OPTIMIZATION OF MANUFACTURE OF ALMOND PASTE COOKIES USING RESPONSE SURFACE METHODOLOGY

STEFANO FARRIS[1] and LUCIANO PIERGIOVANNI

Department of Food Science and Microbiology (diSTAM)
University of Milan
via Celoria 2, 20133, Milan, Italy

Accepted for Publication September 24, 2007

ABSTRACT

The response surface methodology (RSM) was used in the optimization of a modified recipe of almond paste cookies to pinpoint the best combination of the most important factors, in order to obtain an enhanced product with quality characteristics similar to the typical one. In this modified recipe, bamboo fiber and fructose were added as humectant and anticrystallizing agents to extend the shelf life of the cookies. Five quantitative controllable factors were selected for the experimental design: weight of bamboo fiber, fructose/saccharose ratio (F/S) and weight of egg white as ingredients, and baking time and temperature as process parameters. To assess the product quality, texture, moisture content and color were considered as dependent variables. The three second-order polynomial models obtained by RSM and the subsequent optimization step indicated that a formulation with 28.95 g of fiber, 252.5 g of egg white, an F/S equal to 0.1, baking time and temperature of 21.5 min and 185.5C, respectively, represents the best recipe to manufacture a new product with quality attributes very similar to the typical one and, in addition, with an extended shelf life.

PRACTICAL APPLICATIONS

The importance of this work arises from the possibility to obtain new formulations by the enhancement of original recipes of products with a very limited shelf life using a statistical and designed approach. The achieved results led to a higher quality of these products over time, without changing the most important characteristics related to consumer acceptance.

[1] Corresponding author. TEL: +39-02-50-31-6660; FAX: +39-02-50-31-6672; EMAIL: stefano.farris@unimi.it

Journal of Food Process Engineering **32** (2009) 64–87.
DOI: 10.1111/j.1745-4530.2007.00203.x

INTRODUCTION

Almond-based cookies are very popular in all countries. They are made from either ground sweet almonds or sweet almond paste, along with sugar and egg white, although the percentages of these ingredients can vary as a function of the desired final product; sometimes, they can be flavored with bitter almonds, chocolate or liqueurs, and other substances can be added (e.g., water, glucose). They are named in a different way depending on the place of origin: almond macaroon cookies (U.S.A. and England), speculaasjes (Holland), mandelkuchen (Germany), bondkakor (Sweden), kourabiethes (Greece), macarons aux amandes (France), polvorones and perrunillas (Spain), amaretti (Italy), makroud el louse (Algeria), mandelbrot (Israel) and alfafores de almendras (Argentina), for example. Traditionally served with a sweet dessert wine or liqueur, they are now eaten at any time of the day: with tea, during coffee breaks, as a snack, for dessert, with a bowl of ice cream and even given as a welcoming gift.

They can be grouped into two main categories: on one hand, completely hard and dry biscuits, and on the other hand, cookies with a soft and moist internal paste, well differentiated from the hard and dry external crust. The latter is a typical example of multidomain food (Labuza and Hyman 1998). In both cases, quality preservation is of primary importance to promote their diffusion outside of the country, although the extension of their shelf life is strictly related to the specific type taken into account. In fact, for the dry biscuits, a proper barrier packaging could represent the best solution to maintain the original characteristics, avoiding, in particular, moisture gain from the surrounding environment. On the contrary, the quality preservation of multidomain moist cookies appears much more complicated because of the different chemophysical phenomena that take place simultaneously during storage (e.g., water redistribution, sugar crystallization) that lead to severe hardening of the internal paste. This fact strictly limits the shelf life of these cookies, negating their marketing in countries far from the traditional places of manufacturing (Piga *et al.* 2005). Hence, different synergetic tools should be used, such as a suitable packaging solution, the application of an edible layer between crust and paste and the modification of the original recipe. As far as this last aspect is concerned, several ingredients and/or additives can be utilized to inhibit the moisture loss over time, changing the water activity (A_w) of the systems, the effective diffusivity of the water and the viscosity (molecular mobility) in the entrapped amorphous phases. In the case of this kind of cookies, one of the most important tasks is to reduce the A_w gradient between the different components (crust and paste) in order to decrease the rate of hardening. In a previous work (Farris *et al.* 2006), the effect of two different humectant agents (glucose syrup and bamboo fiber) on the quality of almond paste pastries was tested with successful results; both ingredients lowered the initial A_w of the cookies and reduced the A_w gradient between the paste and crust, slowing down the moisture loss and then extending the shelf life. However, severe modification of the original characteristics appeared at the same time (dark color, lost in crispness, viscosity of the internal paste), affecting the typical characteristics of these cookies that are greatly appreciated by consumers. This fact suggests that it is necessary to combine different factors (e.g., the ingredients) in such a way that collateral effects do not appear, but only the goal of shelf-life extension is achieved.

In this research, two different ingredients were added to an original recipe of almond paste cookies. Fructose was chosen as the humectant and anticrystallizing agent (Guilbert 2002). Because of its colligative properties, it can be used to lower the A_w of different foods because of its high water-binding capacity. As a result, this leads to a greater microbial stability, which keeps the same moisture content within the food. Hence, fructose can be used to improve food quality (controlling microbial growth, unwanted crystallizations and improving texture) and to extend the shelf life of different foods (Hanover and White 1993). Bamboo fiber is a cellulose-based insoluble ingredient derived from the fiber-rich parts of bamboo plants. It is characterized by total fiber content greater than 99% (dry basis), a high pectin and hemicellulose content, a length of about 0.25 mm and a density of about 120 g/L. This purified fiber has a water-holding capacity of 8.7 g/g fiber, calculated after centrifugation, showing that it is able to retain water not only by capillary forces but also by specific bonds (data provided by the supplier). For these reasons, the addition of bamboo fiber makes it possible to lower the A_w value of different food systems considerably. It can contribute to beneficial textural properties as it is considered as a texturizing agent, improving mouthfeel. Finally, fibers are credited in having a number of functional properties, including the shortening of intestinal transit time, the slowing down of carbohydrate absorption (controlling the glucose level in diabetic subjects), the control of total cholesterol level in the blood and weight management (Bessesen 2001; De Vries 2003; Lupton and Turner 2003).

Five main factors affecting the quality characteristics of the cookies were selected for the present research: fructose/saccharose ratio (from now on indicated as F/S), weight of bamboo fiber and weight of egg white were selected as ingredients, whereas baking time and baking temperature as processing variables. To pinpoint the best recipe, these five factors needed to be set in a proper combination. To achieve this goal, the design of experiment technique was used. It is a powerful tool that provides the maximum information on the studied system than any other approach by a narrow number of trials, with the advantage of evaluating multiple parameters and their interactions, minimizing costs and time of research (Box *et al.* 1978). In particular, the optimization step was carried out using the response surface methodology (RSM), to discover the conditions that produce the best output (Araujo and Brereton 1996). RSM is the most popular optimization method used in recent years, as shown by the many works based on the application of this technique in chemical and biochemical process (Bas and Boyaci 2007). RSM has also been shown to be a useful tool in the food field, for the development of new products and processes, the enhancement of existing products and processes, the optimization of quality and performance of a product, the optimization of an existing manufacturing procedure and the minimization of production cost, as highlighted by different authors. This technique was used to evaluate the effects of different factors on quality attributes (Elbert *et al.* 2001; Chetana *et al.* 2004; Affrifah and Chinnan 2005; Calzetta Resio *et al.* 2006), to optimize different process conditions (Chen *et al.* 2005; Lee *et al.* 2006; Liu-ping *et al.* 2006; Milan-Carrillo *et al.* 2006) or in the optimization of particular formulations (Jackson *et al.* 1996; Varnalis *et al.* 2004; Colla *et al.* 2006; San Juan *et al.* 2006; Sun *et al.* 2006). However, there are not many examples in the literature concerning the application of RSM to enhance the recipe and the manufacturing procedure of typical food items in order to promote their standardization as well as shelf-life extension. Excellence in formulation and food design may be achieved not only by developing novel food items, but also optimizing and enhancing already existing products (Manzocco and Nicoli 2002).

The aim of this research was to use the RSM technique as a statistical and methodological approach to enhance an already existing recipe, maintaining the quality characteristics of the original one in the new product. At the same time, the development of the mathematical models represents useful tools that are able to predict the effect due to the different settings of factors on the most important characteristics of this kind of cookie (texture, color and moisture content).

MATERIALS AND METHODS

The Manufacture and Storage of the Cookies

Almond paste cookies were made in the department pilot plant, using an already existing procedure and recipe (Piga *et al.* 2005) as shown in Table 1. Table 2 reports the typical mean values for hardness, moisture content and color as the most important attributes of global quality. These values were obtained by analyzing the almond paste cookies provided by 10 different suppliers, using 30 cookies per each lot. For the experimental design, bamboo fiber and crystalline fructose (Chimab s.p.a., Campodarsego, Italy) were added. After baking, the cookies were stored in a climatic chamber (mod. CH

TABLE 1.
INGREDIENTS OF THE ORIGINAL FORMULATION OF THE ALMOND PASTE COOKIES

Ingredients	Amount (% w/w)
Sweet almonds	41.6
Bitter almond aroma	0.4
Egg white	16.4
Saccharose	41.6

TABLE 2.
TYPICAL VALUES OF THE QUALITY ATTRIBUTES OF THE ALMOND PASTE COOKIES

Attribute	Value
Hardness (N × mm)	46 ± 1.75
Moisture content (%)	14 ± 0.80
Color (L^*)	68 ± 3.65

700, Angelantoni Industrie s.p.a., Massa Martana, Italy) under controlled temperature–humidity conditions ($T = 25 \pm 0.5$C and relative humidity $= 40 \pm 2\%$) for 2 h before analyses.

Moisture Content Determination

Gravimetric analysis was performed in triplicate to determine water content (% H_2O on wet basis) following the official method (925.098) of the Association of Analytical Communities (AOAC 1999), using an oven at 105 ± 2.0C for 24 h, by the following equation

$$MC\,(\text{wb}) = \frac{w_{\text{i}} - w_{\text{f,d}}}{w_{\text{i}}} \tag{1}$$

where MC is the moisture content (wet basis), w_{i} is the initial weight (g) of the sample prior to drying and $w_{\text{f,d}}$ is the final dry weight (g) of the sample after drying. The measurements were performed on the internal paste of the cookies, sampling 5 ± 0.1 g of the product from the inner part of the almond paste filling with a laboratory spattle.

Texture Analysis

Hardness was evaluated in the freshly baked cookies using a food texture analyzer (mod. Z005, Zwick Roell, Ulm, Germany). The software "TestXpert

V10.11 Master" (Zwick Roell, Ulm, Germany) was used for data analysis. Textural determination was made on 15 samples for each run by a puncturing test (Bourne 2002). The area under the obtained curve (N × mm), i.e., the total work of the probe to go right through the sample, was considered as an index of global hardness of the cookie (W_{tot}).

Color Measurement

Color analysis was carried out on freshly baked cookies (15 replicates for each run) using a D65 illuminant/10° observer reflection colorimeter (mod. CR 210, Minolta, Osaka, Japan). The cookies were placed on a white standard plate ($L^* = 100$) and the Commission Internationale de l'Eclairage (CIE) $L^*a^*b^*$ coordinates were simultaneously measured, where a^* is the CIE redness/greenness value, b^* is the CIE yellowness/blueness value and L^* is the CIE lightness/darkness value. Only the L^* parameter was considered in the experimental design.

Statistical Analysis

Data were subjected to one-way analysis of variance (ANOVA) using Statgraphics Plus 4.0 software, followed by least significant difference (LSD) multiple range test ($P \leq 0.05$) for comparison of the mean values. Modde software package (Modde 8.0, 2006, Umetrics AB, Umea, Sweden) was used to generate the experimental design, to evaluate raw data and for regression analysis, according to the least square analysis, based on the multiple linear regression.

Experimental Design

In designing this experiment by RSM, a central composite face-centered (CCF) design supporting quadratic models was employed. In general, central composite designs contain an imbedded factorial or fractional factorial design with center points that is augmented with a group of "star points," allowing estimation of curvature. If the distance from the center of the design space to a factorial point is ± 1 unit for each factor, the distance from the center of the design space to a star point is $\pm\alpha$ with $|\alpha| \geq 1$. In particular, for CCF designs, the "star points" are at the center of each face of the factorial space, so $\alpha = \pm 1$. The quantitative controllable factors (independent variables) were five: weight of bamboo fiber (X_1), F/S ratio (X_2) and weight of egg white (X_3) as ingredients, and baking temperature (X_4) and baking time (X_5) as process conditions. Each of these independent variables was assessed at three levels (−1, 0 and +1). Three dependent variables were selected as responses for representing the main parameters of cookie quality: hardness (Y_1), moisture content (Y_2) and color (Y_3). A total of 31 combinations (16 corner points, 10 star points and 5 center points) were chosen in random order in order to avoid any influence of external and/or extraneous factors possibly affecting the results. The experimental design of the coded (x) and actual (X) levels of variables together with the responses is shown in Table 3. As far as the selection of the regression model is concerned, it was assumed that i mathematical functions, f_z ($z = 1, 2 \ldots i$), exist for each response, Y_z, function of l independent factors, X_k ($k = 1, 2, \ldots l$), such that

$$Y_z = f_z(X_1, X_2, \ldots X_l) \quad (2)$$

where $i = 3$ and $l = 5$. Finally, the f_z function was assumed to be approximated by a second-degree polynomial equation

$$Y_z = b_{z0} + \sum_{k=1}^{l=5} b_{zk} X_k + \sum_{k=1}^{l=5} b_{zkk} X_k^2 + \sum_{k \neq j}^{l=5} b_{zkj} X_k X_j + \varepsilon \quad (3)$$

where

b_{z0} = response value with all factors set at medium level (center point);
b_{zk} = linear regression coefficient;
b_{zkk} = quadratic regression coefficient;
b_{zkj} = interaction regression coefficient; and
ε = residual response variation not explained by the model.

Experimental data were then fitted to the selected regression model to achieve a proper understanding of the correlation between each factor and different responses. This was obtained by estimating the numerical values of the model terms (regression coefficients), whose significance was statistically judged in accordance with the t-statistic at a confidence interval of 95%. ANOVA and F-test at a probability (P) of 0.05 were carried out to assess the global validity of the models to explain the actual relationship among factors and responses. Response surface plots were then generated, and the optimal combination of factors was finally pinpointed.

RESULTS AND DISCUSSION

Statistical Analysis

Table 4 shows the linear, quadratic and interaction coefficients obtained from the fitting of the experimental data (Table 3) to the quadratic model for

TABLE 3.
WORKSHEET OF THE CENTRAL COMPOSITE FACE-CENTERED EXPERIMENTAL DESIGN

Experiment	Run order	Independent variable					Dependent variables		
		Fiber (g) $X_1(x_1)$	F/S (g/g) $X_2(x_2)$	Egg white (g) $X_3(x_3)$	Temperature (C) $X_4(x_4)$	Time (min) $X_5(x_5)$	Hardness (N × mm) Y_1	Moisture content (%) Y_2	Color (L*) Y_3
1	19	20 (−1)	0.1 (−1)	250 (−1)	165 (−1)	22 (+1)	19.81	15	85.46
2	4	30 (+1)	0.1 (−1)	250 (−1)	165 (−1)	18 (−1)	15.3	15.27	86.42
3	1	20 (−1)	0.38 (+1)	250 (−1)	165 (−1)	18 (−1)	13.7	18.33	86
4	20	30 (+1)	0.38 (+1)	250 (−1)	165 (−1)	22 (+1)	35.3	17.47	80.66
5	18	20 (−1)	0.1 (−1)	265 (+1)	165 (−1)	18 (−1)	9.9	17	82.91
6	5	30 (+1)	0.1 (−1)	265 (+1)	165 (−1)	22 (+1)	29.95	15.16	86.65
7	17	20 (−1)	0.38 (+1)	265 (+1)	165 (−1)	22 (+1)	19.42	18.99	82.61
8	16	30 (+1)	0.38 (+1)	265 (+1)	165 (−1)	18 (−1)	16.99	18.75	84.12
9	10	20 (−1)	0.1 (−1)	250 (−1)	195 (+1)	18 (−1)	44.27	14.96	75.18
10	14	30 (+1)	0.1 (−1)	250 (−1)	195 (+1)	22 (+1)	56.32	13.59	61.22
11	21	20 (−1)	0.38 (+1)	250 (−1)	195 (+1)	22 (+1)	45.97	15.5	53.24
12	8	30 (+1)	0.38 (+1)	250 (−1)	195 (+1)	18 (−1)	39.02	16.63	63.04
13	13	20 (−1)	0.1 (−1)	265 (+1)	195 (+1)	22 (+1)	32.71	12.31	62.49
14	11	30 (+1)	0.1 (−1)	265 (+1)	195 (+1)	18 (−1)	34.66	15.33	69.41
15	9	20 (−1)	0.38 (+1)	265 (+1)	195 (+1)	18 (−1)	30.1	17.08	57.75
16	3	30 (+1)	0.38 (+1)	265 (+1)	195 (+1)	22 (+1)	46.73	16.01	49.86
17*	12	25 (0)	0.24 (0)	257.5 (0)	180 (0)	20 (0)	31.83	15.99	74.82
18*	6	25 (0)	0.24 (0)	257.5 (0)	180 (0)	20 (0)	31.2	16.58	75.48
19*	7	25 (0)	0.24 (0)	257.5 (0)	180 (0)	20 (0)	29.41	16.61	71.77
20*	15	25 (0)	0.24 (0)	257.5 (0)	180 (0)	20 (0)	29.66	16.64	73.33
21*	2	25 (0)	0.24 (0)	257.5 (0)	180 (0)	20 (0)	31.18	16.26	75.39
22	30	20 (−1)	0.24 (0)	257.5 (0)	180 (0)	20 (0)	30.49	14.85	70.29
23	23	30 (+1)	0.24 (0)	257.5 (0)	180 (0)	20 (0)	34.19	16.08	70.63
24	31	25 (0)	0.1 (−1)	257.5 (0)	180 (0)	20 (0)	33.92	15.44	72.03
25	24	25 (0)	0.38 (+1)	257.5 (0)	180 (0)	20 (0)	35.45	17.61	64.97
26	22	25 (0)	0.24 (0)	250 (−1)	180 (0)	20 (0)	29.01	17.33	74.44
27	27	25 (0)	0.24 (0)	265 (+1)	180 (0)	20 (0)	28.06	18.27	75.24
28	29	25 (0)	0.24 (0)	257.5 (0)	165 (−1)	20 (0)	22.71	17.53	8.8
29	25	25 (0)	0.24 (0)	257.5 (0)	195 (+1)	20 (0)	42.5	15.47	5.45
30	26	25 (0)	0.24 (0)	257.5 (0)	180 (0)	18 (−1)	26.3	17.45	78.58
31	28	25 (0)	0.24 (0)	257.5 (0)	180 (0)	22 (+1)	36.89	15.49	62.73

X represents the actual level of factors.
x represents the coded level of factors.
* Replicates at the center points.
F/S, fructose/saccharose ratio.

TABLE 4.
ESTIMATED SIGNIFICANT COEFFICIENTS OF THE FITTED EQUATIONS FOR THE DIFFERENT RESPONSES

Coefficients	Hardness (Y_1)	Moisture content (Y_2)	Color (Y_3)
b_0	31.5558	–1.92148	0.412381
b_1	3.44944	5.02273 × 10^{-5}*	–0.00357185*
b_2	0.324445*	0.00641684	–0.0688784
b_3	–2.78778	0.00141909	–0.0178407*
b_4	10.5111	–0.00478683	–0.263433
b_5	5.15889	–0.00321875	–0.0662682
b_{11}	0.20372*	–0.00659658	–7.16091 × 10^{-4}*
b_{22}	2.54872	–0.00109152*	–0.0387073*
b_{33}	–3.60128	0.00556382	0.0952176
b_{14}	–0.689373*	0.00140832	–0.00947241*
b_{15}	2.64938	5.58668 × 10^{-4}*	–0.00749038*
b_{24}	–1.03687	–7.49086 × 10^{-4}*	–0.035865
b_{34}	–2.09562	–0.00122518	–0.0109053*
b_{35}	–0.931876*	–0.00121295	–0.0379559

Subscripts: 1 = fiber; 2 = fructose/saccharose ratio; 3 = egg white; 4 = temperature; and 5 = time.
* Not statistically significant coefficient at $P \leq 0.05$ (or 95% confidence interval).

each response. The regression coefficients not significant at 95% level were removed and the model refitted to the data. Only the other ones inside of this confidence interval were selected for developing the models as follows:

$$\text{Hardness}(Y_1) = 31.55 + 3.45X_1 - 2.79X_3 + 10.51X_4 + 5.16X_5 + 2.55X_2^2 - 3.60X_3^2 + 2.65X_1X_5 - 1.04X_2X_4 - 2.09X_3X_4 \quad (4)$$

$$\text{Moisture content}(Y_2) = -1.92 + 0.0064X_2 + 0.0014X_3 - 0.0048X_4 - 0.0032X_5 - 0.0066X_1^2 + 0.0055X_3^2 + 0.0014X_1X_4 - 0.0012X_3X_4 - 0.0012X_3X_5 \quad (5)$$

$$\text{Color}(Y_3) = 0.41 - 0.0069X_2 - 0.26X_4 - 0.0662X_5 + 0.0952X_3^2 - 0.0258X_2X_4 + 0.0379X_3X_5 \quad (6)$$

Once the models were obtained, ANOVA was calculated to verify their capability to represent the data. The ANOVA for all three responses is presented in Table 5. For each model, the mean square regression (i.e., the variance explained by the model) in the first F-test was significantly larger than the mean square residual (i.e., the amount of variance unexplained by the

TABLE 5.
ANALYSIS OF VARIANCE TABLE FOR THE RESPONSE OF HARDNESS, MOISTURE CONTENT AND COLOR

Source of variation	Y_1 (hardness)				Y_2 (moisture content)				Y_3 (color)			
	df	SS	MS	F*	df	SS	MS	F*	df	SS	MS	F*
Total	31	33,062.5	1,066.53		31	114.603	3.69689		31	7.71686	0.248931	
Constant	1	29,912	29,912		1	114.602	114.602		1	6.13264	6.13264	
Tot. corrected†	30	3,150.44	105.015		30	0.00177002	5.90006×10^{-5}		30	1.58423	0.0528076	
Regression	13	3,088	237.538	64.6732	13	0.00168118	0.000129321	24.7453	13	1.50581	0.115832	25.1123
Residual	17	62.4393	3.6729		17	8.88436×10^{-5}	5.22609×10^{-6}		17	0.0784135	0.00461256	
Lack of fit (model error)	13	57.946	4.45738	3.96801	13	8.02086×10^{-5}	6.1699×10^{-6}	2.8581	13	0.0733405	0.00564158	4.44837
Pure error (replicate error)	4	4.49332	1.12333		4	8.63495×10^{-6}	2.15874×10^{-6}		4	0.00507294	0.00126823	
		$R^2 = 0.980$				$R^2 = 0.949$				$R^2 = 0.951$		
		$Q^2 = 0.893$				$Q^2 = 0.741$				$Q^2 = 0.793$		
		$F_{0.95;\,13;\,17} = 2.35$				$F_{0.95;\,13;\,17} = 2.35$				$F_{0.95;\,13;\,17} = 2.35$		
		$F_{0.95;\,13;\,4} = 5.89$				$F_{0.95;\,13;\,4} = 5.89$				$F_{0.95;\,13;\,4} = 5.89$		

* F ratio is the model significance (regression/residual).
† Total variation (of a selected response) corrected for the average. It is partitioned into two parts: one due to the regression and the other due to the residuals.
df, degrees of freedom; SS, sum of squares; MS, mean square.

model). This means that all three models adequately represented the data for hardness, moisture content and color. Furthermore, the lack of fit test showed that model error and replicate error were small and had similar size, meaning that there was no lack of fit; the models were sufficiently accurate for predicting each corresponding response. The values of the coefficients R^2 and Q^2 confirmed these results. In particular, the correlation coefficient R^2 represents the power of fit; it is a measure of how well the regression model fits the raw data. R^2 ranges between 0 and 1, where 1 is for perfect models. Because it can be easily approached to 1 by adding additional terms in the model, the Q^2 coefficient is needed to assess the power of the model. Q^2 represents the power of prediction and it ranges between 1 and –8; in an optimization design, values equal or higher than 0.5 are judged acceptable.

Effects of Factors on Responses

The effects of the different factors on hardness, moisture content and color were assessed by the regression coefficients of the second polynomial order equations (Table 4). Hardness was positively influenced mainly by baking temperature, which promoted water migration from the core of the cookies, resulting in a stiffer and harder texture not suitable to obtain a final product of better quality (Saxena and Rao 1996). In a similar way, an increase in hardness with increasingly linear fiber content occurred, most likely because of an increase in the water-binding capacity of the paste (Zhou *et al.* 2000; Colleoni-Sirghie *et al.* 2004). Moreover, insoluble fibers are credited to modify the texture of foods in which they are incorporated. It has been evaluated by mechanical tests (e.g., penetrometric measures) that increasing fiber amount leads to a concentration-dependent increase in firmness (Thebaudin *et al.* 1997). In addition, bamboo fiber is characterized by mechanical properties (e.g., tensile strength, tensile modulus, elongation at break and hardness) comparable to those of wood, giving the possibility to use this kind of fiber to produce natural rubber composites (Ismail *et al.* 2002). Baking time also had a positive linear effect on hardness response, because of the proportional amount of water removed from the cookies up to the maximum level. In addition, the interaction effect between the fiber weight and baking time was important, because of the effect of prolonged heat treatments on fiber properties. The higher the baking time, the higher the amount of water loss from the internal paste of the cookies, which may have led to a more rigid fiber frame. Alternatively, the formation of fiber–protein complexes during thermal treatments like cooking, boiling or roasting may play an important role in promoting the hardening of the cookies (Caprez *et al.* 1986). Very few studies have investigated the modifications that dietary fibers suffer during thermal processing and hence, further study is required (Rodriguez *et al.* 2006). Finally, the texture of freshly baked cookies depends on two different quadratic effects: positively by the F/S ratio and to a greater, although negative effect of the egg white weight. Moisture content response, instead, was greatly affected by the quadratic effect exerted by the weight of egg white factor. In addition, three main linear effects influenced this response: F/S ratio (positive effect), and baking temperature and baking time (negative effect). Among them, the link between moisture content and fructose can be explained, taking into account the high hygroscopicity of fructose (Hanover and White 1993). It more readily adsorbs water from the external environment than other sugars (saccharose, for example) if ideal packaging is not used. This needs to be considered in order to avoid caking or the lumping phenomena. Finally, baking temperature was undoubtedly the most important factor affecting the color of the cookies. This is because Maillard reaction occurred more at higher baking temperatures. The other important factors include egg white weight as a quadratic effect, and baking time and F/S ratio as linear effects. As far as F/S ratio is concerned, fructose is technically named a reducing sugar, contrary to saccharose (for that termed nonreducing sugar). The presence of the reducing sugars is fundamental in defining the color of baked products, because they represent the substrates of the Maillard reaction (Lerici *et al.* 1990).

Response Surface Plots

A helpful tool for a better understanding of the link between each factor and response is given by the response surface plots, in which the effect of two factors on one specific response is displayed in 3-D view, keeping the other ones on fixed values. Some selected surfaces predicted by Eqs. (4)–(6) are presented in Figs. 1–7.

Hardness. The synergetic effect of baking time and temperature on the increase of hardness is shown in Fig. 1. The unit change in temperature produced a greater change in response than baking time; so, to obtain new cookies according to the optimized recipe with values of hardness similar to the original one, both baking time and temperature should be set at levels close to the maximum. Figure 2 shows the effect of fiber weight and baking time. The two factors affected hardness response in the same direction, but a greater change in hardness was produced with altering the direction of baking time. A further consideration arises from the twisted shape of this response surface, because of the above-mentioned interaction between factors. In this case, the influence of fiber weight on hardness was greater with baking time set at the highest level; hence, the effect of fiber is strictly dependent on baking time. Hence, an increase in hardness can be achieved by setting both baking time and fiber content to a maximum level. The last response surface for hardness

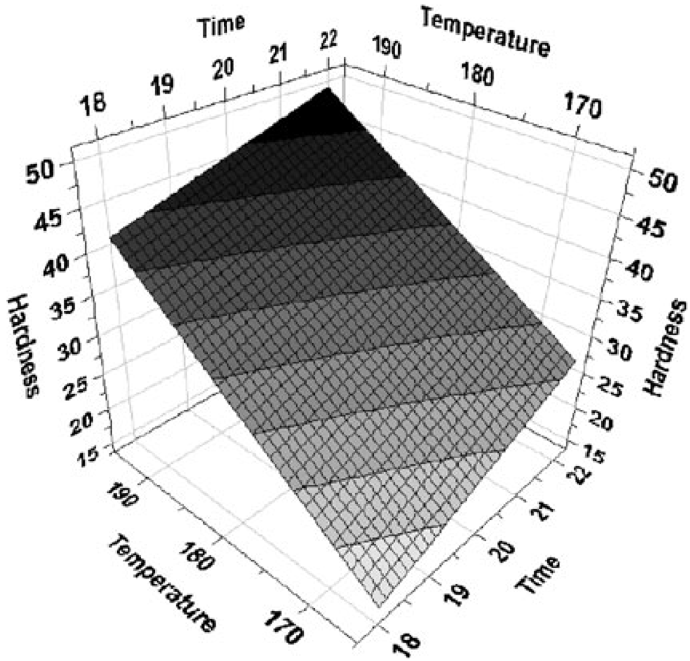

FIG. 1. RESPONSE SURFACE PLOT: EFFECT OF BAKING TIME AND TEMPERATURE ON HARDNESS
Fiber: 25 g; fructose/saccharose ratio: 0.1; egg white: 258 g.

(Fig. 3) was saddle-shaped, because of the quadratic effect of the considered factors (F/S ratio and chiefly egg white). In this case, it is interesting to observe that the highest values in hardness were encountered for an F/S ratio set at the highest or lowest level and egg white always set at medium level. Probably, this is because the last ingredient acted mainly as a plasticizing agent at the highest amounts, leading to the lowest values in hardness ($W_{\text{tot}} \sim 26$ N × mm); however, with egg white set at minimum level, the plasticizing effect disappeared and hardness increased ($W_{\text{tot}} \sim 30$ N × mm), but not enough to be comparable to the original value. Finally, if it is used at medium level, egg white could promote the formation of a lattice that gives strength to the whole matrix (to the crust in particular), resulting in a higher value in hardness ($W_{tot} \sim 35$ N × mm). This is because the egg white proteins, especially ovalbumin, is the most abundant and the only one containing free sulfhydryl (SH) groups. If subjected to heating (80–85C), the egg white proteins denature and active sites become free to create new interactions and new bonds are formed between molecules, which were distant from each other. For instance, the exposed SH groups of ovalbumin undergo oxidation to disulfide bonds (Van

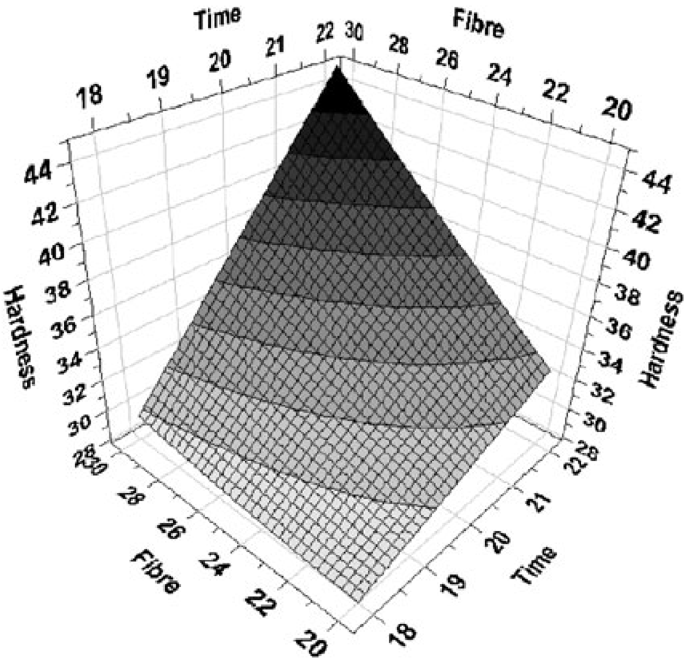

FIG. 2. RESPONSE SURFACE PLOT: EFFECT OF BAKING TIME AND FIBER ON HARDNESS
Temperature: 180C; fructose/saccharose ratio: 0.1; egg white: 258 g.

der Placken *et al.* 2005). The final result is the formation of a new three-dimensional network and the resulting loss in flexibility by the egg white matrix that become stiff and rigid (Gennadios 2002). These hypotheses could be confirmed by in-depth investigation using, for example, the magnetic resonance imaging technique (Labuza and Hyman 1998).

Moisture Content. In Fig. 4, the influence of fiber weight and F/S ratio on moisture content is presented through a bell-shaped curve, where only one quadratic effect exists; moisture content always increased with increasing F/S ratio, but differently depending on the amount of fiber used. According to the developed model, a moisture content close to the original value (~14%) should be measured on the new formulated almond paste cookies using the lowest or the highest amounts of fiber and the minimum amount of fructose compared to saccharose. Figure 5 shows that moisture content depended also on baking temperature and egg white, the latter had mostly a quadratic effect. As predicted, the highest values for moisture content were for the formulation with

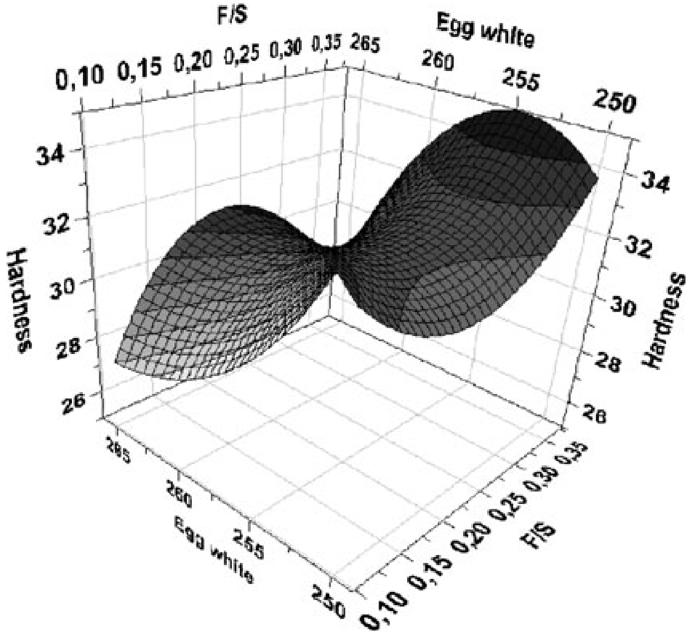

FIG. 3. RESPONSE SURFACE PLOT: EFFECT OF FRUCTOSE/SACCHAROSE RATIO (F/S) AND EGG WHITE ON HARDNESS
Fiber: 25 g; temperature: 180C; time: 20 min.

higher amounts of egg white, with temperature being constant; the higher the baking temperature, the lower the moisture content. Also in this case, the quadratic effect of egg white could be explained by the cross-linking effect of the protein as described above.

Color. As far as the color response, Fig. 6 shows the influence of baking temperature and F/S ratio as main linear effects. The greater change in color was produced by changes in temperature, the F/S ratio also played an important role in promoting nonenzymatic browning. Both the highest baking temperature and F/S ratio led to burnt-like cookies (i.e., the lowest values of L^* parameter). So, the characteristic color of this kind of cookies can be obtained using a small amount of fructose and upper middle temperature values. Other factors affecting the color response were egg white weight and baking time, as quadratic positive and linear negative effect, respectively (Fig. 7). The bell-shaped surface shows that, also in this case, brownness reached maximum with increasing baking time, but differed depending on the amount of egg white used.

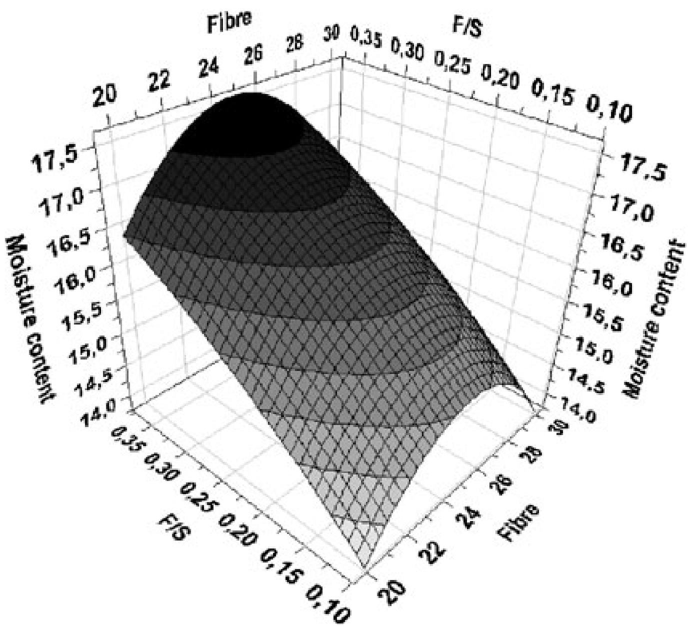

FIG. 4. RESPONSE SURFACE PLOT: EFFECT OF FIBER AND FRUCTOSE/SACCHAROSE RATIO (F/S) ON MOISTURE CONTENT
Egg white: 258 g; temperature: 180C; time: 20 min.

Optimization

On the basis of the above-described results, it can be asserted that the quality of the almond-based cookies is not dependent on a single main factor. All independent variables were important in defining the characteristics of the cookies, even if in a different way, depending on the specific response. So, the next step involved the detection of the best combination of factors that are able to produce the expected characteristics of the final product. All comments arising from the response surface plots were taken into account in the optimization, considering that the optimal solution arises from a compromise among the different responses. In this phase, the criteria of optimization must be selected, that is, a variable response may either be maximized, minimized, excluded or directed towards an interval being the target. In this research, the responses were not maximized or minimized, but each fixed at a target value, corresponding to the actual value of the original cookies (Table 2). Each of these target values ranged from a minimum (L_{min}) to a maximum (L_{max}), as reported in Fig. 8. Moreover, the role of each factor was defined: fiber weight,

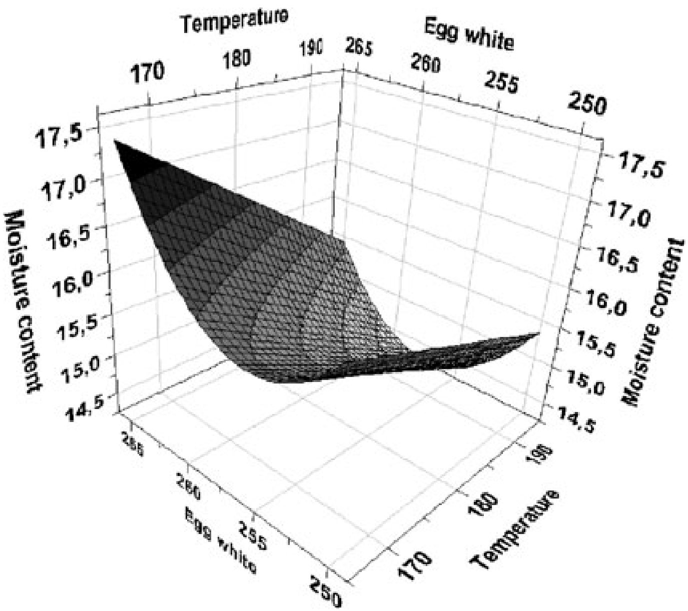

FIG. 5. RESPONSE SURFACE PLOT: EFFECT OF TEMPERATURE AND EGG WHITE ON MOISTURE CONTENT
Fiber: 25 g; fructose/saccharose ratio: 0.1; time: 20 min.

egg white weight, baking time and temperature were allowed to change their value freely; F/S ratio, instead, was fixed at the minimum level (0.1) because of the negative influence of higher levels on all responses, as shown in response surface plots. To obtain an overall desirability function that is able to combine the single desirability of each response, a Nelder–Mead simplex method was used (Nelder and Mead 1965; Walters *et al.* 1991). In particular, for each response (Y_1 = hardness, Y_2 = moisture content and Y_3 = color), the desirability function was:

$$f\left[g\left(Y_z\right)\right] = 100 \times \left(e^{\lambda g(Y_z)} - 1\right) \quad (7)$$

where

$$g\left(Y_z\right) = 100 \times \left[\left(Y_z - P\right)/(V - P)\right] \quad (8)$$

and

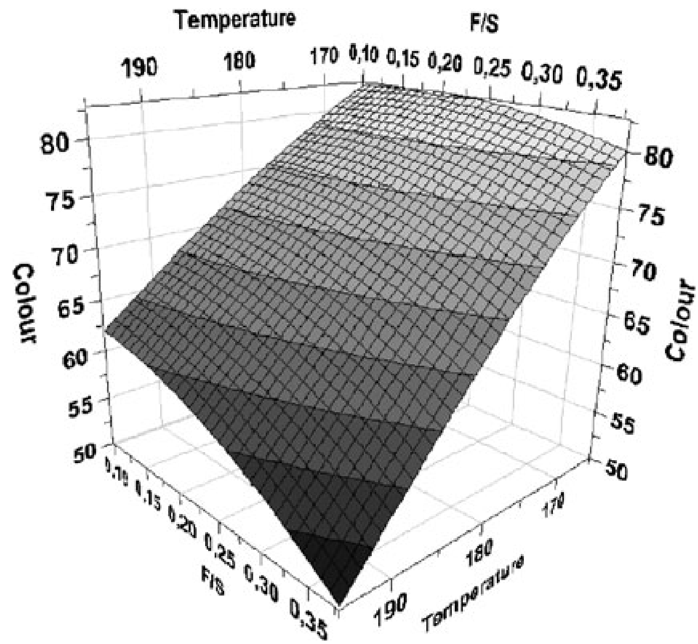

FIG. 6. RESPONSE SURFACE PLOT: EFFECT OF TEMPERATURE AND FRUCTOSE/SACCHAROSE RATIO (F/S) ON COLOR
Fiber: 25 g; egg white: 258 g; time: 20 min.

$$\lambda = -\ln\left(\frac{\dfrac{100}{100 - (90 + 80 \times \log_{10}(\varphi))}}{100 \times \dfrac{(L-P)}{(V-P)}} \right) \quad (9)$$

In Eq. (9)

λ = scaling parameter;
φ = coefficient of importance given to each response. It ranges from 0.1 to 1.0;
P = worst response values computed from the Nelder–Mead simplex method;
L = defined worst acceptable response values (minimum and maximum); and
V = target value.

As a result of the optimization step, the best conditions which were attained for the expected response values were fiber weight: 28.95 g; egg white weight: 252.5 g; F/S ratio: 0.1; baking time: 21.5 min; and baking temperature: 185.5C. According to the models, the predicted values of responses were: hardness

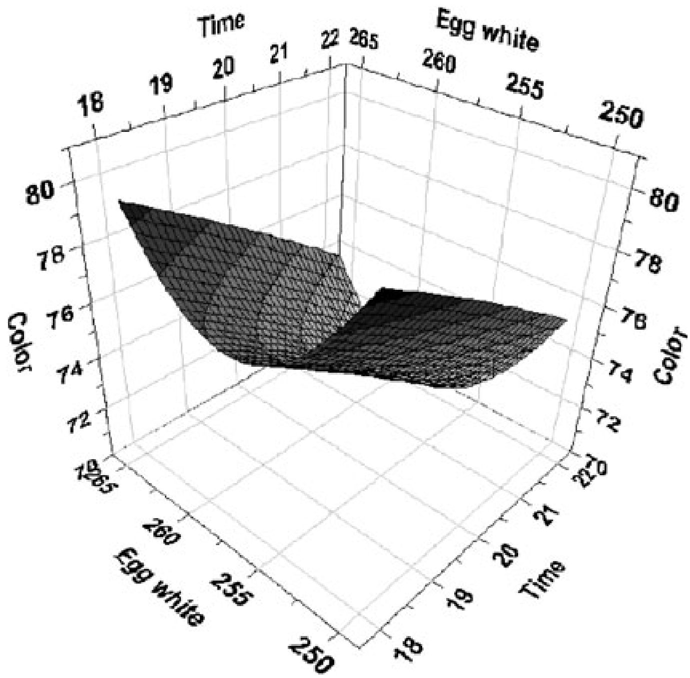

FIG. 7. RESPONSE SURFACE PLOT: EFFECT OF TIME AND EGG WHITE ON COLOR
Fiber: 25 g; fructose/saccharose ratio: 0.1; temperature: 180C.

(W_{tot}): 47.02 N × mm; moisture content (*MC*): 13.89%; color (L^*): 68.5. In Fig. 8, the superimposition of the individual contour plots for each response is presented. The constraints selected in the previous step (i.e., L_{min} and L_{max}) allowed all three responses to meet their target value with the same factor levels. However, this value is inside of an optimum acceptable region (the hatched area) that satisfied all constraints. This means that, in practice, more than a single combination could be used to manufacture almond paste cookies having similar characteristics to the original ones, according to the developed models.

Verification of the Model

Once the best solution had been pinpointed, it was used to manufacture the enhanced cookies. The obtained cookies were analyzed for all three responses, in order to verify the power of prediction of the developed models by comparing theoretical predicted data to the experimental ones. Experiments based on the optimal condition were repeated three times and subjected to one-way ANOVA, with the LSD multiple range test to detect the differences

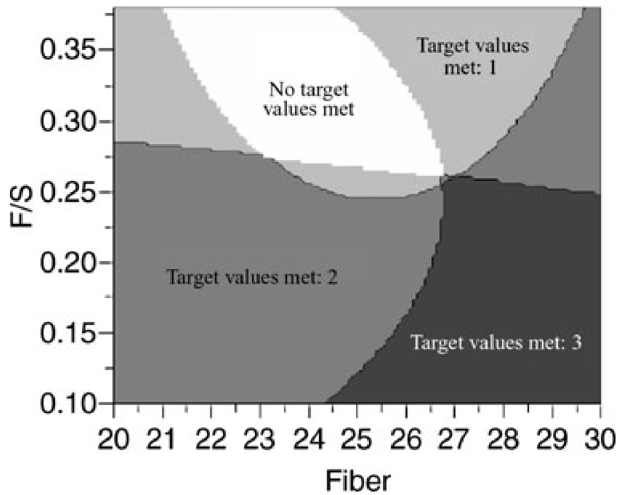

FIG. 8. SUPERIMPOSED CONTOUR PLOTS FOR ALL THE RESPONSES AFFECTED BY FRUCTOSE/SACCHAROSE RATIO (F/S) AND FIBER AMOUNT
The other factors are set according to the optimization. Responses are constrained as follows: hardness (42–50 N × mm); moisture content (12–16%); color as L^* (66–70).

TABLE 6.
PREDICTED AND OBSERVED DATA FOR THE RESPONSES AT OPTIMUM CONDITIONS

Independent variables (responses)	Optimum conditions	
	Predicted value	Observed value
Hardness (N × mm)	47.02	46.85 ± 0.8
Moisture content (%)	13.89	14.05 ± 0.5
Color (L^*)	68.5	67.8 ± 1.5

between predicted and observed values. The obtained results are shown in Table 6. As expected from the high values of R^2 and Q^2, Eqs. (4)–(6) have a satisfactory power of prediction; in fact, experimental values were close to the predicted ones for all three responses, with no statistically significant difference ($P \geq 0.05$ in the F-test at the 95% confidence level).

CONCLUSIONS

RSM was successfully used to pinpoint the best combination of different factors for a modified recipe of typical almond paste cookies. The final goal was to obtain innovative cookies with standardized quality characteristics and with a longer shelf life than the original ones, avoiding those negative effects which take place during manufacture. The statistical approach allowed the achievement of the best recipe (in the investigated experimental region defined by the −1 and +1 level) by the addition of two new ingredients and the handling of the two main process factors. Moreover, the modeling of experimental data allowed the generation of useful equations for general use, to predict the behavior of the system under different factor combinations.

REFERENCES

AFFRIFAH, N.S. and CHINNAN, M.S. 2005. The effect of drying conditions on the development of the hard-to-cook defect steam-treated cowpeas. Int. J. Food Sci. Technol. *40*, 1–11.

AOAC. (1999). *Official Methods of Analysis*, 16th Ed., Association of Analytical Communities, Washington, DC.

ARAUJO, W. and BRERETON, R.G. 1996. Experimental design II. Optimisation. Trends Anal. Chem. *15*, 63–70.

BAS, D. and BOYACI, I.H. 2007. Modeling and optimisation I: Usability of response surface methodology. J. Food Eng. *78*, 836–845.

BESSESEN, D.H. 2001. The role of carbohydrates in insulin resistance. J. Nutr. *131*, 2782S–2786S.

BOURNE, M.C. 2002. *Food Texture and Viscosity: Concepts and Measurement*, 2nd Ed., pp. 113–126, Academic Press, New York.

BOX, G.E.P., HUNTER, W.G. and HUNTER, J.S. 1978. *An Introduction to Design Data Analysis and Model Buildings*, Wiley and Sons, New York, NY.

CALZETTA RESIO, A.N., TOLABA, M.P. and SUAREZ, C. 2006. Effects of steeping conditions on wet-milling attributes of amaranth. Int. J. Food Sci. Technol. *41*(Suppl. 2), 70–76.

CAPREZ, A., ARRIGONI, E., AMADO, R. and NEUCOM, H. 1986. Influence of different types of thermal treatment on the chemical composition and physical properties of wheat bran. J. Cereal Sci. *4*, 233–239.

CHEN, M.-J., CHEN, K.-N. and LIN, C.-W. 2005. Optimisation on response surface models for the optimal manufacturing conditions of dairy tofu. J. Food Eng. *68*, 471–480.

CHETANA, R., MANOHAR, B. and REDDY YELLA REDDY, S. 2004. Process optimisation of Gulab jamun, an Indian traditional sweet, using sugar substitutes. Eur. Food Res. Technol. *219*, 386–392.

COLLA, E., SOBRAL, P.J. and MENEGALLI, F.C. 2006. *Amaranthus cruentus* edible films: Influence of stearic acid addition, plasticizer concentration, and emulsion stirring speed on water vapor permeability and mechanical properties. J. Agric. Food Chem. *54*, 6645–6653.

COLLEONI-SIRGHIE, M., JANNINK, J.L. and WHITE, P.J. 2004. Pasting and thermal properties of flours from oat lines with high and typical amounts of β-glucan. Cereal Chem. *81*, 686–692.

DE VRIES, J. 2003. On defining dietary fibre. Proc. Nutr. Soc. *62*, 37–43.

ELBERT, G., TOLABA, M.P. and SUAREZ, C. 2001. Effects of drying conditions on head rice yield and browning index of parboiled rice. J. Food Eng. *47*, 37–41.

FARRIS, S., LIMBO, S. and PIERGIOVANNI, L. 2006. Effect of two different humectant ingredients on quality of "Amaretti" cookies. In *Proceedings of Shelf Life International Meeting*, Special Issue of Ital. J. Food Scipp. (G. Muratore and F. Licciardello, eds.) pp. 247–254, Chiriotti, Pinerolo, Italy.

GENNADIOS, A. 2002. *Protein-based Films and Coatings*, pp. 66–115, CRC Press, New York, NY.

GUILBERT, S. 2002. Additifs et Agents Depresseur de l'Activitè de l'Eau. In *Additifs et Auxiliares de Fabrication dans les Industries Agroalimentaires*, 3rd Ed. (J.L. Multon, ed.) pp. 237–265, Tec & Doc, Paris, France.

HANOVER, L.M. and WHITE, J.S. 1993. Manufacturing, composition and applications of fructose. Am. J. Clin. Nutr. *58*(Suppl. 5), 724S–732S.

ISMAIL, H., EDYHAM, M.R. and WIRJOSENTONO, B. 2002. Bamboo fibre filled natural rubber composites: The effects of filler loading and bonding agents. Polym. Test. *21*, 139–144.

JACKSON, J.C., BOURNE, M.C. and BARNARD, J. 1996. Optimisation of blanching for crispness of banana chips using response surface methodology. J. Food Sci. *61*(1), 165–166.

LABUZA, T.P. and HYMAN, C.R. 1998. Moisture migration and control in multi-domain foods. Trends Food Sci. Technol. *9*, 47–55.

LEE, W.C., YUSOF, S., HAMID, N.S.A. and BAHARIN, B.S. 2006. Optimising conditions for hot water extraction of banana juice using response surface methodology (RSM). J. Food Eng. *75*, 473–479.

LERICI, C.R., BARBANTI, D., MANZANO, M. and CHERUBIN, S. 1990. Early indicators of chemical changes in foods due to enzymic or nonenzymic browning reactions. 1: Study on heat-treated model systems. Lebensm.-Wiss. Technol. *23*, 289–294.

LIU-PING, F., MIN, Z., GONG-NIAN, X., JIN-CAI, S. and QIAN, T. 2006. The optimisation of vacuum frying to dehydrate carrot chips. Int. J. Food Sci. Technol. *40*, 911–919.

LUPTON, J.R. and TURNER, N.D. 2003. Dietary fibre and coronary disease: Does the evidence support an association? Curr. Atheroscler. Rep. *5*, 500–505.

MANZOCCO, L. and NICOLI, M.C. 2002. Food design: From the methodological approach to the case study of low-calorie syrups. Trends Food Sci. Technol. *13*, 420–427.

MILAN-CARRILLO, J., GUTIERREZ-DORADO, R., PERALEZ-SANCHEZ, J.X.K., CUEVAS-RODRIGUEZ, E.O., RAMIREZ-WONG, B. and REYES-MORENO, C. 2006. The optimisation of the extrusion process when using flour with a modified amino acid profile for making tortillas. Int. J. Food Sci. Technol. *41*, 727–736.

NELDER, J.A. and MEAD, R. 1965. A simplex method for function minimization. Comput. J. *7*, 308–313.

PIGA, A., CATZEDDU, P., FARRIS, S., ROGGIO, T., SANGUINETTI, A. and SCANO, E. 2005. Texture evolution of "Amaretti" cookies during storage. Eur. Food Res. Technol. *221*, 387–391.

RODRIGUEZ, R., JIMENEZ, A., FERNANDEZ-BOLANOS, J., GUILLEN, R. and HEREDIA, A. 2006. Dietary fibre from vegetable products as source of functional ingredients. Trends Food Sci. Technol. *17*, 3–15.

SAN JUAN, E.M., EDRA, E.V., SALES, J.M., LUSTRE, A.O. and RESURRECCION, A.V.A. 2006. Utilization of peanut fines in the optimisation of peanut polvoron using mixture response surface methodology. Int. J. Food Sci. Technol. *41*, 768–774.

SAXENA, D.C. and RAO, P.H. 1996. Optimization of ingredients and process conditions for the preparation of *tandoori roti* using response surface methodology. Int. J. Food Sci. Technol. *31*, 345–351.

SUN, Y., WANG, Z., WU, J., CHEN, F., LIAO, X. and HU, X. 2006. Optimising enzymatic maceration in pre-treatment of carrot juice concentrate by response surface methodology. Int. J. Food Sci. Technol. *41*, 1082–1089.

THEBAUDIN, J.Y., LEFEBVRE, A.C., HARRINGTON, M. and BOURGEOIS, C.M. 1997. Dietary fibres: Nutritional and technological interest. Trends Food Sci. Technol. *8*, 41–48.

VAN DER PLACKEN, I., VAN LOEY, A. and HENDRICKX, M.E. 2005. Combined effect of high pressure and temperature on selected properties of egg white proteins. Innov. Food Sci. Emerg. Technol. *6*, 11–20.

VARNALIS, A.I., BRENNAN, J.G., MACDOUGALL, D.B. and GILMOUR, S.G. 2004. Optimisation of high temperature puffing of potato cubes using response surface methodology. J. Food Eng. *61*, 153–163.

WALTERS, F.H., PARKER, L.R., JR., MORGAN, S.L. and DEMING, S.N. 1991. *Sequential Simplex Optimisation: A Technique for Improving Quality and Productivity in Research, Development, and Manufacturing*, CRC Press, Boca Raton, FL.

ZHOU, M., ROBARDS, K., GLENNIE-HOLMES, M. and HELLIWELL, S. 2000. Effects of enzyme treatment and processing on pasting and thermal properties of oats. J. Sci. Food Agric. *80*, 1486–1494.