draper, 1982) was used to incorporate three independent extrusion variables *viz.,* barrel temperature, screw speed and feed moisture. This design required 20 experimental runs with eight (2^3) factorial points (3 levels for each variables), six star corner points (2 for each variable) and six centre pints. This yielded five levels for each variable

Table 1: Screw configuration in different sections of the extruder (from hopper to die)

Screw section	1	2	3	4	5	6	7	8	9	10
Screw elements*	BAGUE	C2F	C2F	C2F	C2F	C2F	INO0	CIF	CIF	CIF
Length (mm)	20	50	50	50	50	50	5	50	50	25
Pitch (mm)	-	50	33.33	25	25	16.66	-	16.66	12.5	12.5

*Screw elements: BAGUE = Ring, C2F = Conjugated section 2 threads, INO0 = Interface ring between C2F and C1F, CIF = Conjugated section, 1 thread

Fig. 1: Screw configuration

and permitted better assessment of their quadratic effects. The CCRD-coded levels and experimental ranges of 3 independent variables (Table 2) were selected on the basis of preliminary tests.

Colour analysis

The colour measurements of rice grains were performed with a Hunterlab colorimeter (model CM-508d Minolta, Japan) as described by Lambert *et al.* (2006). Each measurement was replicated 10 times and average values considered. Total colour difference (ΔE), chroma values (c*) and Hue angle (°) were calculated from the colour parameters L^* (brightness) a^* (redness) and b^* (yellowness) using

$$\Delta E = \sqrt{(L-L_0)^2+(a-a_0)^2+(b-b_0)^2}$$

$$\text{Chroma } (c^*) = \sqrt{a^2+b^2}$$

$$\text{Hue angle } (^\circ) = \tan^{-1}\left(\frac{b}{a}\right)$$

The control (PR-116) was used as reference to calculate ΔE-values.

Statistical analysis

Responses obtained as a result of the proposed experimental design were subjected to regression analysis to assess the effects of moisture content, screw speed and barrel temperature. Second order polynomial regression models were established for dependent variables to fit experimental data for each response using statistical software Design-Expert 8 (Stat-Ease Inc, Minneapolis, MN, USA).

$$y_i = b_0 + \sum_{i=1}^{3} b_i x_i + \sum_{i=1}^{3} b_{ii} x_i^2 + \sum_{i=1}^{3}\sum_{i=1}^{3} b_{ij} x_i x_j$$

x_i, x_i^2 and $x_i x_j$ are linear, quadratic and interaction effect of input variables which influence the response y, respectively, and b_o, b_i, and b_{ij} were the regression coefficients to be determined. Data was analyzed by multiple regression analysis and analysis of variance (ANOVA) for each response. The adequacy of regression model was checked by correlation coefficients. The lack of fit test was used to judge the adequacy of model fit. To aid visualization of variation in responses with respect to processing variables, series of three dimensional response surfaces plots were drawn.

Optimization

Optimization maximizes a desired quantity or minimizes undesired one. The values of processing variables that produce the desired optimum values are called optimum conditions. The samples were organoleptically evaluated for grain appearance score, based on colour, shape, size, appearance and overall acceptability through a panel of semi-trained judges (n = 40) using 9 point hedonic scale (Larmond, 1970). Of the 20 samples, 8 samples with grain appearance score >7 were selected (Table 2). The range of values for L* (brightness), a* (redness), b* (yellowness), c* (chroma), ΔE (total colour difference) and hue angle were used as criteria to determine optimum extrusion conditions. The response surface of desirability function was used for numerical optimization.

Levels of mono- and di-saccharides

Defatting of rice flour (0.1 g), made from both natural rice (cv. PR-116) and re-fabricated rice grains obtained at optimized extrusion conditions was done with 3.0 mL hexane. After shaking for 15 min., the suspension was centrifuged at 3000 g for 10 min. The pellets were dried under a stream of nitrogen gas. The extraction was done with deionised water (5.0 mL) for 30 min. at 6°C. The suspension was centrifuged at 3000 g for 15 min. The levels of mono- and di-saccharides in diluted supernatants were determined by high performance anion-exchange chromatography with integrated pulsed amperometric detection. All analysis was done in triplicate.

RESULTS AND DISCUSSION

Colour analysis

The data on mean values of colour coordinates of extruded rice grains (Table 2) revealed that the regression, models for L*, a*, b*, c*, ΔE and hue angle were highly significant ($P < 0.01$) with a high correlation coefficient (R^2 = 0.98, 0.99, 0.99, 0.99, 0.99 and 0.98, respectively). None of the models showed significant lack of fit, indicating that all the 2nd order polynomial models correlated well with the measured data. The predicted R-square was found in reasonable agreement with adjusted R-square for all the parameters. Adequate precision compares the range of predicted values at the

Table 2: Effect of extrusion conditions on color coordinates

Run	Extrusion conditions[a] Actual and coded values%			Product responses[b]						GAS*
	Moisture content (%)	Screw speed (rpm)	Temperature (°C)	L* mean ± SE	a* mean±SE	b* mean ± SE	c* mean ± SE	ΔE mean ± SE	Hue angle (°) mean ± SE	
1.	34.0 (-1)	70.0 (-1)	70.0 (-1)	72.2±1.04	2.5±0.10	15.1±0.48	15.3±0.36	8.0±0.12	80.59±1.52	6.4
2.	42.0 (+1)	70.0 (-1)	70.0 (-1)	71.2±1.28	2.8±0.07	15.9±0.45	16.14±0.26	9.2±0.23	80.01±1.00	6.6
3.	34.0 (-1)	130.0 (+1)	70.0 (-1)	72.9±2.06	2.3±0.09	14.8±0.28	14.97±0.21	7.3±0.25	81.16±0.98	6.7
4.	42.0 (+1)	130.0 (+1)	70.0 (-1)	71.6±1.02	2.7±0.09	15.7±0.26	15.93±0.04	8.8±0.70	80.24±1.14	6.4
5.	34.0 (-1)	70.0 (-1)	100.0 (+1)	63.9±1.89	5.6±0.26	18.8±0.19	19.61±0.37	17.5±0.26	73.41±1.25	6.5
6.	42.0 (+1)	70.0 (-1)	100.0 (+1)	60.2±1.21	6.2±0.30	19.3±0.15	20.27±0.99	21.0±0.75	72.19±0.94	6.4
7.	34.0 (-1)	130.0 (+1)	100.0 (+1)	64.3±1.09	5.4±0.20	18.4±0.25	19.17±0.08	16.9±0.23	73.64±1.00	7.3
8.	42.0 (+1)	130.0 (+1)	100.0 (+1)	62.4±1.24	6.0±0.07	19.0±0.09	19.92±0.04	18.9±0.35	72.47±1.32	6.7
9.	31.3 (1.68)	100.0 (0)	85.0 (0)	69.0±1.64	3.3±0.09	16.3±0.10	16.63±0.12	11.4±0.50	78.55±1.41	6.5
10.	44.7 (+1.68)	100.0 (0)	85.0 (0)	66.7±1.33	4.6±0.10	17.9±0.16	18.48±0.09	14.4±0.31	75.58±1.14	6.4
11.	38.0 (0)	49.5 (-1.68)	85.0 (0)	67.2±0.95	4.1±0.10	17.6±0.25	18.07±0.19	13.7±0.21	76.88±1.56	6.8
12.	38.0 (0)	150.4 (+1.68)	85.0 (0)	68.8±1.45	3.8±0.07	16.7±0.36	17.12±0.13	11.9±0.72	77.18±1.45	7.8
13.	38.0 (0)	100.0 (0)	59.8 (-1.68)	74.4±0.63	2.0±0.12	14.2±0.47	14.34±0.44	5.8±0.53	81.89±1.45	6.0
14.	38.0 (0)	100.0 (0)	110.2 (+1.68)	59.4±1.86	6.8±0.07	20.4±0.95	21.5±0.76	22.3±1.58	71.56±0.94	5.5
15.	38.0 (0)	100.0 (0)	85.0 (0)	67.3±1.44	3.8±0.09	16.9±0.46	17.3±0.32	13.2±1.18	77.32±1.38	7.7
16.	38.0 (0)	100.0 (0)	85.0 (0)	68.6±0.68	4.0±0.06	17.6±0.19	18.0±0.22	12.5±0.83	77.19±1.07	7.3
17.	38.0 (0)	100.0 (0)	85.0 (0)	67.5±0.74	3.9±0.12	17.4±0.23	17.83±0.09	13.3±0.30	77.37±0.99	7.4
18.	38.0 (0)	100.0 (0)	85.0 (0)	67.7±2.19	4.1±0.11	17.2±0.30	17.68±0.25	13.1±0.22	76.59±0.94	7.5
19.	38.0 (0)	100.0 (0)	85.0 (0)	68.2±0.96	4.0±0.10	17.3±0.55	17.75±0.39	17.7±0.20	76.98±2.26	7.2
20.	38.0 (0)	100.0 (0)	85.0 (0)	68.0±1.12	3.9±0.10	17.1±0.32	17.54±0.17	12.8±0.13	77.15±1.99	7.6
Control		-		62.88±0.77	2.25±0.15	8.32±0.13	8.62±0.08	-	74.86±1.82	8.4

[a]Coded values of extrusion conditions are in parenthesis; [b]Values of product responses are Means ± standard error of means; *GAS = Grain appearance score

design points to average the prediction error. The adequate precision value of >4 indicated adequate model discrimination. All the parameters showed highly desirable adequate precision (Table 3). Table 2 shows the colour parameters - L*, a*and b* and c* of re-fabricated rice obtained under different extrusion conditions. It also lists the hue angle and total colour different (ΔE) between natural rice (cv. PR-116) and re-fabricated rice grains. The brightness of re-fabricated rice ranged from 59.4 to 74.4, whereas redness and yellowness were in the range of 2.0 to 6.8 and 14.2 to 20.4 respectively. These differences in colour parameters between rice grains prepared under different extrusion conditions were readily observable also by the human eye. The differences in redness and yellowness were also reflected in the values of c*. The fitted models for colour coordinates shown in equation (i) to (vi) (all independent variables in coded values), indicated quadratic effects with barrel temperature for redness and total colour difference (ΔE) only.

$$L^* = 67.57 - 0.862M + 0.468S - 4.564\,T \quad \text{(i)}$$

$$a^* = 3.95 + 0.299M - 0.088S + 1.535\,T + 0.176\,T^2 \quad \text{(ii)}$$

$$b^* = 17.18 + 0.402\,M - 0.199\,S + 1.788\,T \quad \text{(iii)}$$

$$c^* = 17.68 + 0.463\,M - 0.214\,S + 2.099\,T \quad \text{(iv)}$$

$$\Delta E = 12.92 + 0.969\,M - 0.499S + 5.034\,T + 0.436\,T^2 \quad \text{(v)}$$

$$\text{Hue angle} = 76.89 - 0.650\,M + 0.133S - 3.490\,T \quad \text{(vi)}$$

Fig. 2 to 7 shows the response surface plot of different colour coordinates (L*, a*, b*, c*, ΔE and hue angle) v/s two independent variables at a time. The barrel temperature was the most significant factor affecting all the colour coordinates. The effect of temperature on colour parameters of extruded rice was more pronounced for brightness. The negative coefficient of linear term of temperature level (equation i) indicated that brightness decreases with an increase in barrel temperature. The rice obtained at highest extrusions temperature (110°C) was least bright (L* = 59.4). In contrast, the rice obtained at lowest extrusion temperature (59°C) was most bright (L* = 74.4) (Table 2). Similar results were reported by other workers (Kimura *et al.*, 1993; Bhattacharya, 1996; Islam *et al.*, 2003) for raw and parboiled rice.

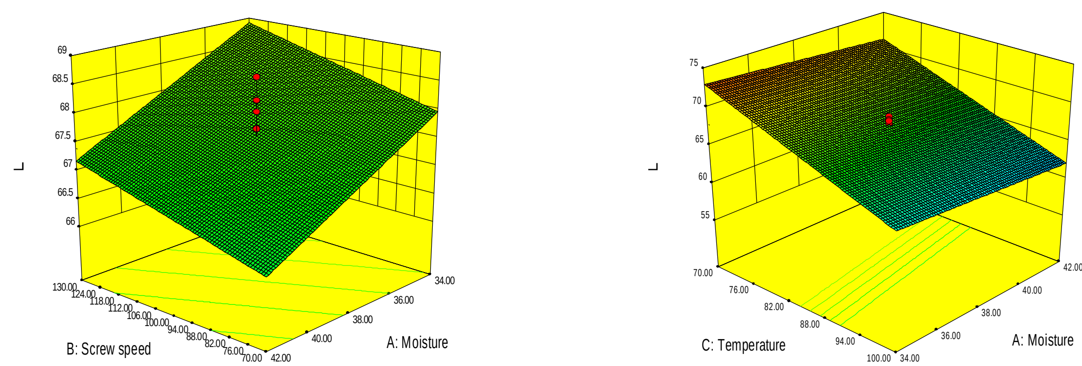

Fig. 2: Effect of moisture, screw speed and temperature on L* value

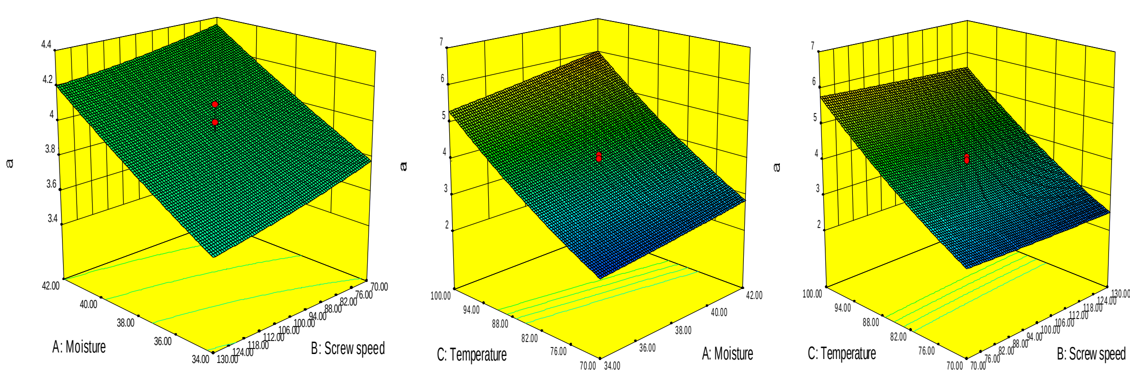

Fig. 3: Effect of moisture, screw speed and temperature on a* value

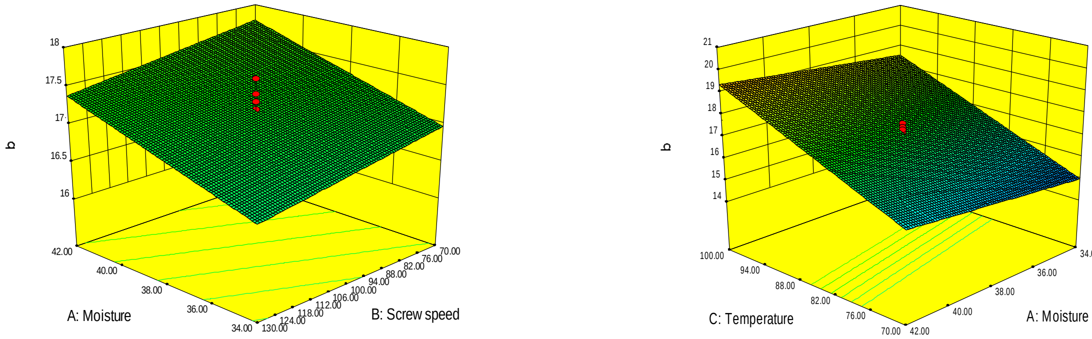

Fig. 4: Effect of moisture, screw speed and temperature on b* value

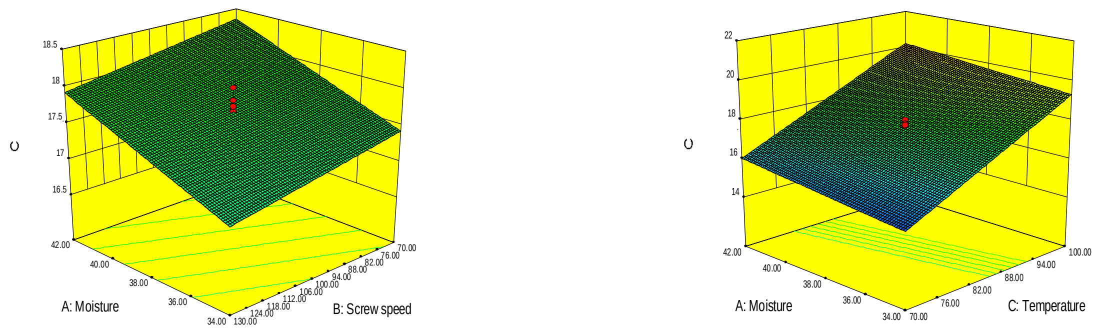

Fig. 5: Effect of moisture, screw speed and temperature on c* value

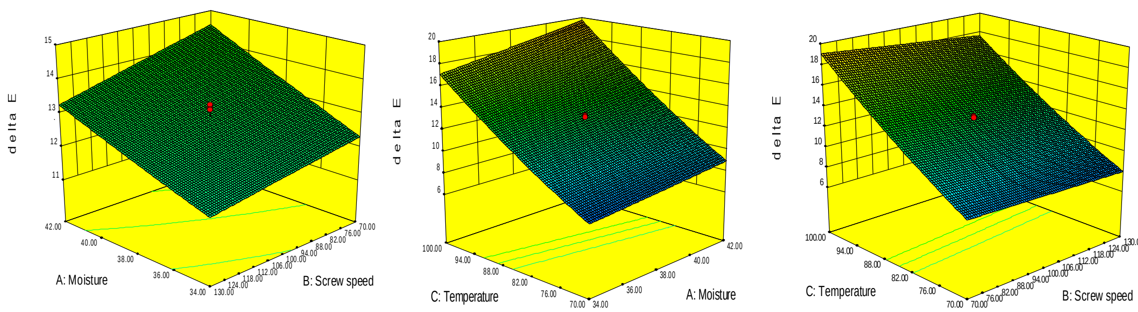

Fig. 6: Effect of moisture, screw speed and temperature on ΔE

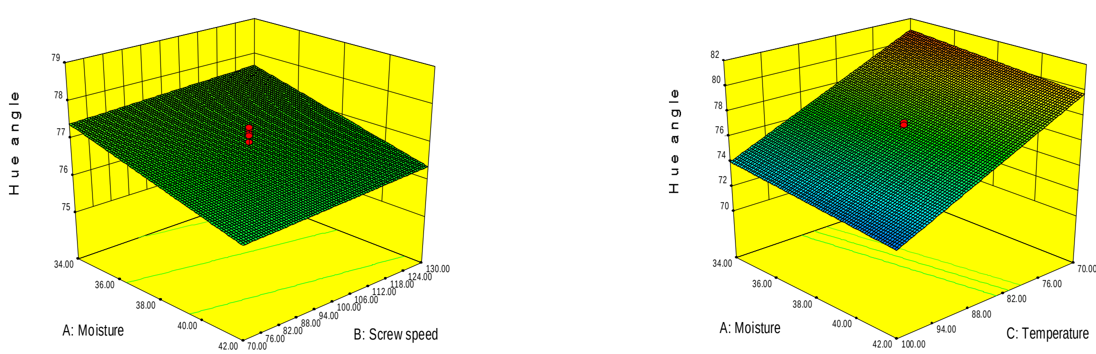

Fig. 7: Effect of moisture, screw speed and temperature on hue angle (°)

The barrel temperature significantly affected redness and yellowness of grains. Due to increased values of a* and b*, the chroma (c*) of re-fabricated grains also increased that ranged from 14.34 - 21.5. However, the effect was reflected more in total colour difference (ΔE) (equation iv and v), since the latter takes into account the changes in brightness.

Table 3: ANOVA and model statistics for the colour coordinates of extruded rice

Term	Response to models					
	L*	a*	b*	c*	ΔE	Hue angle (°)
F-value	244.92	143.48	431.50	552.41	335.89	266.70
P>F	0.0001	0.0001	0.0001	0.001	0.0001	0.0001
Mean	67.58	4.09	17.18	17.68	13.23	76.90
Standard deviation	0.64	0.16	0.19	0.20	0.35	0.46
CV	0.94	3.97	1.10	1.11	2.63	0.60
R-square	0.979	0.992	0.988	0.990	0.997	0.980
Adjusted R square	0.975	0.985	0.985	0.988	0.994	0.977
Predicted R Square	0.965	0.951	0.983	0.986	0.981	0.966
Adequate precision	53.94	45.01	71.03	80.51	68.76	56.55
Lack of fit	0.211	0.085	0.887	0.844	0.318	0.094

The positive coefficients of linear terms of temperature (equations ii to v) indicated that a* (redness), b* (yellowness), c* (chroma) and ΔE (total colour difference) increased with increase in barrel temperature. Out of the 3 independent variables (moisture, screw speed and barrel temperature) considered during the experiment, the effect of barrel temperature was highly significant and more pronounced on total colour difference (ΔE) than other 2 variables (Table 2). Mild extrusion temperature (59°C) resulted in small value of ΔE (5.8) and intermediate temperature caused the hue angle to decrease (equation vi) indicating that if temperature is allowed to increase to a certain level during extrusion, the discolouration could occur (i.e. from yellow to amber) due to Millard type non-enzymatic browning (Table 2). Park *et al.*, (1993) reported significant interaction effect of barrel temperature on extruded products of defatted soy flour and corn starch.

Optimization

The optimum extrusion condition was obtained by employing the desirability function method. The desirability value obtained was 0.802. The moisture content 36%, screw speed 130 rpm and temperature 89°C were predicted as best extrusion conditions by response surface methodology for making the re-fabricated rice with better consumer acceptability. The predicted response values and the actual obtained response values were almost similar. The variation of actual response values was within 5% of the predicted values (Table 4).

Effect of optimized extrusion conditions on maillard precursors

The levels of sucrose, glucose, fructose and maltose in natural (cv. PR-116) and re-fabricated rice obtained at optimized extrusion conditions are presented in Table 5. Sucrose levels were higher than glucose, fructose and maltose levels in both natural and re-fabricated rice. This was in concomitance with the levels already reported (Ali and Bhattacharya, 1980; Lieve *et al.*, 2008). Extrusion-processing reduced maltose content to undetectable level in re-fabricated rice. Further, sucrose was reduced during extrusion process. The decrease in sucrose level was 31.7% in re-fabricated rice extruded at 36% moisture content, 130 rpm screw speed and 89°C barrel temperature. The extrusion process increased the levels of glucose and fructose significantly. The loss of sucrose caused by extrusion process can be explained by conversion of sucrose into glucose and fructose. The differences in glucose and fructose levels in re-fabricated rice can be due to losses in maillard, glucose-fructose isomerisation and degradation reactions during heating in the extruder. The thermal degradation of starch during extrusion may also be responsible for increased sugar content of re-fabricated rice.

Table 4: Predicted response versus actual response

Values	Responses					
	L*	a*	b*	c*	ΔE	Hue angle (°)
Predicted	65.22	4.49	17.78	18.33	11.84	75.83
Actual	63.03	4.68	18.54	19.12	12.34	75.83
Variation (%)	3.36	4.23	4.27	4.30	4.22	0.00

Table 5: Levels of mono- and di-saccharides (sucrose, glucose, fructose and maltose in ppm natural and re-fabricated rice

Type of rice	Sucrose	Glucose	Fructose	Maltose
Natural rice (cv. PR-116)	14500 ± 10.20	230 ± 1.62	150 ± 1.23	190 ± 1.66
*Re-fabricated rice	9900 ± 5.23	410 ± 3.50	220 ± 1.87	nd**

* Re-fabricated rice was obtained at optimized extrusion conditions (36% moisture content, 130 rpm screw speed and 89°C barrel temperature; ** not detectable

Conclusions: All the three important extrusion parameters *viz*., screw speed, feed moisture and barrel temperature significantly affected colour coordinates of twin screw extruded re-fabricated rice. Within the experiment range, the barrel temperature was the most important factor for all the parameters of extruded rice studied. The effect of barrel temperature on L*, b*, c* and hue angle were linear; the effects however, were quadratic with a* and ΔE. The study also revealed that the levels of glucose and fructose in re-fabricated rice were mainly governed by their losses in maillard reactions, glucose formation as a result of thermal starch degradation and isomerization of glucose into fructose during heating in the extrusion process. It can be concluded that extrusion processing can be an ideal method for the production of re-fabricated rice, but being a complex multivariate process, it requires careful control if product quality is to be maintained.

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