

## OPTIMIZATION OF GRAPE PEELS PARTICLE SIZE AND FLOUR SUBSTITUTION IN WHITE WHEAT FLOUR DOUGH

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Received: April, 19, 2018

Accepted: February, 06, 2019

**Abstract:** Grape peels flour (GPF) is regarded as a good source of fiber to enrich baked products. The particle size and the amount of grape peels flour (GPF) added in white wheat flour (WWF) are essential formulation factors which affect dough rheology and bread quality. This study aimed to optimize formulation factors, GPF particle size and level of flour substitution in WWF dough using response surface methodology (RSM) with a full factorial design and multiple responses optimization approach. The results showed that the models developed for response variables, Rheofermentometer characteristics and dynamic rheological properties adequately described the relationships. The optimum value of formulation factors was found to be composite grape peels-wheat flours containing 4.67 % GPF of small particle size when desirability function method was applied.

**Keywords:** *dynamic rheological properties, grape peel, optimization, Rheofermentometer characteristics, white wheat flour*

## INTRODUCTION

In recent decades, research focuses on a new generation of healthy foods to combat many diseases such as diabetes, coronary heart disease and colon cancer. Some researchers have attempted to improve white-bread products with dietary fiber from various sources like agricultural by-products, which offers different technological and nutritional functions [1]. Due to their health potential, grape pomace peels is highly recommended in bread production among the fiber source. Grape peels are an important source of polysaccharides, cellulose and hemicelluloses, acidic pectin substance, sugars, proteins, fat, minerals, phenolics compounds [2, 3], several organic acids and flavors [4]. Grape peels contain many micronutrients such as minerals [5] and large amounts of dietary fiber and antioxidants [6, 7] than white wheat flour. The biological and functional properties of grape antioxidant dietary fiber have been well studied [8 – 12]. The addition of grape dietary fiber to bakery products increase dietary fiber intake but is associated with changes of physical properties. The replacing of wheat flour with grape peels flour (GPF) leads to disruption of the starch-gluten matrix and negatively influences the bread volume. These effects can be reduced by using an appropriate proportion of soluble to insoluble fiber fraction [13]. Inclusion of grape peels flour in white wheat flour can cause structural and sensory changes in bread, leading to lower consumer acceptance. As a result, there are difficulties in producing enriched grape peels bread that maintains the desired functionality and quality equivalent to refined wheat bread. In addition to the qualitative characteristics of the enriched-bread product, the use of GPF also provides many changes in dough properties and processing parameters. The particle size of GPF is an important factor influencing product quality and functionality of flour [14]. Fiber-supplemented dough bread has a significant effect on mixing and viscoelastic properties and fermentation behavior during bread-making. Many studies investigated the effect of bran particle size on the technological properties of the flour and baking product. These highlighted the negative effect of large wheat bran size on bread quality due to the hydration restriction and its integration in the gluten viscoelastic structure [15]. Hydration properties of wheat bran mainly depend on its particle size [16]. Raghavendra et al. (2006) found that as particle size increased, the hydration properties decreased and the oil-holding capacity increased with particle size reduction of coconut residue. Particle size, porosity, preparation process, various surface properties, structures of the fiber and fiber composition are some of the factors which might cause changes in functional properties. For example, an improvement of the functional properties of insoluble dietary fiber from peach and oat with the particle size reduction was found when microfluidization process was applied [18]. Regarding the functional properties of grape dietary fiber, some studies indicated that grape dietary fiber presented high water and oil retention capacity, and high swelling properties [8, 19].

The use of experimental design in the development of enriched foods has increased because it facilitates the investigation of the interaction between the effects of variables through mathematical models and response surface methodology (RSM). The RSM is a statistical technique adequate for optimization of process variables or formulation factors to develop new food products. Some studies reported the application of RSM in the optimization of ingredients or formulations factors [20 – 23], in optimization of fiber particle size and flour replacement in wheat bread [24]. However, there are not data in

the literature related to the use of GPF. Both its addition level and particle size influence on wheat flour dough formulation.

In the literature there are few papers about the effect of different particle size of grape peels flour as a source of dietary fiber at one addition level on dough rheological properties [25]. In our previous works we studied the effect of GPF addition at different levels and different particle sizes on the rheological behaviour of bread dough through empirical and dynamic tests [26, 27]. The addition of large amounts of GPF to dough bread results in a change in rheological properties and consequently on bread quality. Reducing particle size increases water-holding capacity by increasing the surface area, water being essential in bread production because it is involved in the process of starch gelatinization, protein denaturation, formation of flavor characteristics and influences color [28]. Therefore, it is important to optimize a formulation of white wheat flour dough with GPF addition at different levels and of different particle sizes. To the author's knowledge, there is no published study on the optimum GPF level-particle size addition which may be used in bread dough in order to obtain an adequate dough rheological behaviour in terms of rheofermentometric characteristics and dynamic oscillatory rheological properties, essentials to predict quality of GPF enriched-bread products. This result could be used for composite mix development from locally produced grape wine pomace at small scale industry level as value-add products.

## MATERIALS AND METHODS

### Basic ingredients

White wheat flour (WWF) of 480 type (harvest 2016) used in the experiments was obtained from S.C. Dizing S.R.L. (Brusturi, Neamț, Romania). The physico-chemical analyses performed according to the International Association for Cereal Chemistry (ICC) revealed that it contained (in %): moisture 14.10 % (ICC 110/1), protein 10.80 % (ICC 105/1), fat 1.10 % (ICC 105/1), ash 0.47 % (ICC 104/1), wet gluten 27.80 % (ICC 106/1), gluten deformation index 2.5 mm (SR 90:2007) and falling number index 370.5 s (ICC 107/1). The white wine grape pomace peels, *Vitis vinifera* L. provided from the viticulture center Jariște, Odobești ecosystem were analyzed for their chemical characteristics (moisture, protein, fat, ash) according to ICC methods (2010). The grape peels had (in %): moisture 5.80, protein 7.44, fat 3.18, ash 3.70 and fiber 28.50, determined by Near Infrared Reflectance (NIR) spectroscopy technology. The values obtained are the average expression of the analyses made in least duplicate. The dried grape peels were grounded in a domestic blender and sieved through a Retsch Vibratory Sieve Shaker AS 200 basic (Haan, Germany) to obtain grape peels flour (GPF) at three different particle sizes: large,  $L > 500 \mu\text{m}$ , medium,  $200 \mu\text{m} > M > 500 \mu\text{m}$  and small fractions,  $S < 200 \mu\text{m}$ .

### Experimental design and statistical methods

The effect of five levels (0, 3, 5, 7 and 9 %) and of three particle sizes (L, M and S) for the GPF added in WWF on some dough rheological properties, i.e. Rheofermentometer characteristics and dynamic oscillatory rheological properties, as dependent variables, were

investigated using the RSM by means of general factorial design with two independent variables. The complete experimental design required fifteen experimental runs (Table 1).

**Table 1.** Coded and real values of formulation factors used in full factorial design

Run	Coded value		Real value	
	X <sub>1</sub>	X <sub>2</sub>	Grape peels flour [%]	Particle size [μm]
1	-0.333	1.000	3.00	600.00
2	0.556	-1.000	7.00	200.00
3	0.111	-1.000	5.00	200.00
4	0.111	1.000	5.00	600.00
5	0.556	1.000	7.00	600.00
6	-1.000	0.000	0.00	400.00
7	1.000	-1.000	9.00	200.00
8	-0.333	-1.000	3.00	200.00
9	0.111	0.000	5.00	400.00
10	-0.333	0.000	3.00	400.00
11	0.556	0.000	7.00	400.00
12	1.000	1.000	9.00	400.00
13	-1.000	1.000	0.00	600.00
14	1.000	1.000	9.00	600.00
15	-1.000	-1.000	0.00	200.00

Multiple linear regression analysis was applied to fit the data obtained for each response to linear, quadratic and cubic models. The most adequately model was chosen through sequential *F*-test, coefficients of determination ( $R^2$ ), adjusted coefficients of determination ( $Adj.-R^2$ ) and significant probabilities. By analysis of variance (ANOVA) the statistical significance of the coefficients in each predictive model was assessed. The combined effect of the factors, GPF addition level and particle size on the responses, dough fermentation characteristics and dynamic rheological properties was modeled using a polynomial response surface. The full factorial design, model and contour plot generation, test of model adequacy and the optimum level for formulation factors were generated by the Stat-Ease Design Expert 7.0.0 software package. To determine the validity of the model for each response, the experimental and predictive values were compared by paired *t*-test. The optimal value of the factors, level of GPF adding in WWF and GPF particle size was performed by the multiple responses analysis called desirability function [31]. The desirability function involves transformation of each predicted response into an individual desirability function,  $d_n$  which includes the desired and researcher's priorities when building the optimization procedure for each of the factors. The individual desirability functions are then combined into a single composite response, namely overall desirability function,  $D$  computed as the geometric mean of the individual desirability function,  $d_n$  which varies from 0 to 1 [32].

### Dough rheological characteristics during fermentation

Dough fermentation characteristics for the formulated *GPF-WWF* blends (Table 1) were determined using the Chopin Rheofermentometer F3 (Chopin Rheo, Villeneuve-La-Garenne Cedex, France) which gives information about dough height during fermentation, the volume of carbon dioxide retained and released by the dough. Mixing was performed in a Brabender Farinograph for 8 min at 30 °C from 250 g blend flour,

7 g compressed yeast, 5 g salt and water according to the Farinograph water absorption. The dough (315 g), tested at 30 °C for 3 h under a 2000 g cylindrical weight constraint gives the Rheofermentometer curve with following parameters: dough maximum height ( $Hm$ ), maximum height of gaseous production ( $H'm$ ), the time required to reach  $H'm$  ( $T'l$ ), the time at which gas starts to escape from dough ( $Tx$ ), total carbon dioxide volume production ( $Vt$ ) and gas retention coefficient ( $Rc$ ).

### Dough dynamic rheological characteristics

Rheological oscillatory measurements were conducted with a MARS 40 rheometer (Thermo-Haake, Karlsruhe, Germany). The dough samples for rheological investigation were done using standard dough preparation, based on each farinograph water absorption value, but without yeast to avoid the influence of fermentation on the results. The frequency sweep test was followed by a five minute period of relaxation. The investigated dough was subject to frequency sweep test at a temperature of 20°C and shear stress of 15 Pa. The frequency of oscillation of 1-20 Hz was conducted in the plate-plate system geometry with a 2 mm gap. The parameters were selected after a set of measurements that have been performed to determine the linear viscoelastic region. The measured parameters were: storage modulus ( $G'$ ), viscous modulus ( $G''$ ), complex modulus ( $G^*$ ) and loss tangent ( $\tan \delta$ ).

## RESULTS AND DISCUSSION

### Model fitting

The experiments were performed according to the experimental design to determine the combined effect of the factors, GPF level and GPF particle size on rheofermentometric characteristics and dynamic oscillatory rheological properties of dough. The regression models were highly significant for the response variables with  $R^2$  values that varied from 0.66 to 0.95 (Table 2). The predictive models represented well the experimental data with satisfactory  $R^2$  and  $Adj.-R^2$  values. The ANOVA results, including significantly regression coefficients ( $p < 0.05$ ), expressed in terms of coded values (Table 2) showed that the regression models obtained were statistically significant ( $p < 0.05$ ). Based on the predictive models obtained, each response ( $Hm$ ,  $H'm$ ,  $T'l$ ,  $Tx$ ,  $Vt$ ,  $Rc$ ,  $G'$ ,  $G''$ ,  $G^*$  and  $\tan \delta$ ) can be predicted and showed as a response surface.

### Effects of formulation samples on dough rheological characteristics during fermentation

#### Dough maximum height

Table 2 shows the effects of formulation factors, GPF level added in white wheat flour and particle size on dough maximum height,  $Hm$  as their corresponding regression coefficients in the quadratic model. The quadratic model predicts adequately the  $Hm$  as a function of the formulation factors. The regression model indicated that the GPF level and particle size had significant ( $p < 0.05$ ) effects on  $Hm$  characteristic. The ANOVA results showed that the  $R^2$  value of this model was 0.95, while the  $Adj.-R^2$  was 0.93.

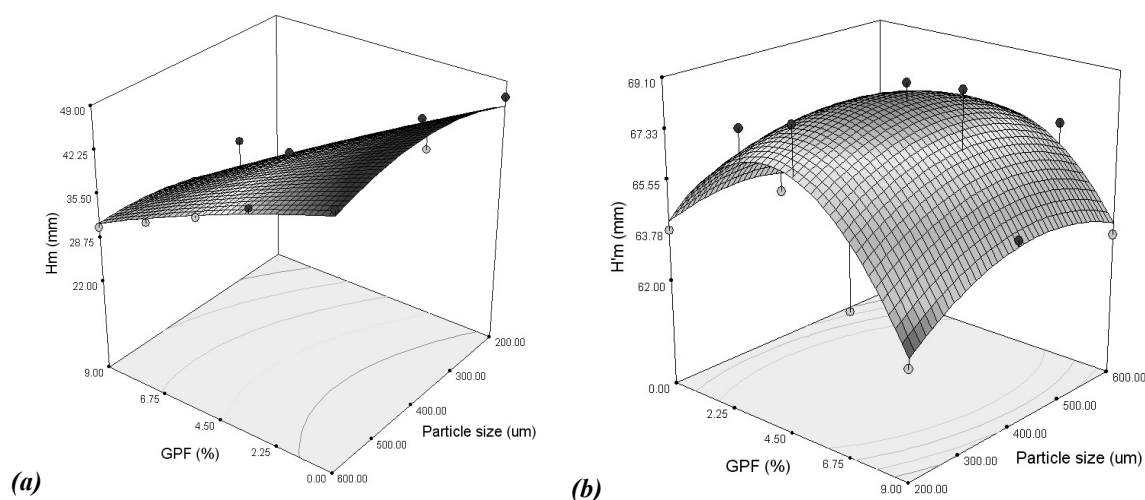
**Table 2.** Effects of formulation factors, expressed as their corresponding coefficients in the predictive models for dough fermentation characteristics and dynamic rheological characteristics<sup>a</sup>

Factors <sup>b</sup>	Characteristics									
	Rheofermentometer						Dynamic rheological			
	<i>Hm</i> [mm]	<i>H'm</i> [mm]	<i>T'l</i> [min]	<i>Tx</i> [min]	<i>Vt</i> [mL]	<i>Rc</i> [%]	<i>G'</i> (10 <sup>3</sup> ) [Pa]	<i>G''</i> (10 <sup>3</sup> ) [Pa]	<i>G*</i> (10 <sup>3</sup> ) [Pa]	<i>tan δ</i> (10 <sup>-3</sup> )
Constant	39.32	68.30	68.39	50.19	1347.61	84.70	26.208	8.764	27.636	340.00
A	-9.35***	-0.067	-5.86*	-8.85**	-52.56**	3.08**	5.522***	1.549***	5.728**	-13.00***
B	2.32	-0.13	-2.86*	-2.69**	-1.05	-0.08	-1.003*	-0.298*	-1.046*	1.97*
A x B	2.08**	0.82*	-5.09*	-0.62	15.77	-0.51*	-0.661	-0.185	-0.686	1.39
A <sup>2</sup>	-0.77	-3.87**	28.78***	-6.35***	-55.32	-3.05*	0.176	0.062	0.187	3.01*
B <sup>2</sup>	-2.98*	-1.34*	-1.00	-1.96*	-69.20**	2.12*	-4.000*	-1.221*	-4.182*	2.85*
A <sup>2</sup> x B	-	-	-	1.70*	-	-	-	-	-	-
A x B <sup>2</sup>	-	-	-	0.84	-	-	-	-	-	-
A <sup>3</sup>	-	-	-	10.80**	-	-	-	-	-	-
B <sup>3</sup>	-	-	-	0.01	-	-	-	-	-	-
<i>R</i> <sup>2</sup>	0.95	0.67	0.80	0.91	0.67	0.69	0.71	0.66	0.71	0.69
<i>Adj.-R</i> <sup>2</sup>	0.93	0.50	0.70	0.79	0.50	0.52	0.55	0.50	0.54	0.52
<i>p</i> -value	< 0.0001	< 0.05	< 0.005	< 0.05	< 0.05	< 0.05	< 0.05	< 0.05	< 0.05	< 0.05

<sup>a</sup>\* *p* < 0.5; \*\* *p* < 0.05; \*\*\* *p* < 0.005<sup>b</sup>A, grape peels flour level added in white wheat flour (%); B, grape peels flour particle size (μm); *R*<sup>2</sup>, *Adj.-R*<sup>2</sup> are measures of model fit; *Hm*, dough maximum height; *H'm*, maximum height of gaseous production; *T'l*, the time required to reach maximum height of gaseous production; *Tx*, the time at which gas starts to escape from dough; *Vt*, total carbon dioxide volume production; *Rc*, gas retention coefficient; *G'*, storage modulus; *G''*, viscous modulus; *G\**, complex modulus; *tan δ*, loss tangent

That values highlight that the predictive model defined well the real behaviour of dough during fermentation in term of dough maximum height. The negative coefficient of GPF addition level indicated that the dough maximum height of GPF-WWF mix increased with decrease of GPF level addition in WWF. This decrease of dough maximum height with GPF level increase may be attributed probably to the gluten dilution as a result of an addition of non-gluten flour [33]. At high level of GPF addition the gluten matrix may present physical interruption which can cause dough weakening. The linear term of particle size and the interaction coefficients have a significant effect (*p* < 0.05) on dough maximum height. *Hm* was not significantly correlated (*p* > 0.05) with quadratic effect of GPF level added in WWF (Table 2), while the quadratic effect of GPF particle size on *Hm* was significant (*p* < 0.05) and highlights the importance of addition of an optimal particle size of GPF to achieve desirable GPF-WWF mix. The *F*-value for *Hm* was significant (*p* < 0.0001). The effect of GPF level and particle size on dough maximum height is shown in Figure 1a. An increase in GPF level added in WWF leads to a decrease of dough maximum height. *Hm* is an indirect estimation of yeast performance and overall microstructure formed in dough system. A lower *Hm* will suggest that the combination of gas produced and the microstructure present in that particular system were no more favorable in sustaining the macrostructure of the proofed dough piece compared to another system with a higher one. If the gas is not efficiently retained in dough, one would expect to obtain no favorable final bread volume.





**Figure 1.** Response surface plot showing the combined effects of grape peels flour (GPF) level and particle size of GPF on: **(a)** dough maximum height ( $H_m$ ); **(b)** maximum height of gaseous production ( $H'm$ )

#### **Maximum height of gaseous production**

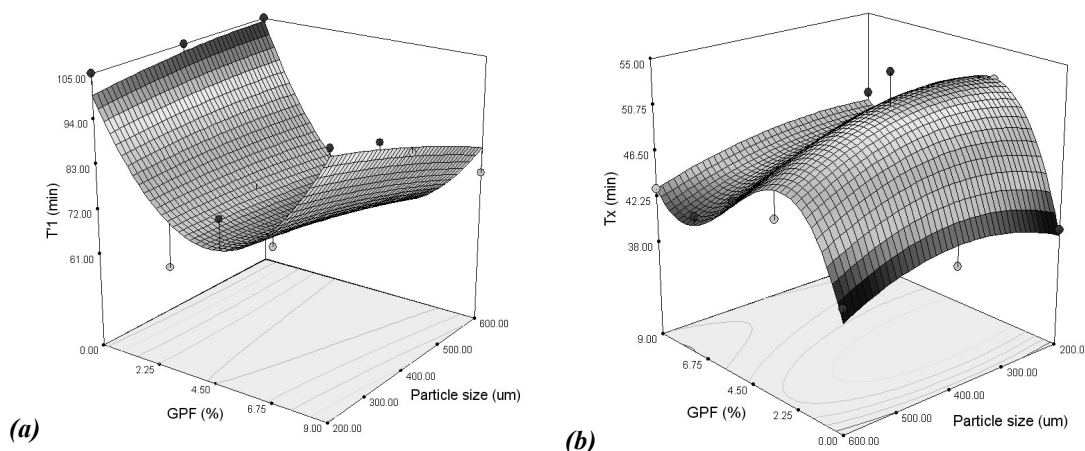
The maximum height of gaseous production,  $H'm$  was significantly ( $p < 0.05$ ) influenced by particle size and the level of GPF addition in WWF. The regression model (Table 2) for  $H'm$  showed a significant effect in quadratic term of GPF level, while the quadratic term of particle size was non-significant ( $p > 0.05$ ). Also, the linear term of GPF level, particle size and the interaction term were non-significant ( $p > 0.05$ ). A negative effect on maximum height of gaseous production was given by particle size of GPF. This can be explained by the disruption of the gluten network when the GPF particles, which are higher than the wheat flour particle, are incorporated in dough system.

The result of the  $F$ -test for ANOVA showed that the regression model for  $H'm$  is statistically significant ( $p < 0.05$ ) and the  $R^2$  value showed that the model explains 67% of the obtained data variation. A contour plot for the maximum height of gaseous production (Figure 1b) showed that the  $H'm$  significantly decreased with an increase of GPF particle size of the mix.

#### **Time at which gas starts to escape from dough**

The factors GPF level added in wheat flour and the particle size affected the time at which gas starts to escape from dough,  $T_x$  significantly ( $p < 0.05$ ). The high value of  $R^2$  (0.91) indicates that the cubic model is a good prediction model of  $T_x$  using the significant factors shown in Table 2. GPF level and the particle size showed negative influences on  $T_x$  while the interaction between the factors was non-significant ( $p > 0.05$ ). High GPF level and large particle size (L) might have inhibited the activity of yeast and decreased the leavening ability during proofing. The GPF level had a significant effect on  $T_x$  in both quadratic and cubic terms ( $p < 0.05$ ). The influence of GPF level was higher than of particle size (Table 2, Figure 2a). The GPF level had a significant negative influence ( $p < 0.05$ ) on  $T_x$  in quadratic term while the cubic term was significantly positive ( $p < 0.05$ ) (Table 2). At the high level of GPF,  $T_x$  decreased

as the particle size increased and the particle size effect was smaller than the one of GPF addition level in WWF (Figure 2a).



**Figure 2.** Response surface plot showing the combined effects of grape peels flour (GPF) level and particle size of GPF on: (a) the time required to reach maximum height of gaseous production ( $T_1$ ); (b) the time at which gas starts to escape from dough ( $T_x$ )

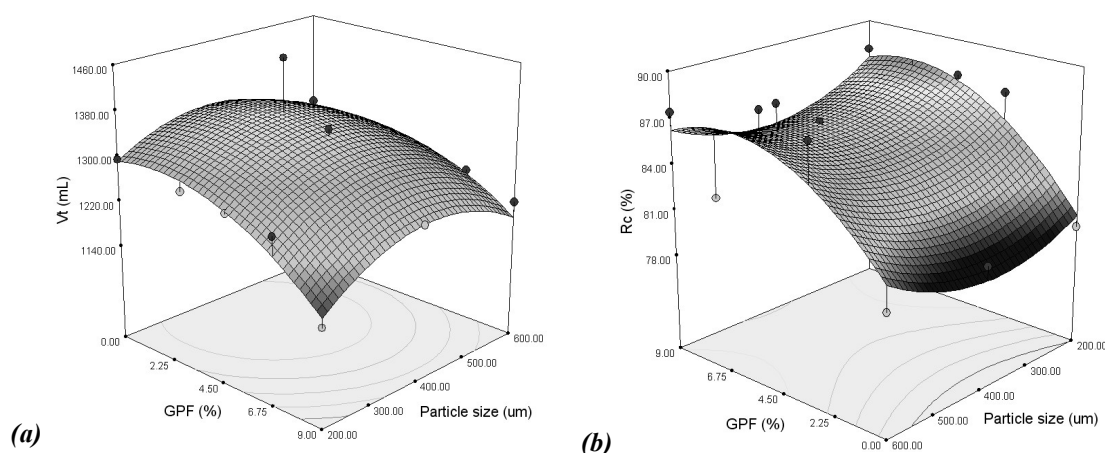
#### ***The time required to reach maximum height of gaseous production***

The quadratic model describes the time required to reach maximum height of gaseous production,  $T_1$  as a simultaneous function of GPF level and particle size addition in WWF. The time required to reach maximum height of gaseous production was significantly affected ( $p < 0.005$ ) by GPF level and particle size added in WWF and by the interaction term between the GPF level added in wheat flour and the particle size. The formulation factor, independently not influence significantly the time required to reach maximum height of gaseous production, however, the quadratic term of GPF level added in wheat flour was found to be significant ( $p < 0.05$ ). The regression model fitted to the experimental results of  $T_1$  characteristic showed higher  $R^2$  value (0.80). The effect of GPF level and particle size added in WWF on the time required to reach maximum height of gaseous production is shown in Figure 2b. Response surface plot showed that an increased level of GPF added in WWF increased the time required to reach maximum height of gaseous production.

#### ***Total carbon dioxide volume production***

The coefficients and their significance of the factors affecting total carbon dioxide volume production ( $V_t$ ) are shown in Table 2. In the model, GPF level added in WWF, as a linear term, had significant ( $p < 0.05$ ) effect on  $V_t$ . However, in the quadratic term, GPF level had no significant effect, but the quadratic term of particle size was significant (Table 2). At the large particle size,  $V_t$  decreased as the particle size increased and the particle size effect was higher than the one of the level of GPF added in WWF. The increase of GPF particle size and addition level influenced yeast activity, having a negative effect on  $\text{CO}_2$  production. This effect may be due to the bioactive compounds from GPF which might inhibit the yeast fermentation. The total carbon dioxide volume production, as shown in Figure 3a, decreased due to the GPF level more so than to the particle size.





**Figure 3.** Response surface plot showing the combined effects of grape peels flour (GPF) level and particle size of GPF on: (a) total carbon dioxide volume production ( $V_t$ ); (b) gas retention coefficient ( $R_c$ )

### Gas retention coefficient

Bread dough having high gas retention coefficient,  $R_c$  are considered to have desirable features. The GPF level added in WWF influenced  $R_c$  positively in the linear term ( $p < 0.05$ ) and negatively in the quadratic terms ( $p < 0.05$ ). GPF level showed most pronounced effect on  $R_c$  compared to the particle size (Table 2). Figure 1f shows the effect of GPF level and of particle sizes added in WWF on gas retention coefficient.  $R_c$  increased with the increase of GPF content (Figure 3b). Whereas at larger GPF particles sizes reverse effect of GPF content was observed on gas retention coefficient.

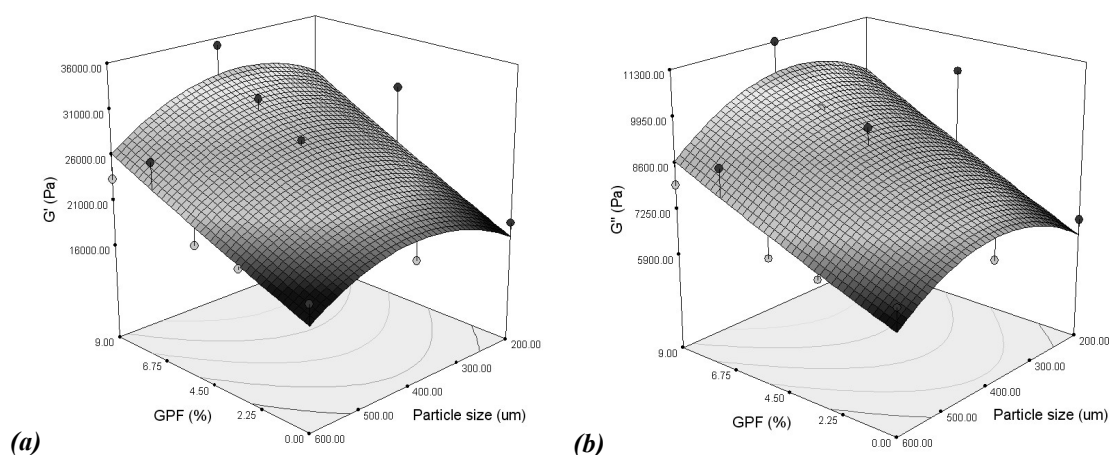
### Effects of formulation samples on dough dynamic rheological characteristics

Oscillatory measurements reported in the LVR were highlighted to be useful in studying the influence and the significance of structural ingredients in dough systems [34]. The doughs are viscoelastic bodies which possessed differing but pronounced elastic components. In bread-making, GPF-WWF mix producing doughs with balanced extensible and elastic properties are required to ensure optimal baking performance. By applying dynamic technique based on use of the rheometer, more specific information on dough physical properties was obtained. For example, the elastic properties can be described by the loss tangent of the dough ( $G''/G'$ ).

### Storage modulus

The storage or elastic modulus,  $G'$  indicated different resistance to the rupture by the action of stress for formulated dough structures. The oscillatory test indicated that when the WWF is replaced with GPF, the  $G'$  parameter is influenced by the GPF levels and particle sizes. GPF level addition conferred significant effect on the elastic modulus  $G'$  through linear positive effect. The effect of GPF on  $G'$  was counteracted due to the negative linear term of particle size, whereas the interaction term between GPF level and particle size was non-significant ( $p > 0.05$ ). The quadratic regression model fitted to the experimental results of elastic modulus showed higher  $R^2$  value (Table 2). The effect of GPF level and particle size on  $G'$  is shown in Figure 4a. Response surface plot showed that an increased level of GPF added in WWF increased the elastic modulus.  $G'$

increase could be explained due to the enhanced elastic properties of polysaccharides in aqueous medium. However, the negative coefficient of the particle size indicates a decreased of  $G'$  modulus probably due to the strong water competition of large GPF particle.  $G'$  modulus decrease can be attributed to the dilution of constituents [35, 36].



**Figure 4.** Response surface plot showing the combined effects of grape peels flour (GPF) level and particle size of GPF on: **(a)** storage modulus ( $G'$ ); **(b)** on viscous modulus ( $G''$ )

### Viscous modulus

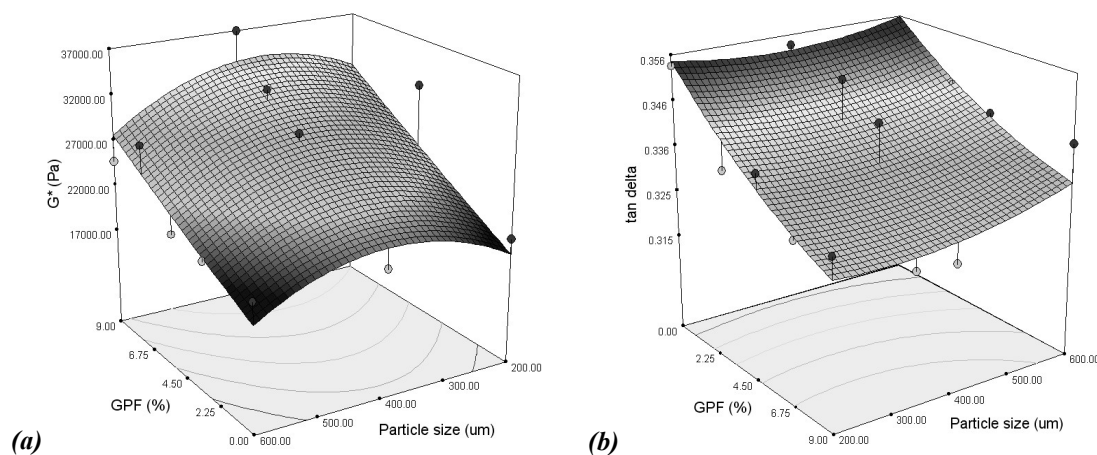
The viscous modulus,  $G''$  was dependent on the presence of GPF levels and particles sizes added in WWF. ANOVA for the quadratic model as fitted to the experimental data showed significance ( $p < 0.05$ ). The positive linear term of GPF level indicated that  $G''$  increased with the increase of GPF level added in WWF. The increased level of GPF leads to an increase of viscous modulus value as shown in Figure 4b. Also, it was observed that the decreased value of particle size increased viscous modulus probably due to the changes that occurred in starch structure. The GPF as sources of fibre competes for water with starch and can have preferential water binding effects depending of the chemical structure, the size of the particle and its porosity [37]. The addition of cellulose increased water absorption in wheat flour dough [38]. The substitution of starch with small particle of GPF may be lead to a decrease in water absorption and rise of viscous modulus.

Frequency sweep test showed that for all formulated dough's the elastic modulus was greater than the viscous modulus in the whole range of frequency, both moduli slightly increased with frequency. These variations suggest a solid elastic-like behaviour of all formulated dough's.

### Complex modulus

Complex modulus,  $G^*$  is a good indicator of stiffness of material when exposed to stresses below the yield stress. Quadratic regression model was fitted for  $G^*$  with satisfactory coefficient of determination (Table 2). ANOVA shows that the selected quadratic model is well adjusted to the experimental data of complex modulus. GPF addition level has a significant effect on  $G^*$  ( $p < 0.05$ ), while the effect of particle size was non-significant ( $p > 0.05$ ). Negative linear effects of GPF particle size and of interaction between GPF addition level and particle size were observed on  $G^*$ . A

decreased value of interaction between GPF level and particle size increased  $G^*$  probably due to the changes which occurred in starch structure. Fibers from GPF composition competes for water with starch showing different water binding effects depending on particle size [39] and the fiber structure. A great number of hydroxyl groups in the fiber structure allows for more water interaction with hydrogen bonding [40]. The increased level of GPF added in WWF leads to an increase of complex modulus values as shown in Figure 5a.



**Figure 5.** Response surface plot showing the combined effects of grape peels flour (GPF) level and particle size of GPF on: **(a)** complex modulus ( $G^*$ ); **(b)** loss tangent ( $\tan \delta$ )

### Loss tangent

The loss tangent or phase angle,  $\tan \delta$  provides information on the phase shift between the stress and strain in oscillation. This parameter, determined as the ratio of the viscous to the elastic properties of the material, at frequency of 1 Hz was significantly ( $p < 0.05$ ) influenced by GPF levels and particles sizes added in WWF. The quadratic model described well  $\tan \delta$  as a simultaneous function of GPF level and particles sizes added in WWF. The individual effect of GPF level was to decrease  $\tan \delta$  values, while the particle size increase  $\tan \delta$  (Figure 5b). The GPF level added in WWF has a significant ( $p < 0.005$ ) individual effect, while the particle size effect on  $\tan \delta$  was not significant at  $p < 0.05$ . However, the increase of GPF amounts raise the effect of increased particle size on the loss tangent further than the sum of individual effects (Table 2). In bread making, mix of flours which produces dough's with balanced tensile and elastic properties is required to provide optimal baking performance. In the literature was reported that dough with small  $\tan \delta$  reflects a rigid and stiff materials and dough characterized as moist and slack showed higher  $\tan \delta$  values than those depicted as short and dried surface appearance [41].

### Optimization of GPF level and particle size added in WWF

The optimum of GPF level and particle size addition in WWF was determined based on the desirability function applied to models fitted in this study. Simultaneously optimization of multiple responses was performed by imposing some constraints such as

$H'm$ ,  $Vt$  and  $Rc$  were desired maximal,  $Hm$ ,  $T'l$ ,  $Tx$ ,  $G'$  and  $G''$  in the range, whereas  $G^*$  and  $\tan \delta$  were specified as minimum desirable. The best combinations between the factors used in this study in order to obtain optimum values for rheofermentometric characteristics and dynamic rheological characteristics of dough were extracted by State-Ease Design Expert software which gives the maximum desirability value and the final conditions. Basis on the calculations, a total desirability value (D) of 0.579 was obtained for the optimum value of factors that indicates a GPF level of 4.66% added in wheat flour and small particle size of 200  $\mu\text{m}$ . Under these optimum factors values, the predicted rheofermentometric characteristics  $Hm$  of 34.20 mm,  $H'm$  of 67.06 mm,  $T'l$  of 70.26 min,  $Tx$  of 50.63 min,  $Vt$  of 1276.92 mL,  $Rc$  of 87.03% and dynamic rheological characteristics,  $G'$  of  $23.435 \cdot 10^3$  Pa,  $G''$  of  $7.904 \cdot 10^3$  Pa,  $G^*$  of  $26.239 \cdot 10^3$  Pa and  $\tan \delta$  of 0.336 were obtained.

## CONCLUSIONS

There are scientific reports presenting the effects of wheat flour substitution with grape peels. However, none of them takes into account the possibility to study the effects of both GPF level and particle size which can be added in WWF. Through this paper, it was highlighted that depending on GPF level and particle size, various responses could be obtained for the dough rheological characteristics. Knowing these results could be helpful to the bread making processors to predict the quality of mix flours based on WWF and dough rheological behavior. Regarding the rheofermentometric characteristics and viscoelastic properties of dough, optimization process revealed that the best results were obtained for small particle size and level of 4.67% GPF added in WWF.

## ACKNOWLEDGEMENTS

This work was supported by a grant of the Romanian National Authority for Scientific Research and Innovation, CNCS/CCCDI – UEFISCDI, project number PN-III-P2-2.1-BG-2016-0136, within PNCDI III.

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