

Crust formation and its role during baking

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| 1 | Crust formation and its role during bread baking |
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8 ABSTRACT

9 The final properties of the crumb and crust differ according to their heat-10 moisture dynamics. Compilations of heating and drying rates reported in 11 the literature are discussed and will serve to validate future models of 12 baking. Their impact on the structural elements in dough films and the 13 porous network are discussed, highlighting the lack of data and the need to 14 reproduce these dynamics inside the instrument of analysis. Some roles of 15 the crust setting during the whole baking process are also presented, 16 suggesting further research in this area. Finally, as the region covered by 17 the crust should be defined as a starting point to future studies, definitions 18 proposed in the literature are discussed.

19 Keywords

crust, bread, baking, water loss, temperature, heating rate, heat and mass
transfer, mathematical model, porosity, permeability, rheology, starch
gelatinization, oven-rise, density, bubble, cell, CO₂ release, pressure

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1 1 Introduction

During baking, heat (HT) and moisture (MT) transport take place in the dough simultaneously and interdependently, and involve three major changes – (for further details see recent review by Mondal & Datta (2007)): (1) Water vaporises at the cell/dough interface, and gases accumulated during fermentation (CO₂, ethanol) or generated by chemical raising agents are also vaporised: the cell volume increases provided that the dough film retains gases and is deformable.

9 (2) Starch gelatinisation and protein coagulation transform the viscous
10 dough into a mainly elastic crumb; these rheological changes limit the cell
11 growth described in (1) and enhance pressure build-up.

12 (3) The structure with gas cells separated by films is transformed into a 13 porous structure with inter-connected pores. In theory ruptured films limit 14 cell growth (1): gas molecules are exchanged between adjacent open cells, 15 and finally transported out of the dough. Dough films rupture when they 16 can no longer withstand over pressure. Film rupture is often associated with 17 the onset temperature of starch gelatinisation (Bloksma, 1990) or a higher 18 temperature. This is also likely to happen if pressure is low, but the dough 19 film presents poor mechanical resistance, as when the protein content in the 20 flour is low, the water content in dough is high, or if the dough is under or 21 over-kneaded (Dobraszczyk & Salmanowicz, 2008).

The crust and crumb come from the same original dough, but their final properties differ according to a distinct local heat-moisture treatment. As soon as the dough is placed in the oven, water evaporates very fast from the surface layers, resulting in a much lower water content (< 20% wet basis)

1 than at the core. The total water loss (WL) from the crust is obviously of economic significance (weight loss). Water distribution between the crust 2 3 and crumb also contributes substantially to the organoleptic perception of 4 the final product. As the water activity (Aw) of sponge cake increased from 5 ~ 0 to 0.75, both initial modulus and critical stress fell by an order of 6 magnitude with both correlating (r>0.90) with a trained panel's assessment 7 of sensory harness (Attenburrow, Goodband, Taylor & Lillford, 1989). A 8 crispy texture is also associated with low moisture content and water 9 activity, when starch and gluten matrix are in a glassy state making cells 10 walls more prone to fracture (Stokes & Donald, 2000). Low water content 11 in the crust will also affect the rheological changes in the dough films and 12 ultimately the cell growth during baking. Starch gelatinisation and protein 13 denaturation are limited when there is restricted access to water. This has 14 an effect antagonistic to the one of the decreasing water content and may 15 prolong the deformability of dough films. The enhanced escape of gases 16 due to the proximity of the boundary to the oven may explain the smaller-17 sized cells which are typical of the crust. These obviously contribute to the 18 mechanical properties of the crust (resistance to rupture during baking, 19 texture in mouth) in addition to the low water content.

As the water content decreases in the crust, the temperature can exceed 100°C, which supports other reactions such as the Maillard reaction responsible for the development of colour and the release of flavours, and also the production of toxic compounds with safety implications (Ahrne, Andersson, Floberg, Rosen & Lingnert, 2007).

1 Reactions specific to the crust also jeopardize the nutritional value of the baking process. On the one hand, the Maillard reaction decreases the 2 3 protein digestibility and the lysine bioavailability of lysine which is the 4 limiting amino-acid in cereal products (O'Brien & Morrissey, 1989). On the 5 other hand, gelatinised starch can be degraded by amylases from the saliva 6 whereas ungelatinised fraction undergoes a much slower metabolism and 7 can remain undigested. Conversely, health benefits (obesity risk reduction) 8 are claimed since bread rolls with a higher proportion of crust have been 9 reported to raise capillary blood glucose more slowly than a corresponding 10 loaf (Glatzel & Rettenmaier, 1962).

All these microstructural changes in the cell size and dough films also contribute to the structural differentiation of the crust. Most previous studies have focused on the crust properties at the end of baking or their changes during storage (Luyten, Pluter & van Vliet, 2004), but the setting of these properties during the baking and post-chilling processes have been little studied to date.

17 The first aim of this review is therefore to present how the well known final 18 properties (once cooled) are created dynamically during baking and how 19 the underlying transformations differ from those in the crumb (section 2). 20 The crust properties differ from a bread making technology to another (pan 21 bread versus hearth bread for instance), but also from one face to the other, 22 depending on the extent of heat and mass transfer at the bottom and top 23 surfaces for instance. Given the very few data available in the literature on 24 the topic, elaborating a typology from this review is out of scope; however,

| 1 | the variety present in the literature will help to identify key factors in t | he |
|---|--|----|
| 2 | crust setting process. | |

3 Improvements in the observation techniques will be highlighted in section 24 and summed up in the conclusion.

As the crust cannot be considered separately from the rest of the bread, but
interacts with other mechanisms, some of its role during baking already
identified in the literature will be presented in section 3.

8 As changes in bread are gradual, proposing a simple definition of the crust
9 is a difficult although necessary starting point for future studies, a second
10 focus of the concluding section (4).

11

Mechanisms specific to the bread surface and contributions to crust formation

14 **2.1 Temperature**

15 Surface temperature quickly reaches 100°C and then approaches more 16 slowly the oven air temperature. This rapid increase in surface temperature 17 is enhanced i) by the low thermal conductivity of the aerated dough limits 18 HT at core; ii) by high radiation originating from the warm oven walls 19 (from 66.2 to 81.5% of overall HT) (Baik, Marcotte & Castaigne, 2000). 20 Surface temperatures exceeding 100°C are explained by the difference in 21 water content between surface and core: water evaporates from the surfaces 22 more quickly than it can be transported from the core; in addition, due to 23 the evaporation-condensation-diffusion mechanism (Wagner, Lucas, Le 24 Ray & Trystram, 2007), water content remains almost constant at the core. 25

Table 1 summarizes the heating rates observed at the surface compared to the centre, as approximated by the slope of the linear segment between the initial temperature and 100°C. The heating rate was high in the crust (up to 14.4°C/min) while not exceeding 3°C/min at core. Note that time derivation of temperature kinetics gave even higher values (20 to 35°C/min) at the onset of baking.

7 Temperature was measured using thermocouples (Zanoni & Peri, 1993; 8 Dogan, 2002; Lostie, Peczalski, Andrieu & Laurent, 2002a) or optic fibres 9 (Wagner, Loubat, Sommier, Le Ray, Collewet, Broyart et al., 2008b) 10 placed at different locations inside the loaf, usually only near the top 11 surface and in the centre. However, this intrusive technique introduces 12 biases, including delicate positioning (1-3mm of accuracy), possible heat 13 conduction along the metallic wire of the thermocouple and constraint to 14 oven-rise. To minimize such bias, surface temperature can be monitored by 15 infra-red sensors (Lostie, Peczalski, Andrieu & Laurent, 2002b; Primo-16 Martin, van Nieuwenhuijzen, Hamer & van Vliet, 2007).

17 2.2 Water content

18 The water content in the crumb remains almost constant compared to the19 initial value while considerable dehydration occurs in the crust (

Figure 1). Water activity can also be used to describe the result of WL and
its effects on water availability, reaction kinetics and mechanical properties
(Czuchajowska, Pomeranz & Jeffers, 1989; Lind & Rask, 1991; Bassal,
Vasseur & Lebert, 1993; Dogan, 2002; Van Nieuwenhuijzen, Tromp,
Hamer & Van Vliet, 2007). Given the low final water content in the crust (

1 Figure 1), a moderate variation in water content can affect Aw considerably 2 (Lind et al., 1991). Aw is also involved in the calculation of water transfer 3 during baking, since the driving force was generally assumed to be the 4 difference in partial vapour pressure between the product surface and the 5 oven atmosphere. Aw was measured at ambient temperatures, and very 6 little information is available for temperatures observed in the crust during 7 baking. Studies have been carried out at 100-150°C for cake dough (Bassal 8 et al., 1993), and at 80-120°C for wet and dried crumb (Jury, Monteau, 9 Comiti & Le-Bail, 2007). Additionally, the assumption of thermodynamic 10 equilibrium between liquid and gaseous phases on which the measurement of Aw is based have also been questioned, given the high heating rates 11 involved during baking (Zhang & Datta, 2006). This approach was 12 13 therefore not included in this review.

14 HT has been considered the main driving force for water vaporisation; in 15 other words, MT from the vaporisation front to the loaf outside, proceeding 16 mainly by convection, was not considered to be a limiting factor (Lostie, 17 Peczalski & Andrieu, 2004; Zhang, Doursat, Flick & Lucas, 2008). HT can 18 be enhanced by increasing the oven air temperature, and eventually the air 19 renewal at the product surface; these variables have consistently been 20 experimentally correlated to WL (Wahlby & Skjoldebrand, 2002). The 21 addition of steam increased the relative humidity of the oven air to 0.7-0.9 (Wiggins, 1998). As the dough surface was cold compared to the 22 23 surrounding air, moisture condensed onto it from the air, up to 1% of loaf 24 weight. Steam injection thus reduced WL at the onset of baking. However, 25 as the surface reached the dew point temperature, steam accelerated temperature rise and WL for long baking times (Eliasson & Larsson, 1993).
 Chevallier, Della Valle, Colonna, Broyart & Trystram (2002) reported
 greater WL with higher air humidity in the oven (20 to 400 g/kg of dry air)
 for biscuits. Internal resistance to HT also affects WL; because this
 involves interlinked mechanisms, it will be discussed in section 3.

6 Water content was generally obtained by weighing the product after 7 stopping the baking process at different times, which is extremely time 8 consuming. Moreover, assessing profiles requires cutting the loaf into 9 samples while still hot, and also deformable. Vaporisation of water from 10 cut surfaces and squeezing crumb samples are thus sources of bias. Another 11 difficulty of the sampling method was separating the crust from the crumb. 12 Thorvaldsson & Skjöldebrand (1996) developed an infrared method for 13 continuous measurement of local water content, but the effect of crumb density on the infrared signal was not corrected, generating bias during the 14 15 oven rise period.

16 Validating the thorough description of the mechanisms of water transport 17 (Zanoni et al., 1993; Lostie et al., 2002a; Lucas, Wagner, Doursat, Flick & 18 Trystram, 2009) would obviously require higher spatial and temporal 19 resolution for profiles of water content; this difficulty has been 20 demonstrated for the crumb (Lucas et al., 2009) and is valid for the crust 21 with even greater acquisition constraints (spatial domain of a few hundred 22 to a few thousand μ m). Note that experimental evidence hardly supports a 23 modelling approach based on uniform temperature and water content as 24 suggested by (Fan, Mitchell & Blanshard, 1999).

1 2.3 Biochemical reactions

2 2.3.1 <u>Starch gelatinisation</u>

3 The degree of starch gelatinisation (DSG) can be observed on final cereal 4 products (once completely cooled) by the loss of birefringence of starch 5 granules or the disappearance of the "Maltese crosses" observed in 6 polarized light (Eliasson et al., 1993; Jenkins & Donald, 1998). DSG has 7 been shown to be directly affected by water (Burt & Russell, 1983; Le 8 Meste, Huang, Panama, Anderson & Lentz, 1992; Jenkins et al., 1998; Fessas & Schiraldi, 2000; Cuq, Abecassis & Guilbert, 2003) and the 9 10 heating rate (Donovan, 1979; Bloksma, 1980), which again both differ 11 between the crust and crumb. If DSG consistently differed between cereal 12 products because of their different compositions and baking times (Eliasson 13 et al., 1993), it also varied within the same product: the greater the distance 14 to the surface, the greater the starch gelatinisation (Luyten et al., 2004). Up 15 to 40% of the starch in the crust did not gelatinize during bread baking 16 (Primo-Martin et al., 2007). This was consistent with DSC measurements 17 carried out with open pans (Fessas et al., 2000): for a global water content decreasing from 44.2 to 30.7% (wb), DSG was about 44% -instead of 66% 18 19 measured for sealed pans at initial water content of 44.2%.

Heat-moisture treatment of starch is known to affect the gelatinisation
temperature. Differential scanning calorimeter (DSC) thermograms
obtained from samples of high water content (>90% wet basis (wb))
exhibited a single endotherm peak (

Figure 2). Beyond this point and at water contents above $\sim 60\%$ wb, the endotherm presented three major endotherms (Eliasson, 1980; Burt *et al.*,

1 1983; Champenois, Colonna, Buléon, Valle & Renault, 1995), the first peak corresponding to cooperative, water-mediated melting of starch 2 3 crystallites, the second peak to the melting of the remaining crystallites, and 4 the third to amylose-lipid complex melting transition (Biliaderis, Maurice 5 & Vose, 1980; Burt et al., 1983; Chevallier, Colonna & Lourdin, 2000; 6 Fessas et al., 2000). As water content progressively decreased below this 7 point, the intensity of the first endotherm was reduced, but remained at 8 almost the same temperature (

Figure 2), until its total disappearance below to 35% of water (Eliasson,
10 1980; Burt *et al.*, 1983; Champenois *et al.*, 1995). Conversely, Sopade,
11 Halley & Junming (2004) reported an increase in the gelatinisation
12 temperature of 1.1°C for a reduction in water content from 50 to 40% wb.
13 At the same time, when water content was lower than 60% the second and
14 third endotherms moved to higher temperatures but at different rates (

15 Figure 2) and finally merged into a single endotherm above 35%wb (Burt 16 et al., 1983; Champenois et al., 1995). In fact, the starch granules and the 17 gluten will compete for water, which will result in an additional increase in 18 the gelatinisation temperature (Eliasson et al., 1993; Wang, Choi & Kerr, 19 2004). The loss of birefringence observed for very low water content 20 (<25%) and at high temperatures was related to the second endotherm (Burt 21 et al., 1983). A peak at 70°C was observed in the thermogram of a freshly 22 sampled crust, corresponding to the starch crystals that did not gelatinise 23 during baking, in addition to the peak corresponding to the amylose-lipid 24 complex (Primo-Martin et al., 2007).

1 Some studies have also shown that the gelatinisation temperature was 2 affected by the heating rate. Although applicable to potato starch, 3 (Donovan, 1979) observed that the greater the heating rate (2 to 10°C/min), 4 the higher the temperature of the endotherm peak $(+2^{\circ}C)$. Likewise, Patel 5 & Seetharaman (2006) observed a shift in the swelling of wheat starch 6 granules at higher temperatures when the heating rate increased from 5 to 7 25°C/min. It must be mentioned here that changes in dough viscosity were 8 also delayed by increasing the heating rate from 3 to 9°C/min (Bloksma, 9 1980).

To conclude, it can be expected that starch granules present in the superficial layers only partially gelatinise, because of the strong reduction in water content and the more rapid heating rates, which both increase the starch melting temperature. This will influence the rheological properties of dough films during baking and possibly cell growth in the crust. As far as the crust is concerned, it would be interesting to characterise this reaction during simultaneous heating and drying.

17 2.3.2 <u>Gluten coagulation</u>

18 The lower the water content, the higher the denaturation temperature of 19 proteins (Eliasson et al., 1993). For ω -gliadin aqueous solutions for 20 instance, it increased from 117 to 157°C by lowering the water content 21 from 20 to 3%wb (Noel, Parker, Ring & Tatham, 1995). Denaturation is 22 followed by aggregation and, for some proteins, gel formation (Eliasson et 23 al., 1993). In contrast to the first stage, this stage is a kinetics-dependent 24 exothermic process: the lower the heating rate, the more advanced the 25 process of aggregation (Myers, 1990).

As a consequence of these combined effects, the bread crust showed a 1 2 minimal decrease in water-extractible proteins, meaning that they were not as aggregated and/or cross-linked as in the crumb (Westerlund, Theander & 3 4 Aman, 1989). This would undoubtedly affect the mechanical modifications 5 of the gluten network in the superficial layers upon heating. The 6 disappearance of soluble protein was almost complete in wheat gluten and 7 glutenin at 25-30% wb of water content heated at very high rates (25°C/s 8 from 40 to 170°C) (Strecker, Cavalieri, Zollars & Pomeranz, 1995); to the 9 light of this last study, results from Westerlund et al. (1989) could be 10 explained to the very low water content encountered in bread crust.

The amount of water to be released was also expected to be lower, with possible limitations on other biochemical reactions including starch gelatinisation (Marston & Wannan, 1976; Wang *et al.*, 2004) and/or to water transport (Thorvaldsson & Skjöldebrand, 1998; Mondal & Datta, 2008).

16 Glass transition (GT) is also an issue given the low water contents
17 associated with high temperatures (Huang, Haynes, Levine & Slade, 1996)
18 (

Figure 2). However, it is commonly accepted that GT usually occurs upon
cooling (Cuq *et al.*, 2003) –also see

Figure 2, which is outside the scope of this review and is not discussed here. Nevertheless local heat-moisture treatment during baking (sections 2.1 and 2.2) undoubtedly affect the consecutive process of GT during postchilling.

1 2.3.3 <u>Maillard reaction</u>

2 Highly reactive compounds are initially produced from this reaction; 3 afterwards, their polymerization yields products of brown colour (Martins, 4 Jongen & van Boekel, 2001) observable at around 105-115°C (Zanoni, Peri 5 & Bruno, 1995a; Broyart, Trystram & Duquenoy, 1998; Wahlby et al., 6 2002). Starch is also consumed during the reaction, leading to a reduction 7 in the starch concentration in the crust compared with the crumb (Cauvain, 8 1998). Finally, some of the desired flavours and aromas of bread, among 9 which 2-Acetyl-1-pyrroline, 4-hydroxy-2,5-dimethyl-3(2H)-furanone, 2,3-10 butanedione, methional, (E)-2-nonenal, methylpropanal, and 2- and 3-11 methylbutanal for baguette crust, are also produced, mainly by the Maillard 12 reaction (Zehentbauer & Grosch, 1998a; b; Poinot, Arvisenet, Grua-Priol, 13 Colas, Fillonneau, Le Bail et al., 2008).

14 Low water activity (optimal between 0.4 and 0.8) (Charissou, Ait-Ameur & 15 Birlouez-Aragon, 2007) and high temperatures (commonly above 105°C as 16 mentioned above) accelerated the Maillard reaction. WL and browning 17 were linearly correlated (Wahlby et al., 2002; Purlis & Salvadori, 2007). 18 Independently of the HT mode (natural or forced convection), the lightness 19 of the bread surface decreased as the oven temperature increased (180 to 20 220°C); at even oven air temperatures, forced convection caused higher 21 browning than natural convection (Purlis et al., 2007).

Different models based on first order kinetics were proposed to predict the kinetics of colour development. They were dependent on surface temperature alone (Zanoni *et al.*, 1995a) or combined to water content, yielding errors of prediction less than 10% for WL higher than 10% (Purlis *et al.*, 2007) or between 1-24% on biscuits at the end of baking (Broyart *et al.*, 1998).

The Maillard reaction may also result in toxic compounds, such as
acrylamide. Several recent reviews have been focalized in the study of
acrylamide formation in cereal products (Sadd & Hamlet, 2005; Konings,
Ashby, Hamlet & Thompson, 2007; Claus, Carle & Schieber, 2008) (for
further details see the cited reviews).

8 Previous studies correlated its formation to the oven air temperature, 9 although the key variable is the exact local temperature at the dough 10 surface which depends on many variables other than the oven air 11 temperature. The amount of acrylamide in the crust of bread, flat bread, dry 12 starch system and dried rye-based flat bread rose with longer baking times 13 and higher oven temperatures (Brathen & Knutsen, 2005). A similar effect 14 was reported by Ahrne *et al.* (2007).

15

16 2.4 Rheological evolution

17 The rheological properties of dough change significantly during baking, 18 mainly as the result of the temperature-induced changes to the molecular 19 structures. Between 26 and 60°C, the viscosity of dough decreased by a 20 factor of 5 (Bloksma, 1990), supporting the deformation of dough films. 21 The water content is a key factor since a variation of 4% induced a 22 variation in viscosity by a factor of 2 (Bloksma, 1990). Above 60°C, 23 viscosity increased by a factor of 10, and the starch granules inflated and 24 the gluten molecules were polymerized, damaging dough extensibility 25 (Bloksma, 1990). Moreover, the higher the heating rate, the lower the minimum viscosity and the higher the temperature where this minimum
viscosity occurred (Bloksma, 1980).

Although starch gelatinisation and protein denaturation are responsible for the loss of extensibility of the crumb, this should not apply to the crust (section 2.3.1): starch gelatinisation is expected to be delayed by the high heating rate and the low water content encountered in the crust and this would favour low levels of viscosity. It is thus most probable that the loss of extensibility of superficial layers during baking is due to dehydration, although this point is not clear in the literature.

10 The rheological properties of bread dough have been widely characterised 11 experimentally but generally not in conditions realistically representing the 12 baking process, especially regarding the crust. Firstly, viscosity and 13 eventually elasticity have been thoroughly measured at ambient and/or 14 constant temperatures, but rarely during dynamic heating. When applied, 15 heating rates have been appropriate to the bread core (Collar, Bollain & 16 Rosell, 2007), and rarely to the crust, the highest comparable values being 9 17 to 11°C/min (Bloksma, 1980; Singh & Bhattacharya, 2005). Secondly, 18 bread dough has been characterised at very low uniaxial shear rates (between 10^{-2} and 10^{-3} s⁻¹) (Bloksma, 1990) and more recently under 19 biaxial extension at rates of 5×10⁻² s⁻¹ (Rouille, Della Valle, Lefebvre, 20 Sliwinski & vanVliet, 2005) and 10⁻¹ s⁻¹ (Dobraszczyk et al., 2008). Most 21 22 of these strain rates failed to reproduce those encountered during baking (1 to 2×10^{-3} s⁻¹) (Bloksma, 1990). With the exception of biaxial extension 23 24 (Dobraszczyk & Morgenstern, 2003), other tests used in combination with 25 dynamic heating only approximately reproduce the expected deformation 1 of dough films over a gas bubble during baking. Finally, rheological 2 measurements applicable to the crust should ideally be conducted under 3 dynamic drying; again most of the data available were obtained at a 4 constant water content, with levels relevant to the dough.

5 In the current mathematical models of baking, the mechanics are usually 6 simplified and the dough considered as viscous only (Lostie et al., 2002b; 7 Lucas et al., 2009). Viscosity was not dependent on water content and was 8 at best temperature-dependent. Nevertheless, fair agreement between 9 simulation and experiment was usually obtained due to the numerous 10 parameters to be adjusted and because the validation was based on overall 11 measurement of the oven-rise (Lostie et al., 2002a; Zhang et al., 2006), and 12 rarely on local measurements (Lucas et al., 2009). The simplified 13 rheological behaviour of dough (viscous rather than viscoelastic) used in 14 these models could be responsible for the discrepancies in total height 15 (Lostie et al., 2002a) or in the intensity of local compression in the crumb 16 (Lucas et al., 2009).

17

18 2.5 Cell growth

Because of the proximity of the boundary with the oven air, gases easily escape the crust and the pressure remains close to the atmospheric pressure (see pressure profiles calculated by Zhang *et al.* (2006) and Lucas *et al.* (2009)). Additionally, the loss of extensibility of dough films is expected to be more rapid, presumably because of the great reduction in water content. Thus, one reason to explain the smaller size of cells in the crust than in the crumb (Figure 3) is that the forces favouring expansion are not sufficient for cell
 growth, although the full mechanism underlying cell growth warrants in
 depth study.

Moreover, the surface layers cannot deform fast enough to accommodate the core expansion. This leads to a domed shaped top surface, and also to similar curvature of the internal structures, *i.e.* elliptical shapes of cells observed during baking (Whitworth & Alava, 2004). In fact, when setting of the surface layers occurred early compared to the heating at core (

9 Figure 4f), two opposite forces were exerted on the intermediate regions,10 which tended to be compressed (

Figure 4d), the extent of compression being related to the time elapsed
between stiffening of dough films and their rupture (Jefferson, Lacey &
Sadd, 2006; Lucas *et al.*, 2009).

14 The dimension, shape and number of gas cells in the crumb have been 15 studied by various imaging techniques; comparatively the crust was subject 16 of few studies (Datta, Sahin, Sumnu & Keskin, 2006; Zhang et al., 2006; 17 Primo-Martin et al., 2007). It must be emphasized that the majority of 18 microscopy techniques are destructive and do not allow dynamic 19 observation of a process such as baking. Other techniques providing 20 dynamic measurements often average the bubble size distribution over the 21 whole dough sample, which is relevant in the case of a quite homogeneous 22 alveolar structure as expected during proving (Leroy, Fan, Strybulevych, 23 Belido, Page & Scanlon, 2008). Another requirement for imaging is high 24 spatial resolution, given that the mean cell size in the crust is around 25 100µm. Among the techniques available, X-ray synchrotron seems to be

the most appropriate. A quantitative, dynamic study of the alveolar structure in the crumb during baking was recently reported (Babin, Della Valle, Chiron, Cloetens, Hoszowska, Pernot *et al.*, 2006). The feasibility for the crust still needs to be tested. It would require warming the sample at higher rates and using higher frequencies of acquisition than those used for the crumb, although the signal to noise ratio would be lower due to low water content and low levels of signal accumulation.

8

9 3 How the crust affects other mechanisms during baking

3.1 Mechanical constraint to crumb expansion, both locally and overall

12 3.1.1 Loss of extensibility

The cessation of overall expansion was experimentally related to the occurrence of a dry surface during sponge cake (Lostie *et al.*, 2002b) and bread (Zanoni *et al.*, 1993) baking. Similarly, protecting dough samples with paraffin oil to prevent dehydration allowed continuous expansion until 100°C, whereas it ceased between 36 and 62°C (Le Meste *et al.*, 1992). Using magnetic resonance imaging (MRI) Wagner, Quellec, Trystram & Lucas (2008a) recently reported that if the crust was set early, and even if

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Figure 4a- c), especially in the colder areas which had not already expanded
(

Figure 4f). These areas expanded to the detriment of others which werethen compressed (

Figure 4d). Such compression was accompanied by the disappearance of the largest bubbles, suggesting that it involved the weakest structures from a mechanical view-point. (Zhang, Lucas, Doursat, Flick & Wagner, 2007) developed a device with a fabric lid which made it possible to stop the oven-rise at different heights. Using MRI they showed that the later the oven-rise was stopped, the deeper the location of the squeezed crumb. Such a trend could be reproduced with a model of baking (

8 Figure 4a-c) which in turn made it possible to relate the occurrence of 9 compression to the mechanical properties of dough films (already ruptured 10 but not already stiffened). Similarly, in the theoretical approach developed 11 by Jefferson et al. (2006), the more a cell is set when its film fractured, the 12 less squashing took place and the lower the final density in that part of the 13 bread. Likewise, Hayman (1998) suggested that crust formation resulted in 14 an increase in the internal pressure in the unbaked portion of the dough, 15 imposing additional stress on the dough films, and ultimately leading to 16 bubble coalescence and to a coarser crumb structure. If no crust was 17 formed, a fine crumb structure was expected, which was evidenced from 18 cross sections of loaves produced with their electrical resistance oven 19 (ERO) for a wide range of wheat flours.

Additionally, MRI and X-ray images showed how crust rupture allowed
prolongation of the local expansion and also contributed to strong spatial
heterogeneity in crumb expansion (Whitworth *et al.*, 2004; Wagner *et al.*,
2008a).

Crust setting has rarely been taken into account in the models of baking. Todo so, the rheological properties of dough should be dependent on both

temperature and water content (section 2.4). Moreover, a proper description would be two-dimensional (Zhang *et al.*, 2006). In current models of unidirectional transport and expansion, the cessation of oven-rise was "successfully" reproduced by artificially imposing nil displacement of the outer boundary after a certain baking time –adjusted on experimental data *e.g.* (Lucas *et al.*, 2009), or by using high values of viscosity at high temperatures *e.g.* (Lostie *et al.*, 2002a).

8

3.1.2 Lower permeability to gases

9 The constraints to crumb expansion may also be generated by low 10 permeability of the crust to gases, through the pressure build-up. Almost no 11 pressure build-up was obtained from baking models when high 12 permeability values with uniform distribution through the dough were used 13 (Zhang et al., 2006). The authors finally retained in their simulations lower 14 permeability for the "crust" (the region of interest was not defined) than for 15 the crumb. The effect of low permeability of the surface layers on the 16 pressure build-up was recently evidenced by Grenier, Le Ray & Lucas 17 (2009).

18 It is well known that the permeability of a porous medium is affected by the 19 pore fraction and structure. Very little information is available in the 20 literature for the dough/crumb, and some of it is controversial. Baker 21 (1939) related the gas flow to cell size in commercial white bread, but no 22 permeability values are available in this study. Dough permeability was 23 directly related to porosity, whatever the water (15-50%wb) and fat (2-8%) contents (Goedeken, 1993). Based on this study, the permeability of typical 24 crumb (75% of porosity) was estimated at around 2.2×10^{-11} m², and of 25

typical crust (40% of porosity) around 10⁻¹² m². These orders of magnitude were used for crumb (10⁻¹¹ m²) and crust (2.5×10⁻¹² m²) in the baking model developed by Zhang *et al.* (2006). All other baking models considered uniform permeability through the product (Lostie *et al.*, 2002a; Lucas *et al.*, 2009), which makes their simulations of CO₂ release and oven-rise questionable.

7 3.1.3 <u>Secondary effect on the gas released (through pressure build-up)</u> 8

9 Zhang et al. (2007) monitored the CO₂ released during baking and 10 observed an induction period followed by a sudden, linear increase in CO₂ 11 release, ending with a plateau zone. They also found that the earlier the 12 oven-rise was stopped, the shorter the induction period. Similar trends 13 could be reproduced by a mathematical model of baking, which showed in 14 addition that film rupture was necessary but not sufficient to demonstrate 15 CO₂ release and that it must be accompanied by a pressure build-up similar 16 to that stemming from an early cessation of the oven-rise. Similarly, Lucas, 17 Le Ray, Peu, Wagner & Picard (2007) found lower CO₂ release for lower 18 oven air temperatures, which delayed the formation of the crust.

19 CO_2 was considered to be the main gas responsible for oven-rise (Bloksma, 20 1990). Leakage of CO_2 obviously implies a decreasing amount of gas in 21 cells and presumably a lower potential for cell growth. Interpreting the CO_2 22 release is however a little more complicated since it also reveals a build-up 23 in internal pressure, which is the driving force for cell growth provided that 24 the dough films are still deformable. The net balance between these two 25 antagonistic mechanisms has not been assessed to date. CO_2 is also regarded as a vector of transport of the aromatic compounds (Eliasson *et al.*, 1993). For all these reasons, the mechanisms governing CO₂ release
 warrant further investigation for a thorough understanding and a clear
 identification of the key factors.

5 **3.2** A barrier to heat and water transport

6 Wahlby et al. (2002) compared the WL of buns reheated with and without 7 crust. Whatever the heating time, WL without crust was three times greater 8 than with crust. This was attributed to the differences in crust and crumb porosity. Breads baked in an impingement or hybrid oven (forced 9 10 convection combined with microwave) lose less water than conventionally 11 baked breads (Patel, Waniska & Seetharaman, 2005). This was attributed to 12 early crust formation in the former case. Although this effect on WL is 13 commonly accepted (Eliasson et al., 1993), the underlying mechanisms 14 have only recently been studied with the aid of models (Lostie et al., 2004; 15 Zhang et al., 2008).

16 As reported in section 2.2, HT governed the progress of the vaporisation 17 front in the surface layers and thus WL during baking. As the front went 18 deeper, the resistance to HT increased and slowed down the WL. Similarly, 19 high porosity in the surface layers increased this resistance (Vanin, Grenier, 20 Doursat, Flick, Trystram & Lucas, 2009) since the same amount of water 21 was distributed over a thicker area. Changes in thermal conductivity could 22 also affect the resistance to HT in the surface layers and in turn the WL 23 (Vanin et al., 2009). Thermal conductivity of the dough/crumb decreased 24 with decreasing water content and increasing porosity (Rask, 1989; Zanoni, 25 Peri & Gianotti, 1995b; Jury et al., 2007). However, conductivity measured on crust and crumb samples has been found to be very close (Jury *et al.*,
 2007), meaning that the effects of porosity and water content may
 compensate for each other.

Water transport to the core by the evaporation-condensation-diffusion mechanism also contributed to the dehydration of superficial layers, but to a lower extent; it was theoretically favoured by higher porosity in the crumb (Zhang *et al.*, 2008).

8 It must be remembered here that the porosity profile which affects the crust 9 thickening through the mechanisms described above is itself greatly 10 affected by fairly early setting of the crust (see section 3.1.1, and also 11 Figure 4a-c). A thorough understanding of these strongly interlinked 12 mechanisms is necessary for fine control of crust formation, WL and local 13 density together (Vanin *et al.*, 2009).

14

15 **4** Conclusion

16 **4.1** Can a definition of the "crust" region be proposed?

An accurate definition of the spatial domain comprising the crust remains
necessary for any further study. Changes in its properties are believed to be
gradual, making it difficult to define a boundary with the crumb beneath.

A crust is commonly referred to as a dry, hard, dense, coloured zone, following exposure to high temperatures: "... a hard, vitreous surface layer formed of collapsed crumb pore walls" (Eliasson *et al.*, 1993); "...a denser, darker surface parts bread, near to its surface" (Jefferson *et al.*, 2006). It must be remembered that low water content and high temperature are

determinants (but not the only) of many of the other features (colour,
 porosity, molecular structures, hardness...).

3 Only a few experimental studies gave their criterion separating the crust 4 from the crumb. The crust sample was often unique, distinguishable from 5 the crumb sample(s) by its darker colour (visual criterion, e.g. (Lind et al., 6 1991)) or its more cohesive, harder structure (mechanical property e.g. 7 (Westerlund et al., 1989)). Many factors, including lower water content, 8 higher density, smaller and more elongated cells, vitreous transition, etc... 9 may contribute together or separately to a distinct mechanical behaviour. 10 Mechanical separation may also originate from constraints created during 11 cooling e.g. (Le Bail, Monteau, Lucas, Chargelègue & Reverdy, 2005) in 12 the case of part-baked breads. (Zanoni et al., 1993) separated the crust by 13 quick freezing which caused rupture between the "crust" and "crumb". All 14 this makes the mechanical criterion not very reliable when comparing 15 results from various studies. The same applies to colour, with the effect of 16 small carbohydrates.

17 Surprisingly, despite these raw criteria, the properties measured on crust18 samples are quite consistent between studies (

Figure 1). Likewise, thickness of the crust has been reported to increase
linearly with baking time (Zanoni *et al.*, 1993; Wiggins, 1998). However,
the number of studies is not sufficient to conclude at this stage.

From a theoretical point-of-view, the crust boundary can be refined by using a threshold value applied to a criterion such as density or water content. Jefferson *et al.* (2006) characterised crust thickness as the distance from the surface where the final density was the same as the original dough density and Zhang *et al.* (2008) from the top surface to the point where the
water flowed most rapidly, *i.e.* the vaporisation front. A criterion with
physical relevance to the threshold should represent a sharp change
between the surface and the core, as water content or temperature do (

5 Figure 4e, f). Porosity presents either no sharp change (

6 Figure 4a) or many sharp changes (

Figure 4c) between the surface and core, which makes this criterion hardly reliable. Corresponding experimental profiles of water content and temperature are few and thus it is a definition that could not be applied at the experimental level to date. All this supports the need to develop a noninvasive, dynamic technique to study the specific behaviour of surface layers, with the special requirement of high spatial resolution.

13 **4.2** Need for further research

Superficial bread layers differ from core layers of the crumb mainly in their dynamics during baking. Lack of published data and information on prevailing mechanisms in the superficial layers of bread have prevented modelling studies from considering crust setting and its effects on heat, mass transport and cell growth in the crumb, as well as the full understanding and control of baking.

On the one hand, this review has shown that the knowledge acquired regarding the crumb cannot be extrapolated to superficial layers with confidence, especially because the mechanisms are highly interrelated, often working antagonistically and because they are highly sensitive to temperature and water content. The identification of prevailing mechanisms thus remains an issue for future studies.

1 On the other hand, we have attempted to fill the gap in the published data 2 through the different enclosed charts. We have also shown that methods of 3 measurement for monitoring the dynamics (temperature, water content) in the superficial lavers present limitations in terms of convenience and 4 5 accuracy, and there is a real need to develop, or sometimes simply 6 implement, non-intrusive, continuous techniques of measurement. The 7 dynamics impact on the microstructures and hence the main properties of 8 the crust. Understanding of the underlying reactions has been inferred from 9 measurements performed on samples of different water content, but very 10 few studies have related heating to drying. There is therefore a real 11 challenge to reproduce these dynamics with miniaturisation inside the 12 instrument of analysis (rheometer or X-ray synchrotron for instance). 13 Aguilera (2005) recently emphasized the possibilities offered by the recent advances in imaging to dynamically study of microstructures during 14 15 processing. All these developments will contribute to providing 16 information of higher spatial resolution and to a clearer definition of the 17 crust region, a prerequisite for further studies in this area.

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| |
| Table and figure list |
| |
| Figure 1: Water content in crust (hollow symbols) and crumb (filled |
| symbols) during baking from different literature sources. |
| |

Figure 2: Dependence on water content of gelatinisation temperatures of
wheat starch-water mixtures and GT of gluten and bread as demonstrated
by DSC from different literature sources.

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Figure 3: Microstructure of bread baked in microwave-infrared
combination oven observed by SEM at ×50 for (a) crumb, (b) crust. Bars
represent 300 μm. Source: Datta *et al.* (2006).Reprinted from Journal of
Food Engineering, 79, Datta, A.K., Sahin, S., Sumnu, G., & Keskin, S.O.,
Porous media characterisation of breads baked using novel heating modes.
106-116, Copyright (2006), with permission from Elsevier.

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Figure 4: Porosity profile for different final total heights (45(a), 50(b) and 55(c) mm) after 45 min of baking. Porosity (d), water content (e) and temperature (f) profiles at different baking times for a final height of 60 mm. Adapted from Zhang *et al.* (2007) and Lucas *et al.* (2009).

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18 Table 1: Heating rates for different cereal products and baking conditions at

- 19 bread surface and core.
- 20
- 21

Table 1

| Product | Mass of dough/ batter (g) | Surfaces exposed to heat transfer | Position of temperature in dough/ batter | Oven air temperature (C°) | Heating rate (°C/min) | Reference |
|-----------------------|------------------------------------|---|--|---------------------------------|---|--|
| Bread | 200 | top bottom | bottom surface centre | 185 | 2.6 2.4 | (Wagner <i>et</i> <i>al.</i> , 2008) |
| Bread | 341 | all | top surface bottom surface at 1cm beneath the top surface | 203 | 11.3 9.5 2.6 | (Zanoni <i>et</i> <i>al.</i> , 1993) |
| Bread | - | all | top surface mid-width centre | 210 | 10.3 * 3.9* 1.8* | (Thorvaldss on <i>et al.</i> , 1996) |
| Pan bread Francala | - | - | surface | 220 | 8.8 14.4 | (Dogan, 2002) |
| Bread | 1.5L | all | top surface | 225 | 6.7 4.4 | (Thorvaldss on <i>et al.</i> , 1998) |
| Bread | 760 | all | side surface centre | 235 | 7.4 2.8 | (Marston <i>et</i> <i>al.</i> , 1976) |
| Biscuit | - | all | centre | 300 | 75.0 ^{**} (400g/kg dry air) | (Chevallier |

| | | | | 300 | 61.7** | et al., 2002) | | |
|-----------------------------------|---------|-----|-------------|-----|-------------------|---------------------|--|--|
| | | | | 500 | (20g/kg dry air) | | | |
| | | | | 240 | 44.7** | | | |
| | | | | | (200g/kg dry air) | | | |
| | | | | 100 | 19.6** | | | |
| | | | | 180 | (20g/kg dry air) | | | |
| Sponge cake | 400-600 | ton | ton surface | 200 | 0.5 | (Lostie et | | |
| Sponge eake | +00-000 | юр | top surface | 200 | 0.5 | <i>al.</i> , 2002b) | | |
| "-" refers to missing information | | | | | | | | |

* data from reheated samples

** with different relative air humidity



Figure 1



Figure 2







Figure 3



Figure 4