# **OPTIMISATION OF MANUFACTURE OF ALMOND PASTE**

## **COOKIES USING RESPONSE SURFACE METHODOLOGY**

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## ABSTRACT

Response surface methodology (RSM) was used in the optimisation 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 fibre and fructose were added as humectant and anti-crystallising agents to extend the shelf-life of the cookies. Five quantitative controllable factors were selected for the experimental design: weight of bamboo fibre, fructose/saccharose ratio and weight of egg white as ingredients; baking time and temperature as process parameters. To assess the product quality, texture, moisture content and colour were considered as dependent variables. The three second order polynomial models obtained by RSM and the subsequent optimisation step indicated that a formulation with 28.95 g of fibre, 252.5 g of egg white, a fructose/saccharose ratio equal to 0.1, baking time and temperature of 21.5 min and 185.5°C, 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.

## **PRATICAL 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 during time, without changing the most important characteristics related to consumer acceptance.

## 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, even if 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 further substances can be added (water, glucose). They are named in a different way depending on the place of origin: almond macaroon cookies (U.S. 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), alfafores de almendras (Argentina), for example. Traditionally served with a sweet dessert wine or liqueur, they are now eaten any time of the day: with a 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; 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 multi-domain 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 multi-domain moist cookies appears much more complicated, due to the different chemophysical phenomena that take place simultaneously during storage (e.g., water redistribution, sugar crystallization) and that lead to a 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, 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 during time, changing the water activity 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 fibre) 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 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 colour, lost in crispness, viscosity of the internal paste), affecting the typical characteristics of these cookies greatly appreciated by consumers. This fact suggested 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 humectant and anti-crystallizing agent (Guilbért 2002). Because of its colligative properties, it can be used to lower the water activity of different foods, due to 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 fibre is a cellulose-based insoluble ingredient derived from the fibre-rich parts of bamboo plants. It is characterized by total fibre 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<sup>-1</sup>. This purified fibre has a water holding capacity (WHC) of 8.7 g g<sup>-1</sup> fibre, 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 fibre 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 mouth feel. Finally, fibres are credited in having a number of functional properties, including, the shortening of intestinal transit time, the slowing down of carbohydrates 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, including: fructose/saccharose ratio (from now on indicated as F/S), weight of bamboo fibre 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, Design Of Experiment (DOE) technique was used. It is a powerful tool which 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 optimisation step was carried out using the response surface methodology (RSM), to discover the conditions that produce the best output (Araujo and Brereton 1996). Response surface methodology is the most popular optimisation 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). Response surface methodology (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 optimisation of quality and performance of a product, the optimisation 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 (Affrifah and Chinnan 2005; Chetana et al. 2004; Calzetta Resio et al. 2006; Elbert et al. 2001), to optimise different process conditions (Liu-ping Min et al. 2006; Lee et al. 2006; Chen et al. 2005; Milàn-Carrillo et al. 2006) or in the optimisation of particular formulations (Jackson et al. 1996; Colla et al. 2006; San Juan et al 2006; Sun et al. 2006; Varnalis et al. 2004). However, there are not many examples in 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 optimising 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 represent a useful tool able to predict the effect due to the different settings of factors on the most important characteristics of this kind of cookies (texture, colour 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 1a. Table 1b reports the typical mean values for hardness, moisture content and colour as the most important attributes of global quality. These values were obtained analysing almond paste cookies provided by ten different suppliers, using 30 cookies per each lot. For the experimental design, bamboo fibre and crystalline fructose (Chimab s.p.a., Campodarsego, Italy) were added. After baking, the cookies were stored in a climatic chamber (mod. CH 700, Angelantoni Industrie s.p.a., Massa Martana, Italy) under controlled temperature-humidity conditions ( $T = 25 \pm 0.5^{\circ}$ C and  $RH = 40 \pm 2$  %) for two hours before analyses.

#### Moisture content determination

Gravimetric analysis was performed in triplicate to determine water content (% H<sub>2</sub>O on wet basis) following official method (925.098) of the AOAC (1999), using an oven at  $105 \pm 2.0^{\circ}$ C for 24 h, by the following equation:

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

where *MC* is the moisture content (wet basis),  $w_i$  is the initial weight (g) of the sample prior drying and  $w_{f,d}$  is the final dry weight (g) of the sample after drying. The measurements were performed on the internal paste of the cookies sampling  $5 \pm 0.1$  g of 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 analyser (mod. Z005, Zwick Roell, Ulm, Germany). The software "TestXpert V10.11 Master" 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  $\times$  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 ( $W_{tot}$ ).

#### **Colour measurement**

Colour 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). Cookies were placed on a white standard plate ( $L^*=100$ ) and the *CIE*  $L^*a^*b^*$  coordinates were simultaneously measured, where  $a^*$  is the CIE redness/greeness 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 ANOVA using Statgraphics Plus 4.0 software, followed by LSD's multiple range test ( $p \le 0.05$ ) 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 squares analysis, based on the multiple linear regression (MLR).

## **Experimental design**

In designing this experiment by RSM, a central composite face-centred (CCF) design supporting quadratic models was employed. In general, central composite designs (CCD) contain an imbedded factorial or fractional factorial design with centre points that is augmented with a group of 'star points' allowing estimation of curvature. If the distance from the centre of the design space to a factorial point is  $\pm 1$  unit for

each factor, the distance from the centre of the design space to a star point is  $\pm \alpha$  with  $|\alpha| \ge 1$ . In particular, for CCF designs the 'star points' are at the centre of each face of the factorial space, so  $\alpha = \pm 1$ . The quantitative controllable factors (independent variables) were five: weight of bamboo fibre  $(X_i)$ , F/S ratio  $(X_2)$  and weight of egg white  $(X_3)$  as ingredients; 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 cookies quality: hardness  $(Y_i)$ , moisture content  $(Y_2)$  and colour  $(Y_3)$ . A total of 31 combinations (16 corner points, 10 star points and 5 centre 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 2. As far as the selection of the regression model is concerned, it was assumed that *i* mathematical functions,  $f_z(z = 1, 2, ...i)$ , exist for each response,  $Y_z$ , function of *l* independent factors,  $X_k$  (k = 1,2,...l), such that:

$$Y_{z} = f_{z} (X_{1}, X_{2}, \dots, X_{l})$$
<sup>(2)</sup>

where: i = 3 and l = 5. Finally, the f<sub>z</sub> function was assumed to be approximated by a second degree polynomial equation:

$$Y_{z} = b_{z0} + \sum_{k=1}^{l=5} b_{zk} X_{k} + \sum_{k=1}^{l=5} b_{zkk} X_{k}^{2} + \sum_{k\neq j}^{l=5} b_{zkj} X_{k} X_{j} + \varepsilon$$
(3)

where:

 $b_{z0}$  = response value with all factors set at medium level (centre point);

 $b_{zk}$  = linear regression coefficient;

 $b_{zkk}$  = quadratic regression coefficient;

 $b_{zkj}$  = interaction regression coefficient;

 $\varepsilon$  = 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 tstatistic 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 3 shows the linear, quadratic and interaction coefficients obtained from the fitting of the experimental data (Table 2) to the quadratic model for 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:

- Hardness 
$$(Y_1) = 31.55 + 3.45 X_1 - 2.79 X_3 + 10.51 X_4 + 5.16 X_5 + 2.55 X_2^2 -$$
(4)

$$-3.60 X_3^2 + 2.65 X_1 X_5 - 1.04 X_2 X_4 - 2.09 X_3 X_4$$

- Moisture content 
$$(Y_2) = -1.92 + 0.0064 X_2 + 0.0014 X_3 - 0.0048 X_4 -$$
 (5)

$$-0.0032 X_5 - 0.0066 X_1^2 + 0.0055 X_3^2 + 0.0014 X_1 X_4 - 0.0012 X_3 X_4 - 0.0012 X_3 X_5$$

$$-Colour(Y_3) = 0.41 - 0.0069 X_2 - 0.26 X_4 - 0.0662 X_5 + 0.0952 X_3^2 -$$
(6)

 $-0.0258 X_2 X_4 + 0.0379 X_3 X_5$ 

Once the models were obtained, an analysis of variance was calculated to verify their capability to represent the data. The ANOVA for all three responses is presented in Table 4. For each model, the Mean Squares Regression (i.e. the variance explained by the model) in the first *F*-test was significantly larger than the Mean Squares Residual (i.e. the amount of variance unexplained by the model). This means that all three models adequately represented the data for hardness, moisture content and colour. 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 - $\infty$ ; in an optimisation 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 colour were assessed by the regression coefficients of the second polynomial order equations (Table 3). 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 increasing linearly fibre content occurred, most likely due to an increase in the water binding capacity of the paste (Colleoni-Sirghie *et al.* 2004; Zhou *et al.* 2000). Moreover, insoluble fibres 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 fibre amount leads to a concentrationdependent increase in firmness (Thebaudin J.Y. et al. 1997). In addition, bamboo fibre 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 fibre to produce natural rubber composites (Ismail et al. 2002). Baking time also had a positive linear effect on hardness response, due to the proportional amount of water removed from the cookies up to the maximum level. In addition, the interaction effect between the fibre weight and baking time was important, due to the effect of prolonged heat treatments on fibre 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 fibre frame. Alternatively, the formation of fibre-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 fibres 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 grater, 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), 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 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 colour of the cookies. This is because Maillard reaction occurred more at higher baking temperatures. The other important factors include egg white weight as quadratic effect, baking time and F/S ratio as linear effects. As far as F/S ratio is concerned, fructose is technically named reducing sugar, contrary to saccharose (for that termed non-reducing sugar). The presence of the reducing sugars is fundamental in defining the colour of baked products, because they represent the substrates of Maillard reaction (Lerici *et al.* 1990).

### **Response surface plots**

A helpful tool for a better understanding of the link between each factors and responses is given by the response surface plots, in which the effect of two factors on one specific response is displayed in 3D view, keeping the other ones on fixed values. Some selected surfaces predicted by Eq. (4), (5) and (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 optimised recipe with values of hardness similar to the original one, both baking time and temperature should be set at levels close to the maximum. Fig. 2. shows the effect of fibre 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, due to the above mentioned interaction between factors. In this case, the influence of fibre weight on hardness was greater with baking time set at the highest level, hence the effect of fibre is strictly dependent on baking time. Hence, an increase in hardness can be achieved by setting both baking time and fibre content to maximum level. The last response surface for hardness (Fig.3) was saddle-shaped, due to 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 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 ( $W_{tot}$ ~26 N x mm); however, with egg white set at minimum level, the plasticizing effect disappeared and hardness increased ( $W_{tot}$ ~30 N x mm), but not enough to be comparable to the original value. Finally, if it is used at medium level, egg white could behave promoting the formation of a lattice which gives strength to the whole matrix (to the crust in particular), resulting in a higher value in hardness ( $W_{tot}$ ~35 N x mm). This is due to egg white proteins, especially ovalbumin that is the most abundant and the only one containing free sulfhydril (SH) groups. If subjected to heating (80-85°C), 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 (SS) bonds (Van 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, MRI (Magnetic Resonance Imaging) technique (Labuza and Hyman 1998).

### Moisture content

In Fig. 4 the influence of fibre 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

fibre 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 fibre and the minimum amount of fructose compared to saccharose. Fig. 5 shows as moisture content depended also on baking temperature and egg white, the latter had a mostly a quadratic effect. As predictable, the highest values for moisture content were for the formulation with higher amounts of egg white, temperature being constant; the higher baking temperature, the lower 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.

### Colour

As far as the colour response, Fig. 6 shows the influence of baking temperature and F/S ratio as main linear effects. The greater change in colour was produced by changes in temperature, the F/S ratio also played an important role in promoting non-enzymatic 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 colour of this kind of cookies can be obtained using a small amount of fructose and upper-middle temperature values. Further factors affecting the colour response are 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, browness reached maximum with increasing baking time, but differed depending on the amount of egg white used.

## Optimisation

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 able to produce the expected characteristics of the final product. All comments arising from the response surface plots were taken into account in the optimisation, considering that the optimal solution arises from a compromise among the different responses. In this phase, the criteria of optimisation 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 (Tab. 1b). 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: fibre weight, 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 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  $Y_3$  = colour), the desirability function was:

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

where:

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

and:

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

In the Eq. (9) :

- $\lambda$  = scaling parameter;
- $\varphi$  = 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);
- V = target value.

As a result of the optimisation step, the best conditions which were attained for the expected response values were: fibre weight: 28.95 g; egg white weight: 252.5 g; F/S ratio:0.1; baking time: 21.5 min; baking temperature: 185.5°C. According to the models, the predicted values of responses were: hardness ( $W_{tot}$ ): 47.02 N × mm; moisture content (MC): 13.89%; colour ( $L^*$ ): 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 analysed 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 a LSD's multiple range test to detect differences between predicted and observed values. The obtained results are shown in Table 5. As expected from the high values of  $R^2$  and  $Q^2$ , equation (4), (5) and (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 ( $p \ge 0.05$  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 standardised 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, modelling of experimental data allowed generating useful equation of general use, to predict the behaviour of the system under different factor combinations.

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19

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## **FIGURE LEGENDS**

Fig. 1. Response surface plot: effect of baking time and temperature on hardness. Fibre = 25 g. F/S = 0.1. Egg white = 258 g.

Fig. 2. Response surface plot: effect of baking time and fibre on hardness. Temperature:  $180^{\circ}$ C. F/S = 0.1. Egg white = 258 g.

Fig. 3. Response surface plot: effect of F/S ratio and egg white on hardness. Fibre: 25 g. Temperature: 180°C. Time: 20 min.

Fig. 4. Response surface plot: effect of fibre and F/S ratio on moisture content. Egg white: 258 g. Temperature: 180°C. Time: 20 min.

Fig. 5. Response surface plot: effect of temperature and egg white on moisture content. Fibre: 25 g. F/S: 0.1. Time: 20 min.

Fig. 6. Response surface plot: effect of temperature and F/S ratio on colour. Fibre: 25 g. Egg white: 258 g. Time: 20 min.

Fig. 7. Response surface plot: effect of time and egg white on colour. Fibre: 25 g. F/S ratio: 0.1. Temperature: 180°C.

Fig. 8. Superimposed contour plots for all the responses affected by F/S ratio and fibre amount. The other factors are set according to the optimisation. Responses are constrained as follows: hardness (42-50 N x mm); moisture content (12-16%); colour as  $L^*$  (66-70).