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1 **Baking process design based on modelling and simulation: Towards**
2 **optimization of bread baking**

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7

8 **Abstract**

9 This paper presents a theoretical approach for optimal design of the baking process.
10 Conventional baking of bread was taken as subject of study, and simulation of
11 previously validated models was used to investigate the process. The proposed approach
12 is based on the definition of two different times for the baking process: a critical time,
13 i.e. a minimum baking time assessed by the complete starch gelatinization in the
14 product, and a quality time, i.e. the time necessary to achieve a target value for a given
15 quality attribute. In this work, browning determined the quality time due to its relevance
16 with regard to sensory and nutritional aspects. As a result, feasible solutions are
17 obtained involving a minimum baking (acceptable products) and a minimum thermal
18 input for a given value of browning, which helps to reduce the formation of acrylamide.
19 Optimum solutions can be then obtained by defining specific objectives; weight loss can
20 be minimized by lowering the value of heat transfer coefficient. Furthermore, obtained
21 results can be helpful to build more efficient ovens.

22 **Keywords:** Heat and mass transfer; Multi-objective optimization; Energy demand;
23 **Process control; Cooking; Drying.**

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24 **Nomenclature**

25

26 a_w water activity27 C_p specific heat ($\text{J kg}^{-1} \text{K}^{-1}$)28 D water (liquid or vapour) diffusion coefficient of product ($\text{m}^2 \text{s}^{-1}$)29 D_{va} water vapour diffusion coefficient in air ($\text{m}^2 \text{s}^{-1}$)30 E_a activation energy of starch gelatinization (J mol^{-1})31 h heat transfer coefficient ($\text{W m}^{-2} \text{K}^{-1}$)32 K rate constant of starch gelatinization (s^{-1})33 K_0 pre-exponential factor in Eq. (19) (s^{-1})34 k thermal conductivity ($\text{W m}^{-1} \text{K}^{-1}$)35 k_b rate constant of browning (min^{-1})36 k_g corrected mass transfer coefficient ($\text{kg Pa}^{-1} \text{m}^{-2} \text{s}^{-1}$)37 k_g^* mass transfer coefficient from Eq. (16) ($\text{kg Pa}^{-1} \text{m}^{-2} \text{s}^{-1}$)38 L^* lightness39 M molecular mass (g mol^{-1})40 P water vapour pressure (Pa)41 Pr Prandlt number42 Q heat uptake in starch gelatinization (J)43 R, r radius (m)44 R_g universal gas constant ($8.314 \text{ J K}^{-1} \text{ mol}^{-1}$)45 RH relative humidity (%)46 Sc Schmidt number47 T temperature (K)48 t time (s)

49 W water (liquid or vapour) content (kg kg^{-1})

50

51 **Greek symbols**

52 α degree of starch gelatinization

53 δ Delta-type function

54 ΔT temperature range of phase change (K)

55 ε emissivity

56 λ_v latent heat of evaporation (J kg^{-1})

57 ρ density (kg m^{-3})

58 σ Stefan-Boltzmann constant ($5.67 \times 10^{-8} \text{ W m}^{-2} \text{ K}^{-4}$)

59

60 **Subscripts**

61 ∞ ambient

62 air air

63 atm atmospheric

64 f phase change

65 s solid or surface

66 sat saturated

67 w water

68 **1. Introduction**

69

70 Baking is the final and most important step in bread production, and can be defined as
71 the process which transforms dough, basically made of flour, water and leavening
72 agents, in a food with unique sensory features by application of heat inside an oven. In
73 particular, white or French bread is the most popular type of bread, and is distinguished
74 for having a crunchy and golden-yellow (or brown) crust, a sponge and light crumb with
75 soft texture and intermediate moisture, and a typical flavour. All these quality aspects
76 are the result of a series of physical and chemical changes produced by simultaneous
77 heat and mass transfer occurring within the product during baking (Mondal & Datta,
78 2008; Purlis, 2010; Sablani, Marcotte, Baik, & Castaigne, 1998; Scanlon & Zghal,
79 2001; Vanin, Lucas, & Trystram, 2009).

80 Optimization of the bread baking process is a subject of great importance for food
81 industry. On the one hand, bread is a staple food and thus its production is relevant from
82 a commercial point of view, besides its cultural relevance. On the other hand, baking is
83 an energy-intensive process due to water evaporation occurring in the product (e.g.
84 latent heat of water vaporization is 2.257 MJ/kg at 100 °C). The energy demand for a
85 conventional baking process is around 3.7 MJ/kg, though it can be higher (up to 7
86 MJ/kg) depending on specific products and operating conditions. In this sense, baking is
87 similar to (conventional) drying, both demanding a high amount of energy in
88 comparison with chilling, freezing, and canning, which need less than 1 MJ/kg (Le Bail
89 et al., 2010). In addition, ovens are often operated in an empirical way by trial-and-
90 error, since information about manipulating the oven settings for an optimum
91 production is still lacking and poorly understood (Broyart & Trystram, 2002). As a
92 result, inconsistency in the quality of bakery products is common in most industrial

93 processes, besides an inefficient use of energy, leading to economical losses (Wong,
94 Zhou, & Hua, 2007).

95 The end point of the bread baking process is generally established by assessing sensory
96 attributes, e.g. surface colour, texture and flavour of bread, which play a key role in the
97 acceptance of the product by consumers (Purlis & Salvadori, 2007). In particular,
98 surface browning is a practical indicator of baking advance, since can be easily
99 monitored during the process by means of in-line sensors, and therefore can be used as a
100 control parameter (McFarlane, 1990). Furthermore, the development of browning
101 caused by the Maillard reaction is associated with nutritional issues, such as acrylamide
102 formation and decrease of nutritional value of proteins (Purlis, 2010). This way of
103 assessment of the bread baking process, i.e. by subjective (sensory) parameters which
104 also depend on type of consumers, culture and even regulations, makes difficult the task
105 of developing a general (and objective) methodology to design, optimize, and control
106 this process.

107 On the other hand, since quality changes depend on transport phenomena, it is essential
108 to perform a comprehensive analysis involving both aspects. In this way, two
109 approaches have been used to optimize or design baking. The first approach includes
110 semi-empirical studies where quality attributes are experimentally determined as a
111 function of operating conditions, with a subsequent application of surface response
112 methodology (Demirekler, Sumnu, & Sahin, 2004; Sevimli, Sumnu, & Sahin, 2005) or
113 nonlinear programming techniques (Dingstad, Egelanddal, Mevik, & Færgestad, 2004;
114 Therdthai, Zhou, & Adamczak, 2002). The second methodology consists in considering
115 transport models coupled with quality kinetic models as a starting point with the aim of
116 describing all changes occurring during the process. Afterwards, process design and
117 optimization can be performed by applying optimization algorithms (Hadiyanto, Boom,

118 van Straten, van Boxtel, & Esveld, 2009; Hadiyanto, Esveld, Boom, van Straten, & van
119 Boxtel, 2008). Besides the advantages, drawbacks, and restrictions of each specific
120 procedure, it is clear the need of adopting a comprehensive point of view with the aim
121 of developing baking strategies considering practical applications. In particular, baking
122 is a special case of food preservation processes and operations, since no microbiological
123 risk has to be considered (as long as good manufacturing practices are carried out), so
124 all objectives to be optimized with regard to the product are quality objectives. Thus,
125 experimental data related to sensory attributes is always necessary to define an objective
126 function, no matter which optimization procedure will be applied.
127 In this context, the objective of this paper was to propose a theoretical approach to
128 design heating strategies with focus on optimization and control of the baking process.
129 For this aim, mathematical modelling and process simulation were implemented to
130 investigate the bread baking process. This work seeks to contribute to a more
131 comprehensive understanding of the baking process in order to design and control the
132 process in a more efficient way. In addition, this investigation can also help to oven
133 designers and manufacturers to build more efficient equipment.

134

135 **2. Theory**

136

137 From the transport phenomena point of view, bread baking is considered as a
138 simultaneous heat and mass transfer (SHMT) process occurring in a porous medium,
139 where phase change (i.e. water vaporization) takes place in a moving front (details are
140 given later in the description of the mathematical model). Amongst all physical and
141 chemical changes that are generated during baking, which actually determine the quality
142 attributes of final product, starch gelatinization and browning development are taken as

143 reference reactions in this work. The complete starch gelatinization ensures the sensory
144 acceptability of the product because determines the transformation of dough into crumb,
145 i.e. a minimum baking (Zanoni, Peri, & Bruno, 1995a). Surface colour is one of the
146 main (and generally the first) quality features considering preference of consumers, and
147 therefore is often used to judge the completion of baking (Ahrné, Andersson, Floberg,
148 Rosén, & Lingnert, 2007). In bakery products, surface colour is an important sensory
149 attribute associated with aroma, taste, appearance, and with the overall quality of food,
150 and certainly has an important effect on the consumer judgment: colour influences the
151 anticipated oral and olfactory sensations because of the memory of previous eating
152 experiences (Abdullah, 2008). Other product quality descriptors such as specific
153 volume, porosity, and mechanical properties are also important in baking design since
154 they are associated with other sensory attributes (e.g. texture). However, these
155 parameters are also affected by product formulation, i.e. type of flour, fat components,
156 and specific additives or improvers, or by a change in baking technology, e.g.
157 introduction of microwave heating (Demirekler et al., 2004; Sevimli et al., 2005).

158 Recently, a technological study of bread baking was presented analyzing simultaneously
159 quality and process aspects (Purlis, 2011). It was found that when surface colour is used
160 to determine the end point of the process, which is a common practice actually, it is
161 possible to not achieve a complete baking due to an incomplete starch gelatinization. In
162 particular, such situation occurs when slightly browned products are sought and intense
163 heating is applied: because of high internal resistance to heat transfer due to low thermal
164 conductivity of bread, surface browning is developed at higher rate than starch
165 gelatinization at product centre. In addition, this is favoured with an increase in bread
166 radius via the diminution of thermal gradient. A control parameter should be established
167 to overcome this problem: a minimum value of 96 °C at the product centre (or coldest

168 point) has been proposed as a practical solution (Purlis, 2011). Therefore, as browning
169 and starch gelatinization have different reaction rates, partly because they are assessed
170 at different locations undergoing different heat and mass transfer processes, operating
171 conditions should be controlled in order to balance such reactions and generate correctly
172 baked products presenting the desired quality attributes.

173 Based on previous hypotheses and results, two different times are identified in the
174 baking process: a *critical time* (CT) and a *quality time* (QT). The CT is the minimum
175 baking time, defined as the time necessary to achieve a complete transition of dough
176 into crumb given by a complete starch gelatinization. The CT has to be assessed at the
177 coldest point of bread, where temperature has to reach 96 °C at least. The QT is defined
178 as the time required to achieve the target value of a given quality attribute, relevant with
179 regard to sensory acceptability of the product. For example, a target value of surface
180 lightness representing the desired surface colour of bread, which can be established by
181 sensory data obtained from preference of consumers. So, the proposed approach
182 establishes that an optimum baking process will present the same value for CT and QT,
183 i.e. at the same time, bread is completely baked and the requirements about sensory
184 attributes are satisfied. In addition, nutritional quality should not be impaired.

185 Obviously, CT and QT can be unequal depending on heat and mass transfer fluxes
186 established by operating conditions and product properties. For a given situation, if CT
187 is greater than QT, the product will present the desired quality attribute (e.g. surface
188 colour) but will remain unbaked since a complete starch gelatinization is not achieved.
189 Alternatively, if the process time is prolonged to overcome this issue, over-baking will
190 generate different values of the chosen quality attribute associated with QT, and even
191 can lead to poor quality products due to excessive thermal input. Prolonged baking
192 times can produce high temperature values at bread surface, leading to nutritional losses

193 (including the formation of toxic compounds) and more weight loss (this is related to
194 mechanical properties of crust and thus texture attributes). On the other hand, if QT is
195 greater than CT, extra time will be needed to accomplish the target value of the chosen
196 sensory attribute, while the product is already baked in terms of dough/crumb
197 transformation. The described situations generated by non-optimum baking processes
198 produce economical losses since unacceptable products are obtained and additional
199 energy is consumed. Therefore, the ultimate objective is to design an optimum baking
200 process based on the proposed approach.

201

202 **3. Methodology**

203

204 The subject of study is conventional baking of French bread (without mould or tin) in a
205 static or batch, indirect (e.g. electric) oven. This is a typical case of traditional bread
206 baking at small and medium scale production, which generally present a low level of
207 process automation and technology, in contrast with continuous baking in large
208 installations equipped with tunnel ovens, which is almost restricted to large scale
209 production of tin bread, as well as biscuits, cakes and similar batter products (Maroulis
210 & Saravacos, 2003). Batch ovens usually have forced convection provided by a fan that
211 recirculates hot air within the baking chamber, which helps to increase the heat and
212 mass flux (and thus transfer coefficients) from air to product. In general, fan velocity is
213 fixed (on/off system) so air velocity and then heat (and mass) transfer coefficient cannot
214 be modified for a given oven and product.

215 To study such process and apply the hypotheses previously proposed, we use the
216 concept of modern food process design (Maroulis & Saravacos, 2003). This concept is
217 based on engineering principles, mathematical modelling, and process simulation; the

218 objective is to economically produce food products, with emphasis on product quality in
219 addition to the conventional engineering considerations of energy, process cost, and
220 environmental impact. In this way, process simulation is performed using a
221 mathematical model for SHMT in bread during baking, which was previously
222 developed and validated using experimental data of the process; discussion about
223 validation and sensitivity analysis regarding the parameters of the model can be found
224 in Purlis and Salvadori (2009a, 2009b, 2010). Kinetic models for starch gelatinization
225 (Zanoni et al., 1995a; Zanoni, Schiraldi, & Simonetta, 1995b) and browning
226 development (Purlis & Salvadori, 2009c) are coupled to the transport model to describe
227 product quality changes as a function of state variables.

228

229 **3.1. Heat and mass transfer model**

230

231 The SHMT model includes the main distinguishing features of bread baking, i.e. the
232 rapid heating of bread core and the development of a dry outer crust. The former has
233 been explained by the evaporation-condensation mechanism (de Vries, Sluimer, &
234 Bloksma, 1989; Sluimer & Krist-Spit, 1987; Wagner, Lucas, Le Ray, & Trystram,
235 2007), while the later is due to the formation and advancing of an evaporation front
236 towards the bread core (Zanoni, Peri, & Pierucci, 1993; Zanoni, Pierucci, & Peri, 1994).
237 So, bread baking is considered as a moving boundary problem (MBP) where SHMT
238 with phase change occurs in a porous medium. Bread is modelled as a system
239 containing three different regions: (1) *crumb*: wet inner zone, where temperature does
240 not exceed 100 °C and dehydration does not occur; (2) *crust*: dry outer zone, where
241 temperature exceeds 100 °C and dehydration occurs; (3) *evaporation front*: between the

242 crumb and crust, where temperature is ca. 100 °C and water evaporates (liquid-vapour
 243 transition).

244 Mathematically, the MBP is formulated using a physical approach, where the enthalpy
 245 jump corresponding to phase change is incorporated in the model by defining equivalent
 246 thermophysical properties (Bonacina, Comini, Fasano, & Primicerio, 1973). Such
 247 definition states that evaporation occurs within a temperature range rather than at a
 248 fixed temperature. Other major assumptions of the model are the following: (1) bread is
 249 homogeneous and continuous; the concept of porous medium is included through
 250 effective or apparent thermophysical properties; (2) heat is transported by conduction
 251 inside bread according to Fourier's law, but an effective thermal conductivity is used to
 252 incorporate the evaporation-condensation mechanism in heat transfer; (3) only liquid
 253 diffusion in the crumb and only vapour diffusion in the crust are assumed to occur
 254 (Luikov, 1975); (4) volume change is neglected. For a detailed description of the model,
 255 including thermophysical properties, the reader is referred to Purlis and Salvadori
 256 (2009a, 2009b, 2010).

257

258 **3.1.1. Governing equations**

259

260 Bread (French type) is considered as an infinite cylinder of radius R , so the problem is
 261 reduced to a single dimension via the axial symmetry assumption. For initial conditions,
 262 uniform temperature and water content are assumed.

263 Heat balance equation:

$$264 \quad \rho C_p \frac{\partial T}{\partial t} = \frac{1}{r} \frac{\partial}{\partial r} \left(r k \frac{\partial T}{\partial r} \right) \quad (1)$$

265 Mass balance equation:

$$266 \quad \frac{\partial W}{\partial t} = \frac{1}{r} \frac{\partial}{\partial r} \left(r D \frac{\partial W}{\partial r} \right) \quad (2)$$

267

268 **3.1.2. Boundary conditions**

269

270 Heat arrives to the bread surface by convection and radiation, and is balanced by
271 conduction inside the bread:

$$272 \quad -k \frac{\partial T}{\partial r} = h(T_s - T_\infty) + \varepsilon \sigma (T_s^4 - T_\infty^4) \quad (3)$$

273 Water migrating towards the bread surface is balanced by convective flux:

$$274 \quad -D \rho_s \frac{\partial W}{\partial r} = k_g (P_s(T_s) - P_\infty(T_\infty)) \quad (4)$$

275 where $P_s = a_w P_{sat}(T_s)$ and $P_\infty = (RH/100) P_{sat}(T_\infty)$.276 At the centre, i.e. $r = 0$:

$$277 \quad \frac{\partial T}{\partial r} = 0 \quad (5)$$

$$278 \quad \frac{\partial W}{\partial r} = 0 \quad (6)$$

279

280 **3.1.3. Thermophysical properties**

281

282 According to the MBP formulation, equivalent thermophysical properties are defined by
283 including the phase transition occurring during the process, thus an equivalent property
284 is valid for dough/crumb and crust. A smoothed Heaviside function with continuous
285 derivative is used to incorporate the phase transition into thermophysical properties,
286 with parameters $T_f = 100$ °C and $\Delta T = 0.5$ °C. In addition, the delta-type function $\delta(T -$

287 $T_f, \Delta T$) that simulates the enthalpy jump (Eq. (7)) is defined by the sum of two
 288 smoothed Heaviside functions with different sign.

289

290 Specific heat:

$$291 \quad C_p(T, W) = C_p^*(T, W) + \lambda_v W \delta(T - T_f, \Delta T) \quad (7)$$

$$292 \quad C_p^*(T, W) = C_{p,s}(T) + WC_{p,w}(T) \quad (8)$$

$$293 \quad C_{p,s} = 5T + 25 \quad (9)$$

$$294 \quad C_{p,w} = 5207 - 7.317T + 1.35 \times 10^{-2} T^2 \quad (10)$$

295

296 Thermal conductivity:

$$297 \quad k(T) = \begin{cases} 0.9/[1 + \exp(-0.1(T - 353.16))] + 0.2 & \text{if } T \leq T_f - \Delta T \\ 0.2 & \text{if } T > T_f + \Delta T \end{cases} \quad (11)$$

298

299 Density:

$$300 \quad \rho(T) = \begin{cases} 180.61 & \text{if } T \leq T_f - \Delta T \\ 321.31 & \text{if } T > T_f + \Delta T \end{cases} \quad (12)$$

301 Density for solid (ρ_s) that appears in Eq. (4) is equal to 241.76 kg m⁻³.

302

303 Mass diffusivity:

$$304 \quad D(T) = \begin{cases} 1 \times 10^{-10} & \text{if } T \leq T_f - \Delta T \\ 1.32 \times 10^{-3} D_{va}(T) & \text{if } T > T_f + \Delta T \end{cases} \quad (13)$$

$$305 \quad D_{va}(T) = 2.302 \times 10^{-5} \frac{p_0}{p} \left(\frac{T}{T_0} \right)^{1.81} \quad (14)$$

306 where $p_0 = 0.98 \times 10^5$ Pa and $T_0 = 256$ K (Eckert & Drake, 1959); $p = P_{am} = 101325$ Pa.

307

308 Water activity:

$$309 \quad a_w(T, W) = \left[\left(\frac{100 W}{\exp(-0.0056T + 5.5)} \right)^{-1/0.38} + 1 \right]^{-1} \quad (15)$$

310

311 The heat transfer coefficient (h) is a model input for process simulation (see Section
312 3.4), and the mass transfer coefficient (k_g) is determined by using the Chilton-Colburn
313 (or heat-mass) analogy and a correction factor (Purlis & Salvadori, 2009b):

$$314 \quad \frac{h}{k_g^*} = \frac{M_{air}}{M_w} P_{atm} C_{p,air} \left(\frac{Sc}{Pr} \right)^{2/3} \quad (16)$$

$$315 \quad k_g = 7.83 \times 10^{-2} k_g^* \quad (17)$$

316 With regard to heat transfer by radiation, the emissivity of bread surface is considered
317 equal to 0.9 (Hamdami, Monteau, & Le Bail, 2004).

318

319 **3.2. Kinetic model for starch gelatinization extent**

320

321 Zanoni et al. (1995a, 1995b) developed and validated a kinetic model of starch
322 gelatinization for bread, which is temperature dependent. The extent of starch
323 gelatinization follows first-order kinetics and the reaction rate constant is temperature
324 dependent according to the Arrhenius equation:

$$325 \quad \frac{d(1-\alpha)}{dt} = -K(1-\alpha) \quad (18)$$

$$326 \quad K = K_0 \exp\left(\frac{-E_a}{R_g T}\right) \quad (19)$$

327 where $K_0 = 2.8 \times 10^{18} \text{ s}^{-1}$ and $E_a = 139 \text{ kJ mol}^{-1}$. The gelatinization degree (α) is defined

328 as:

$$329 \quad \alpha(t) = 1 - \frac{Q(t)}{Q_{max}} \quad (20)$$

330 where $Q(t)$ and Q_{max} are the heat uptakes for partially baked and raw dough,

331 respectively. At initial condition, $\alpha = 0$, i.e. $Q = Q_{max}$ (raw dough).

332 It can be assumed a complete starch gelatinization when the coldest point of the product

333 achieves a value of $\alpha \geq 0.98$ (Therdthai et al., 2002; Zanoni et al., 1995a, 1995b). This

334 parameter is used to verify the assessment of the minimum baking time (CT) by using

335 the core temperature (≥ 96 °C) as a technological solution. It is worth mentioning that

336 this model is applied to crumb but not to crust, where the starch gelatinization process is

337 more complex due to variation in water content (Primo-Martín, van Nieuwenhuijzen,

338 Hamer, & van Vliet, 2007; Vanin, Michon, Trystram, & Lucas, 2010).

339

340 **3.3. Kinetic model for browning development**

341

342 The formation of colour, i.e. browning is the result of non-enzymatic chemical reactions

343 (Maillard reaction and caramelization of sugars) that produce coloured compounds,

344 which are accumulated in the product during baking. This phenomenon is a dynamic

345 process depending on local temperature and water activity, so it should not be

346 decoupled from transport phenomena (Purlis, 2010). Purlis and Salvadori (2009c)

347 developed and validated a kinetic model for browning development based on a non-

348 isothermal kinetic approach and assuming a general mechanism of browning, which can

349 be described by the variation of lightness (L^* parameter of the CIE $L^*a^*b^*$ colour space).

350 Browning advance is described by first-order kinetics, and the rate constant is a function

351 of temperature and water activity of bread:

$$352 \quad \frac{dL^*}{dt} = -k_b L^* \quad (21)$$

$$353 \quad k_b = (7.9233 \times 10^6 + 2.7397 \times 10^6 / a_w) \exp\left(-\frac{8.7015 \times 10^3 + 49.4738 / a_w}{T}\right) \quad (22)$$

354 Browning is initiated when temperature exceeds 120 °C; raw dough has an initial value
 355 of $L^* = 85$ (standard recipe for French bread: 100% wheat flour, 54.1% water, 1.6% salt,
 356 1.6% sugar, 1.6% margarine, 1.2% dry yeast).

357

358 **3.4. Simulations**

359

360 The bread baking process was simulated for several operating conditions. Input
 361 parameters to the SHMT model were oven temperature (180, 190, 200, 210, 220, 230,
 362 and 240 °C), heat transfer coefficient (5, 10, 15, 20, and 25 W m⁻² K⁻¹), and product
 363 radius (0.025, 0.03, and 0.035 m). These values were selected according to reported data
 364 for conventional baking ovens and common industrial practice (Baik, Grabowski,
 365 Trigui, Marcotte, & Castaigne, 1999; Baik, Marcotte, & Castaigne, 2000; Carson,
 366 Willix, & North, 2006; Li & Walker, 1996; Sakin, Kaymak-Ertekin, & Ilicali, 2009;
 367 Therdthai et al., 2002; Zareifard, Boissonneault, & Marcotte, 2009). Initial temperature
 368 and water content were assumed to be uniform and equal to 25 °C and 0.65 kg kg⁻¹ (dry
 369 basis), respectively. Relative humidity (or water vapour pressure) in oven ambient was
 370 assumed to be negligible (i.e. conventional baking without steam injection).

371 The system of nonlinear partial differential equations describing the stated MBP was
 372 solved using the finite element method. The numerical procedure was implemented in
 373 COMSOL Multiphysics 3.2 (COMSOL AB, Sweden) and MATLAB 7.0 (The
 374 MathWorks Inc, USA). The method of lines is used in COMSOL Multiphysics for
 375 discretization of the partial differential equations, so a differential algebraic equation
 376 system is obtained. This new system is solved using an implicit time-stepping scheme

377 (backward differentiation), i.e. a Newton's method together with a COMSOL
378 Multiphysics linear system solver (UMFPACK). The time step taken by the algorithm is
379 variable (COMSOL AB, 2005), but it was ensured to be small enough (< 5 s) to do not
380 miss the latent heat peak corresponding to phase transition. The finite element mesh
381 consisted in 240 elements in all cases. Finally, a medium order Runge-Kutta routine
382 (function *ode45* from MATLAB) was used to solve (numerically) the quality kinetic
383 models from temperature and moisture content profiles obtained through transport
384 model simulation, using the same criterion for time step as before.
385 Baking time used for process simulation was long enough (90 min) to ensure covering a
386 wide range of practical situations. Afterwards, CT was calculated by interpolating the
387 time-temperature curve of product centre for a temperature value of 96 °C. For this time,
388 other variables were determined: surface temperature, water content and water activity,
389 weight loss, surface lightness, and starch gelatinization extent at product centre. Also,
390 the time-temperature curve of product surface was used to assess the *thermal input* (TI),
391 i.e. the combination of temperature and time to which the product is subjected during
392 the process (FoodDrinkEurope, 2011):

$$393 \quad TI = \int_0^{CT} T_s \, dt \quad (23)$$

394 A recursive adaptive Simpson quadrature routine (function *quad* from MATLAB) was
395 used to evaluate numerically the integral in Eq. (23), using the same criterion for time
396 step as before.

397

398 **4. Results and discussion**

399

400 To investigate the proposed approach, bread baking was simulated for 105 operating
401 conditions according to input parameters established in Section 3.4. For each baking
402 condition, the minimum baking time (CT) was determined, and afterwards other
403 variables were calculated. Therefore, all results shown are feasible solutions considering
404 the proposed theory of an optimum baking process. If the target value for the desired
405 attribute is achieved at this time, i.e. $QT = CT$, then feasible conditions become
406 optimum conditions. In other words, if the value of surface lightness reached at CT is
407 the designed target value, then the heating strategy used is optimum. Otherwise, more
408 time will be necessary while the product is already baked, thus consuming extra energy.
409 This non-optimum condition can appear when the end point of baking is established by
410 colour formation, as described before. To analyze this situation with regard to the
411 proposed approach, we will refer to data reported in Purlis (2011). It is worth to note
412 that temperature and moisture content profiles (and other microscopic data) will not be
413 discussed here. The intention is not to avoid a discussion on transport phenomena but
414 concentrate on the engineering aspects of design, optimization, and control of bread
415 baking. A microscopic perspective of the process can be found in the cited literature.
416 Results obtained from simulations are included in Table 1; Figure 1 is introduced to
417 have a visual reference guide of browning development in bread when analyzing the
418 results. Firstly, it is confirmed that a minimum value of 96 °C at the coldest point of the
419 product is an effective control parameter to assess the minimum baking time, which
420 corresponds to a complete starch gelatinization. Nevertheless, not all operating
421 conditions produce a marked development of browning. In some cases, browning is not
422 even initiated since surface temperature does not exceed 120 °C. Although this is an
423 advantageous situation with regard to nutritional quality because toxic compounds
424 associated with browning reactions can not be generated, the products are valueless

425 from a commercial point of view since (French) bread is characterized by a
426 yellow/golden-brown crust. Also, limited dehydration (i.e. low values of weight loss)
427 occurring under these conditions affects sensory attributes associated with texture due to
428 a limited formation of crust. This situation is mainly produced by natural convection
429 heating mode, represented by values of heat transfer coefficient not greater than 10 W
430 $\text{m}^{-2} \text{ K}^{-1}$, approximately (Purlis & Salvadori, 2009b). In addition, a small radius
431 (characteristic length) favours such situation since CT is reduced and there is less time
432 for the development of browning. On the other hand, as h increases and thus forced
433 convection becomes the heating mode, and oven temperatures above $200 \text{ }^\circ\text{C}$ are used,
434 the development of browning is noticeable.

435 This observation (which can be interpreted as a practical recommendation) seems to be
436 in disagreement with (technological) considerations arisen in Purlis (2011): intense
437 heating (e.g., h greater than $15 \text{ W m}^{-2} \text{ K}^{-1}$ and oven temperature above $220 \text{ }^\circ\text{C}$) as a
438 baking strategy was not recommended because unbaked foods could be produced and
439 high values of surface temperature are achieved, thus generating harmful compounds. In
440 fact, rather than a contradiction there is a conceptual difference that lies in the criterion
441 used in both cases to establish the end point of the baking process. In the previous
442 study, a target value of surface lightness determined the end of baking, with the aim of
443 reproducing a common industrial practice. So, such recommendation was founded on the
444 risk of obtaining unbaked foods while surface colour is acceptable. The approach
445 proposed in this work eliminates this possible problem, and the search is now oriented
446 towards optimum conditions of baking. Nevertheless, the nutritional quality issue is still
447 relevant. In this regard, the Confederation of the Food and Drink Industries of the
448 European Union suggests avoiding excessive browning in the crust to reduce
449 acrylamide formation during baking (FoodDrinkEurope, 2011). In addition, it has been

450 found that the thermal input (combination of temperature and heating time) is a key
451 factor in this subject. For instance, a lower temperature combined with a prolonged
452 baking time does not result in lower acrylamide contents if the same browning of the
453 product is to be achieved (Amrein, Schönbacher, Escher, & Amadò, 2004).

454 By applying the proposed theory, it is observed an increasing trend of the thermal input
455 (TI) with browning development, for a given product dimension (note that assessing TI
456 via the evolution of surface temperature instead of oven temperature allows comparing
457 the results obtained by using different values of heat transfer coefficient) (Figure 2). As
458 expected, an increase in radius produces an increase in TI since longer times are needed
459 to achieve 96 °C at bread centre. Therefore, the recommendation of avoiding excessive
460 browning during baking to diminish acrylamide formation is still applicable. The scope
461 of this paper is limited to develop optimum heating strategies and derive some practical
462 recommendations. In this sense, the ultimate decision about the reduction of acrylamide
463 generation via reduction of browning development requires a fundamental change with
464 respect to the production and consumption of baked products, which will be not
465 discussed here (although it is an urgent debate). Nevertheless, an additional
466 consideration is necessary. Thermal input was also calculated for data reported in Purlis
467 (2011), where the end point of the process was determined for three different values of
468 surface lightness, e.g. $L^* = 80, 75, \text{ and } 70$ (results not shown). When comparing these
469 supplementary results with the ones presented in this work, it was found that no further
470 reduction in TI can be done as by applying the proposed approach, for given values of
471 L^* and radius. A further diminution of TI implies that CT is greater than QT, and thus
472 unbaked products are obtained. Although this observation can be derived from previous
473 considerations elaborated in Section 2, now is inferred from numerical results.

474 Different combinations of oven temperature and heat transfer coefficient can produce
475 the same (minimum) thermal input, for fixed values of final L^* and radius. For example,
476 let analyze the case of $L^* = 80$ (approximately), for $R = 0.03$ m and $R = 0.035$ m (results
477 are extracted from Table 1 and summarized in Table 2 for readability). Firstly, the
478 minimum TI value is balanced by opposite variations in oven temperature and heat
479 transfer coefficient, as can be expected from transport phenomena concepts if the
480 driving force has to be balanced to produce the same TI. Secondly, CT shows a
481 diminishing tendency with the increase of h and the balanced diminution of oven
482 temperature, while weight loss presents an opposite trend; final values of surface
483 temperature do not show a marked behaviour in this regard. This observation reveals a
484 higher influence of the heat transfer coefficient than oven temperature to establish more
485 rapidly the evaporation front at the beginning of baking (in the tested range of operating
486 conditions). This would also explain the higher weight loss produced by increasing the
487 heat transfer coefficient, and thus by the earlier formation of the evaporation front in the
488 product. Weight loss by dehydration of the outer zone of the product is the consequence
489 of the advance of the evaporation front towards the core, which also increases the
490 thickness of the crust (Purlis & Salvadori, 2009a; Zandoni et al., 1993, 1994).

491 In summary, the proposed approach of baking optimization could lead to multiple
492 *optimum* solutions or baking strategies to apply, so a new problem is established to
493 decide which baking strategy should be finally applied. Therefore, such solutions are
494 now feasible solutions for the *ultimate decision problem*. In this sense, the developed
495 theory leads to a two-step optimization problem: the first step consists in finding
496 feasible solutions (or multiple *optimum* solutions), and the second step involves the final
497 decision about the baking strategy to be applied. This second step of the global problem
498 requires a variety of considerations, including sensory (subjective) aspects. Indeed, such

499 global problem represents the design of a baking process. In order to be as general as
500 possible, we will limit further discussion to objective factors, focusing on engineering
501 aspects of the baking process. The main factor to analyze is the heat transfer coefficient,
502 i.e. the oven (flow) characteristics. If the value of h can not be modified (e.g. there is
503 already an oven with a characteristic h value), then the problem is simplified from the
504 beginning, and the only way of optimizing the process is by the proposed approach, i.e.
505 equalling CT to QT. This situation can limit the extent of browning development within
506 the space of feasible solutions with minimum thermal input. If possible, an increase in
507 the characteristic length of the product can lead to a wider range of browning since the
508 CT is increased, so more time is available to colour formation (see Table 1).

509 On the other hand, we have the case of a variable h (not fixed a priori), which represents
510 an entire design problem. In this case, other factors become important to make the final
511 decision since multiple solutions can appear, as in previous examples. Two engineering
512 parameters are the weight loss of the product and the energy demand during the baking
513 process. It has been reported that about 20% of total energy related to the baking
514 process is used for evaporation of water in the product (Le Bail et al., 2010). Based on
515 this information, the optimum baking strategy should be the (feasible) one involving the
516 lower value of heat transfer coefficient. Nevertheless, it should be noted that production
517 costs and economy aspects of the process can not be assessed in a general way, and thus
518 the optimum solution may change depending on each particular case. In any case, there
519 is a compromise situation typical of multi-objective optimization problems, which are
520 solved by assigning a relative weight factor to each objective using empirical data.

521 Finally, the results and discussion derived from the proposed theory could also be
522 helpful to develop and improve baking equipment. In this sense, Zareifard et al. (2009)
523 remarked the need of improving oven performance taking into account the quality and

524 appearance of baked products. An interesting alternative to bread manufacturers would
525 be specialized ovens that allow adjusting the heat transfer coefficient. Therefore, in
526 addition to the improved efficiency sought by oven builders, versatility in terms of
527 design, optimization, and control of the baking process would be delivered to baking
528 industry.

529

530 **5. Conclusions**

531

532 Optimal design of a baking process is a complex and challenging problem that involves
533 several aspects including both quality and operating variables, where multiple
534 objectives have to be taken into account. In addition, baked products are mainly
535 evaluated in a subjective or sensory manner, which makes difficult the task of
536 developing a general approach to design, optimize, and control this traditional food
537 process. To deal with these issues, a theoretical approach was developed and applied to
538 the bread baking process.

539 The presented approach establishes a method to obtain firstly feasible heating strategies
540 that ensure a minimum (critical) baking and minimize the thermal input provided to the
541 product, which is essential for reducing the formation of acrylamide during the process.
542 In this sense, is always recommended to avoid an excessive browning in the product.

543 Afterwards, optimum baking strategies can be established according to different
544 objectives. In general terms, minimization of weight loss should be desirable, and can
545 be achieved by using a low heat transfer coefficient when possible. Finally, the
546 investigation shows a balance between the heat transfer coefficient and baking
547 temperature, which can be used to control the process towards optimum conditions, and
548 also design more efficient ovens.

549 Other food processes could be studied under the developed theory by redefining the
550 critical and quality times, as well as identifying the key operating parameters or factors
551 affecting the process. In this sense, the methodology used in this work (modelling and
552 simulation) or the case of study (bread baking) are not restrictive for the application of
553 the presented approach.

554

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556

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560

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679 **Figure captions**

680

681 **Figure 1.** Image gallery of bread samples with the corresponding value of lightness L^*

682 (Purlis & Salvadori, 2009c).

683

684 **Figure 2.** Thermal input (Eq. (23)) as a function of lightness for different values of

685 bread radius (indicated in the figure) and heat transfer coefficient (symbols, in $\text{W m}^{-2} \text{K}^{-1}$)

686 ¹).

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Table 1. Results obtained from simulations of the bread baking process. For all conditions $\alpha \geq 0.98$. Units: h in $W\ m^{-2}\ K^{-1}$, T in $^{\circ}C$, CT in min, WL (weight loss) in %, TI in $^{\circ}C\ min$.

h	T_{∞}	$R = 0.025\ m$				$R = 0.03\ m$				$R = 0.035\ m$						
		CT	WL	T_s	L^*	TI	CT	WL	T_s	L^*	TI	CT	WL	T_s	L^*	TI
5	180	9.45	1.27	105.46	85 ^a	845.93	12.49	1.67	108.15	85 ^a	1154.42	15.95	2.14	111.35	85 ^a	1520.07
	190	8.93	1.52	107.80	85 ^a	817.34	12.00	1.95	110.81	85 ^a	1139.89	15.35	2.50	115.14	85 ^a	1502.99
	200	8.56	1.80	110.71	85 ^a	803.24	11.43	2.77	116.74	85 ^a	1130.40	14.90	3.05	120.16	85 ^a	1506.28
	210	8.21	2.03	113.45	85 ^a	787.59	11.13	2.70	119.08	85 ^a	1112.29	14.43	3.62	125.63	84.45	1506.46
	220	7.83	2.31	117.32	85 ^a	769.25	10.77	3.30	124.81	84.64	1113.68	14.05	4.05	130.88	83.97	1504.41
	230	7.64	2.65	121.60	84.93	770.27	10.39	3.71	129.51	84.35	1104.47	13.61	4.91	138.30	83.10	1519.74
	240	7.44	3.16	126.94	84.66	774.57	10.09	4.89	138.54	83.49	1136.52	13.36	5.96	146.88	81.80	1562.90
	10	180	8.54	2.23	109.62	85 ^a	801.75	11.43	2.91	113.50	85 ^a	1111.00	14.93	3.62	117.60	85 ^a
190		8.08	2.60	112.60	85 ^a	775.15	10.94	3.48	117.78	85 ^a	1093.15	14.43	4.44	123.34	84.62	1501.17
200		7.82	3.03	116.25	85 ^a	770.32	10.71	3.91	122.10	84.83	1097.14	13.91	5.14	129.13	83.95	1492.58
210		7.60	3.64	120.99	84.94	768.83	10.45	4.67	127.95	84.38	1106.45	13.66	5.87	135.52	83.19	1516.31
220		7.38	4.22	125.92	84.67	769.26	10.25	5.29	133.44	83.96	1123.55	13.42	6.65	142.52	82.23	1545.41
230		7.12	4.94	131.81	84.38	765.72	9.90	6.20	140.93	83.23	1124.05	13.11	7.59	150.53	80.59	1576.82
240		7.08	5.20	135.93	84.20	778.25	9.73	7.12	148.83	82.11	1149.22	12.90	8.31	158.09	78.70	1607.71
15		180	7.91	3.08	113.50	85 ^a	769.43	10.77	4.05	118.74	85 ^a	1090.17	14.20	4.93	123.68	84.53
	190	7.68	3.61	117.41	85 ^a	767.18	10.44	4.70	123.74	84.65	1089.42	13.69	5.78	129.90	83.73	1492.45
	200	7.42	4.44	122.99	84.82	766.35	10.21	5.57	130.11	84.12	1105.14	13.36	6.60	136.60	82.79	1511.65
	210	7.24	5.04	128.06	84.52	769.19	9.96	6.40	136.71	83.45	1117.94	13.24	7.28	143.35	81.79	1547.97
	220	6.99	5.85	134.25	84.15	768.29	9.74	7.26	143.89	82.56	1134.85	12.89	8.39	151.98	79.75	1584.71
	230	6.90	6.43	140.13	83.69	779.96	9.59	7.96	150.92	81.47	1158.93	12.72	9.07	159.63	77.78	1620.52
	240	6.78	7.29	147.43	82.95	796.50	9.45	8.70	158.53	80.01	1182.53	12.63	9.49	166.62	75.57	1660.89
	20	180	7.56	3.98	117.80	85 ^a	760.47	10.41	4.94	123.35	84.70	1092.21	13.61	5.96	128.83	83.78
190		7.34	4.75	123.09	84.79	762.43	10.13	5.81	129.57	84.09	1102.63	13.33	6.87	135.86	82.78	1513.50
200		7.17	5.39	128.35	84.48	766.78	9.92	6.59	136.03	83.47	1113.25	13.03	7.93	143.72	81.38	1548.64
210		6.96	6.20	134.61	84.11	770.12	9.69	7.42	142.99	82.55	1132.64	12.81	8.58	150.74	79.92	1580.21
220		6.83	6.90	140.84	83.55	781.47	9.48	8.45	151.23	81.21	1161.33	12.64	9.30	158.55	78.00	1623.51
230		6.69	7.86	148.61	82.71	798.62	9.39	8.97	158.12	79.95	1187.14	12.42	10.27	167.36	74.87	1676.23
240		6.60	8.49	155.45	81.91	815.33	9.33	9.60	165.53	78.04	1229.97	12.33	10.79	175.23	71.88	1718.23
25		180	7.23	4.92	122.24	84.83	752.07	10.05	5.93	128.14	84.16	1091.45	13.29	6.95	133.78	83.00
	190	7.09	5.69	128.01	84.45	762.19	9.91	6.49	133.89	83.68	1106.90	13.09	7.76	140.92	81.91	1535.93
	200	6.92	6.51	134.28	84.04	770.88	9.64	7.74	142.26	82.59	1132.35	12.83	8.56	148.27	80.48	1568.50
	210	6.77	7.33	141.00	83.40	782.12	9.43	8.71	150.11	81.30	1159.14	12.55	9.68	157.16	78.27	1618.89
	220	6.63	8.21	148.32	82.65	797.47	9.26	9.52	157.92	79.85	1186.59	12.41	10.18	164.55	76.33	1662.00
	230	6.54	8.95	155.69	81.77	816.08	9.17	10.12	165.41	78.14	1220.61	12.22	11.05	173.34	73.42	1710.84
	240	6.46	9.51	162.73	80.68	835.66	9.06	10.77	173.43	75.65	1255.87	12.17	11.40	180.84	70.94	1772.20

^a Browning has not been initiated because surface temperature does not exceed 120 $^{\circ}C$, and thus L^* corresponds to its initial value (85).

Table 2

Results corresponding to operating conditions that produce a final value of $L^* = 80$ (approximately). Units: R in m, h in $\text{W m}^{-2} \text{K}^{-1}$, T in $^{\circ}\text{C}$, CT in min, WL (weight loss) in %, TI in $^{\circ}\text{C min}$.

R	h	T_{∞}	CT	WL	T_s	L^*	TI
0.03	15	240	9.45	8.70	158.53	80.01	1182.53
	20	230	9.39	8.97	158.12	79.95	1187.14
	25	220	9.26	9.52	157.92	79.85	1186.59
0.035	10	230	13.11	7.59	150.53	80.59	1576.82
	15	220	12.89	8.39	151.98	79.75	1584.71
	20	210	12.81	8.58	150.74	79.92	1580.21
	25	200	12.83	8.56	148.27	80.48	1568.50

Figure 1 – Purlis

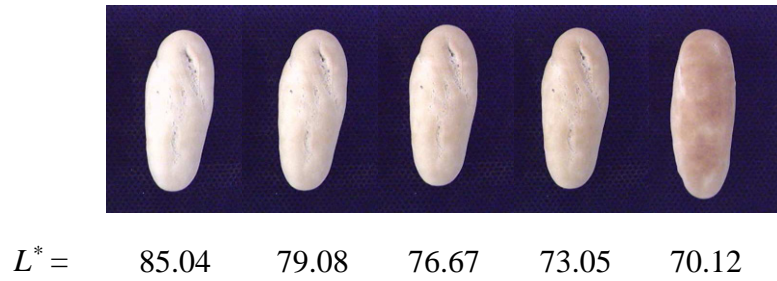


Figure 1. Image gallery of bread samples with the corresponding value of lightness L^* (Purlis & Salvadori, 2009c).

Figure 2 – Purlis

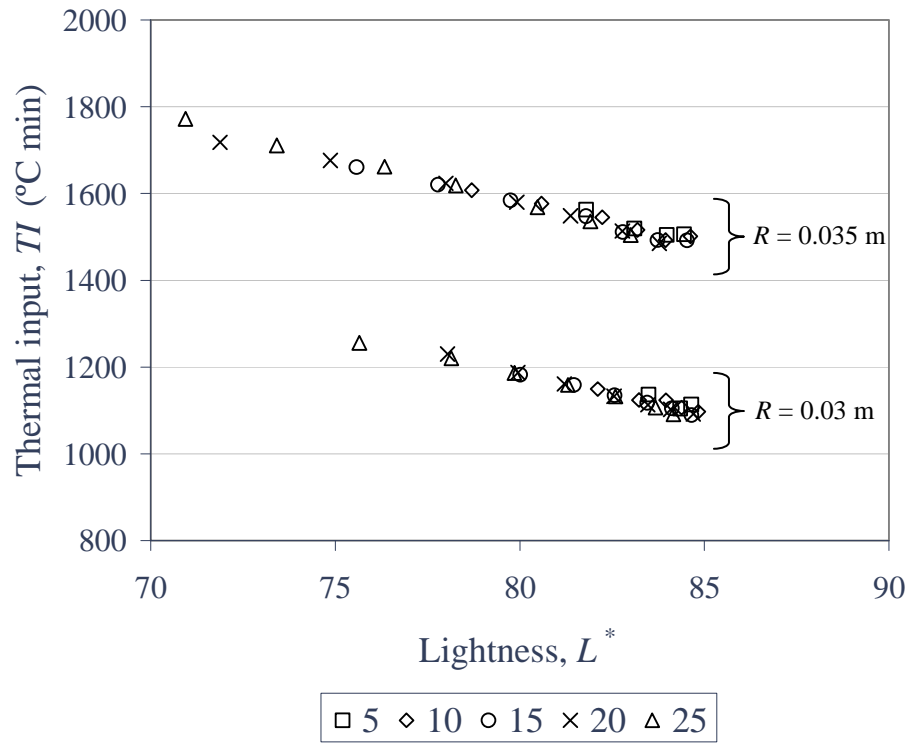


Figure 2. Thermal input (Eq. (23)) as a function of lightness for different values of bread radius (indicated in the figure) and heat transfer coefficient (symbols, in $\text{W m}^{-2}\text{ K}^{-1}$).