

Optimization of wheat flour, pumpkin flour and cranberry pomace blend formulations based on physicochemical properties of value-added cookies

Pranabendu Mitra^{1*}, Razu Thapa¹, Binu Acharya¹ and Md. Abdul Alim²

¹Department of Food and Nutrition, University of Wisconsin-Stout, Menomonie, WI, 54751, USA

²Department of Food Technology and Rural Industries, Bangladesh Agricultural University, Mymensingh 2202, Bangladesh

*Corresponding author: Pranabendu Mitra, Email: mitrap@uwstout.edu, Tel: 1-715-232-5332

Abstract

The objective of the study was to optimize wheat flour, pumpkin flour and cranberry pomace powder blend formulations for developing value-added cookies using RSM combining with CCD to replace wheat flour with pumpkin flour and cranberry pomace powder to increase nutritional health benefits. Twenty different cookies for 15 formulations based on CCD were developed and physicochemical properties (moisture content, water activity, L value, total color difference, spread ratio, piece density and hardness) of the cookies were determined. Regression models, response surface models, numerical optimization based on a desirability function to determine an optimum formulation and graphical optimization by superimposing all contour plots of response surface models to visualize an optimum region were developed using a Design Expert software. The regression models were able to predict the physicochemical properties of cookies as a function of factors with an accuracy of 59-99% depending on the responses. The factors wheat flour, pumpkin flour and cranberry pomace affected the physicochemical properties of cookies significantly ($p < 0.05$). The numerical and graphical optimization revealed that the mixture of wheat flour (45-50%), pumpkin flour (30-35%) and cranberry pomace (24-27%) was the optimum formulation which improved the physicochemical properties of cookies. This optimum formulation can be used for commercial production of value-added cookies.

Keywords: Value-added cookies, optimization, CCD, RSM, pumpkin flour, cranberry pomace

1. Introduction

Refined wheat (*Triticum aestivum*) flour has been used in bakery products from ancient days. Wheat flour is commonly used to produce baked products because the gluten contents of wheat flour develop the viscoelastic property of the dough during baking and maintain the texture of various baked products. A popular baked snack cookie is made of refined wheat flour prepared from an unpalatable dough which is transformed into light porous easily digestible and more appetizing products by baking at 170-250°C for 15-30 min depending on formulations and desired types of cookies (Manley, 2000; Ndife et al., 2014). However, consumer awareness has led to use of whole grains for the improved health benefits. A recent trend showed that consumers inclined towards more nutrients and value-added products. In this regard, protein fortified products with incorporation of antioxidants and dietary fiber are desired to the consumers (Ndife et al., 2014; Jan et al., 2016; Šarić et al., 2016). In recent decade, cookies have gained a very significant importance as a preferred way to use composite flours in cookies to provide balanced nutrients and energy (Rangrej et al., 2015). Considering nutritional and economical contexts of manufacturing of cookies, there is a trend of partial replacement of refined wheat flours with other potential sources of plant flours such as yam, carrot, millet and pumpkin etc. to increase their palatability and nutraceutical properties (Manley,

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2000). Such flours also can be a good source of vitamins and natural color. The different variety of cookies are available in the market because of various options of ingredients and varied formulations of cookies. Incorporation of pumpkin flour combining with fruit pomaces can be a new avenue for value added cookies.

Pumpkin flours contain high amounts of carbohydrates, dietary fibers, minerals, a good source of β -carotene, vitamin A and other functional compounds (Das and Banerjee, 2015). Many food products are being fortified with pumpkin flours because it has been found that direct or indirect consumption of pumpkin flours helps in prevention of skin problem, eye disorder and cancer (Pongjanta et al., 2006). Incorporation of pumpkin flour with wheat flour not only increases the nutritional values but also enhances the flavor of baked products. Other trend is incorporation of fruits and their valuable byproducts (i.e. pomaces) with the baked products in order to make them nutritionally desired and tasty products with polyphenols, flavonoids, antioxidants and natural color (Jan et al., 2016; Šarić et al., 2016; Tańska et al., 2016). Fruits such as blueberries, cranberries and saskatoon berries have high antioxidants capacity because of their high contents of flavonoids, phenolic acids, vitamins and anthocyanins (Meda et al., 2016; Mildner-Szkudlarz et al., 2016). Cranberries have a strong antioxidant capacity because of their high amounts of polyphenols and anthocyanins. They also contain significant amounts of vitamin A & C, minerals (i.e. Ca and Fe) and essential fatty acids. Cranberries are mainly used for cranberry juice commercially. After juice extraction, cranberry juice processing industries produce a huge amount of byproduct pomaces. The management of such pomaces is a big challenge because they cannot be discarded as landfill without proper treatments because environmental problems can arise. Presently, cranberry pomaces are being utilized as cattle feed partially. Cranberry pomaces left after juice extraction contain polyphenols including anthocyanins, vitamins, provitamins, essential unsaturated fatty acids, minerals and dietary fiber and can be incorporated in the formulations to produce value-added functional bakery products (Mildner-Szkudlarz et al., 2016). The cranberry pomaces can be combined with pumpkin flour and wheat flour to develop value added functional cookies. However, this composite formulation affects the physical, mechanical and chemical properties such as color, density hardness, moisture content and water activity of cookies which are important for developing organoleptically acceptable tasty cookies. The optimum ratio of wheat flour, pumpkin flour and cranberry pomace is essential for desired physical, mechanical and chemical properties of cookies.

Response surface methodology (RSM) combining with central composite rotatable design (CCRD) is widely used to determine the optimum independent variables based on dependent or response variables (Myers et al., 2016). In order to determine the relationship between independent variables and response variables a combination of mathematics and statistics is used. The RSM and CCRD significantly determine the effects of processing variables on the response variables and determine the optimum processing conditions based on desired responses (Mitra et al., 2011; Meda et al., 2016). The principles of this optimization technique can be used to determine the optimum conditions for the formulations of wheat flour, pumpkin flour and cranberry pomace powder to develop value-added cookies. The objective of this study was to optimize the combination of wheat flour, pumpkin flour and cranberry pomace powder based on desired moisture content, water activity, color, spread ratio, piece density and hardness of the finished cookies to develop value-added functional cookies.

2. Materials and methods

2.1. Materials

The wheat flour and pumpkin flour of Glean brand were purchased from a local Walmart and cranberry pomaces were donated by Cranberry Partners LLC (Rapids, WI, USA). Cranberry pomaces were dried and ground into fine powder using a coffee grinding machine and the ground powder was sieved using a 60-mesh sieve. All other ingredients white sugar, brown sugar, butter (Great Value brand), eggs, salt (Great Value brand), baking soda (Arm & Hammer, USA) and vanilla extract (McCormick, USA) were purchased from a local Walmart.

2.2. Manufacturing of value-added cookies

The combination of wheat flour, pumpkin flour and cranberry pomace powder for each formulation was varied as per the central composite rotatable design (CCRD) as shown in Table 1. The amounts of all other ingredients were constant for all 15 formulations. The amount of each other ingredient was calculated as percent of total flour (wheat flour + pumpkin flour + cranberry pomace powder, w/w) for each formulation. The dry ingredients flour

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(wheat flour + pumpkin flour + cranberry pomace powder), baking soda (1.5%) and salt (1.5%) were mixed thoroughly with a hand-held mixer. The wet ingredients (butter 70%, sugar 50% and brown sugar 10%) were mixed until a creamy condition achieved. Then the egg (33%) and vanilla extract (2.7%) were added and mixed thoroughly. The dry mix was then added to the wet mix gradually and mixed to prepare the dough of cookies. The dough was cut into small pieces (a tablespoon in size). The small dough pieces were placed on an ungreased baking sheet and the small dough pieces were sheeted for baking. The sheeted cookie doughs were baked at 200°C for 10 minutes with an electric oven (General Electric, USA). After baking, cookies were cooled to room temperature (20°C) and packed in a low-density polyethylene (LDPE) bag for storage and further analysis of physicochemical properties for characterization and optimization of a cookie formulation.

2.3. Quality characterization of value-added cookies

2.3.1. Determination of moisture content of value-added cookies

The moisture content of the value-added cookies was determined using an oven drying method (Horwitz and Latimer, 2005; Timalina et al., 2019). About 5 grams of ground cookies were dried at 140°C for 2 hours to a constant weight of cookies. The moisture content of cookies was calculated from the weight difference between the initial and dried cookie samples and was expressed as a percentage of the initial weight using following equation. Two replications were performed for all samples.

$$\text{Moisture content (\%)} = \frac{\text{Initial weight of cookies} - \text{Dried weight of cookies}}{\text{Initial weight of cookies}} \times 100$$

2.3.2. Determination of water activity of value-added cookies

In order to determine the water activity of cookies the Aqualab Water Activity meter 4TE (Corona, CA) was used. About five grams of ground cookies were placed into a water activity measuring plastic cup. The plastic cup was placed in the sample chamber of the water activity meter and was closed with the outer lid of the water activity meter until a constant reading was obtained. Two replications of each sample were conducted, and the average water activity value was reported.

2.3.3. Determination of color of value-added cookies

The color profile of doughs and cookies were measure using a Hunter Lab Color flex-EZ Colorimeter (Hunter Associates Laboratory Inc., Reston, Virginia, USA). The L value (lightness/darkness), a value (redness/greenness) and b value (yellowness/blueness) of a dough and final cookies were determined (Meda et al., 2016; Timalina et al., 2019). The total color difference (ΔE) was calculated according to the following formula:

$$\text{Total color difference, } \Delta E = \sqrt{(L - L_o)^2 + (a - a_o)^2 + (b - b_o)^2}$$

Where, L_o , a_o and b_o were color values of doughs and L , a and b were color values of value-added cookies. The color measurement for each sample was performed six times and the average value with a standard deviation was reported.

2.3.4. Determination of spread ratio of value-added cookies

Spread ratio of cookies was determined according to AACC method 10-50D (AACC, 2001). The average diameter in millimeters were obtained by placing six cookies edge to edge horizontally and rotating at 90° angle using a Vernier Caliper. The thickness of cookies was measured five times at four different spots by rotating cookies at different angles. The spread ratio was calculated using following formula.

$$\text{Spread ratio} = \frac{\text{Diameter of cookies(mm)}}{\text{Thickness of cookies(mm)}}$$

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2.3.5. Determination of piece density of value-added cookies

Rapeseed displacement method (AACC, 2001; Mitra et al., 2019) was used to measure the piece density of the value-added cookies. Two cookies were weighed and placed in a graduated 1000 mL cylinder. Rapeseeds were used to fill the volume of the cylinder up to 1000 mL by repeated tapping of the cylinder until the samples came to a completely settled level. The cookies were taken out from the cylinder and the volume of rapeseeds was measured. The volume of cookies (mL) was measured by subtracting the volume of rapeseed from the cylinder volume (1000 mL). Two replicates for each sample were done and the average density of cookies was reported. The piece density of cookies was determined using following equation.

$$\text{Piece density}(g/mL) = \frac{\text{mass of cookies}(g)}{\text{volume of cookies}(mL)}$$

2.3.6. Determination of hardness of value-added cookies

The hardness (N) of the value-added cookies were determined using an Instron Machine (Instron Corporation, Norwood, USA). One single cookie was placed on the lower ram and the upper ram attached with a 35mm stainless steel probe with a 500 N load cell was compressed to break the cookie to 60% strain of the cookie. A strain level of 60-70% provided a complete information on the behavior of the sample without excessive densification. Before starting the test, the anvil height of the frame was adjusted which was depending upon the total standing height of the specimen. In order to perform the compression test, the pre-test speed 2 mm/s, test speed 1 mm/s and the post-test speed 2 mm/s were set. The Blue Hill 3 software was used to generate force-deformation curve. The maximum force of the force-deformation curve was considered as hardness (N) of the cookies (Meda et al., 2016; Timalisina et al., 2019). Five replications of each sample were conducted and the average value with a standard deviation of hardness of the cookies was reported.

2.4. Statistical analysis and modeling

2.4.1. Experimental design

In order to study the effect of independent variables (wheat flour, pumpkin flour, cranberry pomace powder) of cookie dough formulations on the response variables (moisture content, water activity, L value, total color difference, spread ratio, piece density and hardness,) and arrive at an optimum formulation the response surface methodology (RSM) combined with the central composite rotatable design (CCRD) was used. The CCRD was used to determine the experimental points. In this process, three independent variables (factors) and their five levels (coded values -1.68, -1, 0, +1 and +1.68 as shown in Table 1) were used. The experimental design comprised of 20 experimental points ($n = 2k + 2^k + m$, where, n = total experimental points, input variables, $k = 3$ and center point, $m = 6$ replications), which consisted of 8 factorial points, 6 axial points and six replicated center points (Mason et al., 2003; Myers et al., 2016; Mitra et al., 2011) as shown in Table 1.

2.4.2. Prediction modeling and optimization of cookie formulations

The response surface methodology (RSM) was used to determine the correlation between independent variables (wheat flour, pumpkin flour and cranberry pomace powder) and response variables (moisture content, water activity, L value, total color difference, spread ratio, piece density and hardness) of the value-added cookies and to optimize the factors/independent variables on response variables (physicochemical properties of cookies). Data analysis was carried out assuming a third order polynomial cubic equation for each of the response variables as a function of independent variables of wheat flour (X_1), pumpkin flour (X_2) and cranberry pomace powder (X_3). The third order polynomial cubic model to fit the coded variables was as follow:

$$Y = B_0 + B_1X_1 + B_2X_2 + B_3X_3 + B_{11}X_1^2 + B_{22}X_2^2 + B_{33}X_3^2 + B_{12}X_1X_2 + B_{23}X_2X_3 + B_{13}X_1X_3 + B_{111}X_1^3 + B_{222}X_2^3 + B_{333}X_3^3 + B_{123}X_1X_2X_3 + B_{112}X_1^2X_2 + B_{113}X_1^2X_3 + B_{122}X_1X_2^2 + B_{133}X_1X_3^2 + B_{223}X_2^2X_3 + B_{233}X_2X_3^2$$

Where, Y was the response variable, $B_0, B_1, B_2, B_3, B_{11}, B_{22}, B_{33}, B_{12}, B_{23}, B_{13}, B_{111}, B_{222}, B_{333}, B_{123}, B_{112}, B_{113}, B_{122}, B_{133}, B_{223}$ and B_{233} were constants and regression coefficients of the model, and X_1, X_2 and X_3 were the independent variables (factors).

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Table 1. Experimental design (CCRD) and experimental results of moisture content, water activity, L value, total color difference, spread ratio, piece density and hardness of value-added cookies

Run	Wheat flour (%) X_1	Pumpkin flour (%) X_2	Cranberry pomace (%) X_3	Moisture content (%)	Water activity	L value	Total color difference	Spread ratio	Piece density (g/mL)	Hardness (N)
1	(-1) 45	(-1) 25	(-1) 15	12.70 ± 0.02	0.68 ± 0.00	34.28 ± 0.07	9.89 ± 0.21	2.12 ± 0.25	0.46 ± 0.03	213.89 ± 35.77
2	(-1) 45	(-1) 25	(1) 25	10.84 ± 0.04	0.64 ± 0.00	36.72 ± 0.10	18.56 ± 0.20	2.5 ± 0.08	0.55 ± 0.14	368.40 ± 13.16
3	(-1) 45	(1) 35	(-1) 15	11.70 ± 0.09	0.67 ± 0.01	35.73 ± 0.07	8.46 ± 0.14	3.21 ± 0.31	0.48 ± 0.12	219.67 ± 32.54
4	(1) 55	(-1) 25	(1) 25	9.12 ± 0.20	0.61 ± 0.00	37.08 ± 0.07	11.89 ± 0.17	3.22 ± 0.27	0.58 ± 0.06	306.8 ± 107.70
5	(-1) 45	(1) 35	(1) 25	7.59 ± 0.36	0.56 ± 0.01	37.18 ± 0.23	12.28 ± 0.40	3.01 ± 0.21	0.48 ± 0.06	192.53 ± 37.67
6	(1) 55	(1) 35	(1) 25	10.16 ± 3.07	0.54 ± 0.00	34.39 ± 0.44	14.41 ± 0.53	2.99 ± 0.12	0.58 ± 0.08	319.25 ± 48.39
7	(1) 55	(1) 35	(-1) 15	8.77 ± 0.06	0.55 ± 0.00	34.39 ± 0.44	8.78 ± 0.60	2.77 ± 0.12	0.58 ± 0.10	295.08 ± 62.72
8	(1) 55	(-1) 25	(-1) 15	6.10 ± 0.01	0.41 ± 0.01	36.86 ± 0.85	12.16 ± 1.16	3.57 ± 0.22	0.61 ± 0.09	496.06 ± 1.26
9	(-1.68) 41.6	(0) 30	(0) 20	7.56 ± 0.05	0.50 ± 0.00	35.14 ± 0.18	10.02 ± 0.44	2.83 ± 0.23	0.59 ± 0.00	470.16 ± 22.09
10	(1.68) 58.4	(0) 30	(0) 20	9.76 ± 0.23	0.61 ± 0.00	35.24 ± 0.09	13.14 ± 0.39	5.73 ± 0.31	0.56 ± 0.07	329.60 ± 89.20
11	(0) 50	(-1.68) 21.6	(0) 20	9.46 ± 0.04	0.62 ± 0.00	35.17 ± 0.04	8.16 ± 1.46	3.79 ± 0.20	0.71 ± 0.15	335.45 ± 64.23
12	(0) 50	(1.68) 38.4	(0) 20	9.42 ± 0.00	0.59 ± 0.00	34.99 ± 0.08	3.70 ± 0.67	3.71 ± 0.29	0.61 ± 0.02	456.18 ± 41.44
13	(0) 50	(0) 30	(-1.68) 11.6	11.02 ± 0.35	0.63 ± 0.01	35.07 ± 0.23	7.88 ± 0.51	3.53 ± 0.35	0.61 ± 0.07	235.01 ± 56.89
14	(0) 50	(0) 30	(1.68) 28.4	12.19 ± 0.16	0.67 ± 0.00	26.69 ± 0.05	6.98 ± 0.39	2.86 ± 0.13	0.55 ± 0.05	202.73 ± 22.59
15	(0) 50	(0) 30	(0) 20	10.58 ± 0.34	0.56 ± 0.01	33.38 ± 0.43	9.08 ± 0.32	3.02 ± 0.11	0.56 ± 0.04	281.59 ± 39.36
16	(0) 50	(0) 30	(0) 20	9.79 ± 0.09	0.53 ± 0.01	32.71 ± 0.01	8.44 ± 0.39	3.22 ± 0.15	0.55 ± 0.00	281.59 ± 39.36
17	(0) 50	(0) 30	(0) 20	9.24 ± 0.07	0.52 ± 0.01	29.82 ± 0.08	7.27 ± 0.12	3.07 ± 0.10	0.54 ± 0.00	273.97 ± 41.28
18	(0) 50	(0) 30	(0) 20	9.77 ± 0.14	0.53 ± 0.01	30.42 ± 0.07	7.71 ± 0.17	3.40 ± 0.21	0.56 ± 0.02	351.04 ± 26.34
19	(0) 50	(0) 30	(0) 20	9.42 ± 0.13	0.53 ± 0.01	31.18 ± 0.18	7.36 ± 0.24	3.45 ± 0.18	0.54 ± 0.01	273.97 ± 41.28
20	(0) 50	(0) 30	(0) 20	9.98 ± 0.89	0.53 ± 0.00	31.18 ± 0.08	8.24 ± 0.23	3.24 ± 0.22	0.55 ± 0.01	354.12 ± 36.82

$X_1 = (W - 50)/5$, $X_2 = (P - 30)/5$, $X_3 = (C - 20)/5$; W = Wheat flour, P = Pumpkin flour, C = Cranberry pomace powder

The Design-Expert software was used to solve the third order polynomial cubic equation to develop regression models (response prediction equation), response surface models (effect of factors when varying two and keeping third constant) and numerical and graphical optimization of independent variables to determine the suitable cookie formulations. The regression models were justified using the analysis of variance (ANOVA). The three-dimensional response surface models were produced by presenting the response as a function of two factors keeping the third constant at the center point. The optimum regions were visualized by graphical optimization with the Design-Expert software. Numerical optimization was conducted based on a desirability function by a trial and error method using Design-Expert software.

3. Results and discussion

3.1. Regression prediction modeling with experimental data

Value-added cookies were developed using 15 different formulations by varying wheat flour, pumpkin flour and cranberry pomace powder as per the CCRD experimental conditions as shown in Table 1. The physicochemical properties (moisture content, water activity, L value, total color difference, spread ratio, piece density and hardness) of the cookies were determined and the results were presented in Table 1. The observations from Table 1 indicated that the moisture content (12.70-6.10%), water activity (0.68-0.41), L value (37.18-26.69), total color difference (18.56-3.70), spread ratio (5.73-2.12), piece density (0.71-0.46 g/mL) and hardness (496.06-192.53 N) of cookies varied with the dough formulations of cookies. However, the variability of those properties of cookies were not same. The results indicated that the combination of wheat flour, pumpkin flour and cranberry pomace powder affected the physicochemical properties of cookies significantly ($p < 0.05$). The L value (lightness) of the value-added cookies decreased with the pumpkin flour and cranberry pomace powder. Cookies became darker with increased incorporation of pumpkin flour and cranberry pomace powder. The results of the physicochemical properties of value-added cookies were in the agreement with other studies (Ahmad et al., 2016; Mildner-Szkudlarczyk et al., 2016; Šarić et al., 2016; Tańska et al., 2016). The water activity was under 0.6 (on set water activity of microbial growth) for 9 out of 15 formulations when cookies were developed with the combination of wheat flour, pumpkin flour and cranberry pomace powder (Table 1). The value-added developed cookies with under 0.6 water activity were shelf-stable and could be stored for a longer period in room conditions. Because the microorganism cannot grow under 0.6 water activity of food products due to scarcity of available water to grow (dos Santos et al., 2017).

The experimental results (Table 1) of the physicochemical properties of cookies were fitted into the third order polynomial cubic equation to develop the regression models to predict the physicochemical properties (response variables) of cookies as a function of independent variables (factors) of wheat flour (X_1), pumpkin flour (X_2) and cranberry pomace powder (X_3). In this process, the CCRD was combined with response surface methodology (RSM) to determine the coefficients and constants of the third order polynomial cubic equation using a Design-Expert software. To eliminate insignificant terms of the third order polynomial equation the backward algorithm was used. Regression predictive models for the physicochemical properties (response variables) of the value-added cookies as a function of coded independent variables are shown in Table 2.

Table 2. Regression predictive models for moisture content, water activity, L value, total color difference, spread ratio, piece density and hardness of value-added cookies

Response variable	Regression prediction models
Moisture content (%)	$= 9.67 + 0.6541X_1 - 0.0119X_2 + 0.3478X_3 + 0.9950X_1X_2 - 0.4850X_2X_3 + 1.30 X_1X_3 - 0.4173X_1^2 - 0.1415X_2^2 + 0.6240X_3^2 + 0.0775X_1X_2X_3 - 0.0556X_1^2X_2 - 0.5428X_1^2X_3 + 1.74X_1X_2^2$
Water activity	$= 0.5292 + 0.0327X_1 - 0.0089X_2 + 0.0119X_3 + 0.0200X_1X_2 - 0.0350X_2X_3 + 0.0425X_1X_3 + 0.041X_1^2 + 0.0217X_2^2 + 0.0377X_3^2 - 0.0175X_1X_2X_3 + 0.0064X_1^2X_2 - 0.0069X_1^2X_3 - 0.0877X_1X_2^2$
L value	$= 31.55 - 0.0748X_1 - 0.2601X_2 - 2.49X_3 - 0.8837X_1X_2 - 0.4587X_1X_3 + 1.72X_1^2 + 1.68X_2^2 + 3.01X_1^2X_3$
Total color difference	$= 8.35 + 0.2414X_1 - 1.18X_2 - 0.2676X_3 + 0.8562X_1X_2 - 0.8913X_1X_3 + 0.1312X_2X_3 + 2X_1^2 + 1.34X_1X_2X_3 + 2.5X_1^2X_3$
Spread ratio	$= 3.44 + 0.8622X_1 + 0.0319X_2 - 0.0788X_3 - 0.3287X_1X_2 - 0.0256X_2^2 - 0.2219X_3^2 - 0.6484X_1X_2^2$
Piece density	$= 0.5444 - 0.0089X_1 - 0.0182X_2 + 0.0025X_1X_2 + 0.0258X_2^2 + 0.0564X_1X_2^2$
Hardness	$= 303.97 - 41.79X_1 + 35.89X_2 - 9.6X_3 - 2.31X_1X_2 + 3.97X_2X_3 - 36.55X_1X_3 + 26.15X_1^2 + 24.71X_2^2 - 37.85X_3^2 + 49.38X_1X_2X_3 - 80.73X_1^2X_2 + 4.89X_1^2X_3 + 94.63X_1X_2^2$

The validity of the developed regression models for moisture content, water activity, L value, total color difference, spread ratio, piece density and hardness of cookies was confirmed by the statistical analysis of variance (ANOVA). The ANOVA results for the response variables of the cookies are shown in Table 3. The p values for all response variables were less than 0.05 or equal to 0.05. The p value results indicated that the individual terms of the

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regression models affected the response variables (physicochemical properties of cookies) significantly (Mason et al., 2003; Mitra et al., 2011). The F value of all response variables except total color difference was greater than F value at a critical point ($F_{critical}$). The F value (2.94) of total color difference of value-added cookies was close to F value at a critical point (3.18) as shown in Table 3. Since the F values of the response variables exceeded the F value at a critical point the null hypothesis was rejected. The higher F values of the regression models indicated that the models were able to predict response variables significantly (Mason et al., 2003; Mitra et al., 2011; Meda et al., 2016). Adequate precision value for all the response variables were greater than 4 (Table 3), which according to Design Expert software indicated enough model discrimination. The adequate precision higher than 4 showed one of the adequate signals of the accuracy of the model for the prediction of response variables (physicochemical properties of value-added cookies) because the adequate precision measured the range of the predicted values at a design point to the average prediction error (Meda et al., 2016; Timalsina et al., 2019). The coefficient of determination (R^2) value ranges from 0 to 1 with a score being closer to 1 is better (Mason et al., 2003; Mitra et al., 2007a,b). R^2 for moisture content, water activity, L value, total color difference and hardness of the value-added cookies was close to 1. But the R^2 values for spread ratio and piece density of the cookies were 0.66 and 0.59 (Table 3), respectively. Although the models for spread ratio and piece density had lower R^2 the regression models could predict those two properties of cookies with a significant accuracy because of their lower p value and the stronger adequate precision (Meda et al., 2016). It could be concluded from the ANOVA analysis (Table 3) that the cubic regression models for response variables of value-added cookies agreed with the experimental results for predicting physicochemical properties of value-added cookies as a function of wheat flour, pumpkin flour and cranberry pomace.

Table 3: Analysis of variance (ANOVA) for the regression models of moisture content, water activity, L value, total color difference, spread ratio, piece density and hardness of value-added cookies

Model Source	Sum of Squares	DF	Mean Square	F-value	$F_{critical}$	p-value	Adequate Precision	R^2
Moisture Content (%)	45.51	13	3.50	31.96	2.58	0.00	23.83	0.99
Water Activity	0.09	13	0.01	18.72	2.58	0.01	17.08	0.98
L value	123.53	8	15.44	8.78	3.44	0.00	10.55	0.87
Total Color Difference	145.33	9	16.15	2.94	3.18	0.05	6.22	0.73
Spread Ratio	6.25	7	0.89	3.35	3.78	0.03	8.88	0.66
Piece Density (g/mL)	0.03	5	0.01	3.94	5.05	0.02	6.53	0.59
Hardness (N)	130100	13	10004.14	4.86	2.58	0.03	8.72	0.91

3.2 Response surface modeling: effect of factors on responses

The effect of the factors (wheat flour, pumpkin flour and cranberry pomace powder) on the responses (moisture content, water activity, L value, total color difference, spread ratio, piece density and hardness) of value-added cookies can be understood by the response surface models. The relationship between factors and responses were visualized as 3-dimensional graphs generated by varying two factors and keeping the third factor constant for a response.

Response surface graphs indicated the direction of change made in response variables (physicochemical properties of cookies) with the change of independent variables (factors). The response surface graphs are shown in Figure 1. Among the seven response variables of value-added cookies, water activity and L value were not shown in the Figure 1 because those responses of value-added cookies showed a similar trend of moisture content and total color difference, respectively. The response surface models produced 3-dimensional graphs for each response showing the effect of wheat flour and pumpkin flour keeping cranberry pomace powder constant (1A), the effect of pumpkin flour and cranberry pomace powder keeping wheat flour constant (1B) and the effect of cranberry powder and wheat flour keeping pumpkin flour constant (1C) on the response variables of moisture content, total color difference, spread ratio, piece density and hardness of value-added developed cookies.

The wheat flour showed a negative linear effect on moisture content and water activity, a slight quadratic effect on total color difference and L value and a linear effect on spread ratio, piece density and hardness of the value-added developed cookies (Figure 1A and Figure 1C). The results indicated that the increased wheat flour to 55% in cookies decreased the moisture content and water activity to 6.10% and 0.41 and increased spread ratio,

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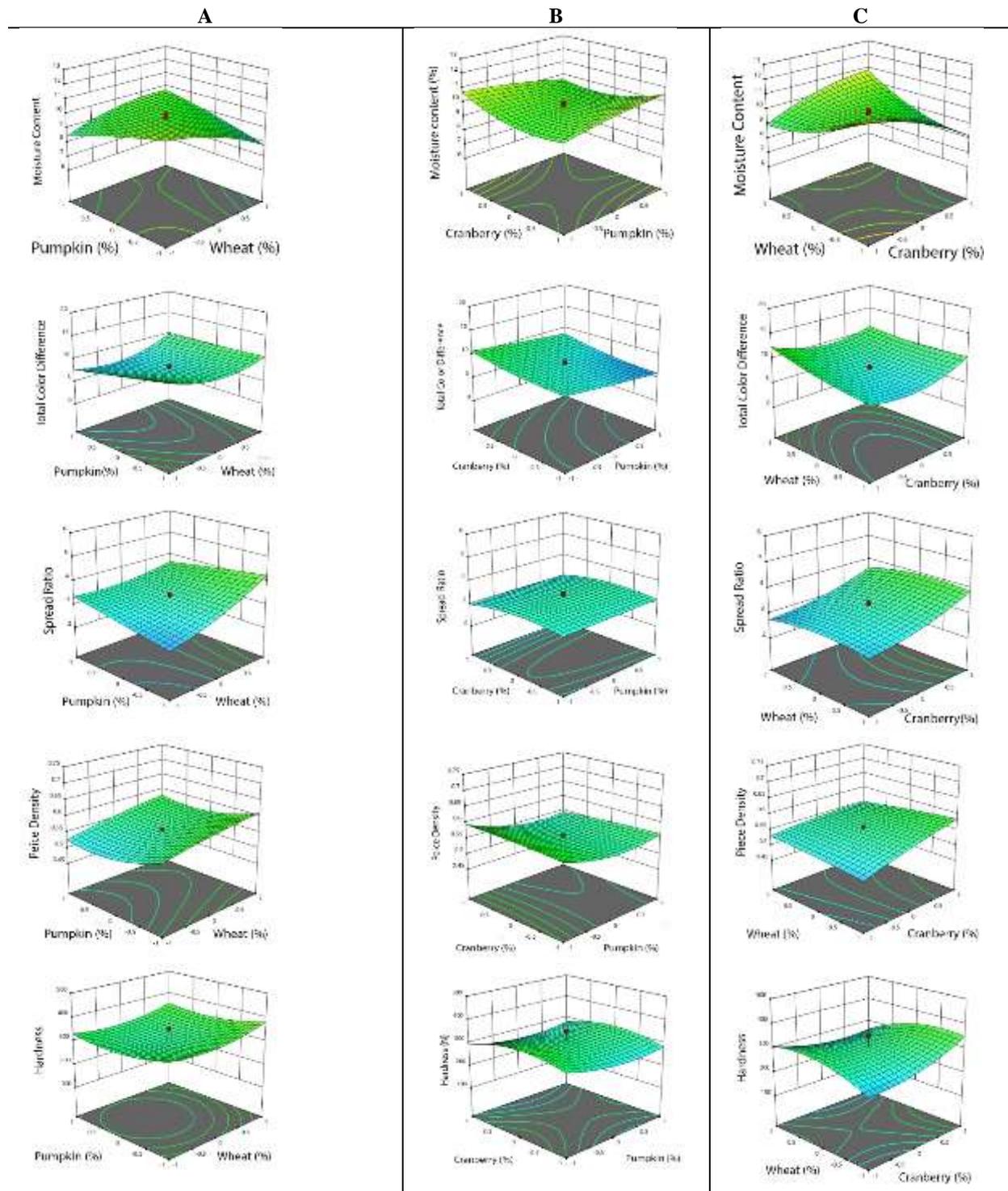


Figure 1. Effect of wheat flour and pumpkin flour (A), effect of pumpkin flour and cranberry pomace powder (B) and effect of wheat flour and cranberry pomace powder (C) on the moisture content, total color difference, spread ratio, piece density and hardness of value-added cookies.

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piece density and hardness of the value-added developed cookies to 3.57, 0.61 and 496.06, respectively. The total color difference of the cookies slightly decreased with up to 50% wheat flour and further increased with increased wheat flour. A study on carrot and wheat flour mixed cookies showed that the water holding capacity of wheat flour of the cookies reduced when the particle size of wheat flour was reduced (72-120 mesh). However, the water holding capacity increased when carrot pomace powder with a higher particle size was mixed with the wheat flour (Ahmad et al., 2016). The lower particle size of wheat flour might cause a lower water holding capacity, subsequently, the increased wheat flour decreased the moisture content of the developed cookies. Initially the whitish wheat flour might cause the slight decreased of total color difference of the cookies but further increased wheat flour formed the brown color during baking at high temperature due to Millard reactions, dextrinization of starches and caramelization of sugars (Jan et al., 2016; Jukić et al., 2019). This phenomenon might cause the quadratic effect of wheat flour on the total color difference of the developed cookies. The spread ratio of cookies is important for the quality and packaging design of cookies. The spread ratio depends on the viscosity of the dough of cookies. The anti-plasticizing effect of sugar and the effect of shortening regulate the gluten proteins cross-linking during baking that control the viscosity of the doughs. Starch gelatinization is occurred in a limited extent due to low moisture content and high sugar content of doughs. Non-gelatinized starches, proteins and shortening granules develop non-continuous structural network of cookies (Manley, 2000; Jukić et al., 2019). These might cause the increased spread ratio of value-added developed cookies with increased wheat flour. The results were in the agreement with other studies (Jan et al., 2016; Jukić et al., 2019). The reduced moisture content of dried saskatoon berries (Meda et al., 2016) and ginger candy (Bhaktaraj et al., 2019) increased the compaction and reduced porosity, subsequently, increased the piece density of the products. Similarly, the increased wheat flour contents with a reduced moisture content might increase the compaction and decreased the porosity of the value-added cookies. Hence, the piece density of value-added cookies increased with wheat flour. Hardness of products is most important textural properties for the quality assessment of food products. The moisture content is inversely related to the hardness and density is linearly related to the hardness of the products (Meda et al., 2016; Bhaktaraj et al., 2019; Timalisina et al., 2019). The lower moisture content and higher piece density of value-added cookies might increase the hardness of the cookies with wheat flour.

The incorporation of pumpkin flour showed the moisture content was inversely related to pumpkin flour, whereas, the total color difference and spread ratio of the value-added cookies were linearly related to pumpkin flour (Figure 1A and Figure 1B). But the pumpkin flour showed quadratic effect on the piece density and hardness of the cookies (Figure 1A and Figure 1B). The baking process at a high temperature might manipulate the water holding capacity of pumpkin flour which increased the water loss during baking with the increased pumpkin flour (Mildner-Szkudlarz et al., 2016). This might be the increased percentage of pumpkin flour in cookies which resulted in decreased moisture contents in value-added cookies. The yellowish color of pumpkin flour influenced the total color difference during baking. This might be the increased total color difference with pumpkin flour of the cookies (Pongjanta et al., 2006; Jukić et al., 2019). Besides added shortening in the cookie formulations, the pumpkin flour contained pumpkin oil (Jukić et al., 2019). The additional pumpkin oil melted during baking and made the dough less viscous. This phenomenon might cause the increased spread ratio of the cookies with pumpkin flour. The piece density of the cookies followed the similar trend (quadratic) of the hardness of cookies because piece density and hardness of food products were correlated (Bhaktaraj *et al.*, 2019). The results were in the agreement with other studies (Pongjanta et al., 2006; Das and Banerjee, 2015; Jukić et al., 2019).

The cranberry pomace powder showed a linear effect on moisture content, total color difference, spread ratio, piece density and hardness of the value-added developed cookies (Figure 1B and Figure 1C). The fiber content and porosity of cranberry pomace might increase the water holding capacity of cranberry pomace and the water of the doughs might be entrapped into the pore spaces of cranberry pomace. This might cause the less water loss from the doughs due to evaporation during baking (Ahmad et al., 2016; Mildner-Szkudlarz et al., 2016), subsequently, the value-added cookies had increased moisture content with increased cranberry pomace. The reddish cranberry pomace might dominate on the whitish wheat flour and increased the total color difference of cookies with the increased cranberry pomace. The trend of the total color difference of the value-added developed cookies was similar to other pomace-based cookies and muffins products (Mildner-Szkudlarz et al., 2016; Šarić et al., 2016; Tańska et al., 2016). The higher moisture content of dough and less gluten development (Ahmad et al., 2016) because of higher fiber from cranberry pomace of dough might decreased the viscosity of doughs. It might be decreased viscosity or less viscous dough that cause the increased spread ratio of value-added cookies during baking. The higher moisture content of the cookies might increase the piece density of cookies with cranberry pomace. A study on

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ginger candy showed that the piece density of ginger candy increased with the increased moisture content of ginger candy (Bhaktaraj et al., 2019). The sugar content with moisture of doughs might build strong composites with the fiber and other ingredients of cranberry pomace which increased the hardness of value-added cookies. Addition of carrot pomace in cookies (Ahmad et al., 2016), raspberry and blueberry pomace in cookies (Šarić et al., 2016) and raspberry and cranberry pomace in muffins (Mildner-Szkudlarz et al., 2016) was found to increase the hardness of the products.

Table 4. Results of numerical optimization using desirability function of value-added cookies

Parameter	Goal	Lower limit	Upper limit	Lower weight	Upper weight	Importance
Wheat	Minimize	-1	1	1	1	3
Pumpkin	Maximize	-1	1	1	1	3
Cranberry pomace	Maximize	-1	1	1	1	3
Moisture content	Minimize	6.1	12.7	1	1	3
Water activity	Minimize	0.41	0.68	1	1	3
L value	In range	26.69	37.18	1	1	3
Total color difference	In range	3.7	18.56	1	1	3
Spread ratio	In range	2.12	5.73	1	1	3
Piece density	Minimize	0.46	0.71	1	1	3
Hardness	Minimize	192.53	496.06	1	1	3

Solutions

No	Wheat flour (%)	Pumpkin flour (%)	Cranberry pomace powder (%)	Moisture content (%)	Water activity	L value	Total color difference	Spread ratio	Piece density (gm/mL)	Hardness	Desirability
1	-1	0.950	1	7.63	0.56	36.43	10.14	3.18	0.51	219.32	0.82
2	-1	0.958	1	7.64	0.56	36.46	10.12	3.20	0.51	217.55	0.82
3	-1	0.940	1	7.62	0.56	36.40	10.17	3.17	0.51	221.41	0.82
4	-1	0.930	1	7.61	0.56	36.36	10.21	3.15	0.51	223.62	0.82
5	-1	0.916	1	7.59	0.56	36.31	10.25	3.13	0.51	226.65	0.82

3.3. Optimization of value-added cookies formulation

3.3.1. Numerical optimization using a desirability function

The response variables moisture content, water activity, L value, total color difference, spread ratio, piece density and hardness of value-added cookies were optimized numerically using a desirability function. The desirability of cookies was a combination of all response variables into a single response by a mathematical technique (Meda et al., 2016; Timalina et al., 2019). The desirability function ranges from 0 – 1 which is calculated as geometric mean, and the mean value close to 1 shows the highest desirability. The desirable level for each response variable can be selected as minimum, maximum, in range or target which can be varied as per the objectives of the study. The objective of our study was to minimize wheat flour, maximize pumpkin flour and cranberry pomace powder to minimize moisture content, water activity, total color difference, piece density and hardness and to get L value and spread ratio in range. The optimum conditions were determined by conducting several trials. The final optimum condition was obtained when the wheat flour was minimized, and pumpkin flour

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and cranberry pomace powder were maximized considering the highest incorporation of pumpkin flour and cranberry pomace in the cookies to improve the value addition in terms of nutrition and economy.

Comparing all trial tests, the five best (based on a desirability value) solutions are shown in Table 4. It can be concluded from Table 4 that the solution number 2 (desirability 0.82) gave an optimum condition to develop desirable quality value-added cookies. The optimum results (Table 4) indicated that the desirable formulation had wheat flour (45%), pumpkin powder (34.79%) and cranberry pomace powder (25%) with a desirability of 0.82. The cookies with an optimum formulation had moisture content (7.64%), water activity (0.56), L value (36.46), total color difference (10.12), spread ratio (3.20), piece density (0.51g/mL) and hardness (217.55 N).

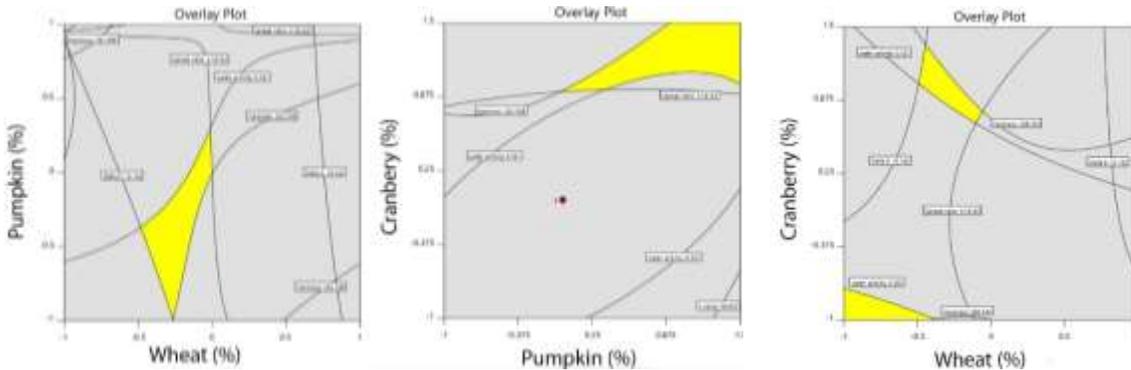


Figure 2. Graphical optimization (shaded area) of wheat flour, pumpkin flour and cranberry pomace for the moisture content, water activity, L value, total color difference, spread ratio, piece density and hardness of value-added cookies

3.3.2. Graphical optimization using overlay of contours of response variables

A significant comprehension can be developed to determine the optimum region of the cookie formulations by the graphical optimization with the combination of numerical optimization. The numerical optimized solution (Table 4) was visualized graphically to determine the optimum regions where all constraints were satisfied. The contour plots of all response variables (moisture content, water activity, L value, total color difference, spread ratio piece density and hardness) as a function of two factors (wheat flour and pumpkin flour, pumpkin flour and cranberry pomace powder and cranberry pomace powder and wheat flour) keeping the third variable constant at the center point were overlaid all together and visualized the optimum regions (shaded areas in the overlaid plots) as shown in Figure 2 (Meda et al., 2016; Timalisina et al., 2019). The contour plots of all response variables with shaded regions (Figure 2) showed an optimum ranged 45-50% wheat flour, 30-35% pumpkin flour and 24-27% cranberry pomace powder.

4. Conclusions

The optimization based on numerical and graphical solutions revealed that 45-50% wheat flour of cookies could be replaced by combining 24-27% cranberry juice industry waste of dried cranberry pomace and 30-35% pumpkin flour to develop value-added cookies because of their higher nutrient contents (i.e. high fiber, antioxidant, vitamins and minerals etc.). The response surface methodology (RSM) combining with central composite rotatable design (CCRD) was very effective to develop regression predictive models response surface models to determine the relationship between independent variables (% wheat flour, % pumpkin flour and % cranberry pomace powder) and response variables (moisture content, water activity, L value, total color difference, spread ratio, piece density and hardness). The regression modeling results indicated that the models could predict the response variables as a function of independent variables with an accuracy of 59-99% depending on the responses. The response surface models indicated that the moisture content and water activity of cookies decreased with wheat flour and pumpkin flour and increased with cranberry pomace powder. The L value increased with wheat flour but decreased with pumpkin flour and cranberry pomace. Total color difference decreased with wheat flour and increased with pumpkin flour and cranberry pomace powder. The spread ratio, piece density and hardness increased with wheat flour,

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pumpkin flour and cranberry pomace powder. The results could lead to develop highly nutritious wheat flour cookies by supplementing a combination of up to 50% pumpkin flour and cranberry pomace. The results can be valuable in decision making for bakery industries to take nutritional and economic benefits of pumpkin flour and cranberry pomaces as alternative/supplement to wheat flour.

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