Color development in an extrusion-cooked model system

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Abstract: Rice-glucose-lysine blend was extruded using a co-rotating twin-screw extruder. The effects of different extrusion conditions on the browning properties of extrudates were analyzed and compared using the CIE Lab system of measurement. Extrusion process variables included moisture content, screw speed, barrel temperature, and screw geometry. The influence of product temperature on the browning property of extrudates was significant ($P<0.05$). The color parameters were related to product temperature by a 4th degree polynomial ($P<0.05$). Hunter color scale values (L^*, a^*, b^*, $-L^*a^*/b^*$, Whiteness Index, and Yellowness Index) from extruded samples were analyzed to relate to extrusion process variables. Product temperature and browning properties were modeled and tested at various screw configurations and extrusion conditions. Product temperature and browning property models were verified using different screw geometries and other processing conditions with reasonable accuracy.

Keywords: twin-screw extrusion, browning, hunter color scale, Maillard reaction, empirical modeling

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1 Introduction

In many areas of the food industry, extrusion is an important manufacturing method. Uniformly processed products can be obtained in a continuous operation with variable thermal and mechanical energy inputs. Extrusion significantly increases overall palatability of foods by enhancing flavor, color, texture, and appearance characteristics. These changes are mainly related to unique extrusion processing and non-enzymatic browning during extrusion. Understanding the reaction kinetics in food extrusion processing is essential for predicting food quality and controlling extrusion. Food color changes after thermal processing can be used to predict the food quality deterioration resulting from exposure to heat.

Non-enzymatic browning reactions are important phenomena that occur during food processing and storage and, depending on the system, can be either desirable or undesirable[1]. Non-enzymatic browning can involve different compounds and proceed via different chemical pathways. It includes a wide number of reactions such as Maillard reaction (MR), caramelization, maderization, and ascorbic acid oxidization. Non-enzymatic browning is stated to be dependent on the temperature and water activity of the food[2-5]. Non-enzymatic browning may develop at varying reaction rates in samples due to processing which may cause differences in temperature and moisture distribution within the food and subsequently localized concentration of reactants[6]. Fluorescence and browning development in MR are generally used as indicators of the reaction rate and MR product formation[7]. Many authors use color as a quality control indicator of processes because brown pigments increase as browning and caramelization reactions progress[8,9].

With high temperature, high pressure, and low moisture content of the feed, extrusion cooking often results in a colored product, even though the residence

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time is low. As in traditional cooking processes, flavor and color are generated during cooking by a number of reactions which are controlled by the composition, temperature, and residence time. However, in extrusion cooking, these reactions are accelerated because of shear forces. Although the effects of extrusion conditions on the properties of extruded starch-based foods were previously studied using a one-variable-at-a-time approach or response surface methodology[10-14], data on the effects of extrusion cooking conditions (such as the extrusion process variables) on the color of the product were scarce. According to Bhattacharya, et al., researchers used single-screw extruders to report the effect of extrusion variables on product characteristics excluding product color, whereas reports on color changes by using twin-screw extruders were scarce[15].

The objective of this research was to investigate the influence of the extrusion variables (moisture content, screw speed, barrel temperature, and screw geometry) on the color properties of rice-glucose-lysine blend using the CIE Lab system of measurement. The systematic evaluation and mathematical modeling of extrusion color development were performed in order to apply it as an indicator for the impact of thermal and mechanical processing conditions on the extrudate quality.

2 Materials and methods

2.1 Preparation of extruded model system

Rice flour (Rivland Partnership, Houston, Texas) at 9% moisture content (wet basis), D-(+)-glucose, anhydrous (ACROS Organics, New Jersey), and L-lysine mono hydrochloride (ICN Biomedicals, Inc., Aurora, Ohio) were mixed (96.9%, 3%, 0.1% by weight) to give a homogeneous extruder feed. Particle size distribution of rice flour was provided by Rivland to be: 95%-100% through U.S. 50 mesh, 45%-65% through U.S. 100 mesh, and 25%-40% through U.S. 140 mesh.

2.2 Extrusion test

The experiments were carried out with a co-rotating twin-screw extruder (DNDL-44/28D twin-screw extruder, Buhler Inc. Uzwil/Switzerland). The extruder was comprised of seven barrels, each of 4 L/D (screw length/screw diameter), and a die holder plate of 0.5 L/D. The total machine L/D was 28.5. Four separate temperature controlled oil circulating units were connected to the extruder barrels to maintain a preset temperature. Cooling water was connected to the first barrel (feed barrel). The second and last barrels had their own circulating oil heater to control temperatures. Barrels 3 and 4 were connected to one circulating oil heater. Barrels 5 and 6 were connected to another circulating oil heater. A temperature probe and a pressure transducer were inserted into the extruder channel right before the die to measure the product temperature and pressure at the entrance of the die. The extruder had digital displays for shaft torque (% and Nm), product temperature (PT, ℃), and pressure (bar) developed during extrusion. Shaft torque is a percent number of a maximum torque. Specific mechanical energy input (*SME*) is calculated from Equation (1):

$$SME=\frac{M_d\omega}{\dot{m}} \tag{1}$$

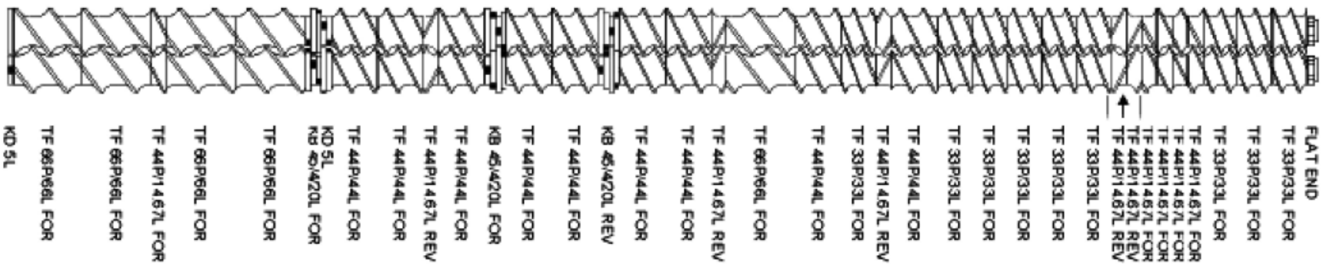
Where *SME* is specific mechanical energy input, Wh/kg; M_d is torque, N·m; ω is angular velocity, radian/s, 1 r/min = 2π/60 radian/s; $\dot{m}$ is throughput, kg/h [16].

Screw elements "TFxxP/xxL For" represent forward conveying elements with different pitches and length (Figure 1 (a, b, c, and d) and Table 1). Screw elements "KB45/4/20L For and Rev" were kneading elements with forward and reverse directions, respectively. Each kneading element consisted of 4 adjacent bilobal discs (thickness = 5 mm) with stagger angles of +45° (forward) and -45° (reverse). Screw elements "TF44P/14.67L For and Rev" were both one-third cut of forward and reverse conveying elements with a pitch of 44 mm. Screw configurations were chosen to cover the range of the pressure and torque of the extruder (Figure 1). Screw configurations with more reverse elements (screw configurations 1 and 3) would result in higher shaft torque and higher shearing during extrusion. This corresponds to higher mechanical energy input. Screw configuration without reverse elements (screw configurations 2 and 4) would result in higher pressures. Different numbers of kneading elements and conveying elements were also considered in the design of screw configurations.

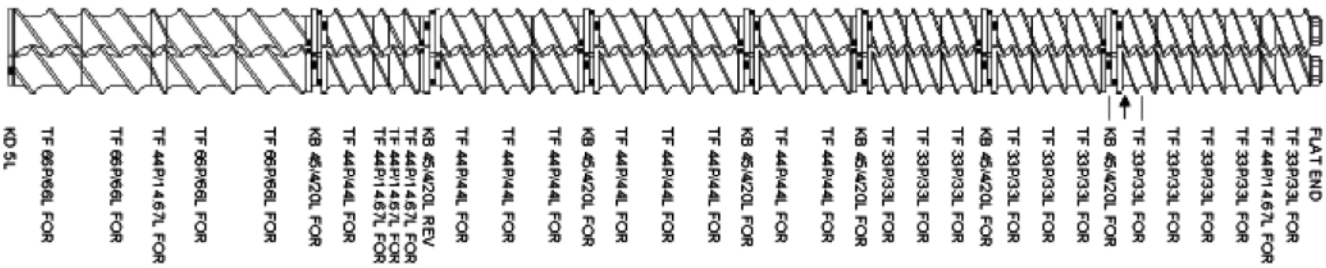
Rice-glucose-lysine blend was added to the extruder with a loss-in-weight feeder (K-ML-KT20, K-Tron AG. Industrie Lenzhard, Niederlenz, Switzerland) at a solid feed rate of 40 kg/h. Water was added directly to the feed barrel from an electromagnetic flow metering system (Proline Promag 50, Endress + Hauser Flowtec AG, Reinach, Switzerland). This system permits precise control of the water flow rate. An extruder die was used throughout the studies with the die geometry: two circular die holes of 3 mm diameter with a length of 6 mm. The extrusion conditions with variables of moisture content, screw speed, barrel temperature, and screw profile are shown in Tables 1 and 2. The steady state of processing conditions was achieved as indicated by constant pressure, temperature, and torque readings.

Table 1 Screw configurations

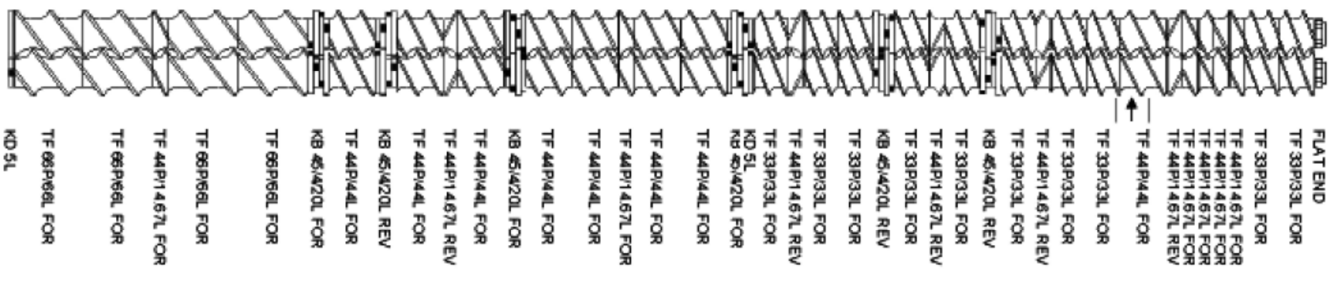
Type of screw element	Length/mm	Pitch/mm	Direction	Number of screw elements			
				Screw configuration #1	Screw configuration #2	Screw configuration #3	Screw configuration #4
TF66P/66L For	66	66	Forward	5	4	4	4
TF44P/44L For	44	44	Forward	9	9	8	9
TF33P/33L For	33	33	Forward	9	11	10	12
KB45/4/20L For	20	Polypack	Forward	2	6	3	8
KB45/4/20L Rev	20	Polypack	Reverse	1	1	3	1
TF44P/14.67L For	14.67	44	Forward	5	5	5	0
TF44P/14.67L Rev	14.67	44	Reverse	5	0	5	0



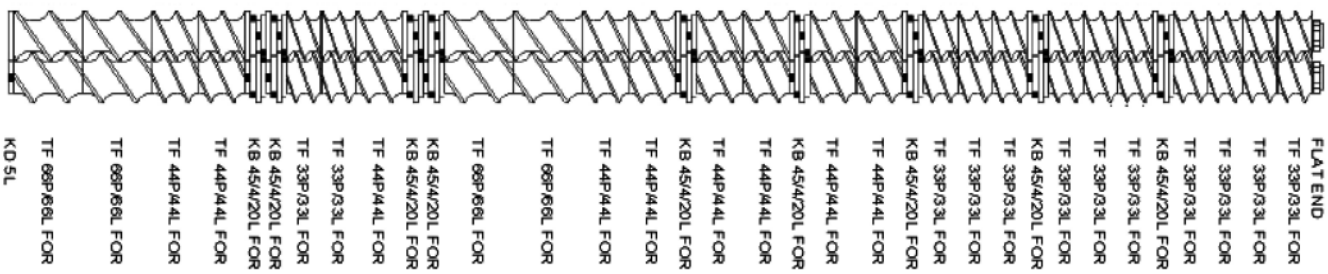
a. Extruder barrels and screw configuration #1



b. Screw configuration #2



c. Screw configuration #3



d. Screw configuration #4

Figure 1 Extruder barrels and four different screw configurations

2.3 CIE system of color measurement

The color parameters determined for the extrudates included L^*, a^*, b^*, *WI*, and *YI* values using a HunterLab colorimeter (MiniScan XE Plus, Virginia, USA) with illuminant D_{65}, a 10^0 standard observer. Samples were set in the lab atmosphere to equilibrate to ambient moisture content over a one-week period. Then extrudates were milled through a 0.5 mm sieve. The reported color parameters are the mean values of five observations.

Whiteness index (*WI*) and yellowness index (*YI*) of the CIE color system were calculated as follows:

$$WI = Y + 800(x_n - x) + 1700(y_n - y) \quad (2)$$

Where Y, x, and y are the luminance factor and chromaticity coordinates of the specimen, and x_n and y_n are the chromaticity coordinates for the CIE standard illuminant used.

$$YI = \frac{100(1.28X - 1.06Z)}{Y} \quad (3)$$

Where X, Y, and Z are the CIE Tristimulus values.

The color concentration can be translated in the L^*, a^*, and b^* parameters of the CIE color system or by a combination of these values, such as $-L^*a^*/b^*$, which expresses a total color change of the food product[17].

2.4 Statistical analysis

Extrusion process conditions (Table 2) were used to evaluate the effects of screw speed, moisture content, barrel temperature, and screw configuration on product temperature and browning properties of extruded rice blend. SAS Institute software was used to analyze the data for relationships between extrusion processing independent and dependent variables with respect to the prescribed properties of the extrudates using ANOVA with significance level established at $P<0.05$. Regression analyses were performed using MATLAB® (version 6.0, the Mathworks, Inc. Natick, MA) software for relationships between extrusion processing independent and dependent variables.

Table 2 Extrusion processing conditions for different extrusion runs

Extrusion runs	Screw configu -rations	Barrel temper -ature/℃	Screw speed / r • min^{-1}	Moisture content /%
#1	Screw #1	70	350/450/580	22.5/24.5/28
#2	Screw #1	100	350/450/580	22.5/24.5/28
#3	Screw #1	130	350/450/580	22.5/24.5/28
#4	Screw #2	70	350/450/580	22.5/24.5/28
#5	Screw #2	100	350/450/580	22.5/24.5/28
#6	Screw #2	130	350/450/580	22.5/24.5/28
#7	Screw #3	70	350/450/580	22.5/24.5/28
#8	Screw #3	100	350/450/580	22.5/24.5/28
#9	Screw #3	130	350/450/580	22.5/24.5/28
#10	Screw #4	70	350/450/580	22.5/24.5/28
#11	Screw #4	100	350/450/580	22.5/24.5/28
#12	Screw #4	130	350/450/580	22.5/24.5/28

3 Results and discussion

The colorimeter provides the values of three-color components: L^* (black-white component, luminosity) and the chromaticness coordinates, a^* (green to red component), and b^* (yellow to blue component). Whiteness and yellowness indexes signify the color changes of the extruded rice blend compared to its uncooked status. The mean L^*, a^*, b^*, *WI*, and *YI* values prior to extrusion cooking were 95.12, -0.25, 6.53, 53.52, and 11.74, respectively for the rice blend. The L^* and *WI* of the extruded sample were expected to be lower than the rice blend before cooking, while a^*, b^*, and *YI* were expected to be higher than the rice blend before cooking due to the darkened color caused by browning. Therefore, lower values of L^* and *WI* and higher values of a^*, b^*, and *YI* correspond to darker color of extrudates after cooking.

The difference in color change between extrudates can be attributed to the different extrusion conditions. It was found that screw speed, moisture content, barrel temperature, and screw geometry had significant influences on the changes of extrudate color ($P<0.05$). Severe extrusion conditions, which correspond to low values of L^* and *WI* and high values of a^*, b^*, and *YI*, are usually caused by low moisture content, high screw speed, high barrel temperature, and screw configuration with more reverse elements.

Among system parameters, product temperature (*PT*) and specific mechanical energy input (*SME*) had decisive effects on the functionality of extruded food products when reaction occurs during extrusion[18]. The effects of extrusion process parameters on the system parameters were examined thoroughly by researchers[19-25]. Results of this research also followed some well-known relationships between extrusion process and system variables. Results of this research also indicated that decreasing moisture content and increasing screw speed resulted in the increases of *PT* and *SME*. Increasing barrel temperature increased *PT* and decreased *SME*. Screw configurations with more reverse elements, which increased shearing, also increased *PT* and *SME*. *PT* and *SME* are good indicators for the changes of extrusion process variables.

There are essentially transfers of heat either thermal (from barrel heating and cooling) or mechanical (from shearing by the movement of the screws) during extrusion.

The final quality of the extrudate depends on how the thermal performance of the extruder is controlled and what the thermomechanical history of the product is inside the extruder. The product temperature is a reflection of these energy sources. Using extrusion process variables to describe browning properties is complicated due to the number of process parameters and their interactions. Thus, efforts to establish the relationship between extrudate color and product temperature are required. In this research, the extrusion product temperature was used to quantify the extrudate color and previously developed models were used to predict product temperature. Thus, the color developments were indirectly related to and predicted by the extrusion process variables.

3.1 Effect of *PT* on color development

A polynomial regression analysis of extrusion run #1 gave the following *L** and *PT* relationship:

$$L^* = K_0 + K_1 * PT + K_2 * PT^2 + K_3 * PT^3 + K_4 * PT^4 \quad (4)$$

Where *L** is luminosity value, *PT* is product temperature, ℃, and K_i is constant with K_0=4456.8, K_1=−137.22, K_2=1.60, K_3=−0.0082, and K_4=2E-05.

The regression analysis for the determination of the desired constants yielded a regression coefficient (R^2) of 0.99, indicating a good fit of the L^* value. A plot of *L** versus *PT* data is illustrated in Figure 2. This relationship was verified by using different barrel temperatures, different screw geometries, and other processing variables (Table 3). This 4th polynomial relationship between product temperature and color parameters was strongly held for extrudates (R^2>0.95, Table 3) by screw configuration with more reverse element (screws #1 and #3), even with lower barrel temperature (70℃). The color parameters and *PT* relationships were relatively weak (R^2 <0.8) when the rice-glucose-lysine blend was extruded with the screw configuration with less reverse element (screws #2 and #4, extrusion runs #4 and 10) and at the low barrel temperatures (70℃). Higher barrel temperatures (100 and 130℃) helped to improve the 4th polynomial relationships between color parameters and *PT* when products were extruded with screw configurations with less reverse element (Table 3, R^2> 0.90 of extrusion runs #5 and 6, #11 and 12).

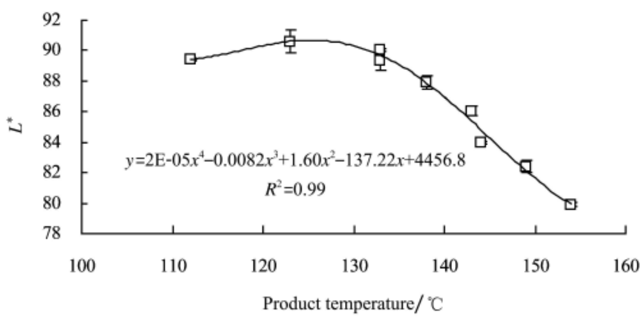


Figure 2 Product temperature vs. *L** of extrusion run #1 with screw configuration #1 at barrel temperature of 70℃ (error bars are standard deviations)

Table 3 Regression coefficients of the 4th degree polynomial relationships between Hunter color values and *PT* for different extrusion runs

Extrusion runs	*L**	*a**	*b**	-*L***a**/*b**	*WI*	*YI*
#1	0.99	0.99	0.97	0.99	0.97	0.98
#2	0.99	0.99	0.95	0.99	0.96	0.97
#3	0.98	0.98	0.96	0.98	0.96	0.97
#4	0.73	0.86	0.88	0.77	0.87	0.84
#5	0.99	0.94	0.99	0.93	0.99	0.99
#6	0.97	0.97	0.95	0.97	0.95	0.96
#7	1.00	1.00	1.00	1.00	1.00	1.00
#8	0.96	0.97	0.95	0.96	0.96	0.96
#9	0.97	0.98	0.97	0.97	0.98	0.97
#10	0.3	0.73	0.95	0.41	0.93	0.94
#11	0.94	0.89	0.94	0.94	0.94	0.94
#12	0.97	0.96	0.96	0.96	0.98	0.97

Figure 2 represented a typical relationship between color parameters and product temperature. *L** values decreased at higher product temperatures (*PT*>130℃, Figure 2). The changes of *L** were not significant when *PT* was less than 130℃ (*P*>0.05). Most of the product temperatures of extrusion runs #4 and #10 were less than 140℃ and resulted in no significant changes of color parameters (Table 3 and Upper part of the line in Figure 2). The data with no significant changes of color parameters helped to construct the 4th polynomial relationship between color parameters and *PT* (upper level part of line in Figure 2 and lower level part of line in Figure 3). The *L** value of the non-extruded blend was 95.12, while the *L** values of extruded samples were between 73.5 and 92.0. High barrel temperature and screw speed with low moisture and screw configurations with more reverse element resulted in low *L** values. Increases in product temperature (>130℃) decreased the brightness of the samples (Figure 2). After 165℃, the decrease in *L** values was low.

The *a** values indicating the greenness to redness of the extruded samples varied from 0.32 to 8.40, while the *a** value of the non-extruded blend was -0.25. The low *a** values were obtained from extrusion conditions with low barrel temperatures and screw profiles with less reverse elements. The changes of *a** values were significantly related to product temperatures ($P<0.05$).

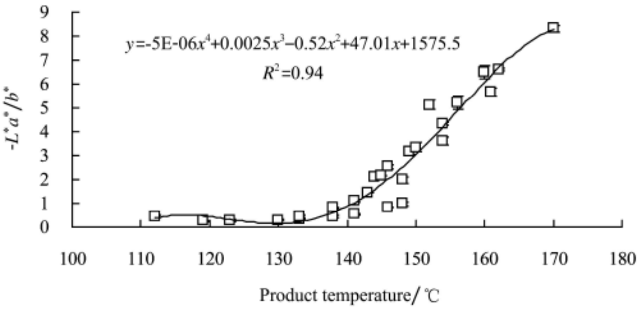


Figure 3 Product temperature vs. –*L***a**/*b** of screw configuration #1 at barrel temperature of 70, 100, and 130℃ (extrusion runs #1 - 3, error bars are standard deviations)

The *b** values indicating the yellowness of the extruded samples varied from 9.0 to 25.5, while the *b** value of the non-extruded blend was 6.5. The *b** value of extruded samples showed a marked increase in yellowness. The high *b** values were obtained from extrusion conditions with high barrel temperatures and screw speeds, low moisture contents, and screw profiles with more reverse elements. The changes of *b** values were significantly related to product temperatures ($P<0.05$).

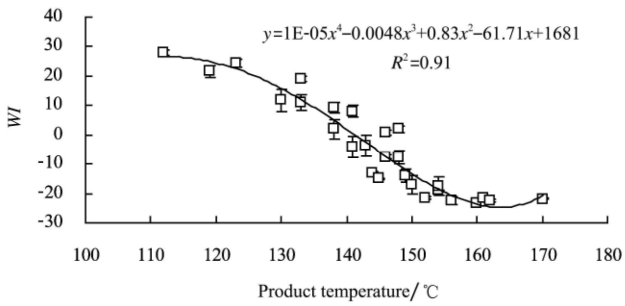


Figure 4 Product temperature vs. *WI* of screw configuration #1 at barrel temperature of 70, 100, and 130℃ (extrusion runs #1 - 3, error bars are standard deviations)

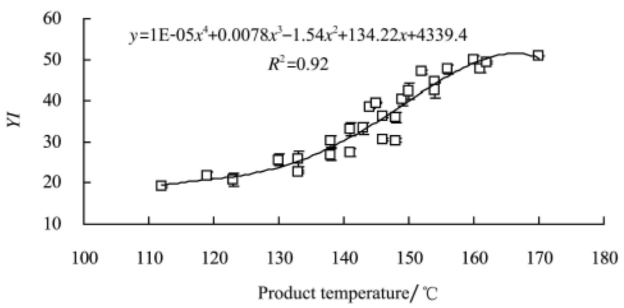


Figure 5 Product temperature vs. *YI* of screw configuration #1 at barrel temperature of 70, 100, and 130℃ (extrusion runs #1 - 3, error bars are standard deviations)

It is surprising that color parameter data with different barrel temperatures can be used to construct a single regression line (Figures 3, 4, and 5). A combination of mechanical and thermal energy input, product temperature is the correct factor to affect browning reactions during extrusion and efforts to predict product temperature are necessary to quantify and predict the color development of extrudate during extrusion.

3.2 Empirical modeling of product temperature

A nonlinear regression analysis of extrusion runs with the correction for the shear rate, moisture content, screw geometry, flow rate, and barrel temperature gave the following product temperature and processing variables relationship:

$$Exp(1/PT) = k\dot{\gamma}^n e^{\frac{A}{T_b}} e^{BMC} \left(\sum_{All\_screw\_elements}^{i} \frac{1}{C_{1pi}}\right)^C q^D \tag{5}$$

Where PT is product temperature, ℃; $\dot{\gamma}$ is shear rate, s^{-1}; T_b is barrel temperature, ℃; MC is moisture content, %; C_{1pi} is screw factor, q is flow rate, kg/h; n, A, B, C, and D are constant.

A detailed description of this model can be found in Lei et al[26]. A plot of the modeled product temperature data using Equation (5) versus the experimental product temperature data from extrusion runs #1 to #6 is illustrated in Figure 6 (R^2 = 0.96 and slope = 0.998). As can be seen, the prediction of the product temperature is quite good for all data points.

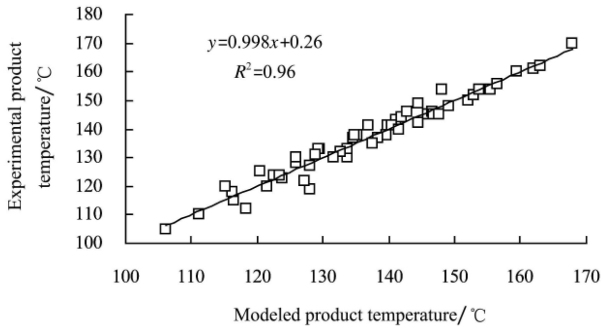


Figure 6 Modeled vs. experimental temperature of extrudates of extrusion runs #1-6 using Equation 5

Model validations of the product temperature model were verified using different screw geometries and other processing variables (Table 1 extrusion runs #7 and #12). A plot of the observed product temperature versus the predicted product temperature (data from extrusion runs #7 and #12) using Equation (5) is shown in Figure 7. Although the slope of 0.93 obtained by linear regression is less than one, the y axial intersection compensates for the slope and gives very good matches of predicted product temperature and experimental product temperature. The corresponding R^2 of 0.94 also indicates that the predictions are very good.

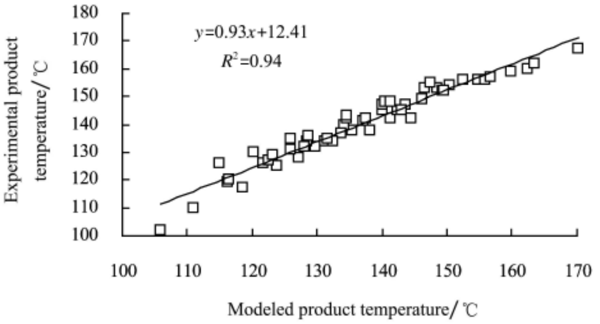


Figure 7 Predicted vs. experimental temperatures of extrudates of extrusion runs #7-12 using Equation 5

3.3 Prediction of color development

A polynomial regression analysis of extrusion runs #1-3 gave the following color parameters and *PT* relationships:

$$-L^*a^*/b^* = -1575.5 + 47.01PT - 0.52PT^2 + 0.0025PT^3 - 5\times10^{-6}PT^4 \quad (6)$$

$$WI = 1681 - 61.71PT + 83PT^2 - 0.0048PT^3 + 1.1\times10^{-5}PT^4 \quad (7)$$

$$YI = -4339.4 + 134.22PT - 1.54PT^2 + 0.0078PT^3 - 1.02\times10^{-5}PT^4 \quad (8)$$

The regression analysis for the determination of the desired constants yielded a regression coefficient (R^2) of 0.94, 0.91, and 0.92 respectively for $-L^*a^*/b^*$, *WI*, and *YI*, indicating a good fit of the color parameter data. Plots of the $-L^*a^*/b^*$, *WI*, and *YI* versus *PT* data are illustrated in Figures 3, 4, and 5. As can be seen, a single regression line was constructed for color parameters vs. *PT*, reflecting the changes of screw speed, moisture content, and barrel temperature. These regression correlations provided a simple way to quantify the color development of extrudates but intrinsically contained the information of the effects of extrusion process variables.

Model validations of these 4th polynomial relationships were verified using different screw geometries and other processing variables (Table 1 extrusion runs #4 -12). With the success of modeling the product temperature, the color of extrudates can be predicted from extrusion process parameters instead of using experimental product temperatures. Product temperatures were predicted using model Equation (5), then these calculated product temperatures were used in the color model Equations 6, 7, and 8 to predict the color development of extrusion. Plots of the observed browning changes versus the predicted color parameters (data from extrusion runs #4 - 12) using Equations 5, 6, 7, and 8 are shown in Figures 8, 9 , and 10, respectively, for $-L^*a^*/b^*$, *WI*, and *YI*. The slopes of near 1.0 obtained by a linear regression with the corresponding $R^2 > 0.90$ indicates that the predictions are good (Figures 8, 9, and 10). The model verification indicates that the quantification of the color was successful by means of the 4th polynomial relationship and using product temperature, which results from the input of mechanical and thermal energy, reflecting the changes of screw speed, moisture content, barrel temperature, and screw geometry. The results show that the CIE Lab system of measurement provides a simple approach for assessing the color of extrudates, which were simply linked to extrusion system parameters by means of a 4th polynomial relationship and can be predicted from both the extrusion process and system variables.

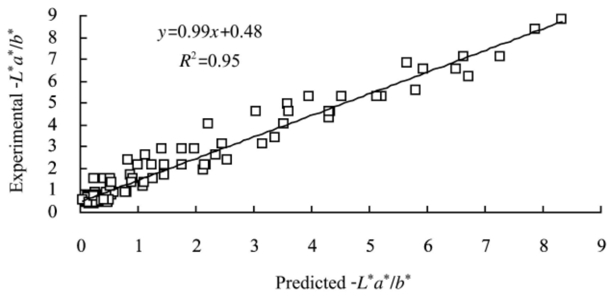


Figure 8 Predicted vs. experimental $-L^*a^*/b^*$ values for extrusion runs #4-12 using Equations 5 and 6 by predicted product temperature

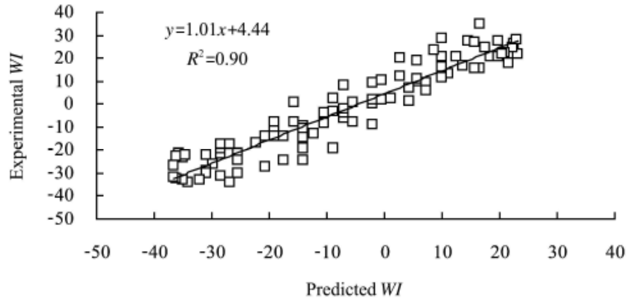


Figure 9 Predicted vs. experimental *WI* values for extrusion runs #4-12 using Equations 5 and 7 by predicted product temperature

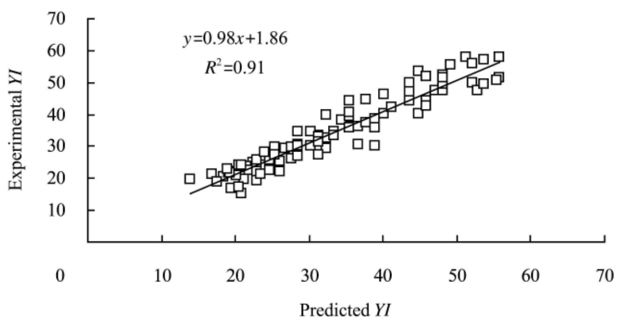


Figure 10 Predicted vs. experimental *YI* values for extrusion runs #4-12 using Equations 5 and 8 by predicted product temperature

4 Conclusions

Rice-glucose-lysine blend was extruded using a co-rotating twin-screw extruder. The effects of different extrusion conditions on the browning properties of extrudates were analyzed and compared using the CIE Lab system of measurement. Extrusion process variables included moisture content, screw speed, barrel temperature, and screw geometry. The influence of product temperature on the browning property of extrudates was significant. Hunter color scale values (L*, a*, b*, $-L^*a^*/b^*$, Whiteness Index, and Yellowness Index) from extruded samples were analyzed to relate to extrusion process variables. Product temperature and browning properties were modeled and tested at various screw configurations and extrusion conditions. The color development during extrusion process was linked to extrusion system parameters by means of a 4th polynomial relationship and can be predicted from both the extrusion process and system variables. Product temperature and browning property models were verified using different screw geometries and other processing conditions with reasonable accuracy. Extrusion tests indicate that the developed predictive models can be used for extrusion processing. The results also indicate a rapid response way for assessing the color development during extrusion process can be established for quality control.

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