

# Optimization of White Wheat Flour Dough Rheological Properties with Different Levels of Grape Peels Flour Addition

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## Abstract

The use of grape peels flour to improve the nutritional value of wheat flour bread has received considerable interest due to their high fibers content and bioactive compounds compared to white wheat flour. The aim of this study was to establish the optimal combination of grape peels particle size (PS) and flour replacement (FR) level with grape peel flour on the dynamic and empirical rheological properties of mix flour dough to develop innovative bakery products. The results obtained highlighted that the small PS resulted in increased Farinograph water absorption and Amylograph peak viscosity and decreased dough development time. The FR showed a significant ( $p < 0.05$ ) effect on Rheofermentometer parameters. The dynamic moduli increased when increasing FR and decreasing PS, while the loss tangent decreased with FR increase. The white wheat flour with 3.80% small particle size of GPF was found to be the best formulation in order to achieve the desirable dough rheological properties.

**Keywords:** grape peels flour, optimization, particle size, rheology, white wheat flour

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## Introduction

The addition of dietary fiber from various sources in wheat-based bakery foods is a solution to the existing problem regarding the human nutrition and health. Dietary fiber intake (25-50 g per day) has been associated with prevention or treatment of some diseases such as, regulation of the intestinal tract, diabetes, several types of cancer, etc. (Kendall *et al.*, 2010; Rodríguez *et al.*, 2006). Due to the relationships between food and health, the development of functional foods like fiber enriched-bread is increasingly explored in various works. Bread, as a regular basis, represents an interesting vehicle for dietary fiber incorporation. Agro-industrial by-products derived from fruits and vegetables processing can

become ingredients of value added products due to their nutrients and extranutritional compounds, with the associated health benefits (Gomez and Martinez, 2017). Dietary fibers from fruits have better quality than those from cereals (Figuerola *et al.*, 2005).

Grapes pomace, a by-product of wine industry, consists mainly of peels and seeds, represents approximately 20-25% of the weight of the grapes processed for wine production (Yu and Ahmedna, 2013). Grape peels and seeds are composed of water, proteins, lipids, carbohydrates, vitamins and minerals, being also a rich source of high-value compounds, such as phenolics antioxidants (flavonoids, phenolics acids and stilbenes) and dietary fiber (Nerantzis and Tataridis, 2006; Deng

*et al.*, 2011; Trošt *et al.*, 2016). The composition depends of the type of by-product, type of grape, cultivation and climatic conditions, as well as the processing conditions (Călinoiu *et al.*, 2018; Mironeasa, 2017; Karovičová *et al.*, 2015). Grape fiber met the definition of antioxidant dietary fibre (ADF) which was first proposed by Saura-Calixto (1998) with the specification that 1 g of ADF should have DPPH free radical scavenging capacity equivalent to at least 50 mg vitamin E and dietary fiber content higher than 50% dry matter from the natural constituents of the material.

Some studies have indicated the addition of grape peels in foods to increase the nutritional value and/or to improve the sensory properties, extending the shelf-life of food products. Grape peels could be directly used in the form of dried powder as a source of fiber and antioxidants or, indirectly, through the production of phenolics extracts. Dehydrated grape peels from the juice industry was added to aged and young red wines as a way to counteract the color loss before bottling (Pedroza *et al.*, 2013). A new tomato-based product was developed using dried and ground grape peels in tomato pure (Lavelli *et al.*, 2014). Grape peel flour has been used as a source of polyphenolic compounds in yoghurt formulation (Marchiani *et al.*, 2016).

A few studies have reported the enrichment of wheat-based bakery products with grape peels flour. Mildner-Szkudlarz *et al.*, (2011) incorporated milled red grape peels as a source of dietary fiber and phenolics compounds in rye sourdough bread. Acun and Gül, (2013) evaluate the effect of grape peel flour on the physical, biochemical, sensory characteristics and affordability of cookies. Maner *et al.*, (2015) evaluate the influence of different levels of grape peels powder on the physico-chemical and sensory parameters of cookies. Bender *et al.*, (2017) evaluate the effects of incorporation of peels flour from two grape varieties, red and white, in wheat flour on sensory and technological properties of the muffins. Kuchtová *et al.*, (2016) assess the influence of white grape peels variety on the sensory properties and overall acceptability of enriched cookies. In another study, Kuchtová *et al.*, (2018) reveal the effect of the incorporation of grape peel on the rheological properties of wheat dough, and on the quality parameters and sensory properties of the prepared cookies. Mironeasa *et al.*, (2018) evaluate the effects of grape peels flour

(GPF) addition, from white wine grape pomace, on mixing, pasting and fermentation characteristics of white wheat flour dough. Thus the application of grape peels by-product into baked products has resulted in increasing dietary fiber and total phenolic content, improving nutritive value, antioxidant activity, changing dough viscoelastic properties.

Based on these reported studies, it can be affirmed that grape peel should be a valuable raw material for nutritional enrichment of cereals products. However, previous research did not highlighted the optimal particle size and addition level which can be replaced in wheat flour dough in order to obtain wheat flour dough with the best rheological properties. Grape peel flour particle size and addition level has an essential impact on dough rheological behaviour and finite product quality. The incorporation of grape peels, as a source of fibers in white wheat flour bread to ensure beneficial physiological impact, causes disruption of the continuous matrix, affecting dough and bread manufacture. The trend of the effects on dough rheological parameters depends on the grape peels flour particle size in the mix and on the level of flour substitution. The optimum combination of these process factors on the responses, in terms of rheological properties, can be found by using response surface methodology (RSM) and simultaneous optimization method. The aim of this investigation was to find the optimum grape peel flour particle size (PS) and flour replacement (FR) in the formulation of grape peel-wheat flour with the purpose of achieving the best rheological properties.

## Materials and methods

### Basic ingredients

White wheat flour of 550 type (harvest 2016) was provided from S.C. Dizing S.R.L. (Brusturi, Neamț, Romania) and consisted of 13.80% moisture (ICC 110/1), 11.05% protein (ICC 105/1), 1.30% fat (ICC 105/1), 0.55% ash (ICC 104/1), 28.00% wet gluten (ICC 106/1), 4.00 mm gluten deformation index (SR 90:2007), and 351.50 s falling number index (ICC 107/1). Red wine grape pomace, *Vitis vinifera* L. provided from the viticulture center Jarișteea, Odobesti ecosystem was used in this study. The grape peels,

manually separated from dried grape pomace in open air; analyzed according to ICC methods (2010) have following chemical characteristics (in %): moisture 5.70 (ICC method 110/1), protein content 12.70 (ICC 105/1), fat content 5.51 (SR EN ISO 659:2009) and ash 5.66 (SR ISO 2171:2009). Fiber analysis by Near Infrared Reflectance (NIR) spectroscopy technology, a non-destructive and rapid technique (Fărcaș *et al.*, 2014) showed that the by-product contains 45.90%. 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 statistics**

The combinations of five levels (0, 3, 5, 7 and 9%) of white wheat flour replacement (FR) and three (L, M and S) particle sizes (PS) of grape peels flour (GPF) on dough empirical rheological properties evaluated using Farinograph, Amylograph, Rheofermentometer and dynamic oscillatory rheological properties, as dependent variables or responses, were investigated using the RSM by means of full factorial design with two independent variables, FR and PS. The dependent variables that described dough rheological properties consisted of Farinograph water absorption (WA), dough development time (DT), stability (ST), degree of softening (SDg), gelatinization temperature (Tg), peak viscosity (PVmax), temperature at peak viscosity (Tmax), maximum height of gaseous production (H'm), gas retention coefficient (Rc), storage modulus ( $G'$ ), viscous modulus ( $G''$ ), loss tangent ( $\tan \delta$ ) and complex modulus ( $G^*$ ).

A polynomial quadratic regression equation (Eq. 1) was used to describe the impact of two factors, FR and PS on the twelve responses, in terms of dough rheological properties and multiple linear regression analysis was applied to fit the data obtained for each response to linear, quadratic and cubic models.

$$Y = b_0 + b_1 \cdot X_1 + b_2 \cdot X_2 + b_{11} \cdot X_1^2 + b_{22} \cdot X_2^2 + b_{12} \cdot X_1 \cdot X_2 \quad (\text{Eq. 1})$$

where,  $Y$  is the response variable,  $b_0, b_1, b_2, b_{11}, b_{22}$  and  $b_{12}$  – regression coefficients for intercept,

linear, quadratic and interaction effects respectively of  $X_1$  and  $X_2$  independent variables (wheat flour replacement and particle size of grape peels flour).

Analysis of variance (ANOVA) was carried out on each response to identify the most adequately model, chosen through sequential  $F$ -test, coefficients of determination ( $R^2$ ), adjusted coefficients of determination ( $Adj.-R^2$ ), and the statistical significance of the coefficients in each predictive model. To determine the validity of the model for each response, the experimental and predictive values were compared by paired  $t$ -test ( $p < 0.05$ ). The combined effect of the factors, FR and PS on the response variables was modeled using a polynomial response surface. Stat-Ease Design-Expert 7.1 software (trial version) was employed to carry out the experimental design, the model analysis, generation of the response surface plots, test of model adequacy and the optimum level of formulation factors. The multiple responses analysis was applied to models fitted in this study to found the optimal value of the factors, level of wheat flour replacement and grape peels flour particle size. In the optimization process, each predicted response is converted 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, named overall desirability function,  $D$  computed as the geometric mean of the individual desirability function,  $d_n$  which varies from 0 to 1 (Wu *et al.*, 2011). Using the predictive tool, the minimum particle size of grape peels flour, as factor and maximum dough stability, minimum degree of softening, maximum gas retention coefficient, minimum complex viscosity, as responses where chosen for numerical optimization. The level of flour replacement and all remaining responses were kept within range.

#### **Empirical dough rheological properties**

Farinograph test was performed on a Brabender Farinograph®-E (Brabender OHG, Duisburg, Germany) with a 300 g capacity, according to ICC method 115/1, in order to determine dough rheological properties during mixing. Farinograph characteristics, water absorption (WA, %), as the amount of water required to reach a dough consistency of 500 BU, dough development time (DT, min), as the time

to reach maximum consistency, dough stability (ST, min), as the time which dough consistency is kept at 500 BU and degree of softening (SDg) at 10 min, as the difference between maximum dough consistency and that after 10 min were obtained from the recorded curve.

The gelatinization properties of the mix flour were determined using the Brabender Amylograph®-E (Brabender OHG, Duisburg, Germany) according to ICC method 126/1 and from the Amylograph curve, the following parameters were obtained: gelatinisation temperature ( $T_g$ , °C), peak viscosity (PVmax, BU) and temperature at peak viscosity ( $T_{max}$ , °C).

Dough rheological properties during fermentation process was recorded using the Chopin Rheofermentometer F3 (Chopin Rheo, Villeneuve-La-Garenne Cedex, France) which indicates maximum height of gaseous production ( $H'm$ , mm) and gas retention coefficient (Rc, %) in the dough at the end of the test.

#### **Dynamic rheological properties**

The dynamic rheological properties of dough were determined using a MARS 40 rheometer (Thermo-Haake, Karlsruhe, Germany) equipped with a plate-plate geometry system with a diameter

of 40 mm and a gap between plates of 3 mm, selected based on the viscosity range of dough. The dough samples for rheological measurements were done using standard dough preparation, without yeast, based on each farinograph water absorption value by mixing until full dough development. Vaseline was applied to prevent drying of the exposed samples edge during tests. A rested time of 5 min to allow relaxation and stabilize temperature was applied to all samples before testing.

Stress sweep tests in the range of strain from 0.01% to 1%, at constant oscillation frequency of 1 Hz were firstly conducted in order to define the linear viscoelasticity region, according to some indications (Miś, 2011; Lazaridou *et al.*, 2007). Frequency sweep tests were performed in the range of 1 to 10 Hz at a strain value of 0.15% and temperature of  $20.0 \pm 0.1^\circ\text{C}$  to determine the storage modulus ( $G'$ ), loss modulus ( $G''$ ), loss tangent ( $\tan \delta = G''/G'$ ) and complex modulus ( $G^*$ ).

#### **Sensory analysis**

The sensory analysis of optimum composite flour was conducted to determine the quality of raw material from the organoleptic characteristics point of view. A semi-trained panel of 17 members selected from University community performed the sensory evaluation of grape peel-wheat mix

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

Run	Coded value		Real value	
	$X_1$	$X_2$	FR (%)	PS ( $\mu\text{m}$ )
1	0.556	0.000	7.000	400.000
2	-1.000	-1.000	0.000	200.000
3	0.111	1.000	5.000	600.000
4	1.000	1.000	9.000	600.000
5	-0.333	0.000	3.000	400.000
6	-1.000	0.000	0.000	400.000
7	0.111	0.000	5.000	400.000
8	0.556	-1.000	7.000	200.000
9	0.556	1.000	7.000	600.000
10	1.000	-1.000	9.000	200.000
11	-1.000	1.000	0.000	600.000
12	-0.333	-1.000	3.000	200.000
13	0.111	-1.000	5.000	200.000
14	1.000	0.000	9.000	400.000
15	-0.333	1.000	3.000	600.000

FR, level of white wheat flour replaced with grape peels flour; PS, grape peels flour particle size added in white wheat flour



flour at optimum formulation. The mix flour formula was evaluated on the basis of their smell, taste and colour in according to the Romanian standard SR 90:2007.

## Results and discussion

### Adequacy of the model

The experiments were performed according to the experimental design to determine the combined effects of the factors, FR and PS on dough rheological properties. The quadratic regression models can predict adequately the Farinograph, Amylograph, Rheofermentometer and dynamic rheological parameters evaluated as a function of the formulation factors. The models were highly significant for most of the response variables, with reasonable  $R^2$  values that varied from 0.69 to 0.96 (Table 2). The predictive models represented well the experimental data with satisfactory  $R^2$  and  $Adj.-R^2$  values. The ANOVA results, including significantly ( $p < 0.05$ ) and non-significantly ( $p > 0.05$ ) regression coefficients, expressed in terms of coded values,  $R^2$ ,  $Adj.-R^2$  and  $p$  values are shown in Table 2.

### Effects of the level of flour replacement and grape peels particle size on wheat flour dough rheological properties during mixing, pasting and fermentation

Table 2 shows the effects of formulation factors, FR and PS on Farinograph properties, WA, DT, ST and SDg of grape peels-wheat composite flour dough exhibited as their corresponding regression coefficients in the quadratic model. The ANOVA results showed that all the models were highly significant (Table 2) and can predict adequately the relationship between the independent variables and the response variables. The effects of factors FR and PS on Farinograph properties were shown in the response surface plots from Figure 1a-d.

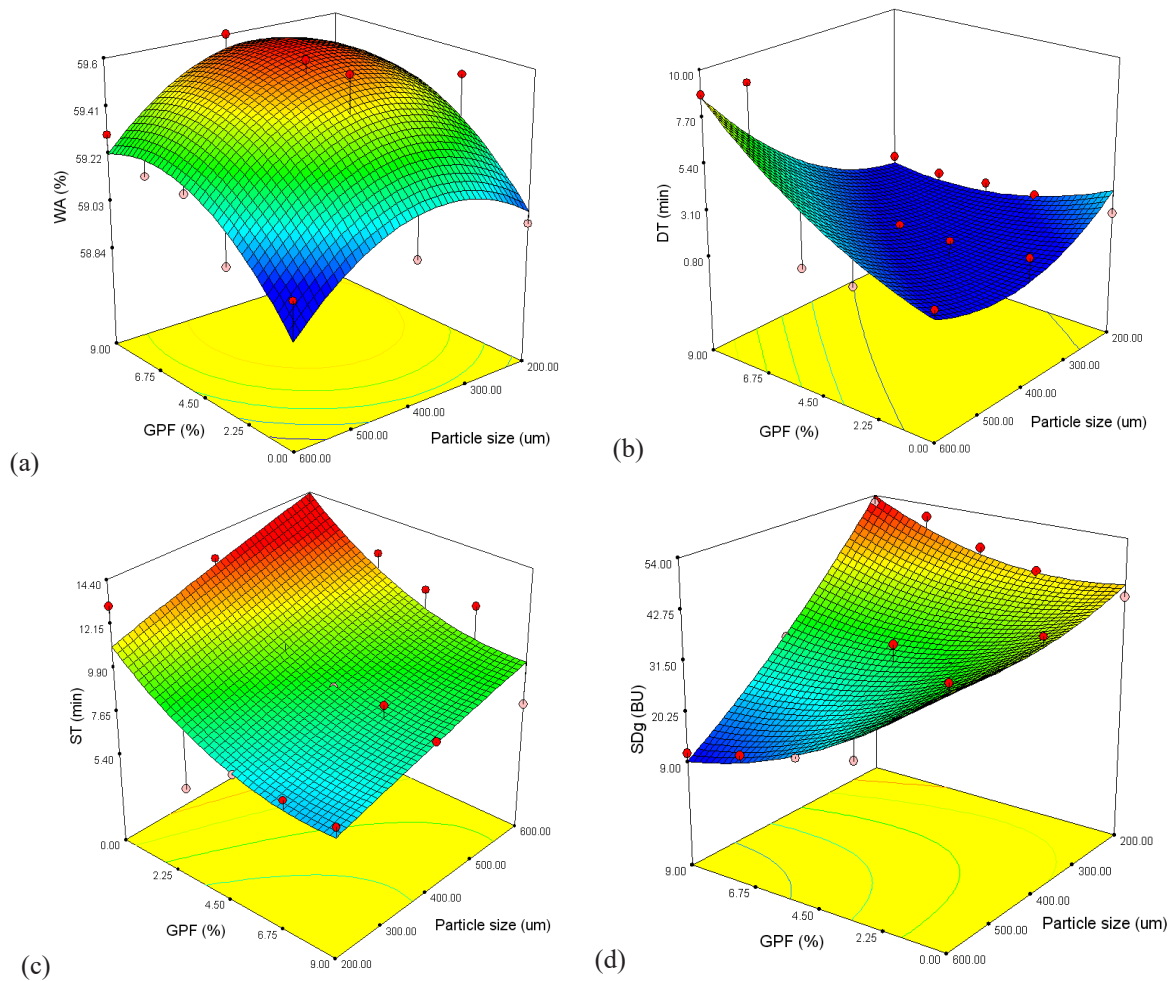
The equation for WA indicated that FR and PS had significant ( $p < 0.05$ ) effect on it. The  $R^2$  (0.80) and  $Adj.-R^2$  (0.69) values showed that the quadratic regression model defined well the real behavior of dough among WA. The positive coefficient of FR showed that the WA increased with FR increase, while the negative coefficient of PS showed that WA decreased with PS increase. The effects of FR and PS on WA of grape peels-wheat composite flour dough are shown in Figure

**Table 2.** Effects of formulation factors, expressed as their corresponding coefficients in the predictive models for dough properties during mixing and pasting<sup>b</sup>

Factors <sup>a</sup>	Properties						
	Farinograph				Amylograph		
	WA (%)	DT (min)	ST (min)	SDg (BU)	Tg (°C)	PVmax (BU)	Tmax (°C)
Constant	59.93	1.33	9.24	28.69	60.85	1396.25	89.69
FR	0.19***	1.28*	- 2.22**	- 4.96***	0.18***	365.86***	0.65**
PS	- 0.10**	1.36**	1.52**	- 12.33***	- 0.00*	- 100.56***	0.11*
FR x PS	0.00*	2.19**	-0.13*	- 10.05***	0.07*	- 48.58*	0.06*
FR <sup>2</sup>	- 0.18**	0.95*	1.33*	5.02**	0.19**	7.32*	- 0.35*
PS <sup>2</sup>	- 0.22**	1.57*	- 0.09*	2.80*	0.00*	30.20*	- 0.15*
R <sup>2</sup>	0.80	0.69	0.70	0.96	0.72	0.94	0.76
Adj.-R <sup>2</sup>	0.69	0.52	0.53	0.94	0.56	0.91	0.63
p-value	< 0.01	< 0.05	< 0.005	< 0.0001	< 0.05	< 0.0001	< 0.05

<sup>a</sup> FR, level of white wheat flour replacement with grape peels flour (%); PS, grape peels flour particle size (mm);  $R^2$ ,  $Adj.-R^2$  are measures of model fit; \*  $p > 0.05$ ; \*\*  $p < 0.05$ ; \*\*\*  $p < 0.005$ .

<sup>b</sup> WA, water absorption; DT, development time; ST, stability; SDg, degree of softening; Tg, gelatinization temperature; PVmax, peak viscosity; Tmax, temperature at peak viscosity



**Figure 1.** Response surface plot showing the combined effects of grape peels flour (GPF) level and particle size of GPF on Farinograph parameters: (a) water absorption (WA); (b) dough development time (DT); (c) stability (ST) and (d) degree of softening (SDg)

1a. The surface plot indicates that with the FR increase and PS decrease, the WA increased. The WA value depends on PS of GPF, large particle sizes decreasing the values of dough WA. These results may be attributed to the larger PS phenomenon of GPF, with lower surface area, in comparison to that of white wheat flour. A decrease in WA with the increase of PS may be due to the chemical structure of the fibers from GPF added, the porosity of the fibers and the association between molecules, in agreement with the results reported about dietary fiber (Thebaudin *et al.*, 1997). WA was not significantly correlated ( $p > 0.05$ ) with interaction effect between FR and PS, while the quadratic effect of FR and PS on WA was significant ( $p < 0.05$ ). These results may be attributed to the decrease of the osmotic pressure outside of the protein micelle which will intensify the water

osmotic absorption. This is a consequence due to the chemical structure of fibers from GPF added, the porosity of fibers and the association between molecules, as previously reported about dietary fiber (Thebaudin *et al.*, 1997). A possible reason for the increase in water absorption with the decrease of GPF particle size might be the hydroxyl groups in phenolics compounds which directly interact with proteins, and consequently influencing WA according to the results obtained by Zhang *et al.*, (2010) who added tannic acid to dough.

The effect of FR and PS on DT, presented as their corresponding regression coefficients in the quadratic regression models, is shown in Table 2. The model can be suitable to predict DT as a function of the FR and PS factors. As for DT parameter, the coefficient of determination ( $R^2$ ) is 0.69, indicating that the model explains only 69%

of the observed data variation. The formulation factor had significant effects on DT of grape peels-wheat composite flour dough. The ANOVA results showed that linear term of PS and the interaction coefficient between FR and PS were significant, while the linear term of FR was insignificant ( $p > 0.05$ ) (Table 2). The PS and the interaction effect between FR and PS have a positive linear effect on DT. The effect of FR and PS factors on DT value as response surface plot is shown in Figure 1b.

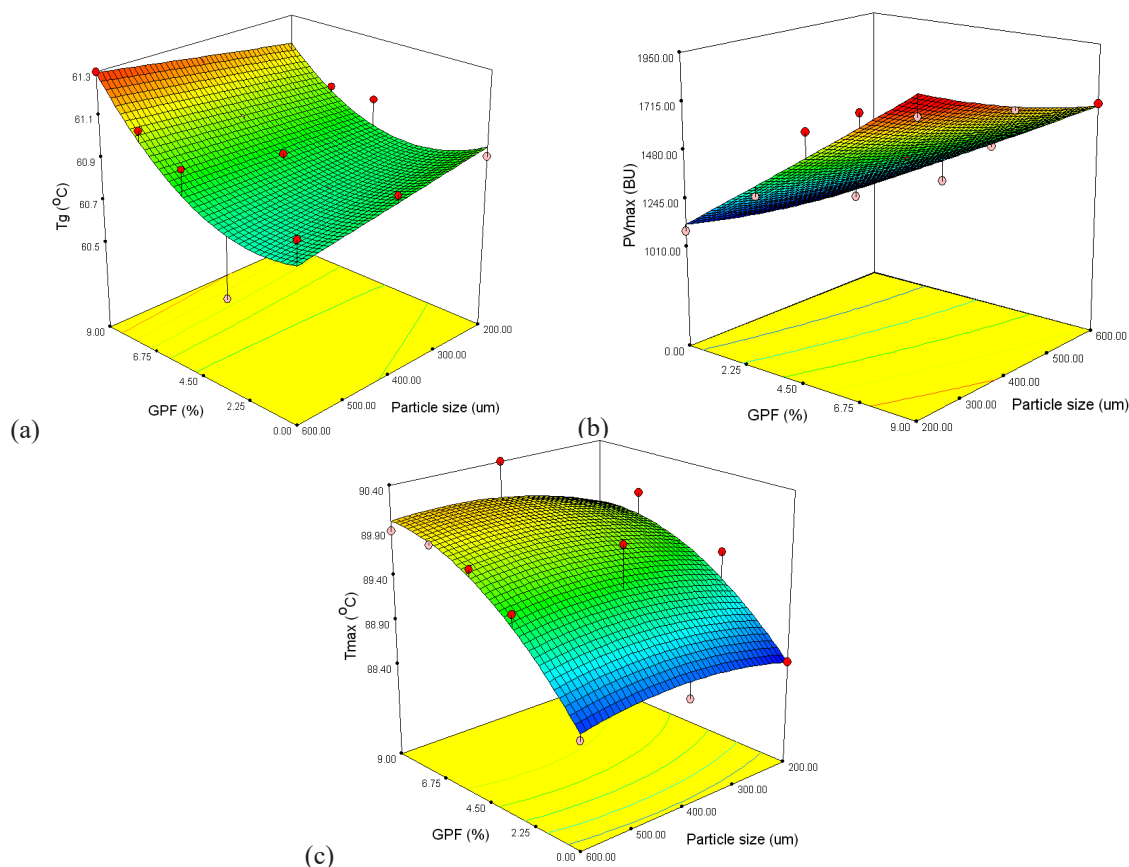
The effect of FR and PS factors on DT value as response surface plot is shown in Figure 1b. The response surface plot revealed that DT increased as the PS increased. The increase in DT may be explained by the increase of WA which indicates that dough absorbs more water and, therefore, it requires more mixing time. Greater PS needs longer hydration time and thus, a higher DT.

Dough ST was significantly influenced ( $p < 0.005$ ) by the FR and PS and the quadratic model fitted to the experimental results of ST, explaining 70% of the obtained data variation (Table 2). The statistical analysis of the model coefficients showed that the linear terms were significant, while the interaction coefficient and quadratic terms were non-significant ( $p > 0.05$ ) (Table 2). A negative effect on ST was provided by the linear term of FR, suggesting that the increased level of FR with GPF in white wheat flour induced a decrease in ST value (Figure 1c). The decrease in ST with the increase of FR level may be related to the lower water availability in the dough system, the water absorption capacity of dough decreasing with the increase of FR level. Also, the gluten dilution, as a result of an addition of non-gluten flour, diminished the dough viscoelastic properties (Mohammed *et al.*, 2014) and, therefore, the ST decrease.

At high FR level the gluten matrix may appear with physical interruption which can lead to the dough weakening. A decrease of dough stability with the FR level increase was reported in previously study (Mironeasa *et al.*, 2018; Bono, 2014). The positive sign of the significant coefficient included in the quadratic model showed that ST increased with PS increase. According to Figure 1c, large PS of GPF led to a higher value for ST, probably due to the amount of fibers and other constituents present in the large PS which can stabilize the hydrophobic interactions between gluten proteins.

The ANOVA results indicated that FR, PS and the interaction between these factors (FR x PS) have significant negative effect on SDg (Table 2). Also, the quadratic coefficient of FR had a significant effect ( $p < 0.05$ ) on SDg, while the quadratic coefficient of PS did not show significant effect ( $p > 0.05$ ) on SDg. For SDg, the quadratic regression model obtained was an adequate one, with a high coefficient of determination ( $R^2 = 0.96$ ). Compared to the FR, PS of GPF which replaces white wheat flour has a higher influence on SDg of the composite flour dough. The response surface plot revealed that the SDg decreased as the FR and PS increased (Figure 1d). The decrease in SDg may be correlated with the presence of fiber from GPF which has the ability to increase the dough viscosity, L and M particle sizes increase more the viscosity as compared to S particle size (Mironeasa *et al.*, 2018). The decrease in SDg can be related to a higher amount of insoluble fibers comparatively with soluble fiber from mix flour dough which can have a strengthening effect on the dough. Probably, with PS increase the amount of insoluble fibers is higher than the soluble ones and it decreases the softening degree of dough. Also, an increase in PS can lead to an increase of insoluble to soluble dietary fiber ratio, as an increase of soluble to insoluble dietary fiber ratio with decreased of particle size was reported (Cappa *et al.*, 2015).

The effect of FR and PS on the gelatinization temperature (Tg), peak viscosity (PVmax) and temperature at peak viscosity (Tmax) exhibited as their corresponding regression coefficients in the quadratic models are shown in Table 2. The quadratic regression model represents accurately the experimental results of Tg, the  $R^2$  value (0.72), confirming the adequacy of the model. The linear and quadratic terms of FR influenced significantly Tg parameter, while the PS was not significant ( $p > 0.05$ ). The effect of FR and PS of GPF which replace wheat flour can be seen in Figure 2a, indicating an increase in Tg with FR increase. A possible reason for this effect can be due to the varying gelatinization temperature of the fiber fractions from GPF (Mironeasa *et al.*, 2018). The increase in Tg value lead to a delaying starch gelatinization process which can be related to the decrease of water absorption by the starch granules due to the compounds from mix flour. Grape peels contain high amounts of mineral elements (Oprea *et al.*, 2018). The increase in calcium content from mix



**Figure 2.** Response surface plot showing the combined effects of grape peels flour (GPF) level and particle size of GPF on: (a) gelatinization temperature (Tg); (b) peak viscosity (PVmax) and (c) temperature at peak viscosity (Tmax)

flour by incorporating GPF stabilizes the alpha-amylase enzyme activity, unable to function in calcium absence (Mironeasa *et al.*, 2012; 2016) and destabilizes the wheat flour starch component.

The quadratic model which was obtained for the Amylograph PVmax showed high correlation coefficient ( $R^2 = 0.94$ ). It is easily observed from the Table 2 that PVmax was influenced significantly ( $p < 0.005$ ) by the FR and PR formulation factors. The effect of FR and PS on PVmax is shown in Figure 2b. The response surface plot revealed an increase of FR with the decrease of PS of GPF which replaced wheat flour increased PVmax, leading to the increase of the viscosity of the dough gel formed. The results are in agreement with those reported by Bono (2014) and Mironeasa *et al.*, (2018). The small PS of GPF lead to a higher PVmax and lower tendencies to retrogradation, it's appeared to interfere with the starch matrix during cooling. The increase in PVmax may be explained to the fact the amylolytic activity in mix flour decreased by small PS of GPF addition in white wheat flour.

When large particle size of GPF was added, the PVmax value decreases as a consequence of alpha-amylase activity increase. In addition, the large PS can destabilize wheat flour starch component.

Through ANOVA, the quadratic model was found to represent adequately the experimental data for Tmax, the  $R^2$  value (0.76), confirming the adequacy of the model. The linear coefficient of FR indicated a significant positive influence on the Tmax, while the linear term of PS was not significant. The effect of FR and PS can be seen in Figure 2c, indicating an increase in Tmax.

The maximum height of the gaseous production ( $H'm$ ) is a critical parameter in fermentation process and is related to the maximum height of dough development and to the height of dough development at the end of the test. Table 3 shows the effects of FR and PS formulation factors on maximum height of gaseous production ( $H'm$ ). The quadratic model predicts adequately the  $H'm$  as a function of the formulation factors. The regression model indicated that the linear and



quadratic terms of FR had significant ( $p < 0.05$ ) effects on  $H'm$  parameter; while PS was non-significant effect ( $p > 0.05$ ). Also, the interaction term between factors was non-significant ( $p > 0.05$ ). The ANOVA results showed that the model obtained was an adequate one with a good  $R^2$  value of 0.72. A response surface plot, showing the effect of FR level and PS on  $H'm$ , is represented in Figure 2a. As it can be seen, the  $H'm$  significantly decreased as the FR level increased, probably due to the increase of the osmotic pressure on the yeast cells, which can lead to an inhibitory effect on the yeast metabolism. Also, the interactions between gluten and the fibrous fraction from GPF may prevent the free expansion of dough during proofing (Mironeasa *et al.*, 2018).

The decrease of  $H'm$  parameter can be due to the effect of disruption of hydrogen and hydrophobic bonds, the structure of gluten network depending on non-covalent (hydrogen and hydrophobic) bonds as well of disulfide bonds. Hydrogen bonding with water increases by gluten hydration. When these bonds are disrupted it will affect the fermentation properties of dough (Visireddy, 2011). The function of gluten depends on his molecular weight, the formation of covalent and non-covalent bonds between glutenin molecules and the interactions between

glutenin and other flour constituents (Goesaert *et al.*, 2005).

The retention coefficient ( $R_c$ ) defined as the retention volume divided by the total gaseous release, increased by reducing the surface tension. The effect of FR level and PS of GPF which replaced white wheat flour on  $R_c$  expressed as their corresponding regression coefficients in the quadratic regression model are shown in Table 3.  $R_c$  was significantly dependent ( $p < 0.005$ ) on the level of FR. Also, the quadratic term of FR factor was found to have a significant negative influence on  $R_c$ . The ANOVA shows that the selected quadratic model is well adjusted to the experimental data for  $R_c$ . The regression model was highly significant, showing higher  $R^2$  value (0.95).

The response surface obtained for  $R_c$  (Figure 2b) showed that the increasing of FR level increased  $R_c$  parameter. This may be due to the reduced surface tension through the increase FR and PS which positively affects the fermentation properties by increasing the retention coefficient. The increase in  $R_c$  parameter may be related to the decrease in hydrogen and hydrophobic bonds, the disruption of these bonds positively contributes to the retention coefficient (Visireddy, 2011). A high  $R_c$  value can be related to the gas efficiently

**Table 3.** Effects of formulation factors, expressed as their corresponding coefficients in the predictive models for Rheofermentometer and dynamic rheological properties<sup>b</sup>

Factors <sup>a</sup>	Properties					
	Rheofermentometer		Dynamic rheological			
	$H'm$ (mm)	$R_c$ (%)	$G'$ (Pa)	$G''$ (Pa)	$\tan d$	$G^*$ (Pa)
Constant	60.35	89.43	40888.61	13384.00	0.330	43024.05
FR	-2.83**	5.66***	11807.24***	3709.42**	-0.004***	12375.69***
PS	1.03*s	0.19*	-414.58*	-160.59*	0.000*	-443.98*
FR x PS	0.96*s	0.22*	-876.30*	-258.18*	0.000*	-913.14*
FR <sup>2</sup>	4.84**	-7.71***	728.25*	96.02*	-0.006***	722.05*
PS <sup>2</sup>	0.09*s	0.72*	-515.00*	-86.20*	0.002*	-516.68*
$R^2$	0.7221	0.9258	0.7641	0.7369	0.7659	0.7618
Adj.- $R^2$	0.5678	0.8846	0.6331	0.5908	0.6359	0.6295
$p$ -value	< 0.05	< 0.0001	< 0.05	< 0.05	< 0.05	< 0.05

<sup>a</sup> FR, level of white wheat flour replacement with grape peels flour (%); PS, grape peels flour particle size (mm);  $R^2$ , Adj.- $R^2$  are measures of model fit; \* $p > 0.05$ ; \*\* $p < 0.05$ ; \*\*\* $p < 0.005$ .

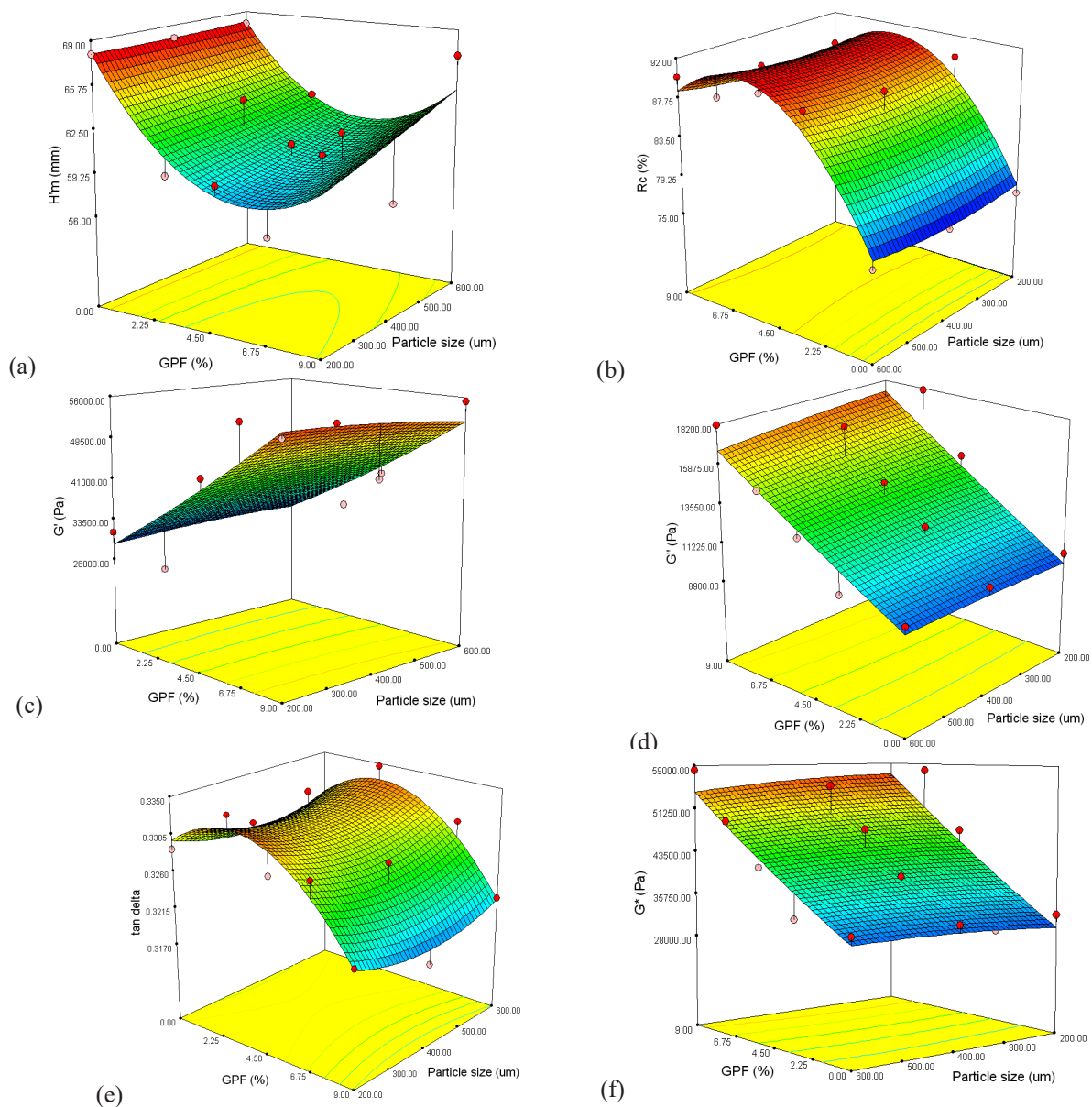
<sup>b</sup>  $H'm$ , maximum height of gaseous production;  $R_c$ , gas retention coefficient;  $G'$ , storage modulus;  $G''$ , viscous modulus;  $\tan d$ , loss tangent and  $G^*$ , complex modulus.

retained in dough and one would expect to obtain an improving loaf volume in bread.

### ***Effects of the level of flour replacement and particle size on dough dynamic rheological properties***

The effect and the significance of structural ingredients, like grape peels in dough system can be revealed through oscillatory measurements in the LVR. Different resistance to the rupture by stress action on formulation dough structures can be evaluated by measuring the elastic or storage

modulus,  $G'$ . The linear term of FR level with GPF influenced significantly ( $p < 0.005$ )  $G'$  modulus, while the PS was not significant (Table 3). The quadratic regression model are well adjusted to the experimental data of  $G'$ , the coefficient of determination value ( $R^2 = 0.76$ ) confirming the adequacy of the model. The influence of FR and PS on  $G'$  modulus can be seen in Figure 3c, indicating an increase in  $G'$ , the dough presenting a rise elasticity due to the FR more so than to the PS. The PS factor was found to have a negative influence on the  $G'$ , insignificantly at  $p < 0.05$ .



**Figure 3.** Response surface plot showing the combined effects of grape peels flour (GPF) level and particle size of GPF on: (a) maximum height of gaseous production ( $H'm$ ), (b) gas retention coefficient ( $R_c$ ), (c) elastic modulus ( $G'$ ), (d) viscous modulus ( $G''$ ), (e) loss tangent ( $\tan \delta$ ) and (f) complex viscosity ( $G^*$ )

The viscous modulus ( $G''$ ) is significantly ( $p < 0.05$ ) influenced by the FR level and PS. The positive linear term of FR revealed that the level of FR increase with GPF lead to increase of viscous modulus. PS and the interaction term between factors have a non-significant ( $p > 0.05$ ) effect on  $G''$  (Table 3). The quadratic regression model which was obtained for  $G''$  showed good correlation coefficient ( $R^2 = 0.73$ ). Response surface plot showed that an increased level of GPF in WF increased viscous modulus. The effect of FR level and PS on  $G''$  is shown in Figure 3d. The response surface plot revealed that the magnitude of  $G''$  increase is proportional with the increase of FR level and decrease of particle size of GPF. This behavior can be attributed to the different proportion of the insoluble and soluble fractions of dietary fiber present in the GPF, the changes in viscosity are more influenced by fiber structure than by chemical composition (Rosell *et al.*, 2009). In this study the water was adjusted based on Farinograph water absorption of wheat flour and wheat flour replaced with GPF. The additional water added to dough to compensate for addition of GPF results in excess free water as fibers from GPF take longer to hydrate, in agreement with the results reported by Singh *et al.*, (2012) for corn bran. The GPF particle size played an important role as well. As the particle size decreased, the specific surface area, porosity and capillary attraction of fiber increased, requiring smaller time to hydrate and dough viscosity may increase. Comparatively to the L and M particle sizes, S particle size requires smaller time to hydrate due to large specific surface area and porosity, decreasing particle size can lead to viscosity increase.

The ANOVA results revealed that loss tangent ( $\tan \delta$ ) was not significantly associated ( $p > 0.05$ ) with PS, but the linear and quadratic coefficient of FR level indicated a significant negative influence on the  $\tan \delta$  (Table 2). As for  $\tan \delta$ , a good coefficient of determination ( $R^2 = 0.76$ ) was obtained for the quadratic model, statistically significant at  $p < 0.05$ , revealed that this model is a predictive one for this parameter. As can be seen in Figure 3e, the response surface plot showed a decrease in  $\tan \delta$  as the level of FR increased.

The complex modulus ( $G^*$ ), as total resistance to deformation of dough, was significantly influenced ( $p < 0.05$ ) by the FR level and PS of GPF which replaced white wheat flour. For  $G^*$ ,

the quadratic regression model obtained was an adequate one, indicating a good  $R^2$  value of 0.76. The regression model for  $G^*$  showed a significant effect in linear term of FR, while the linear term of PS did not show significant effect ( $p > 0.05$ ) on  $G^*$  (Table 3). The response surface plot (Figure 3f) indicated an increase in total resistance to deformation of dough with FR level increase. We may presume that these increases may be attributed to the high content of fiber from GPF, especially insoluble fiber, which replaced white wheat flour knowing that the dough with fiber enrichment formed stiffer structure than the wheat sample (Ahmed, 2014). Insoluble fiber compounds of GPF can interact with the gluten matrix through hydrogen bonds, increasing dough stiffness or acting as filler in the viscoelastic matrix.

#### ***The optimization of flour replacement and grape peels particle size***

The models fitted in this study for dough rheological properties were used for simultaneous optimization by applying the desirability function. Dough rheological properties are essential factors to improve dough machinability, especially in composite flour dough with different grape peel particle sizes. The multiresponse optimization was applied to minimize the GPF particle size, degree of softening and complex viscosity and to maximize dough stability and gas retention coefficient simultaneously. The results for optimization suggested that mix flour based on white wheat flour containing 3.80% of GPF of 200  $\mu\text{m}$  particle size could be a good composite flour to achieve the best grape peel-wheat composite flour in terms of dough rheological properties investigated. The values predicted by the equations of the model for each response in terms of rheological properties during mixing, pasting and fermentation process were found to be as follows: water absorption, WA of 59.38%, dough development time, DT of 1.70 min, dough stability, ST of 7.99 min, degree of softening, SDg of 43.15 BU, gelatinization temperature, Tg of 60.84°C, peak viscosity, PVmax of 1463.14 BU, temperature at peak viscosity, Tmax of 89.33°C, maximum height of gaseous production, H'm of 60.12 mm, and gas retention coefficient, Rc of 88.94%. For the dynamic rheological properties the predicted response variables at optimum formulation showed an elastic modulus,  $G'$  of 38845.54 Pa, viscous modulus,  $G''$  of 12847.55 Pa,

loss tangent,  $\tan \delta$  of 0.33 and complex modulus,  $G^*$  of 41624.94 Pa.

The results obtained suggested that grape peels flour with particle size of 200  $\mu\text{m}$  could replace white wheat flour at a level of 3.80% to achieve the best grape peel-wheat composite flour in terms of dough rheological properties investigated.

### Sensory analysis

The results of sensory analysis revealed that the optimum grape peel-wheat composite flour formulation was characterized by a pleasant, fruity-acidic and easily grape specific smell. The taste was also pleasant, a little sweet, and specific to wheat flour taste with light red grape notes. The colour was darker than the control sample, without grape peel flour, with brown particles uniformly distributed in the wheat flour.

This optimum grape peel-wheat composite flour formulation can be used to enrich cereal products such as bread, cookie, cakes, muffins, breakfast cereal, snacks and pasta allowing the incorporation of natural functional ingredients and also adding extra health benefits. Further studies are needed for the application of GPF related to the sensory parameter of final products.

### Conclusion

Response surface methodology with a multiple response optimizations represent a statistical tool which allowed us to propose the optimal grape peels particle size and level of grape peels flour that can replace white wheat flour in order to obtain wheat flour dough with the best rheological properties. Based on this, the grape peels flour could be used by the food processors to obtain the desired properties. White wheat flour replacement at different levels and particle sizes with grape peels flour decreased dough stability, softening degree and changed the viscometric and fermentation properties of grape peels-wheat flour dough. The peak viscosity, temperature at peak viscosity and gas retention coefficient increased with the increase of level of flour replacement. The level of flour replacement and particle size limited in a significant way the water absorption from the dough system. According to our results indicated by the replacement of white wheat flour

with grape peels flour, especially at high levels and small particle size, dough became stronger by an increase of viscous modulus and a decrease of loss tangent.

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