

1 Article

2 Use of response surface methodology to investigate 3 the effects of sodium chloride substitution with 4 potassium chloride on dough rheological properties

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9 Received: date; Accepted: date; Published: date

10 **Abstract:** Bakery products are one of the main sources of dietary sodium intake of the world's
11 population. During the last decade the sodium intake has increased worldwide and now a days the
12 WHO World Health Organization recommends us to reduce sodium intake up to 2 g Na/day. KCl
13 is the leading substitute for reducing sodium in bakery products. Due to this fact the main purpose
14 of our study was to investigate the impact of sodium reduction on dough rheological properties by
15 reformulation the dough recipe using two types of salts namely NaCl and KCl in different amounts
16 addition in wheat flour. In order to establish their combination for obtaining optimum dough
17 rheological properties the response surface methodology (RSM) by the Design Expert software was
18 used. The effect of combined NaCl and KCl salts were made on mixing, viscometric and
19 fermentation process by using Farinograph, Extensograph, Amylograph and Rheofermentometer
20 devices. On dough rheological properties KCl and NaCl presented a significant effect ($p < 0.01$) on
21 water absorption, stability, energy, dough resistance to extension, falling number and all
22 Rheofermentometer analyzed values. Mathematical models were achieved between independent
23 variables, the KCl and NaCl amounts, and the dependent ones, dough rheological values. The
24 optimal values obtained through RSM for the KCl and NaCl salts were of 0.37 gKCl/100g and 1.31 g
25 NaCl/100 g wheat flour fact that leads to a 22% replacement of NaCl in dough recipe.

26 **Keywords:** KCl; NaCl; rheological properties; multiple criteria optimization; desirability functions.

27

28 1. Introduction

29 A high dietary sodium intake may leads to cardiovascular, bone demineralization and cancer
30 diseases [1, 2]. According to the American Heart Association, the cardiovascular diseases are the
31 leading cause of mortality globally [3]. The close association between hypertension values and
32 sodium intake is an important issue from a public health perspective. Nowadays World Health
33 Organization (WHO) recommends us to not exceed a sodium consumption of 5 g per day [4] and
34 wants to reduce sodium intake up to 2 g Na/day [5]. In addition to the beneficial effects on health,
35 sodium reduction also contributes to an annual decrease of medical expenditures. Bread is considered
36 worldwide to be an essential food for human nutrition. However, it might represent an important
37 source of sodium intake. The increased consumption of bakery products increases the risk of diseases
38 associated with sodium consumption [7,8]. The sodium sources in bakery products are provided by
39 ingredients such as sodium bicarbonate, a baking agent, widely used in baking and sodium chloride
40 (NaCl) which is one of the main ingredients used in the bakery manufacturing process [9]. The sodium
41 chloride additions in bakery recipe are very important from the a technological point of view [10]. Its
42 reduction may leads to negative effects on technological process of bakery products and the quality
43 of the finished products [11-14]. From the technological point of view, the sodium chloride addition

44 increases dough strength and stability, its capacity to retain gases and at low levels yeast activity
45 [10,11]. To bakery products NaCl increases the shelf-life due to the inhibition effect on microbial
46 growth, it improves bread texture and its sensory properties [13,14]. Due to the effect of sodium
47 chloride on the technological process and the quality of the bakery products its substitution in order
48 to reduce the sodium content from the bakery products is a problematic issue. Previous studies
49 have shown that the potassium chloride (KCl) is the leading substitute for reducing sodium in bakery
50 products [9,10,15-17]. Potassium chloride is a natural ingredient obtained from rock and sea salts
51 with extraction methods similar to those of sodium chloride. The effect of potassium chloride
52 consumption in the human diet is associated with a low risk of high blood pressure and other diseases
53 associated with it, the effect being contrary to the intake of sodium chloride [18,19]. It has an intrinsic
54 salty taste but with a metallic and bitter after taste when high levels are incorporated in bakery
55 recipes [20]. Therefore, the complete replacement of sodium chloride in bakery products it is not
56 recommended. Its use in food products maybe only in combination with sodium chloride in order to
57 obtain products of a high quality [10, 16]. Reducing sodium by replacing it with potassium chloride
58 has to be done gradually because of its influence on technological process and quality of the bakery
59 products [21]. The Response Surface Methodology (RSM) has been used in several food related
60 papers and applications. In the literature some applications of RSM to flour, dough and bread,
61 demonstrate its effectiveness. In particular, Cappelli et al. (2020) developed optimization charts
62 regarding the milling process of wheat and for flour characterization [22]. Moreover, Cappelli et al.
63 (2018) published predictive models of the rheological properties of doughs specifically developed
64 with RSM [23]. The aim of this study was to analyze the effect of partial sodium chloride substitution
65 with potassium chloride on the technological process of the bakery products. For this purpose, we
66 used KCl and NaCl in different combinations by using response surface methodology (RSM) in order
67 to analyze their effect on dough rheological properties and to obtain their optimum formulation from
68 the technological point of view.

69 2. Materials and Methods

70 2.1. Materials

71 Refined wheat flour (harvest 2019) was provided by the S.C Mopan S.A. (Suceava, Romania).
72 The NaCl and KCl were purchased from the Romania market. A high quality wheat flour was used.
73 This is confirmed by the characteristics analyzed by the Romanian and international standard
74 methods: 0.65 g/ 100g ash (ICC 104/1), 14.0 g/100 g moisture (ICC 110/1), gluten deformation index 6
75 mm (SR 90:2007), 12.67 g/100g protein (ICC 105/2), wet gluten 30 g /100g (ICC106/1), falling number
76 442 s (ICC 107/1) [24].

77 2.2. Dough rheological properties during mixing and extension

78 In order to analyze dough rheological properties during mixing a Farinograph device
79 (Brabender, Duisburg, Germany, 300 g capacity) was used. The dough rheological properties during
80 extension were analyzed using the Extensograph device (Brabender, Duisburg, Germany). The
81 Farinograph values analyzed through ICC method 115/1 were: water absorption (WA), dough
82 stability (ST), dough development time (DDT) and degree of softening at 10 minutes (DS). The
83 Extensograph values analyzed through ICC method 114/1 were: resistance to extension (R_{50}),
84 maximum resistance to extension (R_{max}), energy (E) and ratio number (R/E) at a proving time of 135.

85 2.3. Dough viscometric rheological properties

86 In order to analyze the dough viscometric rheological properties an Amylograph (Brabender
87 OGH, Duisburg, Germany) and Falling Number (Perten Instruments AB, Sweden) devices were used.
88 Amylograph trials were performed according to ICC method 126/1: gelatinization temperature (T_g),
89 temperature at peak viscosity (T_{max}) and peak viscosity (PV_{max}). With respect to falling number trials,
90 the ICC method 107/1 was applied.

91 2.4. Dough rheological properties during fermentation

92 The dough rheological properties during fermentation were determined by using an
 93 Rheofermentometer device (Chopin Rheo, type F3, Villeneuve-La-Garenne Cedex, France). The
 94 fermentation parameters analyzed according to AACC method 89-01.01. were: maximum height of
 95 gaseous production (H'_m), volume of the gas retained in the dough at the end of the test (VR), total
 96 CO_2 volume production (VT) and retention coefficient (CR).

97 2.5. Experimental design and statistical analysis

98 In order to analyze the simultaneous effects of the KCl and NaCl amounts on the rheological
 99 properties of the wheat flour dough, the response surfaces methodology (RSM) was used. RSM has
 100 important application in the design, development and formulation of new products, to optimize the
 101 formulations factors [25-27] or to determine the optimum conditions for the process [28], showing the
 102 effect of the factors on the responses. The RSM is one of the most popular methods for evaluating the
 103 significance of the effects of independent variables on system responses and to decide which ones
 104 may be consider in the final model [22-24]. Results optimization by the RSM method involved three
 105 main steps: statistical design of the experiment, then, determination of the mathematical models
 106 coefficients and finally, prediction of the responses and checking the adequacy of the mathematical
 107 model within the design of the experiment (DOE) using the Design Expert software, trial version 12
 108 (Stat-Ease, Inc., Minneapolis, USA). For this study two independent variables were chosen as follows:
 109 the influence of the variations of the potassium chloride amount ($A=X_1$) and sodium chloride ($B=X_2$)
 110 on the rheological parameters (dependent variables) of wheat flour dough. The experimental designs,
 111 with the real and coded values of the independent variables, are shown in Table 1.

112 **Table 1.** Real and coded values of independent variables.

113 used in the experimental design

Run	Real value		Coded value	
	KCl ¹ (g/100g)	NaCl ¹ (g/100g)	X ₁	X ₂
1	0.3	0.3	-1	-1
2	1.5	0.3	1	-1
3	0.3	1.5	-1	1
4	0.9	0.9	0	0
5	0.9	0.9	0	0
6	0.9	0.9	0	0
7	1.5	0.9	1	0
8	0.9	0.9	0	0
9	1.5	1.5	1	1
10	0.9	1.5	0	1
11	0.9	0.9	0	0
12	0.9	0.3	0	-1
13	0.3	0.9	-1	0

114 ¹KCl - potassium chloride; NaCl - sodium chloride.

115
 116 The rheological parameters determined to Farinograph were: WA - water absorption (Y_1); DT -
 117 development time (Y_2); ST - stability of dough (Y_3); DS - degree of softening (Y_4). The rheological
 118 parameters determined to Extensograph were: E - Energy (Y_5); R₅₀- resistance to extension up to 50
 119 mm (Y_6); R_{max} - maximum resistance (Y_7); R/E - (Y_8). Moreover, the Falling Number index values (Y_9)
 120 has been determined. The rheological parameters determined to Amylograph were: T_g -
 121 gelatinization temperature (Y_{10}); PV_{max} - peak viscosity (Y_{11}); T_{max} - temperature at peak viscosity (Y_{12}),
 122 H'_m - height under constraint of dough at maximum development time (Y_{13}), VT - total volume of
 123 CO_2 produced during fermentation (Y_{14}), VR - volume of the gas retained in the dough at the end of

124 the test (Y_{15}) and CR - retention coefficient (Y_{16}). In order to minimize the measurement errors of the
 125 experimental data, the rheological values obtained for the wheat flour samples with different levels
 126 of KCl and NaCl addition according to our experimental design were carried out twice. In the
 127 statistical processing their average values were used.

128 The predicted responses of the system ($Y_{1...n}$) (Eq. 1) in factorial screening experiments have
 129 been defined by a mathematical model:

130

$$Y = f(X_1, X_2) = \beta_0 + \sum_{i=1}^n \beta_i \cdot X_i + \sum_{i=1}^{n-1} \sum_{j=i+1}^n \beta_{ij} \cdot X_i \cdot X_j + \sum_{i=1}^n \beta_{ii} \cdot X_i^2 + \varepsilon \quad (1)$$

131 where: β_0 is the constant coefficient; β_i is the linear coefficient; β_{ij} is the interaction coefficient;
 132 β_{ii} is the quadratic coefficient; n is the number of factors studied and optimized in the experiment; X_i
 133 and X_j are the coded values of the independent variables and ε is the residual associated with the
 134 experiment. The residuals associated with the experiment were used to calculate the standard
 135 deviation values for each dependent variable. The significance of the model terms is evaluated by
 136 ANOVA, which performs a comparison of the variation in the response with the variation due to
 137 random error, at the probability value (p -value) of 95%. The suitability of the mathematical models
 138 has been checked by the F -tests and for the accuracy of the fitted polynomial equation was determined
 139 by adjusted the coefficient of determination (Adjusted R^2). The significant model terms were
 140 evaluated by the probability value (P -value) at 95% confidence interval. The P -value is the probability
 141 that the given statistical model is the same as or higher than the obtained results when the null
 142 hypothesis is true. The non-significant coefficients were eliminated from the polynomial equations.
 143 In order to illustrate the dependence between the dependent and the independent variables, the
 144 three-dimensional graphical representation of the response surfaces was made.

145 3. Results and discussions

146 3.1. Fitting models

147 Following the statistical processing of the experimental data regarding the effects of
 148 independent variables on the predictive models for dough rheological properties during mixing of
 149 KCl-NaCl mixtures, the most fitting models (quadratic models) were obtained for the following
 150 parameters: water absorption (WA), dough development time (DT), dough stability (ST), degree of
 151 softening at 10 min (DS), the Falling Number value (FN), peak viscosity (PV_{max}), temperature at peak
 152 viscosity (T_{max}), height under constraint of dough at maximum development time ($H'm$), total volume
 153 of CO_2 produced during fermentation (VT), volume of the gas retained in the dough at the end of the
 154 test (VR) and retention coefficient (CR).

155 3.2. The mixing and extension rheological properties for the mixes samples

156 Applying the ANOVA method to the mixing and extension values, it was observed that KCl has
 157 a significant effect ($p < 0.01$) on the rheological parameters as E, R_{50} , R_{max} , R/E, Tg, $H'm$, VT, VR, CR,
 158 while NaCl has a significant effect ($p < 0.01$) on WA, ST, E, R_{50} , R_{max} , R/E, PV_{max} , $H'm$, VT, VR.

159 As it may be seen from Figure 1a both types of salt led to a significant decreased ($p < 0.01$) of
 160 WA value. This may be due to the fact that in the presence of salt ions the electrostatic repulsion
 161 between gluten molecules are reduced as a consequence of their ability to partially shields the present
 162 charges between molecules. Thus, the gluten proteins aggregate in a higher extent due to the increase
 163 level of hydrophobic interactions between molecules fact that leads to a decrease of the water uptake
 164 ability [2].

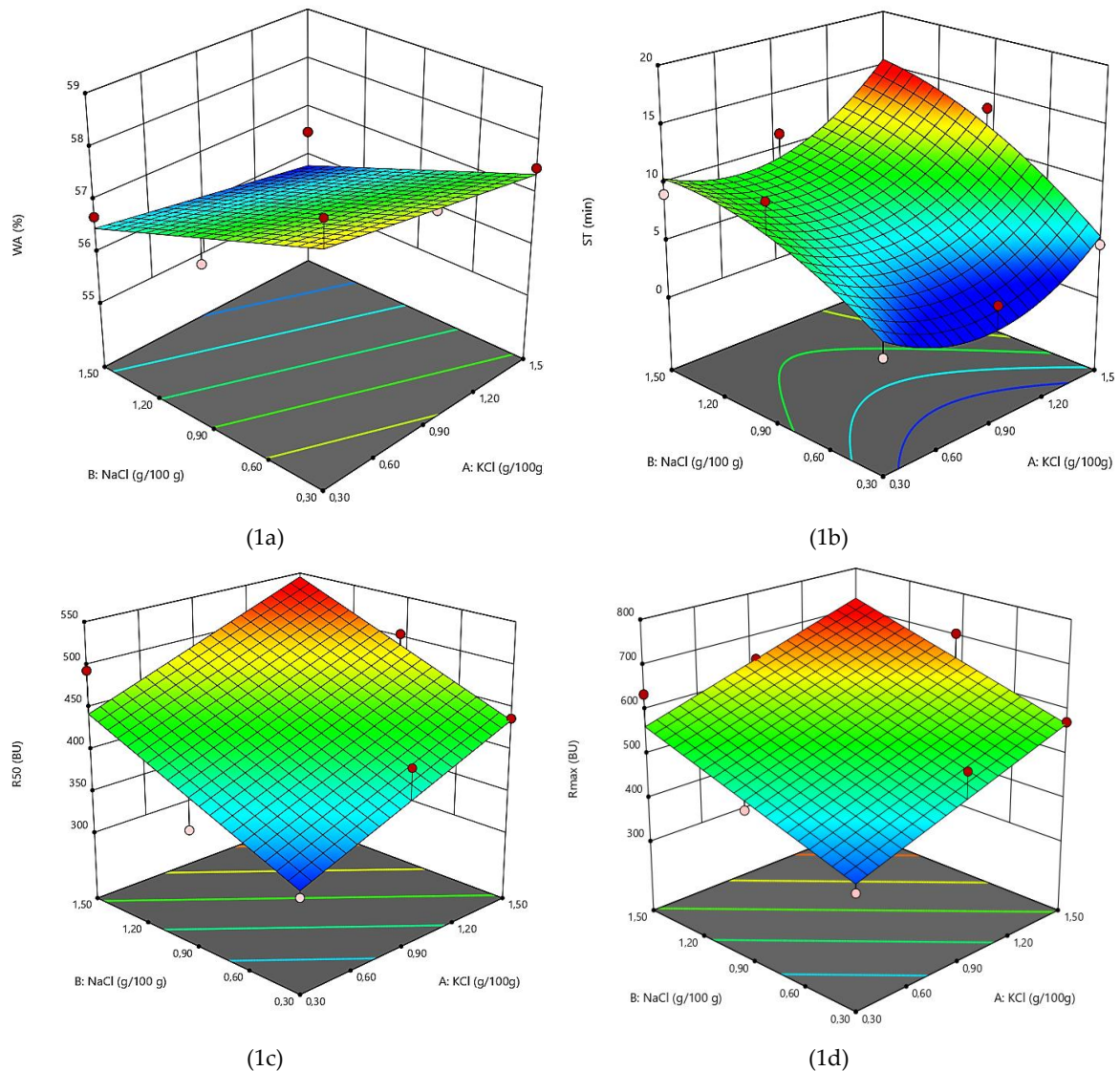


Figure 1. The graphical representations of the Farinograph and Extenograph parameters: 1a. water absorption (WA); 1b. stability of dough (ST); 1c. resistance to extension up to 50 mm (R₅₀); 1d. maximum resistance to extension (R_{max}).

165 From the two types of chloride salts it seems that NaCl presented a highly significant effect ($p < 0.01$)
 166 on WA value than KCl salt ($p < 0.01$). These results were similar with those reported by
 167 Tuhumuryet al. [32]; Jeckle et al. [2] which concluded that the intensity of chloride salts on WA value
 168 depends on cation position in Hofmeister series K⁺ being positioned before Na⁺. A decrease of WA
 169 value with the increase of the salt level addition has also been reported by different researchers
 170 [5,9,33-36].

171 The graphical representation of dough stability (ST) value in relation with the level of KCl and
 172 NaCl addition is shown in Figure 1b. It may be seen a significant increase ($p < 0.01$) of this value with
 173 the increase level of the both independent variables addition. This indicates a strengthening effect of
 174 chloride salts on wheat dough. Gluten proteins presents a surface hydrophobicity and contains
 175 almost 35% hydrophobic amino acids which promotes a protein aggregation in a more extent way
 176 when chloride salts are incorporated [9] leading to a higher dough stability. A significant increase of
 177 dough stability with the addition of chloride salts in wheat flour has also been reported by different
 178 researchers [2, 12, 37].

179 The effects of chloride salts on the Extenograph parameters curve are similar. According to
 180 Tuhumury et al. [32] this may be due to the fact that Na⁺ and K⁺ are nearby situated in the Hofmeister
 181 series and that way they exhibit similar effects on wheat dough properties. All the models for the

182 Extensograph values were linear. Both independent variables presented a significant positive effect (p
 183 < 0.01) on energy (E), resistance to extension (R_{50}), maximum resistance to extension (R_{max}) and ratio
 184 number (R/E). The effects of chloride salts on Extensograph values are related to their effect on gluten
 185 proteins. Their strengthen effect on dough due to the increase amount of hydrophobic interactions
 186 between molecules conducted to an increase value of E, R_{max} , R_{50} and R/E. These results were similar
 187 with those reported by McCann and Day [33]; Miller and Hosney [9]; Tuhumury et al. [32]; Ortolan
 188 et al. [38] which concluded that chloride salts increased the resistance to extension as it may be seen
 189 in Figures 1 c,d.

190 3.3. The viscometric rheological properties of the mixes samples

191 The effect of NaCl and KCl addition in wheat flour on dough viscometric properties, expressed
 192 as their corresponding regression coefficients and models, are shown in Table 2. From model analysis
 193 the most significant models were those for quadratic model ($Adjusted R^2=0.82$) Falling Number value
 194 (FN) followed by those for quadratic model ($Adjusted R^2=0.63$), peak viscosity (PV_{max}) and 2FI model
 195 ($Adjusted R^2=0.63$) for gelatinization temperature (T_g) which was less significant (Table 3).

196 **Table 2.** Effects of independent variables, expressed as their corresponding coefficients on the
 197 predictive models for dough rheological properties during mixing of KCl-NaCl mixtures.

Factors ^b	Parameters							
	Farinograph			Extensograph (proving time 135 min)				
	WA(%)	DT (min)	ST (min)	DS (UB)	E (cm ²)	R_{50} (BU)	R_{max} (BU)	R/E
Constant	56.75	1.55	6.85	54.86	106.54	439.62	566.85	4.11
A	-0.35**	0.0167	1.55*	0.666	14.33***	51.00***	81.83***	0.57***
B	-0.80***	-0.05	4.07***	-5.67**	15.00***	54.67***	78.67***	0.50***
A x B	0.22	0.025	1.00	4.00	-	-	-	-
A ²	0.019	0.319***	3.78**	-10.52**	-	-	-	-
B ²	0.37	0.019	-1.67	11.48**	-	-	-	-
AdjustedR ²	0.76	0.70	0.75	0.60	0.74	0.82	0.79	0.79
p-value	0.0072***	0.031**	0.0079***	0.0355**	0.0005***	<0.0001***	0.0002***	0.0005***

198 ^a Significant at $p < 0.01$ ***, at $p < 0.05$ **, at $p < 0.1$ *.

199 ^b A - KCl (g/100g); B - NaCl (g/100g); Adj. R² is measure of fit of the model.

200 WA – water absorption; DT – development time; ST – dough stability; DS - degree of softening; E - Energy;
 201 R_{50} – resistance to extension up to 50 mm; R_{max} - maximum resistance, R/E –ratio number.

202
 203 No significant model was obtained for T_{max} . Similar results were reported by Samutsri and
 204 Suphantharika [39] who concluded that different types of chloride salts did not presented any
 205 significant effect on pasting temperature on starch from rice. A positive effect on all viscometric
 206 properties was provided by the linear regression coefficients, suggesting that the increase levels of
 207 NaCl and KCl addition in wheat flour will lead to an increase in the viscometric values as it may be
 208 seen from Figure 2.

209

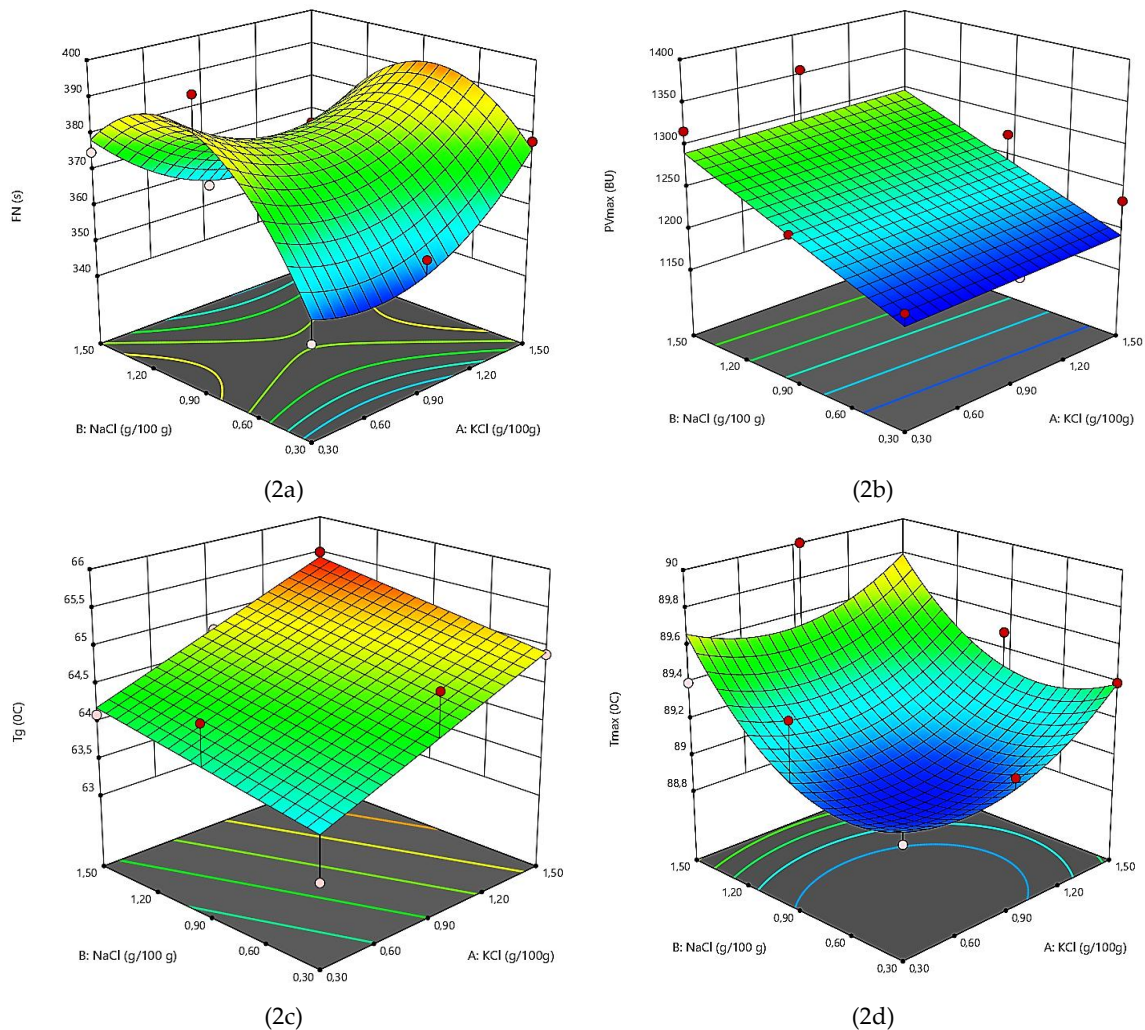


Figure 2. The graphical representations of the Falling Number and Amylograph parameters: 1a. Falling Number value (FN); 1b. peak viscosity (PV_{max}); 1c. gelatinization temperature (T_g); 1d. temperature at peak viscosity (T_{max}).

210 The increase of the FN and PV_{max} values with the increase of the NaCl and KCl addition (Figures
 211 2a,b) may be due to the action of chloride salts on the protein part from the **amylases** structure. **This**
 212 **fact reduces the activity of these enzymes with influence on dough rheological properties.**

213 Previous studies have shown that in the optimal range of pH activity of amylases, the chloride
 214 salts favor their activity in dough system, whereas outside of the pH range, it reduced its activity due
 215 to the shielding effect of the reactive groups of enzymes by the ions from the system as H^+ , Na^+ , K^+ ,
 216 Cl^- [40]. In general, wheat flour has a pH between 6.0-6.8, fact that makes **it** ~~her~~ slightly acidic and
 217 even close to the neutral pH value. Chloride salts presents an alkaline pH. Therefore, a mix between
 218 chloride salts and wheat flour will lead to higher pH values. In Amylograph and Falling Number
 219 methods wheat flour is mixed with distilled water with a pH value around 7.00 and different levels
 220 of chloride salts. Therefore, the mixes formed **from of** wheat flour, distilled water and chloride salts
 221 will present pH values outside the optimal range of pH amylases activity which is around 5.2÷5.4
 222 value [41]. Due to the alkaline pH of chloride salts the pH mixes analyzed to Amylograph and Falling
 223 Number will be even more outside of the optimal range of amylase activity with the increase level of
 224 chloride salts addition. Therefore, the amylases activity from the mixes from wheat flour, distilled
 225 water and chloride salts will decrease with the increase level of chloride salts addition. This fact leads
 226 to an increase in PV_{max} to the Amylograph device and to FN value to the Falling Number device
 227 which expresses α -amylase activity [42,43].

228 For T_g value a positive effect was provided by KCl and NaCl as it may be seen in Figure 2c this
 229 data being similar with those reported by different researchers [44-46]. This behavior are due to the

230 fact that this types of chloride salts decreased solubility of hydrophobic chains and enhanced water
 231 structure. When NaCl and KCl are incorporated in wheat flour dough it decreases water activity and
 232 increases the energy for physical and chemical reactions which involves water fact that delays the
 233 starch gelatinization process [5,31,45].

234 3.4. The fermentation rheological properties of the mixes samples

235 All the dependent variables analyzed through Rheofermentometer parameters was significantly
 236 affected ($p < 0.01$) by the levels of NaCl and KCl addition in wheat flour. Quadratic models (Table 3)
 237 for Rheofermentometer values showed a significant effect of the linear terms of NaCl and KCl with
 238 an highly coefficient of determination (R^2) which varies mostly between 0.70 to 0.91.

239 **Table 3.** Effects of independent variables, expressed as their corresponding coefficients on the
 240 predictive models for dough rheological properties during fermentation, gelatinization properties
 241 and α -amylase activity of KCl-NaCl mixtures.

Factors ^b	Parameters							
	FN (s)	Tg (°C)	PV _{max} (BU)	T _{max} (°C)	H'm (mm)	VT (mL)	VR (mL)	CR (%)
Constant	378.52	64.56	1221.66	89.00	61.95	1251.93	1117.07	89.28
A	1.50	0.62***	3.33	0.1167	-7.35***	159.50***	124.00***	2.12***
B	2.83	0.25*	52.67***	0.25	-5.58***	115.33***	-88.00***	1.62**
A x B	-10.25**	-0.1	-21.25	-0.05	-0.6750	-15.25	-24.00	-0.45
A ²	12.19**	-	14.21	0.1879	-4.38**	-95.26**	-63.24**	2.22**
B ²	-23.81***	-	29.21	0.2879	-1.78	-61.76	-40.24	1.42
Adjusted R ²	0.82	0.63	0.633	0.40	0.90	0.87	0.91	0.75
p-value	0.0028***	0.0071***	0.0143**	0.1193	0.0004***	0.0008***	0.0003***	0.0073***

242 ^a Significant at $p < 0.01$ ***, at $p < 0.05$ **, at $p < 0.1$ *.

243 ^b A - KCl (g/100g); B - NaCl (g/100g); Adj. R² is measure of fit of the model.

244 FN - Falling Number; Tg - gelatinization temperature; PV_{max} - peak viscosity; T_{max}- temperature at peak
 245 viscosity; H'm - height under constraint of dough at maximum development time; VT - total volume of CO₂
 246 produced during fermentation; VR - volume of the gas retained in the dough at the end of the test; CR -
 247 retention coefficient.

248
 249 The contours plots from Figure 3 for the Rheofermentometer values showed that the maximum
 250 height of gaseous production (H'm), total CO₂ volume production (VT) and volume of the gas
 251 retained in the dough at the end of the test (VR) significantly decreased ($p < 0.01$) with an increase in
 252 KCl and NaCl levels addition in wheat flour. The retention coefficient value (CR) increased with the
 253 increase level of KCl and NaCl addition in wheat flour.

254

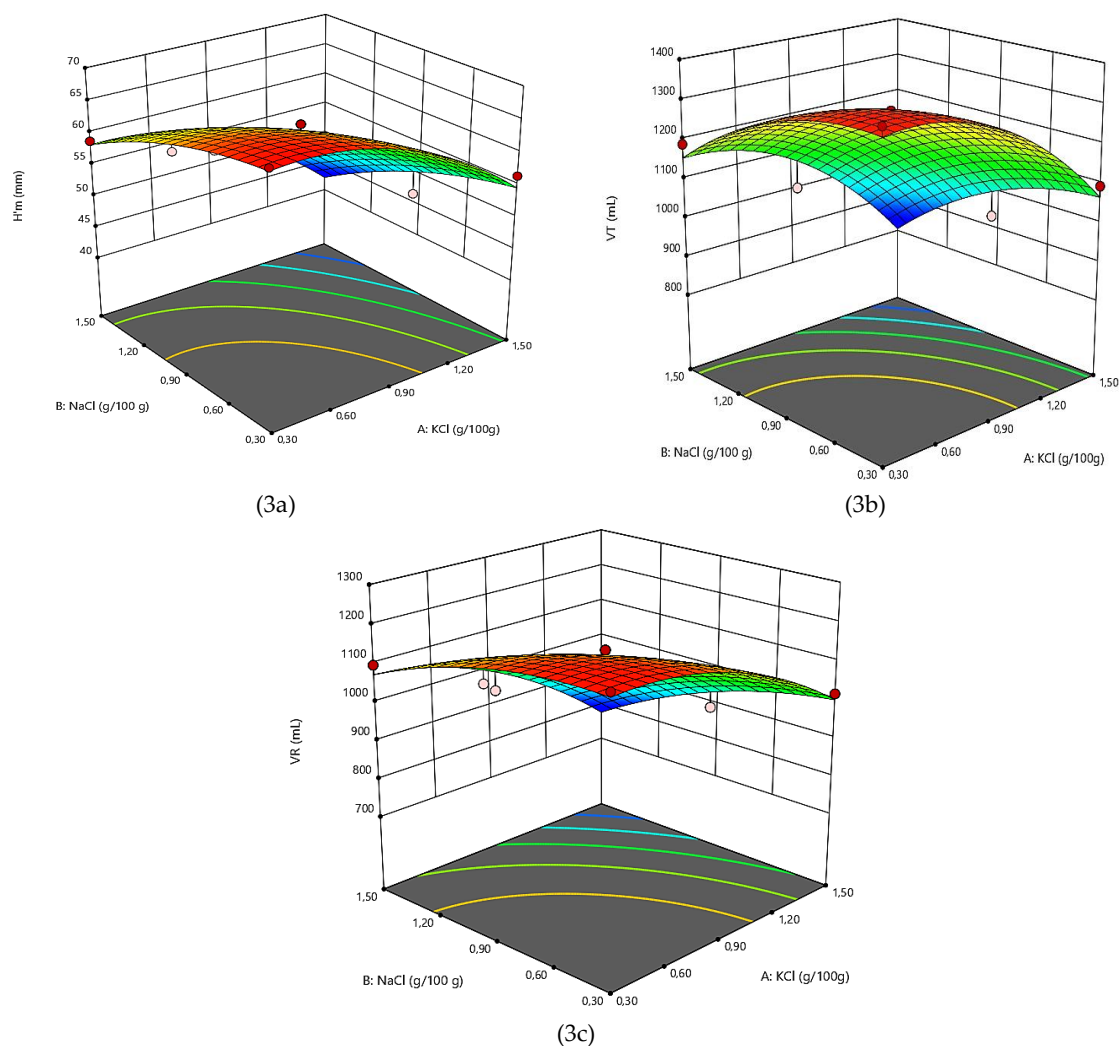


Figure 3. The graphical representations of the Rheofermentometer parameters: 3a. maximum height of gaseous production ($H'm$); 3b. total CO_2 volume production (VT); 3c. volume of the gas retained in the dough at the end of the test (VR).

255 $H'm$ is strongly affected by the yeast fermentation and also by the dough structure [2, 47]. Thus,
 256 by chloride salts addition the gluten network become more strength and less extensible fact that will
 257 lead to a lower dough expansion during fermentation. Also, chloride salts represses yeast activity by
 258 its osmotic pressure effect [13], fact that will lead to less CO_2 production and lower $H'm$ values as #
 259 may be seen from Figure 3a. The decrease of the $H'm$ together with the increased level of KCl and
 260 NaCl addition is in agreement with many previously made studies which reported that the addition
 261 of any type of chloride salts decreased the values of Rheofermentometer parameters [2, 9, 13, 34, 46].

262 The repressing effect of salt on yeast leads also to lower VT values as it may be seen from the
 263 Figure 3b and as a consequence, to lower VR values [44]. However, contrary to the negative effect of
 264 NaCl and KCl on $H'm$, VT and VR values it's presented a significantly ($p < 0.01$) positive one on CR
 265 value. This behavior is due to the gluten network improvement which becomes more strength by
 266 chloride salt addition and with a higher ability in retaining the gas released by fermentation [48].

267 3.5. Optimization of the KCl and NaCl formulation

268 An important objective of this research was to calculate the optimal values of the rheological
 269 parameters of the dough. For this purpose the Derringer desirability function (Equation 2), a multi-
 270 criterion decision-making method, was used [49, 50]. The optimization process using the numerical
 271 method by the Design-Expert was performed.

$$D = (d_1^{r_1} \cdot d_2^{r_2} \cdot \dots \cdot d_n^{r_n})^{\frac{1}{\sum r_i}} \quad (2)$$

272 where: d_1, d_2, \dots, d_n are the desirability indices for each dependent variables and r_1, r_2, \dots, r_i are the
 273 relative importance of the dependent variables. A non-zero value of D from zero implies that all
 274 responses are in desirable range and for a D value close to 1 the response values are close to the
 275 desirable values. By applying the desirable function methodology, the optimal values of the
 276 independent variables were obtained.

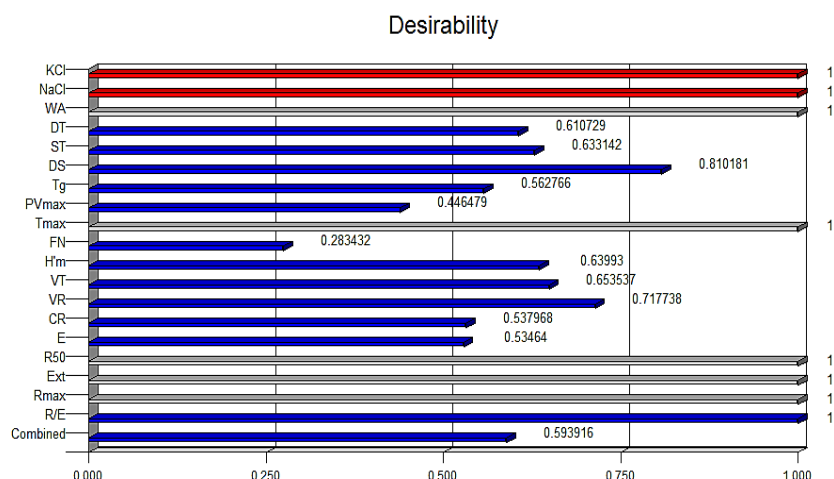


Figure 4. Desirability function scores for the independent variables (A and B) and the studied dependent variables.

277 The optimum values of the amount of KCl are 0.37g/100g wheat flour, and the optimal amount
 278 of NaCl is 1.31 g/100g wheat flour. For these optimal solutions, the optimal values for the dependent
 279 variables were obtained: WA - 56.626%, DT - 1.805 min., ST - 10.471 min., DS - 43.454 UB, Tg - 64.149
 280 °C, PV_{max} - 1281.438 BU, T_{max} - 89.513OC, FN - 383.978 s, H'm - 59.714 mm, VT - 1192.988mL, VR -
 281 1098.555mL, CR - 92.027 %, E - 104.475 cm², R₅₀ - 433.33 BU, Ext - 138.923 mm, R_{max} - 549.359 BU and
 282 R/E - 3.95with a desirable function score of 0.594.

283 3.6.Strategy approach for bakery products reformulation for sodium amount reduction related to our KCl-NaCl 284 optimum values

285 Bakery products are one of the main dietary sources of salt in the most European countries.
 286 It seems that the highest consumption of bakery products occurred in the Eastern and Central Europe.
 287 In Northern Europe other products such of those of animal origin are the most consumed ones being
 288 the main dietary sources of salt intake. Nowadays the daily salt intake in most EU countries ranges
 289 from 7 to 13 g per day (with the lowest intake values in Northern Europe countries and the highest
 290 ones in Central and Eastern countries)fact that exceed the World Health Organization
 291 recommendation data [51]. Due to this fact many European countries recommends foods
 292 reformulation in order to reduce the salt content from itsand runs many nutrition action plansfor this
 293 purpose. For exemple, the Ministry of Health from some EU countries recommended reducing salt
 294 in bakery products with 15% up tp 2015 in Austria, with 10% up to 2012 in Italy, with 20% up to 2014
 295 in Spain, etc. [16]. The maximum target for the salt level that want to be achived varies from one
 296 country to another. For exemple of 2.35% for bread products from Hungary from 2019and of 1.4% for
 297 bakery products from Spain. These high differences between EU counties targets are related to the
 298 usual levels that normally exist in this countries in bread products. So, for exemple the level of salt
 299 from popular Hungarian bakery products are arround of 3% [52] whereas in Spain the mean salt
 300 content from the bakery products are arround of 2% [53]. Besides the fact that salt reduction in bakery
 301 products affect its tehnological process, fact developed in a quite large extense during this study, it
 302 also affect bakery products quality. According to our study, a 22% replacement of NaCl in dough

303 recipe through KCl is the optimum one in order to obtain bakery products of a very good quality.
304 According to the data from the international literature a 20% sodium reduction in bakery products
305 did not affected bread quality from the sensory (including taste) point of view [51]. Regarding the
306 use of KCl as a NaCl substitute previous studies has shown that its addition up to 20-30% did not
307 affected the taste of the bakery products. However, levels higher than 30% of KCl addition in wheat
308 flour led to a metallic and bitter aftertastes fact that not recommend it in bread making [10]. Therefore,
309 our results are favorable for obtaining very good bakery products from technological and sensory
310 point of view. As we mentioned before, nowadays different countries are trying gradual reduction of
311 sodium levels from different foodstuffs. But this reduction is limited due to the consumers
312 acceptance who are not willing to give up to their eating habits especially from the sensory point of
313 view. The use of KCl as an ingredient to reduce sodium in foodstuffs is expected to increase in the
314 coming years [54]. This use it is also in accordance with the many years recommendations of the
315 WHO that people must reduce Na intake and increase K intake. Despite WHO rigorous
316 recommendation very little progress are being made worldwide in this direction [55]. Our optimum
317 values obtained through RSM methodology reduces with 22% the sodium content from the bread
318 recipe and increased the K level through KCl addition to around 200 mg/100 g bread. Our study
319 proposes a formulation which lead to bakery products of a good quality in accordance with WHO
320 recommendation of sodium reduction intake from foodstuffs. Also, the propose of sodium reduction
321 is by NaCl substitution with KCl, a natural ingredient which is also an agreed one by World Health
322 Organization.

323 4. Conclusions

324 According to the obtained data, it seems that both chloride salts has a similar effect on dough
325 rheological properties. With respect to mixing properties, both types of salts presented a positive
326 effect on dough stability, and to the energy, and to dough extensibility resistance to extension values.
327 During heating the chloride salts increased dough viscosity, fact reflected in an increase of PV_{max} and
328 FN values. During fermentation the both chloride salts decreased the $H'm$, VT, VR
329 Rheofermentometer values and increased the CR value. The mathematical models obtained for the
330 response variables were significant ones with a high values of Adjusted $R^2 > 0.70$ (except for DS, PV_{max}
331 and T_{max}), $p-value < 0.05$ (except for T_{max}) showing for most dependent variables no lack of fit. The
332 optimum values, obtained with the numerical method, were for KCl - 0.37 g/100g wheat flour and
333 for NaCl - 1.31 g/100g wheat flour. The use of the potassium chloride as a substitute of sodium
334 chloride in bakery products has a double advantage, namely the reduction of sodium content as well
335 the increase of potassium amount from the final products. Our optimum values obtained through
336 RSM methodology lead to the best technological parameters and also reduced the amount of sodium
337 from the bakery products with 22%, a decreased level that according to the international literature
338 data did not affects the sensory characteristics of the food products.

339

340 **Author Contributions:** A.V., S.G.S. and G.G.C. contributed equally to the study design, collection of data,
341 development of the sampling, analyses, interpretation of results, and preparation of the paper. All authors read
342 and approved the final manuscript.

343 **Acknowledgement:** This work was supported from contract no. 18PFE/16.10.2018 funded by Ministry of
344 Research and Innovation within Program 1 - Development of national research and development system,
345 Subprogram 1.2 - Institutional Performance - RDI excellence funding projects.

346 **Conflict of Interest:** The authors declare no conflict of interest.

347 References

- 348 1. De Wardener, H.E.; MacGregor, G.A. Harmful effects of dietary salt in addition to hypertension. *J. Hum.*
349 *Hypertens.* **2002**, *16*, 213-223.
- 350 2. Jekle, M.; Necula, A.; Jekle, M.; Becker, T. Concentration dependent rate constants of sodium substitute
351 functionalities during wheat dough development. *Food Res. Int.* **2019**, *116*, 356-353.

- 352 3. Mozaffarian, D., Benjamin, E.J., Turner, M.B., American Heart Association Statistics Committee and Stroke
353 Statistics Subcommittee. Heart disease and stroke statistics—2015 update: a report from the American
354 Heart Association. *Circulation*, **2015**, *131*, 29–322.
- 355 4. Zandstra, E.H.; Lion, R.; Newson, R.S. Salt reduction: Moving from consumer awareness to action. *Food*
356 *Qual. Prefer*, **2016**, *48*, 376–381.
- 357 5. Lopes, M.; Cavaleiro, C.; Ramos, F. Sodium reduction in bread: a role for glasswort (*Salicornia ramosissima*
358 *J. woods*). *Compr Rev Food Sci F*. **2017**, *16*, 1056–1071.
- 359 6. Bibbins-Domingo, K., Chertow, G.M., Coxson, P.G., Moran, A., Lightwood, J.M., Pletcher, M.J., Goldman,
360 L. Projected effect of dietary salt reductions on future cardiovascular disease. *N. Engl. J. Med.* **2010**, *362*, 590–
361 599.
- 362 7. Raffo, A.; Carcea, M.; Moneta, E.; Narducci, V.; Nicoli, S. Influence of different levels of sodium chloride of
363 a reduced sodium salt substitute on volatiles formation and sensory quality of wheat bread. *J Cereal*
364 *Sci.* **2018**, *79*, 519–526.
- 365 8. Valerio, F.; Conte, A.; Di Base, M.; Lattanzio, V.M.T.; Lonigro, S.L.; Padalino, L.; Pontonio, E.; Lavermicocca,
366 P. Formulation of yeast-leavened bread with reduced salt content by using a *Lactobacillus plantarum*
367 fermentation product. *Food Chem.* **2017**, *221*, 582–589.
- 368 9. Miller, R. A.; Hosney, R. C. Role of Salt in Baking. *Cereal Foods World*, **2008**, *53*, 4–6.
- 369 10. Silow, C.; Axel, C.; Zannini, E.; Arendt, E.K. Current status of salt reduction in bread and bakery products-
370 A review. *J Cereal Sci.* **2016**, *72*, 135–145.
- 371 11. Raffo, A.; Carcea, M.; Moneta, E.; Narducci, V.; Nicoli, S. Influence of different levels of sodium chloride of
372 a reduced sodium salt substitute on volatiles formation and sensory quality of wheat bread. *J Cereal*
373 *Sci.* **2018**, *79*, 519–526.
- 374 12. Diler, G.; Le-Bail, A.; Chevallier, S. Salt reduction in sheeted dough: a successful technological approach.
375 *Food Res. Int.* **2016**, *88*, 10–15.
- 376 13. Lynch, E.J.; Dal Bello, F.; Sheehan, E.M.; Cashman, K.D.; Arendt, E.K. Fundamental studies on the reduction
377 of salt on dough and bread characteristics. *Food Res. Int.* **2009**, *42*, 885–891.
- 378 14. Moreau, L.; Lagrange, J.; Bindzus, W.; Hill, S. 2009. Influence of sodium chloride on colour, residual
379 volatiles and acrylamide formation in model systems and breakfast cereals. *Int. J. Food Sci. Technol.* **2009**, *44*,
380 2407–2416.
- 381 15. Belz, M. C. E.; Ryan, L.A. M.; Arendt, E. K. The Impact of Salt Reduction in Bread: A Review. *Crit Rev Food*
382 *Sci. Nutr.* **2012**, *52*, 514–524.
- 383 16. Belc, N.; Smeu, I.; Macri, A.; Vallauri, D.; Flynn, K. Reformulating foods to meet current scientific
384 knowledge about salt, sugar and fats. *Trends Food Sci Technol* **2019**, *84*, 25–28.
- 385 17. Van Buren, L.; Dötsch-Klerk, M.; Seewi, G.; Newson, R.S. Dietary impact of adding potassium chloride to
386 foods as a sodium reduction technique. *Nutrients* **2016**, *8*, 235.
- 387 18. Binia, A.; Jaeger, J.; Hu, Y.; Singh, A.; Zimmermann, D. Daily potassium intake and sodium-to-
388 potassium ratio in the reduction of blood pressure: A meta-analysis of randomized controlled trials. *J.*
389 *Hypertens.* **2015**, *33*, 1509–1520.
- 390 19. Aburto, N.J.; Hanson, S.; Gutierrez, H.; Hooper, L.; Elliott, P.; Cappuccio, F.P. Effect of increased
391 potassium intake on cardiovascular risk factors and disease: Systematic review and meta-analyses. *BMJ*
392 **2013**, 346.
- 393 20. Braschi, A.; Gill, L.; Naismith, D.J. Partial substitution of sodium with potassium in white bread: feasibility
394 and bioavailability. *Int. J. Food Sci. Nutr.* **2009**, *60*, 507–521.
- 395 21. Zandstra, E.H.; Lion, R.; Newson, R.S. Salt reduction: Moving from consumer awareness to action. *Food*
396 *Qual. Preference* **2016**, *48*, 376–381.
- 397 22. Cappelli, A.; Guerrini, L.; Parenti, A.; Palladino, G.; Cini, E. Effects of wheat tempering and stone rotational
398 speed on particle size, dough rheology and bread characteristics for a stone-milled weak flour. *J Cereal Sci.*,
399 **2020**, *91*, 102879.
- 400 23. Cappelli, A., Cini, E., Guerrini, L., Masella, P., Angeloni, G., & Parenti, A. (2018). Predictive models of the
401 rheological properties and optimal water content in doughs: An application to ancient grain flours with
402 different degrees of refining. *J Cereal Sci.*, **2018**, *83*, 229–235.
- 403 24. Popa, N.C.; Tamba-Berehoiu, R.; Popescu, S.; Varga, M.; Codină, G.G. Predictive model of the
404 alveographic parameters in flours obtained from Romanian grains. *Rom Biotech Lett* **2008**, *14* (2), 4234–4242.

- 405 25. Mironeasa, S.; Iuga, M.; Zaharia, D.; Mironeasa, C. Optimization of grape peels particle size and flour
406 substitution in white wheat flour dough. *Scientific Study & Research. Chemistry & Chemical Engineering,*
407 *Biotechnology, Food Industry*, **2019**, *20*, 29-42.
- 408 26. Codină, G. G.; Mironeasa, S. Use of response surface methodology to investigate the effects of brown and
409 golden flaxseed on wheat flour dough microstructure and rheological properties. *J Food Sci Tech Mys.*
410 *Journal of food science and technology*, **2016**, *53*, 4149-4158.
- 411 27. Mironeasa, S.; Iuga, M.; Zaharia, D.; Mironeasa, C. Optimization of white wheat flour dough rheological
412 properties with different levels of grape peels flour addition. *Bulletin of University of Agricultural Sciences*
413 *and Veterinary Medicine Cluj-Napoca. Food Science and Technology*, **2019**, *76*, 27-39.
- 414 28. Wang, Y.; Gao, Y.; Ding, H.; Liu, S.; Han, X.; Gui, J.; Liu, D. Subcritical ethanol extraction of flavonoids from
415 *Moringa oleifera* leaf and evaluation of antioxidant activity. *Food Chem*, **2017**, *218*, 152-158.
- 416 29. Candiotti, L.V.; De Zan, M.M.; Cámara, M.S.; Héctor C.G. Experimental design and multiple response
417 optimization. Using the desirability function in analytical methods development *Talanta***2014**, 123–138.
- 418 30. Behera, S. K.; Meena, H.; Chakraborty, S.; Meikap, B.C. Application of response surface methodology
419 (RSM) for optimization of leaching parameters for ash reduction from low-grade coal. *International Journal*
420 *of Mining Science and Technology*. **2018**, 28-4, 621-629.
- 421 31. Wang, Y.; Li, X.; Zhang, B.; Zhao, Z. Optimization of multiple hydraulically fractured factors to maximize
422 the stimulated reservoir volume in silty laminated shale formation, Southeastern Ordos Basin, China. *J. Pet.*
423 *Sci. Eng.* **2016**, *145*, 370–381.
- 424 32. Tuhumury, H.C.D.; Small, D.M.; Day, L. Effects of Hofmeister salt series on gluten network formation: Part
425 I. Cation series. *Food Chem.***2016**, *212*, 789-797.
- 426 33. McCann, T. H.; Day, L. Effect of sodium chloride on gluten network formation, dough microstructure and
427 rheology in relation to breadmaking. *J Cereal Sci.***2013**, *57*, 444–452.
- 428 34. Beck, M.; Jekle, M.; Becker, T. Impact of sodium chloride on wheat flour dough for yeast-leavened products.
429 I. Rheological attributes. *J Food Sci Agric.***2012**, *92*, 585–592.
- 430 35. Uthayakumar, S.; Batey, I. L.; Day, L.; Wrigley, C. W. Salt reduction in wheat-based foods-technical
431 challenges and opportunities. *Food Aust.***2011**, *63*,137-140.
- 432 36. Voinea, A.; Stroe, S.G.; Codină, G.G. The Effect of Sodium Reduction by Sea Salt and Dry Sourdough
433 Addition on the Wheat Flour Dough Rheological Properties. *Foods* **2020**, *9*, 610.
- 434 37. Bernklau, I.; Neußer, C.; Moroni, A. V.; Gysler, C.; Spagnolello, A.; Chung, W.; Jekle, M. Becker, T.
435 2017. Structural, textual and sensory impact of sodium reduction on long fermented pizza. *Food Chem*,
436 **2017**, *234*, 1, 398-407.
- 437 38. Ortolan, F.; Corream G.P.; da Cunha, R.L.; Steel, C.J. Rheological properties of vital wheat glutes with
438 water or sodium chloride. *LWT-Food Sci. Tehcnol***2017**, *79*, 647-654.
- 439 39. Samutsri, W.; Suphantharika, M. Effect of salts on pasting, thermal, and rheological properties of rice starch
440 in the presence of non-ionic and ionic hydrocolloids. *Carbohydr. Polym.***2012**, *87*, 1559-1568.
- 441 40. Cordeiro, C.A.M.; Martins, M.L.L.; Luciano, A.B. Production and properties of α -amylase from
442 thermophilic *Bacillus* sp. *Braz. J. Microbiol.* **2002**, *33*, 57-61.
- 443 41. Sinani, A.; Sana, M.; Seferi, E.; Sheahaj, M. The effect of α -amylase in rheology features of some wheat
444 cultivars and their harmonization for producing baking according to customer requirements. *Global Journal*
445 *of Biology Agriculture & Health Sciences*. **2014**, *4*, 58-64.
- 446 42. Struyf, N.; Verspreet, J.; Courtin, C.M. The effect of amylolytic activity and substrate availability on sugar
447 release in non-yeasted dough. *J. Cereal Sci.* **2016**, *69*, 111–118
- 448 43. Codină, G.G.; Dabija, A.; Oroian, M. Prediction of pasting properties of dough from Mixolab measurements
449 using artificial neuronal networks. *Foods* **2019**, *8*, 447.
- 450 44. Nicola, T.W.J.; Isobe, N.; Clark, J.H.; Matubayasi, N.; Shimizu, S. The mechanism of salt effects on starch
451 gelatinization from a statistical thermodynamic perspective. *Food Hydrocoll.***2018**, *87*, 593-601.
- 452 45. Moreira, R.; Chenlo, F.; Torres, M.D. Effect of sodium chloride, sucrose and chestnut on rheological
453 properties of chestnut flour doughs. *Food Hydrocoll.***2011**, *25*, 1041-1050.
- 454 46. Salvador, A.; Sanz, T.; Fiszman, S. M. Dynamic rheological characteristics of wheat flour-water doughs.
455 Effect of adding NaCl, sucrose and yeast. *Food Hydrocoll.***2006**, *20*, 780-786.
- 456 47. Codină G.G., Voica, D. The influence of different forms of bakery yeast *Saccharomyces cerevisiae* type strain
457 on the concentration of individual sugars and their utilization during fermentation **2010**, *15* (4), 5417-5422

- 458 48. Pasqualone, A.; Caponio, F.; Pagani, M.A.; Summo, C.; Paradiso, V.M. Effect of salt reduction on quality
459 and acceptability. *Food Chem.* **2019**, *289*, 575-581.
- 460 49. Badwaik, L.S.; Prasad K.; Seth D. Optimization of ingredient levels for the development of peanut based
461 fiber rich pasta. *J. Food Sci Technol.* **2014**, *51*(10), 2713-2719.
- 462 50. Candioti, L.V.; De Zan, M.M.; Cámara, M.S.; Héctor C.G. Experimental design and multiple response
463 optimization. Using the desirability function in analytical methods development *Talanta* **2014**, 123–138.
- 464 51. Kloss, L.; Meyer, J.D.; Graeve, L.; Vetter, W. Sodium intake and its reduction by food reformulation in the
465 European Union - A review. *NFS Journal*, **2015**, *1*, 9-19.
- 466 52. Lásztity, R. (1995). *The Chemistry of Cereal Proteins* (Second Edition). CRC Press
- 467 53. Farinós, P. N.; Sanz, S.S.; Dal Re, M.A.; Boyo, Y.J.; Robledo, T.; Castrodeza, J.J.; Amado, C.J.; Villar, C. Salt
468 content in bread in Spain, 2014. *Nutricion Hospitalaria*, **2017**, *35* (3), 650-654.
- 469 54. Van Buren, L.; Dötsch-Klerk, M.; Seewi, G.; Newson, R.S. Dietary Impact of Adding Potassium Chloride to
470 Foods as a Sodium Reduction Technique. *Nutrients* **2016**, *8*, 235.
- 471 55. Iwahori, T.; Miura, K.; Ueshima, H. Time to Consider Use of the Sodium-to-Potassium Ratio for Practical
472 Sodium Reduction and Potassium Increase. *Nutrients* **2017**, *9*, 700.
- 473



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