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Ingredient functionality in multilayered dough-margarine systems and the resultant pastry products: a review

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ABSTRACT

Pastry products are produced from heterogeneous multilayered dough systems. The main ingredients are flour, water, fat and sugar for puff pastry and the same plus yeast for fermented pastry. Key aspects in pastry production are (i) building laminated dough containing alternating layers of dough and bakery fat and (ii) maintaining this multilayered structure during processing to allow for steam entrapment for proper dough lift during baking. Although most authors agree on the importance of gluten and fat for maintaining the integrity of the different layers, detailed studies on their specific function are lacking. The exact mechanism of steam entrapment during dough lift and the relative contribution of water set free from the fat phase during baking also remain unclear. This review brings together current knowledge on pastry products and the factors determining (intermediate) product quality. Its focus is on flour constituents, fat, water and (where applicable) yeast during the different production stages of pastry products. Future

research needs are addressed as the knowledge on biochemical and physical changes occurring in flour constituents and other ingredients during pastry production and their effect on product quality is currently inadequate.

KEYWORDS

Pastry, puff, croissant, wheat flour constituents, margarine, laminated dough, dough lift

ABBREVIATIONS USED

FA, fatty acids; GO, glucose oxidase; MAG, monoacylglycerols; SS, disulfide; SSL, sodium stearoyl lactylate; TG, transglutaminase; T_m, gelatinization temperature.

1. INTRODUCTION

The term "pastry" groups various fat-rich and often sweet bakery products. Pastry products are consumed worldwide as numerous regional variants. They include shortcrust pastry (as in pies, "quiche"), phyllo pastry (as in "strudel", "baklava"), choux pastry (as in "éclair", "profiterole"), puff pastry [as in (British) pasty, "vol au vent"] and fermented pastry (e.g. Danish pastry, "croissant") (Baking Industry Research Trust, 2013). This review focuses on puff pastry and fermented pastry.

The production of puff and fermented pastry products is very similar. Their characteristic flaky texture is achieved by producing and baking multilayered heterogeneous dough-margarine systems (Hay, 1993). **Table I** shows typical formulations for puff pastry, Danish pastry and croissant. Essential ingredients for puff pastry are water, (wheat) flour, bakery fat and salt (Hay, 1993). Typically, sugar, milk powder, eggs and/or improvers are also part of the recipe (Bent, 2007; Cauvain and Young, 2009). The ingredient list of fermented pastry products evidently also includes yeast (Baardseth *et al.*, 1995; Cauvain and Young, 2009). Variations between different fermented pastry products such as Danish pastry and "croissant" typically depend on differences in the relative ratios of the individual ingredients (Bent, 2007).

Most cereal product research has focused on the role of the abovementioned ingredients in the production of different bakery products such as bread (Goesaert *et al.*, 2005 and references cited therein), cake (Wilderjans *et al.*, 2010; Luyts *et al.*, 2013) and cookies (Kaldy *et al.*, 1991; Pareyt *et al.*, 2008; Pareyt *et al.*, 2009; Pauly *et al.*, 2013). In contrast, their roles in puff and fermented pastry products remain largely unclear as reflected by the lack of extensive literature on the topic. We here provide an up-to-date overview of the role of the main ingredients and their

constituents during dough formation, fermentation (where relevant), baking and ageing of pastry products. Where possible, we identify the current knowledge gaps and speculate on ingredient functionality based on insights for other bakery products. Before doing so, an outline of the pastry production process is given. Finally, some commonly used pastry improvers and how they affect pastry product characteristics are discussed.

2. PASTRY PRODUCTION PROCESS

2.1 Ingredient mixing and predough formation

The manufacturing of puff and fermented pastry products starts with producing what is referred to as "predough". Predough is usually prepared according to a "straight dough" method in which all ingredients are mixed together in a single step (Stauffer, 1990; Bent, 2007). Part of the bakery fat used in pastry production is incorporated in predough (Hay, 1993). This fat is here further referred to as "in-dough" fat. In an industrial environment, the source of in-dough fat is typically the fat present in the recovered 'pastry trimmings' (see also Section 3.1.3).

Predough mixing typically takes place in a relatively cool environment (18-20°C) (Stauffer, 1990). To allow sufficient yeast activity in predough at such temperatures, rather high yeast levels are added (Stauffer, 1990). The temperature of the ingredients and more in particular that of the added water is important for obtaining the relatively low temperature. After mixing, for quality and processing reasons outlined further, the predough temperature is to be similar to that of the fat folded in during subsequent lamination (see Section 2.2), i.e. the "roll-in" fat (Cauvain and Young, 2009). Typically, the mixed dough temperature approaches 20°C (Bennion *et al.*,

⁴ ACCEPTED MANUSCRIPT

1997). According to the New Zealand Bakery Industry Research Trust (2014), predough mixing of croissant dough takes longer than that of Danish pastry dough.

2.2 The lamination process

The second step in pastry manufacturing, i.e. lamination, produces multilayered dough consisting of alternating layers of predough and fat. The roll-in fat is usually margarine (Cauvain and Young, 2009).

Traditionally, two different methods are used for folding the roll-in fat into dough. In the English method (**Figure Ia**), predough is first spread out or sheeted. The roll-in fat is flattened to a thickness comparable to that of the predough and put on top of the laminated predough, thereby covering two-thirds of the surface of the latter. The part of the predough that remains free from roll-in fat is then folded into the middle and covers about half of the roll-in fat. Finally, the other end (one predough and one fat layer) is folded on top. This way, after such first folding, two fat and three dough layers result (**Figure Ia**) (Bennion *et al.*, 1997; Podmore, 2002; Bent, 2007). Alternatively, in the French or envelope method, a rectangular layer of fat is wrapped in the laminated predough by folding the corners of the predough to the middle on top of the fat. This method results in one fat layer surrounded by two layers of predough (**Figure Ib**) (Bennion *et al.*, 1997; Podmore, 2002; Bent, 2007).

Bent (2007) described three industrial methods of building the layered structure of pastry dough. These are the "sandwich", the "co-extrusion" and the "curling" methods. All three are based on extrusion of a fat layer on top of a single or between two predough layers.

After folding the roll-in fat into predough, multiple layers of predough and fat are created by alternately sheeting/laminating (i.e. reducing the dough thickness by sheeting) and folding the layered dough (Cauvain and Young, 2009) (**Figure II**). Croissant and Danish pastry dough is typically laminated until 16 to 50 (theoretical) fat layers are obtained. The number of fat layers obtained for puff pastry is typically higher (130-250 theoretical fat layers) (Bent, 2007; Cauvain and Young, 2009). The reason for this lies in the absence of yeast in puff pastry (see Section 3.3).

The number of fat layers affects product volume and crumb structure (Bent, 2007). Although a low number of layers can yield pastry with large specific heights, it can also result in irregular crumb structure with large voids. In contrast, a large number of relatively thin layers can lead to interconnections between different dough layers and result in loss of dough lift (Sievert *et al.*, 2007). Bousquieres *et al.* (2014) recently demonstrated that a higher number of layers increases their thickness homogeneity, but also induces more fragmentation of the fat layers. Based on the above, it seems that an optimal number of layers can be defined when targeting certain crumb properties and the largest specific volume. According to Bennion *et al.* (1997) and Cauvain and Young (2009), increasing the number of laminations increases dough lift to reach a maximum before it again decreases. Baking laminated puff pastry dough with less than 162 (calculated) fat layers results in irregular dough lift and produces puff pastry with poor crumb structure (Bennion *et al.* (1997).

The temperature during processing is strictly controlled, as it affects the integrity of the fat layers: (too) low temperatures make the fat brittle, whereas (too) high temperatures result in melting (portions) of the fat (Cauvain and Young, 2009). To make sure that roll-in fat does not

melt during processing, it is common to introduce resting phases at lower temperature between the consecutive sheeting steps. This also allows relaxation of the developed gluten network and avoids dough shrinkage during baking (Bennion *et al.*, 1997; Sievert *et al.*, 2007).

2.3 Further processing of multilayered dough: fermentation and/or baking

The multilayered dough is typically rolled and cut into dough pieces from which individual products are formed (Calvel, 1990; Sievert *et al.*, 2007).

For fermented pastry, the individual multilayered dough pieces are then fermented, during which yeast activity increases the pastry dough volume (see Section 3.3). After fermentation, the multilayered dough is baked. During baking, water is converted to steam which is trapped in the layered structure, increases the pressure underneath each dough layer, and ultimately expands the dough. This process is referred to as "pastry lift" or "dough lift" (**Figure III**) (Kazier and Dyer, 1995; Sievert *et al.*, 2007; Cauvain and Young, 2009). Dough lift of puff pastry is sometimes called "puffing" (Sievert *et al.*, 2007). Although puff pastry contains no added leavening agent(s), it can rise up to eight times its original thickness (Lai and Lin, 2006). To ensure adequate lifting, the integrity of the layers should be as complete as possible as interconnections between different dough layers allow steam to escape (Bennion *et al.*, 1997; Sievert *et al.*, 2007). Baking of laminated dough yields the characteristic, flaky texture of croissant, Danish and puff pastry (Kazier and Dyer, 1995).

In an alternative puff and fermented pastry production process, known as the Scotch, Dutch or Blitz method, no distinction is made between in-dough and roll-in fat. The entire fat fraction is added with the other ingredients and mixed in gently, ensuring that large distinct lumps of fat are still present throughout the dough. Although this method is quicker, the resulting pastry dough

does not always rise evenly, because it does not contain continuous sheets of fat (Bennion *et al.*, 1997; Podmore, 2002; O'Brien, 2004; Bent, 2007).

3. INGREDIENT FUNCTIONALITY IN PASTRY PRODUCTION

3.1 Predough making

Pastry predough mixing strongly resembles that of bread dough. Both produce homogeneous dough. The transitions of flour, water, salt, sugar, fat and, where relevant, yeast in pastry predough can at least to a certain extent be compared to those in bread dough. However, different relative levels of the ingredients result in differences in their functionality.

3.1.1 Flour

Quantitatively, (wheat) flour is the most important ingredient in pastry dough (**Table I**). Wheat flour for pastry production typically contains *ca.* 11.5 to 14.5% protein (Hay, 1993; Bent, 2007; Sievert *et al.*, 2007). However, protein levels as low as 8-10% have also been reported (Hay, 1993).

Proteins

The properties and structure of wheat flour proteins have been described extensively elsewhere (Shewry and Tatham, 1997; Belton, 1999; Gianibelli *et al.*, 2001; Wieser, 2007). They have been divided into gluten and non-gluten proteins based on their function in dough making. The importance of non-gluten proteins in dough formation remains largely unknown. In general, the gluten proteins are accepted to be determinants of the bread making potential of wheat flour, while the non-gluten proteins are thought to exert only a negligible effect (Veraverbeke and

Delcour, 2002). Non-gluten proteins mainly consist of monomeric proteins that either are metabolic (*e.g.* enzymes) or structural proteins (Veraverbeke and Delcour, 2002). Triticins, a group of polymeric wheat storage proteins, are also included in the group of non-gluten proteins (Veraverbeke and Delcour, 2002). Gluten proteins consist of monomeric gliadins and polymeric glutenin proteins (Shewry *et al.*, 1992; Wieser, 2007). Gliadin proteins form a heterogeneous mixture of α-, γ- and ω-gliadin proteins (Lagrain *et al.*, 2008). The α- and γ- gliadins contain cysteine residues that form intramolecular disulfide (SS) bonds (3 and 4 in α- and γ-gliadins, respectively). In contrast, ω-gliadins lacks cysteine residues (Shewry and Tatham, 1997; Lagrain *et al.*, 2008). Glutenin polymers are made up of high and low molecular weight glutenin subunits (Delcour *et al.*, 2012) that form both intra- and intermolecular SS bonds (Lagrain *et al.*, 2007; Wieser, 2007).

Gluten proteins strongly influence both the water absorption and the viscoelastic properties of wheat flour bread dough (Attenburrow *et al.*, 1990; Wieser, 2007) and, based on the similarities mentioned earlier, also pastry predough. This is related to their ability to form a network upon hydration and mixing (Scanlon and Zghal, 2001).

Belton (2005) subdivided dough formation in two stages: a hydration stage followed by energy input through deformation (kneading). In the hydration stage, gluten proteins can absorb up to twice their own weight of water (Manley *et al.*, 2011). In the second stage, depolymerisation and (re)polymerisation reactions take place during mixing (Weegels *et al.*, 1997; Belton, 2005). This eventually results in a viscoelastic gluten network (Scanlon and Zghal, 2001; Belton, 2005; Joye *et al.*, 2009). Differences in the structures of glutenin and gliadin proteins provide them with different functionalities during dough formation. Whereas glutenin proteins build up the

polymeric protein network that provides cohesiveness and elasticity to dough (Tatham and Shewry, 1985; Veraverbeke and Delcour, 2002), (globular) gliadin proteins do not participate in network formation (Belton, 1999; Goesaert *et al.*, 2009). They act as plasticizers of the glutenin network, thereby contributing to dough viscosity and extensibility (Cornec *et al.*, 1994; Veraverbeke and Delcour, 2002; Kovacs *et al.*, 2004; Wieser, 2007).

Compared to optimally mixed bread dough, pastry predough is usually "underdeveloped" (Cauvain and Young, 2009), since mixing is stopped as soon as homogeneous dough has been formed (O'Brien, 2004). This allows for further dough development during lamination. During this step, the pressure exerted by the sheeting rolls transfers additional energy to dough (O'Brien, 2004; Cauvain and Young, 2009). However, to our knowledge, no publically available data describe the extent to which predough should be mixed or further dough development occurs. It is also unclear whether predough mixing disrupts all flour particles. Moreover, the use of chilled water during predough making (see Section 3.1.2) influences gluten development, as mixing temperature impacts dough hydration time, consistency, and the amount of mixing energy (Farahnaky and Hill, 2007; Huang et al., 2008). It would therefore be of interest to study the effect of water temperature on gluten development during mixing as well as during the further production stages. This would possibly allow defining an "optimal degree of underdevelopment" at the predough stage. In the interest of obtaining optimally developed dough after the final laminating step, we reasonably assume that the use of flour that is (more) tolerant to overmixing is advantageous.

Starch

With a level of about 70 to 75%, starch is wheat flour's main constituent (Goesaert *et al.*, 2005). It occurs in the form of semi-crystalline granules that are part of the flour particles. Starch granules essentially consist of amylose and amylopectin (Tester *et al.*, 2004). Amylose consists of about 500 to 6,000 D-glucopyranosyl units which are mainly linked through $\alpha(1\rightarrow 4)$ linkages, and contains a small portion of $\alpha(1\rightarrow 6)$ interlinkages. Although amylose molecules are thus not completely linear, their behavior approximates that of linear polymers (Karim *et al.*, 2000). Amylopectin is much larger (3×10^5 to 3×10^6 D-glucopyranosyl units) and highly branched as a result of $\alpha(1\rightarrow 6)$ -linkages in its structure (Buléon *et al.*, 1998; Vandeputte and Delcour, 2004; Goesaert *et al.*, 2005). Starch composition, physical and chemical properties have been extensively studied (French, 1973; Buléon *et al.*, 1998; Hug-Iten *et al.*, 1999; Vandeputte and Delcour, 2004) and reviewed (Tester *et al.*, 2004; Goesaert *et al.*, 2005; Delcour and Hoseney, 2010) elsewhere.

Predough viscosity is mainly determined by its gluten proteins (Letang *et al.*, 1999). Still, at room temperature, intact starch granules can absorb water (up to 30% of their own dry weight in excess water), which causes them to swell slightly (Goesaert *et al.*, 2005). According to Hug-Iten *et al.* (1999), this swelling affects dough consistency. Whatever be the case, in dough starch is mainly present in the form of slightly swollen granules which fill up the spaces in the viscoelastic gluten network (Scanlon and Zghal, 2001; Goesaert *et al.*, 2005).

Milling of wheat into flour damages some of the starch granules. Damaged starch can absorb up to its own weight in water (*i.e.* about three times as much as undamaged starch) (Manley *et al.*, 2011). Higher levels of damaged starch increase the water absorption capacity of dough (Dexter *et al.*, 1994; Miller and Hoseney, 1997; Martinez *et al.*, 2013). Hence, when using flour with

higher levels of damaged starch, more water needs to be added to puff pastry dough to obtain desirable dough development and consistency than when using flour having lower levels of such starch (Sliwinski *et al.*, (2004). Of further note is that Miller and Hoseney (1997) stated that starch damage is accompanied by release of dextrins which (partly) dissolve in the dough aqueous phase and increase its viscosity.

3.1.2 Water

Dough mechanical behavior strongly depends on its water content (Belton, 2005; Farahnaky and Hill, 2007; Koksel and Scanlon, 2012). Less water is added in pastry predough than in bread dough. While the roll-in bakery fat also contains water, this is only released when the fat melts during baking. Water distribution in pastry dough thus differs strongly from that in bread dough. This specific water distribution in pastry may be essential to dough lift during baking (see Section 3.4.3).

The temperature of the water used controls the predough temperature. As indicated earlier, the temperature of the mixed predough is to be comparable to that of the roll-in fat to facilitate processing. To that end, cold water or an ice-water mixture is used (Cauvain and Young, 2009).

3.1.3 In-dough fat

During production of laminated dough, bakery fat is added at two distinctive moments: in-dough fat is added during predough mixing, whereas roll-in fat is used as the laminating medium (Lai and Lin, 2006). In industrial puff and fermented pastry production, in-dough fat is commonly added as "pastry trimmings" recovered from trimming the multilayered dough sheets during processing. Typically, roll-in fat in pastry trimmings of a particular production run serves as the in-dough fat in the next. Pastry trimmings can either be added directly to the mixer or fed back

during the sheeting stages. To avoid unwanted product variation, their life time, temperature and level of addition should be strictly controlled (Bennion *et al.*, 1997; Cauvain and Young, 2009). Higher levels of dough fat soften puff pastry dough, but also lower dough lift during baking (Cauvain and Young, (2006). Different other effects have been attributed to the fat incorporated in predough. These include lubrication (O'Brien, 2004; Lai and Lin, 2006) and even improving dough elasticity (Bent, 2007). However, as far as we can judge, little if any analytical data support the latter statements. Moreover, lubrication may interfere with the formation of the gluten network (Cauvain and Young, 2006) and thus reduce dough elasticity. This effect has also been reported for sugar-snap cookie dough (Pareyt *et al.*, 2008).

3.2 Pastry dough lamination process

3.2.1 Flour

Proteins

Because of the layered character of laminated dough, each fat layer interrupts the gluten network (Baardseth *et al.*, 1995). Hence, the gluten network is not continuous over the entire dough as is the case in bread dough, but the gluten proteins in laminated dough are rather present as thin gluten films between the dough layers and do not form a three-dimensional structure (Baardseth *et al.*, 1995). In our opinion, because of the resemblance of predough and bread dough, it seems more logical to present the gluten network in laminated dough as consisting of separate, three-dimensional gluten networks within each dough layer, and preferentially oriented in the (final) sheeting direction. Therefore, in what follows, the term "layered gluten network" will be used rather than the term "gluten film".

According to Hay (1993), the formation of thin, well defined layers in laminated dough significantly influences dough lift. In the view of Bent (2007), flour with a protein content of 12 to 13.5% allows obtaining desired dough properties in fermented laminated dough. According to Sievert *et al.* (2007), while flour with a protein level exceeding 13.0% can tolerate more lamination, it will not necessarily result in better pastry.

3.2.2 Roll-in fat

Roll-in fat helps creating the flaky layers of pastry products and provides them with tenderness and flavor (Lai and Lin, 2006). Its most important function is to form and maintain a barrier between the different dough layers during subsequent sheeting and folding (Baardseth et al., 1995). Otherwise, interconnections between different dough layers, and the resulting too high dough strength, would allow water vapor to escape to the outside through micropores and capillaries during baking (Bennion et al., 1997; Sievert et al., 2007) and, thereby, decrease dough lift. Roll-in fat can be butter, margarine or shortening (Kazier and Dyer, 1995). Butter and margarine are water-in-oil emulsions. They contain (stabilized) water droplets dispersed in an oily medium (McClements, 2010) that also contains solid fat, whereas shortening consists of 100% triacylglycerols (Kazier and Dyer, 1995). Although butter has high consumer acceptance (Gassenmeier and Schieberle, 1994), its low melting point [typically 32°C for a roll-in butter (Lai and Lin, 2006)] can cause it to partly oil out during pastry dough production. Therefore, butter is hard to use in processing, as problems can arise during sheeting and fermentation and the integrity of the layers can be lost (Cauvain and Young, 2009) if no adequate temperature control is in place. Pastry margarines are commonly used as roll-in fat (Kazier and Dyer, 1995) because they facilitate dough handling (Gassenmeier and Schieberle, 1994). They should meet

some specific requirements (Simovic *et al.*, 2009; Pajin *et al.*, 2011). One of them is their melting range. Natural fats, as complex mixtures of different compounds, gradually soften upon heating (i.e. a melting range) rather than displaying a true melting point (O' Brien, 2004). Several melting point determination procedures have been standardized for fats and oils, such as those determining "softening", "Mettler dropping" or "slipping" points, "capillary (tube)", "Ubbelobde", and "Wiley" melting points, and others (Deman *et al.*, 1983; O' Brien, 2004). The outcome of each of these strongly depends both on the method itself as well as on the applied conditions (Deman *et al.*, 1983). Bennion *et al.* (1997) stated that roll-in fats usually have a slipping melting point of at least 40°C. Bockish (1998), without specifying the method used, claimed that the melting point of a roll-in fat should be considerably higher than that of bakery margarine and range between 40°C and 44°C. O'Brien (2004) added that, to prevent "oiling-out" of margarine before baking, its melting point must be at least 3°C higher than the fermentation temperature and that a highest melting point of at least 45°C is necessary for puff pastry roll-in margarine.

A good specification for roll-in margarine requirements seems to be the solid fat content (i.e. the proportion of solid to liquid fat) at a given temperature (O' Brien, 2004). The most important features of a roll-in fat are its plasticity and firmness (O'Brien, 2004) at the process temperature. These properties are determined by the solid fat content (Lefebure *et al.*, 2013). According to Cauvain and Young (2006), a larger proportion of solid fat at a given temperature goes hand in hand with larger pastry lift. Sievert et al. (2007) noted that roll-in margarines with high solid to liquid ratios that show little changes over a broad temperature range are needed in puff pastry production. Still, roll-in fat should have a minimal plasticity to be spread or rolled out in the

(layered) dough so that the formed roll-in fat layers do not break during repeated sheeting and folding operations (O'Brien, 2004). The plasticity of the fat should therefore be comparable to that of the dough. When the fat is more firm than the dough (or the dough is too soft), the dough can rupture during machining (O' Brien, 2004; Lai and Lin, 2006). In contrast, when the fat is (too) soft, it does not withstand mechanical stress of the sheeting process and can migrate into the dough (Lai and Lin, 2006). When this occurs, the resulting baked products are typically small and have a poor crumb structure (Bent, 2007). According to Sievert et al. (2007), migration of fat into the dough layers interferes with the (further) development of the gluten network in these layers during sheeting. However, since it is unclear to what extent gluten should be developed during predough making and to what extent it further develops during sheeting and folding, one could also argue that the effect of fat migration in the dough layers might be related more to it no longer being present as a barrier between the layers than to its interference with (further) gluten development. Apart from the above, both storage and processing temperatures of bakery fat are critical (Bent, 2007; Cauvain and Young, 2009) because they both affect its solid fat content (Simovic et al., 2009; Manley et al., 2011). Ideally, fat should be kept at a temperature which is both constant and similar to that of the processing environment. Evidently, the process temperature must also be kept constant (Cauvain and Young, 2009).

Vicario *et al.* (2003) characterized the fatty acid (FA) profile of different bakery products. They found high levels of saturated FA (mainly palmitic acid) and mono-unsaturated FA (mainly oleic acid) and low levels of polyunsaturated FA (linoleic and linolenic acids) in croissants.

Moreover, pastry products can contain trans-FA, which have been linked to cardio-vascular diseases (Vicario *et al.*, 2003; Simovic *et al.*, 2009). Therefore, much research has been

conducted towards developing alternative roll-in fats with a lower (or no-) trans- and lower saturated FA content (Cavillot *et al.*, 2009; Simovic *et al.*, 2009; de Cindio and Lupi, 2011; Pajin *et al.*, 2011; Garcia-Macias *et al.*, 2012) or a lower fat content in general (Kazier and Dyer, 1995; Gabriele *et al.*, 2008; Garcia-Macias *et al.*, 2011). Today, partly as a result of increasing consumer awareness, trans-FA are typically banned.

3.3 Pastry fermentation

The recipe for predough for Danish pastry and croissant products contains yeast (*Saccharomyces cerevisiae*). During mixing, yeast consumes oxygen available to it in dough by respiration. Once all oxygen has been consumed, yeast switches to anaerobic fermentation (Rollini *et al.*, 2007). It then converts glucose, sucrose, and maltose into mainly ethanol and carbon dioxide (CO₂) (Attfield, 1997). Before the CO₂ produced can leaven dough, the dough's aqueous phase must become saturated with CO₂ (Delcour and Hoseney, 2010). Then, CO₂ diffuses to preexisting gas cells (nuclei) (Shah *et al.*, 1998; Scanlon and Zghal, 2001). Indeed, yeast action does not form new gas cells (Gan *et al.*, 1995; Gandikota and MacRitchie, 2005) because the pressure required for a single CO₂ molecule to create a new gas bubble in a system where the interfacial tension does not change, would be infinite (Delcour and Hoseney, 2010). In bread production, the nuclei in dough originate from air entrapped during mixing (Shah *et al.*, 1998; Gandikota and MacRitchie, 2005).

For pastry production, we reasonably assume that air is not only incorporated during predough mixing and that it is also entrapped between the different layers during the folding stages. During proofing of (straight dough) bread, sheeting, punching and molding operations are carried out to redistribute gas cells to optimize their number and size distribution (Scanlon and Zghal, 2001;

Gandikota and MacRitchie, 2005). In contrast, during fermentation of pastry dough, no sheeting, punching nor molding is executed, and gas cells most likely already (re)distribute during lamination.

During fermentation, croissant and Danish pastry dough volume nearly doubles (Baking Industry Research Trust, 2014). Fermentation is commonly at lower temperatures than for bread dough, although similar fermentation temperatures (*i.e.* 30-32°C) can be applied when combined with a (lower) relative humidity of 70-80% (Cauvain and Young, 2009).

Yeast activity is said to negatively impact dough lift as the produced CO₂ can destroy the integrity of the layered structures, which then allows for too early escape of steam during baking and reduces the lift (Cauvain and Young, 2006). The balance between expansion from yeast during fermentation and from steam production during dough lift significantly affects the flaky texture of the end product (Cauvain and Young, 2006). To ensure adequate dough lifting despite the negative impact of yeast, less layers are created during croissant and Danish than during puff pastry production. This ensures layer integrity (Cauvain and Young, 2009).

3.4 Pastry baking

3.4.1 Flour

Proteins

During baking, the transient gluten network is transformed into a permanent network (Goesaert et al., 2009). Heat-induced sulfhydryl/SS interchange reactions induce formation of intermolecular SS bonds between glutenin molecules, and, at higher temperatures, also between gliadin and glutenin. Indeed, at such temperatures, some gliadin is incorporated into the polymeric glutenin network structure (Lagrain et al., 2005; Lagrain et al., 2008; Delcour and

Hoseney, 2010). During bread baking, with proceeding polymerization, the gluten network becomes more rigid and strongly influences the crumb texture (Scanlon and Zghal, 2001). In addition, starch gelatinization can actively draw water from the amorphous gluten fraction, which increases the rigidity and decreases the flexibility of the gluten network (Delcour and Hoseney, 2010).

While Hay (1993) found a positive relation between flour protein content and the height and specific volume of puff pastry, to the best of our knowledge, there is no literature discussing the structural changes of gluten proteins during baking for pastry products. We assume that within the layers, gluten polymerization occurs more or less in the way it occurs during bread baking. However, one must take into account that the layers during pastry baking are stretched extensively because of the large macroscopic deformation during dough lift. Also, the (molten) fat phase possibly interferes with gluten polymerization (see also Section 3.4.2). However, whether and to what extent both phenomena effectively impact gluten network formation during baking of pastry dough and affect the quality of the baked product is unclear.

Starch

As stated above (see Section 3.1.1), starch granules absorb some water at room temperature and thereby swell to a limited extent. As long as a certain temperature, known as the gelatinization temperature (T_m), is not exceeded, this granule swelling is reversible (French, 1973; Goesaert *et al.*, 2005). However, heating of wheat starch in the presence of sufficient water to temperatures exceeding T_m induces an irreversible loss of the molecular order inside the starch granules due to melting of the amylopectin crystallites (French, 1973; Goesaert *et al.*, 2005). In dough, starch gelatinizes over a wide temperature range (Delcour and Hoseney, 2010). T_m strongly depends on

the dough formula [e.g. its water content, and the presence of anti-plasticizing components (Slade and Levine, 1991)] and ranges from ca. 55°C for a flour-water dough, as deduced from changes in its rheology (Dreese et al.(1988) to 67°C for a flour-water dough with 2.0% sodium chloride (NaCl), 6.0% sucrose and 3.0% shortening (Ghiasi et al., 1983). In pastry dough, T_m is expected to be higher because of the lower water content and the presence of sugar (Slade and Levine, 1991; Weegels et al., 1997). Sodium chloride, in concentrations comparable to those in bread and pastry, also increases T_m (Wootton and Bamunuarachchi, 1980; Chiotelli et al., 2002). During bread baking, according to Hug-Iten et al. (1999; 2003), amylose leaches out of the starch granules, although the granular structure is largely retained due to the limited amount of water present (Hug-Iten et al., 1999; Hug-Iten et al., 2003). Whether and to what extent granular structure is distorted and amylose leaching occurs in pastry baking is not clear either. However, retention of the granular structure might be even more pronounced in pastry for reasons outlined above.

3.4.2 Roll-in fat

During baking, with increasing temperature, the roll-in fat gradually melts (Kazier and Dyer, 1995). Part of the molten fat phase can migrate into the dough matrix (Sievert et al., 2007). Here, the fat possibly interferes with gluten polymerization, as has been described for sugar-snap cookies (Pareyt et al., 2010). Indeed, according to Cauvain and Young (2006), in puff pastry the oil fraction of bakery fat can lubricate the gluten proteins and limit their water uptake. The authors proposed that solid fat crystals physically hinder protein cross-linking. However, one may well argue that oil exhibits similar effects.

3.4.3 Water

During pastry baking, water is converted to steam which creates leavening (Kazier and Dyer, 1995; Sievert *et al.*, 2007; Cauvain and Young, 2009). It is generally accepted that steam is trapped in the layered structure and lifts the dough layers (Sternhagen and Hoseney, 1994; Cauvain and Young, 2009). The water for steam production can be present in the dough layers but may also be released from the roll-in fat. Indeed, when molten, the continuous fat phase no longer stabilizes the water droplets which are then released from the emulsion (Borwankar *et al.*, 1992). However, according to O'Brien (2004), the use of shortenings in puff pastry production does not yield pastry products inferior to those prepared with roll-in fats containing water. Based on this statement, it seems that not the water released from the margarine is responsible for dough lift, but rather that present in the dough layers. However, the above as well as the extent to which water released from roll-in fat contributes to dough lift is unclear and deserves more indepth study because of its importance for product volume.

Most authors do not (further) elaborate on the mechanism of steam entrapment and those that do, are of different opinion. According to some authors (Kazier and Dyer, 1995; Podmore, 2002), the (hydrophobic) fat layers form layers "impervious" to the produced steam. In their view, the steam remains inside each dough layer, forcing it to expand because of the pressure it develops underneath each fat layer. However, significant steam production only starts at higher temperatures, when all roll-in fat is already in a molten state. However, according to Sievert *et al.* (2007) steam from the dough layers of puff pastry dough migrates into the oil layers (since all fat is molten at that point), where it inflates gas bubbles. In their opinion, steam migrates into the oil phase because of the smaller pressure differential needed to inflate a bubble of steam in liquid

fat than in the dough phase. The steam migration then increases pressure between the dough layers and subsequently causes dough to lift. Because of the resemblance between pastry dough layers and bread dough, which also expands during baking, we here reason that pastry lift may well be the result of both phenomena, with the gas cells in dough inflating the dough layers and those present between dough and fat layers acting between the layers and/or as nuclei for the gas released when the dough layers become continuous due to gas cell opening (**Figure IV**). Moreover, the relative importance of both mechanisms might differ between puff pastry and fermented pastry, as layer thickness, homogeneity and integrity, which depend on degree of lamination (Bousquieres *et al.*, 2014), also affect the gas retention capacity of the dough layers. During baking of puff pastry, only about half of the water vapor contributes to dough lift. The other half escapes through micropores and capillaries of interconnected dough layers after the structure has been set (Sievert *et al.*, 2007).

3.5 Pastry cooling and storage

During cooling and storage of bakery goods, the product properties evolve. However, pastry products are generally not stored for long periods. Moreover, there is an ongoing trend toward using frozen pastry products which are baked and consumed soon thereafter.

3.5.1 Flour

Proteins

Whether and how wheat flour proteins contribute to quality loss of bakery products is not well understood. In an early interpretation, Willhoft (1973) and Kim and D'Appolonia (1977) suggested that gluten proteins serve as anti-firming agent by diluting the starch fraction. However, others (Martin *et al.*, 1991; Gerrard *et al.*, 1997) suggested they contribute to bread

firming by cross-linking with starch granules through hydrogen bonds. Recent views describe gluten proteins as contributing to bread crumb firming due to loss of plasticizing water, which reduces the flexibility of the gluten network (Goesaert *et al.*, 2009). In bread, water migration from crumb to crust as well as redistribution of moisture between gluten and starch can decrease the moisture content of the gluten fraction below the critical point where it no longer is (fully) plasticized, causing an increased stiffness of the gluten network (see Section 3.5.2) (Bosmans *et al.*, 2013). However, whether such water migration occurs to the same extent in pastry products is unclear.

Starch

The role of starch during storage of bakery products has been extensively studied, with a major focus on bread firming (Willhoft, 1973; Kim and D'appolonia, 1977; Pisesookbunterng *et al.*, 1983; Martin *et al.*, 1991; Hug-Iten *et al.*, 1999; Hug-Iten *et al.*, 2003; Bosmans *et al.*, 2013). Amylose and amylopectin clearly have different roles in the firmness and firming of bakery products. In bread, (leached) amylose forms a continuous, partially crystalline network immediately after baking, which incorporates the swollen and deformed granule remnants (Zobel and Kulp, 1996; Goesaert *et al.*, 2009). This permanent network and the gluten network are responsible for the (desirable) initial crumb texture (Goesaert *et al.*, 2005). However, as stated above, the extent to which starch granules rupture and amylose leaching (if any) occurs during pastry baking is not clear. Amylopectin retrogradation, i.e. the transformation of amorphous amylopectin into a more ordered (partial) crystalline structure, is slower than amylose crystallization and contributes to (undesirable) firming of bread crumb during storage over several days to weeks (Willhoft, 1971; Pisesookbunterng *et al.*, 1983; Biliaderis, 1992).

Moreover, the formation of crystalline amylopectin requires incorporation of water into the crystal structure. This water originates from the gluten fraction (Willhoft, 1971; Goesaert *et al.*, 2009) and/or from the amorphous starch fraction (Bosmans *et al.*, 2013) which then become less plasticized.

Sternhagen and Hoseney (1994) showed an increase in the amount of retrograded amylopectin during storage of Danish pastry but emphasized that amylopectin retrogradation is not the only mechanism of pastry firming. While the additional mechanisms remain unclear, they presumably include changes in the gluten fraction and moisture redistribution.

3.5.2 Water

In bread and presumably also in pastry products, water redistribution occurs during storage. In fresh bread, a moisture gradient from the inside (high moisture content) to the outside (low moisture content) is present throughout the product as a result of (an opposite directed) heat transfer from the oven to the baking product. The moisture gradient induces moisture migration from crumb to crust (Davidou *et al.*, 1996; Baik and Chinachoti, 2000; Gray and BeMiller, 2003). Water from the amorphous starch and gluten is lost. The latter then become less plasticized and more rigid (Davidou *et al.*, 1996; Gray and BeMiller, 2003). Crumb hardness increases whereas the crust becomes softer (leathery) (Piazza and Masi, 1995). Apart from migration of water from crumb to crust, migration from crust to the environment occurs at the same time (Gray and BeMiller, 2003).

Water also redistributes between gluten and starch in the product. For bread, starch retrogradation actively draws moisture from the amorphous gluten network (Willhoft, 1971; Gray and BeMiller, 2003; Goesaert *et al.*, 2009) and possibly also from the amorphous starch

fraction (Bosmans *et al.*, 2013) to incorporate it in its crystal structure. As stated by Tolstoguzov (1997) and recently confirmed in ¹H-NMR studies by Bosmans *et al.* (2013), water migration from gluten to starch in bread may also originate from diffusion, due to the thermodynamic immiscibility of gluten and starch.

For pastry products, both crumb to crust moisture migration and water redistribution between gluten and starch during product storage possibly play an important role. However, to the best of our knowledge, there is literature data neither on initial moisture distribution nor on the presence of moisture gradients in pastry products immediately after baking. Nevertheless, we assume water migration from crumb to crust to a lesser extent than in bread, as the moisture gradient between the so-called crumb and crust is less pronounced for pastry products. Furthermore, the higher fat content of pastry than of bread might impact moisture migration as well, as increasing fat levels decrease moisture diffusion in e.g. sponge cake (Roca et al., 2006). However, Roca et al. (2006) also demonstrated that increased porosities go hand in hand with increased moisture diffusivity in such cake. The presence of large pores in the baked pastry product may well influence moisture migration from product to the "environment" (i.e. to the pores). This may explain why pastry products typically become harder faster during storage than does bread. Based on the findings of Sternhagen and Hoseney (1994), who reported that amylopectin retrogrades during storage of Danish pastry, one could expect molecular scale moisture migration to occur as well, since both phenomena are associated, as outlined above (see Section 3.5.1).

3.5.3 In-dough fat

According to Lai and Lin (2006), in-dough fat improves the eating quality of the baked product. Sternhagen and Hoseney (1994) evaluated the texture of crumb of Danish pastry prepared with different amounts of in-dough fat (7, 10 and 13% on flour weight basis) and reported reduced crumb hardness immediately after baking with increasing amounts of in-dough fat.

3.5.4 Roll-in fat

Sternhagen and Hoseney (1994) studied the shelf life of Danish pastry containing roll-in fat levels of 15% to 25% (of dough weight), but found no significant relation between the level of roll-in fat and shelf life. Baardseth *et al.* (1995) demonstrated that initial crumb of Danish pastry is increasingly softer with increasing levels of roll-in fat, but did not study the increase in crumb hardness (i.e. crumb firming) during pastry storage.

4. IMPROVING AGENTS IN PASTRY PRODUCTION

A range of improving agents are commonly used in the baking industry. They allow better control of the production process, improve product quality and/or increase shelf-life. We here focus on those improving agents that have been mentioned in the peer reviewed literature on pastry production.

4.1 α-amylases

The most frequently used enzymes impacting starch functionality are α -amylases. They hydrolyze α -1,4 linkages. Different effects of α -amylase addition to bread dough have been described. However, amylases from different sources hydrolyze starch in a different manner (Bijttebier *et al.*, 2007) and the perceived effects will thus depend on the action pattern of the amylase used. First, α -amylase addition may result in the production of shorter chain dextrins

(Rosell *et al.*, 2001), thus increasing the level of fermentable sugars (promoting yeast fermentation) and reducing sugars (promoting Maillard reactions, thus intensifying bread crust flavor and color) in bread dough (Goesaert *et al.*, 2009) and most likely also in pastry. α-Amylase activity may also reduce bread dough viscosity during baking (Weipert, 1990), resulting in a prolonged oven rise and an increased bread loaf volume (Goesaert *et al.*, 2009). Finally, certain amylases can function as anti-firming agents, delaying bread crumb firming (Blaszczak *et al.*, 2004; Goesaert *et al.*, 2009).

The effects of α -amylases in pastry might however differ from those in bread. Sternhagen and Hoseney (1994) reported increased crumb hardness when adding a bacterial α -amylase known to reduce bread crumb firmness to a Danish pastry dough recipe. They attributed this dissimilarity to differences in ingredient functionality and hypothesized that the roll-in fat masks the effects of the α -amylase. However, whether and how other ingredients influence α -amylase activity was not further examined.

4.2 Transglutaminase

Many enzymes or redox agents can affect protein network formation. Their impact during bread making has been studied extensively (Bollecker *et al.*, 2000; Angioloni and Rosa, 2007; Caballero *et al.*, 2007; Decamps *et al.*, 2014) and reviewed (Joye *et al.*, 2009) elsewhere. In contrast, how such additives affect pastry production has not been studied thoroughly and, much as for starch modifying additives, their effects on pastry might differ from that in bread production.

Transglutaminase (TG, EC2.3.2.13) is (one of) the most studied enzymes in pastry applications. It catalyzes acyl-transfer reactions, thereby introducing covalent isopeptide crosslinks in proteins

between glutamine residues (acyl donors) and various primary amines (acyl acceptors) including amino groups of lysine residues (Gerrard et al., 2001; Hozova et al., 2002; Bauer et al., 2003). Gerrard et al. (2000) showed that inclusion of TG in a puff pastry dough recipe substantially increases pastry lift. The authors observed increased pastry height and volume (up to 30%) upon TG addition. These observations were confirmed by Kuraishi et al. (2001) and explained in terms of gluten crosslinks formed and, hence, by the impact of TG on the gluten network structure and viscoelastic properties (Bauer et al., 2003). That way, the positive effect on pastry lift has been attributed to better preservation of dough layer integrity by the stronger gluten network obtained when using TG. A stronger gluten network thus seems to result in more pronounced pastry lift (Gerrard et al., 2000; Gerrard et al., 2001). Hozova et al. (2002) also reported a positive effect of TG inter alia on sensory characteristics of pastry.

4.3 Glucose oxidase

Another enzyme tested for its suitability in pastry production is glucose oxidase (GO) (Rasiah *et al.*, 2005). In the presence of O₂, GO catalyzes the oxidation of C1 of β-D-glucose, a reaction in which H₂O₂ and _D-glucono-δ-lactone are produced. D-glucono-δ-lactone spontaneously hydrolyzes to gluconic acid, whereas the formed H₂O₂ may indirectly oxidise the thiol groups of two cysteine residues, forming SS bonds and thus crosslinking the gluten proteins (Miller and Hoseney, 1999; Joye *et al.*, 2009; Decamps *et al.*, 2012). However, the occurrence of additional (side-) mechanisms such as crosslinking of arabinoxylan has been suggested (Izydorczyk and Biliaderis, 1995; Primo-Martin *et al.*, 2005). Rasiah *et al.* (2005) added GO to a croissant dough recipe, but observed that doing so did not increase the volume of the final product. They stated that the enzyme cannot introduce SS crosslinks in the glutenin fraction and that, therefore, its

effect is rather limited. In contrast, GO introduced a low number of non-SS crosslinks, possibly dityrosine linkages, in the glutenin fraction, as well as SS and non-SS bonds in the albumin and globulin fractions. A slightly positive influence on pastry crumb was reported (moister, thicker, larger gas cells) (Rasiah *et al.*, 2005).

However, as far as we are aware, the use of GO in pastry applications holds risks in terms of the formation of off flavour components. More work is needed in this area, and should confirm or nullify the hypothesis that the off flavours formed result from the interplay between GO action and (part of) the lipid population.

4.4 Surfactants

Surfactants or surface-active agents contain both a hydrophilic and hydrophobic part and are thus amphiphilic molecules (Pareyt et al., 2011). They can decrease interfacial tension between two initially immiscible phases (liquid, solid or gas) by positioning themselves at the interface (Belitz et al., 2009). Sternhagen and Hoseney (1994) examined the effect of the surfactants sodium stearoyl lactylate (SSL) and monoacylglycerol (MAG) during storage of Danish pastry. SSL is used in bread making as a dough strengthener, whereas MAG is used as a crumb softener (Huschka et al., 2011; Pareyt et al., 2011). In addition, SSL, like MAG, also forms amylose inclusion complexes (Van Steertegem *et al.*, 2013). However, neither SSL nor MAG slow down the firming (rate) of Danish pastry. SSL even increases crumb hardness (Sternhagen and Hoseney, 1994). This again shows that classical bread improving substances can have different effects when added during pastry production.

5. CONCLUSION AND RESEARCH NEEDS

²⁹ ACCEPTED MANUSCRIPT

Flour (constituents), fat, water, and in the case of fermented pastry also yeast strongly influence the pastry making process as well as the quality of the baked pastry product. They undergo a series of complex biochemical and physical transformations during pastry production which lie at the basis of the specific product characteristics related to satisfactory product quality. Much as in bread making, the wheat gluten protein fraction has a prominent role in product quality. Differences in its functionality between bread and pastry making originate from the lamination process and the resulting (intermediate) product heterogeneity, which prevents the formation of a continuous gluten network throughout the entire product, but rather yields different non-connected continuous gluten layers at the dough stage. During baking, at least part of this layered structure is lost when the roll-in fat melts.

Apart from flour constituents, roll-in fat plays multiple roles in pastry production. It (i) helps building the layers, (ii) is crucial in preserving layer integrity during lamination, and (iii) can contribute to dough lift by providing water for steam formation during baking. However, the latter effect seems less important than that of the water originating from the predough layers.

While yeast may increase pastry volume during fermentation, it can decrease dough lift during baking by damaging layer integrity as a result of CO₂ production.

The use and working mechanism of several bread making non-essential ingredients is not well documented in the case of pastry production and, based on the limited information available, their mode of action may in some cases differ strongly from that in bread making.

In conclusion, it seems that there is insufficient knowledge on the (bio)chemical and physical changes occurring in flour constituents and other ingredients during pastry production, how different ingredients affect each other's transformations during the different processing steps and

how the latter affect product quality. Also, much is to be learned about the role improving agents play in influencing these interactions. However, these improving agents might offer a possibility to explore ingredient functionality by evaluating the effect of using selective improving agents, altering only one ingredient (constituent) at a time.

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Table I: Typical ingredient levels (on flour weight basis) for puff pastry, Danish pastry and croissant. Compiled from (a) Bennion et al. (1997), (b) Lai and Lin (2006) and (c) Bent (2007).

		Puff pastry	Danish pastry	Croissant
	Flour	100	100	100
	Water	58 ^a , 50 ^b	45 ^b , 50-52 ^c	50 ^b , 52- 55.4 ^c
	In-dough fat	5 ^a , 17 ^b	12 ^b , 6.3-12.5 ^c	5 ^b , 2-8 ^c
Predough	Salt	1 ^a , 0 ^b	1.5 ^b , 1.1-1.56 ^c	1 ^b , 1.8-2 ^c
	Yeast		2 ^b , 6-6.25 ^c	1.5 ^b , 4-5 ^c
	Eggs	0 ^a , 7 ^b	15 ^b , 5-25 ^c	10 ^b , 0-24 ^c
	Sugar(syrup)	0 ^a , 3 ^b	15 ^b , 9.4-25 ^c	8 ^b , 2-10 ^c
Roll-in fat		50-100 ^a , 85 ^b	50 ^{b,c}	45 ^b , 32-45 ^c

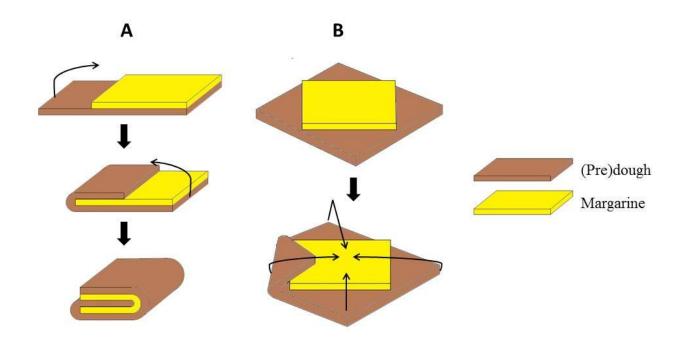


Figure I: Methods of folding bakery fat into predough in pastry making: (A) English and (B) French method.

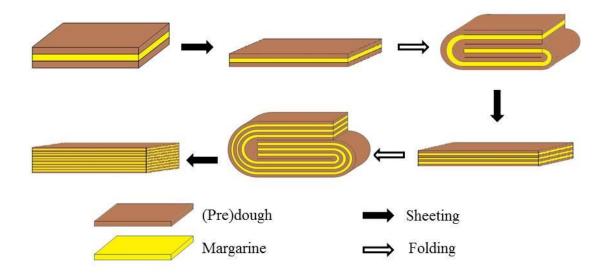


Figure II: The lamination process, including dough sheeting and folding.

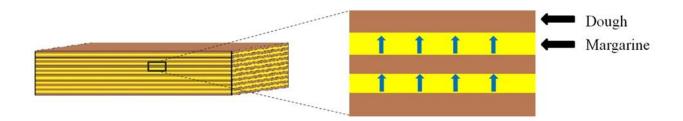


Figure III: Steam trapping in the liquid fat phase during baking, resulting in the dough lift. Upright arrows represent pressure development between the dough layers [adapted from Kazier & Dyer (1995)].

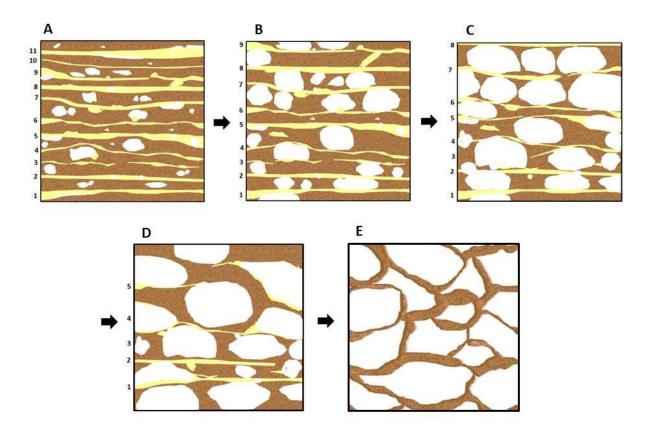


Figure IV: Schematic representation of the possible mechanism of gas cell development during pastry fermentation and baking: (A) laminated dough; gas cells within dough and margarine layers are stretched in the final sheeting direction [drawing based on confocal laser scanning microscopy images by Bousquieres *et al.* (2014)], (B) fermentation step; gas cells within the dough layers expand, (C) early baking phase; further expansion of gas cells within the dough layers stabilized by the gluten network, (D) later baking phase; formation of continuous dough layers due to gas cell opening, steam migration to and inflation of gas cells within the molten fat phase, (E) fully baked product with interconnected (porous) gas cells.