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T. Lucas, G. Collewet, J. Bousquières, C. Deligny. The size of eye-shaped bubbles in Danish pastry in relation to the size of fat fragments; a reverse engineering approach of the alveolar structure. Journal of Food Engineering, 2018, 237, pp.194-203. hal-01871051

HAL Id: hal-01871051 https://hal.science/hal-01871051

Submitted on 10 Sep 2018

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Accepted Manuscript

The size of eye-shaped bubbles in Danish pastry in relation to the size of fat fragments; A reverse engineering approach of the alveolar structure

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PII: S0260-8774(18)30186-9

DOI: 10.1016/j.jfoodeng.2018.04.025

Reference: JFOE 9239

To appear in: Journal of Food Engineering

Received Date: 12 January 2018

Revised Date: 18 April 2018

Accepted Date: 22 April 2018

Please cite this article as: Tiphaine, L., Guylaine, C., Josselin, Bousquiè., Cécile, D., The size of eyeshaped bubbles in Danish pastry in relation to the size of fat fragments; A reverse engineering approach of the alveolar structure, *Journal of Food Engineering* (2018), doi: 10.1016/j.jfoodeng.2018.04.025.

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11 **1. Introduction**

Puff pastry, Danish pastry and croissants are characterized by a unique alveolar structure with large,
 eye-shaped bubbles that are always aligned almost horizontally, providing a typically light and flaky
 texture at eating. The high capacity of lift is another particular feature of these bakery products.

15 This singular structure results from alternating thin layers of fat and base dough formed by 16 successive folding and sheeting steps (also called "turns"). In previous studies, which were mostly 17 dedicated to puff pastry, the fat layers were believed to be almost continuous when the number of 18 turns was low and to form an impervious barrier to the water vapor produced in the adjacent dough 19 layers during baking, hence facilitating lift. The higher the number of turns, the higher the number of 20 fat layers, and the better gas retention and lift. However, as the number of turns increases, the 21 individual thickness of the fat layers decreases and the initially continuous fat layer is also more likely 22 to fracture into fragments. Extreme fat fragmentation was thought to be responsible for loss of gas 23 retention and the decrease in lift reported for high numbers of turns. The optimal number of turns 24 for maximizing the lift is between 130 and 250 for puff pastry (Telloke, 1991) and 20-50 for Danish 25 pastry (Cauvain and Telloke, 1993).

26 Recently, Bousquieres et al. (2014a) estimated the size of fat fragments in Danish pastry for the first 27 time and the absence of a clear correlation with pastry lift raised doubts about the above-mentioned 28 governing mechanism. Likewise, MRI monitoring of proving Danish pastry also showed bubbles 29 growing inside the fat layers with the eye shape typical of puff and Danish pastry (Deligny et al., 30 2017). Pixels with very low signal intensity (gas gives no signal with MRI) forming pools of rather large 31 dimensions were the first criterion facilitating their visualization; the second criterion was their outline, whose signal intensity was much higher than the average signal from the base dough and 32 33 was attributed to fat. Finally, pictures of the alveolar structure of the finished products are rarely 34 reported or statistically analyzed, making the concepts of coarse crumb texture or over-laminated pastry very subjective; however, it should be noted that one recent study includes such pictures as a 35 36 function of fat consistency (Renzetti et al., 2016). All this underlines the need for both better 37 characterization of the alveolar structure in finished pastry and a better understanding of the growth 38 of eye-shaped bubbles specific to puff and Danish pastry. This is all the more worthwhile since, due 39 to nutritional or health concerns, the fat usually used for layering is being replaced. Interest in the 40 topic is confirmed by the increasing number of studies e.g. Silow et al. (2016), plus two recent 41 reviews (Wickramarachchi et al., 2015; Ooms et al., 2015).

The aim of the present study was first to characterize the size distributions of large eye-shaped 43 44 bubbles (> 1 mm in width) in Danish pastry after baking under different numbers of turns (the theoretical numbers of fat layers ranging from 4 to 128). The second aim was to link the size of these 45 46 eye-shaped bubbles to the other sizes measurable at earlier processing steps in a reverseengineering inspired approach. Our starting hypothesis was that the size of the eye-shaped bubbles 47 48 previously observed by MRI in the fat layers at advanced proving times is correlated to that 49 measured after baking in the present study. The growth of bubbles in the fat could be limited in 50 some way by the dimensions of the fat fragments, dimensions that are established after the lamination step; this constitutes the second and final step of the comparison of the sizes of the eye-51 52 bubbles in Danish pastry. The whole approach provided the basis for re-visiting the mechanisms governing the growth of eye-shaped bubbles in Danish pastry. 53

54 **2. Materials and Methods**

55 2.1. Production of laminated pastry

The recipe for the base dough contained 54% standard bread flour (water content 14%, ash 0.5%, 56 proteins 14%, Minoterie du château), 33% water, 5% sugar (commercial brand Beghin Say), 3% 57 58 glucose-fructose syrup, 2.4% yeast (dry yeast Saf Instant Or, Lesaffre), 1.6% guar (Cargill), 1% salt. Water temperature T_{water} was adjusted according to the equation: $T_{water} = 48 - (T_{air} + T_{flour})$ 59 where T_{air} is the temperature of the room in which the dough was mixed (maintained at 17°C) and 60 T_{flour} the temperature of the flour when all ingredients were weighed except water. The dough was 61 62 mixed in a Moretti Spiry 8 Spiral Dough Mixer with a vertical spiral axis for 19 min at 97 rpm (speed 63 of axis) and 10 rpm (speed of bowl); the dough was then allowed to rest and cool to 16 °C at the core for 30 min. 64

The fat used for the layering was an anhydrous milk fat (Corman, reference 0029551) with a melting point (MP) of 34 °C and 45% of solids at 15 °C (temperature at which it was stored at receipt). Some pastries with 16 theoretical fat layers were prepared with anhydrous milk fat with a MP of 40 °C and 55% of solids at 15°C. In preliminary trials, fat consistency has been estimated by cone penetrometry at different operating temperatures and fat temperature before lamination has been adjusted

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empirically so to produce good quality products (numerous, large-sized eyes according to theindustrial target).

72 Sheeting was performed in a temperature-controlled room, operating at 17°C. The dough sheeter 73 used for this study was a reversible Rondo Automat 600 equipped with two stainless steel rollers of 74 identical diameter (84 mm), both running at 138 rpm. The original "book" was composed of one 7 75 mm thick layer of fat between two base layers of dough (each 15 mm thick) giving a total thickness of 76 37 mm with 16.6% layering fat relative to the mass of dough. The book was progressively thinned to 77 about 10 mm in 2 mm steps. The pastry was folded in half, and left to rest for 15 min at 12 °C. The 78 operation was repeated several times to obtain pastries with 4, 8, 16, 32, 64, and 128 fat layers. 79 Forty-eight and 81 fat layers were obtained with the three-fold turn method applied to a pastry sheet 80 with 16 fat layers and one fat layer respectively. The number of fat layers mentioned in the rest of the paper refers to the theoretical values; the actual values were shown to be on average around 7% 81 82 lower than the theoretical ones (Bousquieres et al., 2014a). At the last folding step, sheeting was continued to a thickness of 4 mm. More details can be found in Deligny and Lucas (2015). 83

After the last laminating step, a large rectangle (about 70 × 30 cm) was cut out of the sheet (Bousquieres, et al., 2014b). Eight smaller rectangles (10 × 5 cm) were cut out of the rectangle, 4 for proving and baking (see next section) and 4 for confocal laser scanning microscopy (CLSM). To allow cross comparison, each sample rectangular removed for baking was taken in the vicinity of a sample rectangular removed for CLSM, the distance between the two samples not exceeding 1 cm.

89 2.2. Proving and baking Danish pastry samples

Four rectangular samples were placed in a proving chamber (Bongard) at 30 °C (±2 °C) at saturated humidity for 2 h and then for 11 min in a deck oven (MIWE CO 11208) set at 190 °C (deck) and 210 °C (vault). Steam (800 mL) was injected during the first few minutes of baking. The pastries were left to cool for 10 min at ambient temperature before measurements (total height and photos of cross section) were taken.

95 2.3. CLSM of just laminated pastry

96 The procedure is described in detail in Bousquieres et al. (2014b), and only the important points are97 summarized below.

Fluorophores were used to visualize fat in the fat layers (Nile Red, Sigma Aldrich) and the gluten
 network in the dough layers (Alexa Fluor 546, Life Technologies) separately. Slices (20 μm thick) of

Danish pastry were cut with a cryo-microtome (Microm Model HM 500 OM) set at -20 °C. The slices were covered with 15 µL of the fluorophore solution (0.17 mg/mL of Alexa and 0.5 mg/mL of Nile Red prepared in dimethylformamide as solvent) and stored for 24 h to ensure diffusion of the fluorophores before observation.

A Nikon (Model A1) CLSM with a green laser was used for observations. The fluorescence emission of
 the two dyes was detected between 563 nm and 643 nm and spectral deconvolution was performed
 by using spectra obtained in regions of interest (ROI) selected in the dough and fat layers. Images
 were acquired at ×20 magnification, giving a 1.24 µm/pixel resolution and a 2.49 µm thick optical
 slice.

ImageJ and Scilab software was used to prepare images manually and to automatically measure the
width of the fat fragments. Other morphological features were also measured, further details are
provided in Bousquieres et al. (2014a-b).

Eight images were acquired per batch; the batches for 8-16, 32-48 fat layers were repeated 3 and 2 times, respectively. Samples with more than 64 fat layers were not studied. Average and standard deviation were calculated over the whole set of images available, thus accounting for variability in width at a very small scale (4 x 3.8 mm²).

116 2.4. MRI during proving

For the MRI probe, just before proving, optical fibers were placed in a single rectangular sample and the atmosphere of the MRI device was set at 30 °C (±1°C) with humidity close to saturation. Further details about the MRI device can be found in Lucas et al. (2010). MRI was performed with a Siemens Avanto 1.5T, equipped with a head antenna, a spin-echo sequence (TE 7 and 32 ms, TR 400 ms, pixel side 0.5 mm, slice thickness 3 mm, 9 scans, bandwidth, 230 Hz/pixel, acquisition time for the pair of images was 4 min 48 s).

MRI images were acquired and then converted into maps of the proportion of fat, gas-free doughand gas.

The bubbles in the dough are expected to be around 1 mm in diameter and the fat layers to be 130 to 20 µm thick when increasing the number of fat layers from 8 to 64 (Bousquieres et al., 2014a) and cannot be resolved with MRI, which yields partial volume, i.e. many voxels are filled with fat, dough and gas. The proportions of fat, dough and gas were thus re-calculated in each voxel. In NMR, the water protons in the dough and the protons in the fat relax at different rates. With MRI, this results in different image contrasts at using different acquisition protocols. Knowing the signal of pure

components under the same acquisition conditions, the proportion of each component can then be re-calculated from the images. Given the absence of a signal for gas, only two images were acquired to obtain the three proportions. A regularization term was added to the optimization procedure to reduce the noise while allowing considerable spatial variations. More details can be found in Deligny et al. (2017) and Collewet et al. (2016).

136 2.5. Pictures of cross sections of Danish pastries

137 Each baked sample was cut crosswise at mid-length. A Nikon D40X camera was used to photograph 138 the cross section with a ruler placed next to the sample. Image J software was used to manually 139 measure the width of each eye-shaped bubble. Bubbles much smaller than 1 mm were not easy to 140 discern with the naked eye in the pictures because there is not enough contrast, and were consequently not counted. To check, the size of bubbles that formed in the thick layers of dough (e.g. 141 142 those with 4 or 8 fat layers) were tentatively counted (Fig. 1); the size of the bubbles was estimated 143 at 1.2±0.7mm, a value that was undoubtedly overestimated since only the bubbles visible with the naked eye were counted. It was concluded that, consistent with the general aim of the present study, 144 145 this method was suitable for counting the eye-shaped bubbles typical of Danish pastry. Fifteen to 20 146 pictures were used for each number of layers of fat, except for 32 fat layers, for which only 7 photos 147 were used.

148 **3. Results**

The optimal number of fat layers reported in the literature for maximum lift ranges from 20 to 50 for Danish pastry (Cauvain & Telloke, 1993). The numbers tested in the present study closest to those in the literature were 16 and 48; these were used to characterize the lower and upper boundaries of this optimal range and were considered as the reference for comparison with other numbers of fat layers in the following analysis. Note that in this study, pastry lift was maximum with 48 fat layers, as previously characterized with the same preparation of Danish pastry by Deligny and Lucas (2015).

155 3.1. Size of eye-shaped bubbles after baking

156 Fig. 1 shows typical internal structures of Danish pastry with different numbers of fat layers.

157 With 4 to 8 layers of fat, large eye-shaped bubbles (> 20 mm, typical of craft pastries) were present,158 and thick layers of dough were also often observed.

As the number of layers of fat increased, the overall size of the bubbles decreased. Eventually, they became rounder and were no longer oriented along the horizontal direction of layering (Fig. 1), typical features of over-lamination. In the preparation conditions used in the present study, pastry with 64 layers of fat was considered as the lower boundary of over lamination.

163 In the optimal range (16-48 fat layers), bubbles were predominantly eye-shaped, and aligned in the 164 layering direction. With 16 fat layers, bubbles larger than 20 mm were still present and the crumb 165 texture was coarse, while with 32 and 48 fat layers, the bubbles were smaller and the crumb texture 166 was more regular. Next objective will be to quantify these features.

For this purpose, Fig. 2a provides further details about the number of bubbles per width class with different numbers of fat layers. Since the largest bubbles were at least 100 times less frequent than the smallest ones, the numbers were cumulated starting from the largest ones and logically, the xaxis in the plot indicates the bubble width in *decreasing* order (Fig. 2b). Size classes are presented and discussed below in this order.

The decrease in the number of the largest bubbles (>20 mm) with an increase in the number of fat layers was confirmed, and more importantly, remained below unity (in other words, these bubbles were not often observed) when the number of fat layers was equal to or greater than 32. This result confirmed the visual inspection of the photos shown in Fig. 1 and the appearance of a much finer crumb texture with more than 32-48 fat layers.

Bubbles that were 10 to 20 mm wide were rare and did not differ much in number (in total 4±2 bubbles per pastry cross-section) when the number of fat layers was between 8 and 48. However, in over-laminated pastry, their number decreased (1±1 with 64 fat layers). In conclusion, this size range was a feature of Danish pastry, although surprisingly, it was not very sensitive to the number of turns in the optimal range.

Bubbles that were five to 10 mm wide were still elongated (Fig. 1). Occurring rarely with 8 fat layers (3 on average), around 11 bubbles were counted with 16 fat layers and 23 with 48 fat layers (Fig. 2a). The number of bubbles started decreasing in over-laminated pastry (16 bubbles with 64 fat layers). This increase in number undoubtedly contributed to the more regular crumb texture as evaluated subjectively from the photos in Fig. 1 when the number of fat layers increased from 16 to 48. It should be noted that the quasi disappearance of very large bubbles (see above) also contributed to the regularity of the crumb texture obtained at the upper boundary of the optimal range. Finally, the

number of 5-10 mm wide bubbles correlated best with the total height of the pastry once baked (Fig.
3). Interestingly, their number decreased when the number of fat layers increased from 48 to 64, as
did the total height of the pastry. This correlation is further discussed below.

192 The smallest bubbles (< 5 mm wide) were rather round in shape (Fig. 1). Their number increased 193 from 3 to about 72 with an increase in the number of fat layers from 8 to 48. Because of their shape, these bubbles hardly contributed to the typical features of Danish pastry but when combined with 194 195 larger eye-shaped bubbles in sufficient number, they made the crumb texture more regular (32-48 196 fat layers). Beyond this point, the increase of these small bubbles in number was accompanied by the 197 progressive decrease in the above-mentioned large elongated bubbles and the loss of alignment typical of Danish pastries. All this reinforced the bread-like crumb texture of the over-laminated 198 199 dough (Fig. 1). In conclusion, crumb regularity can also be achieved in this size class of Danish pastry 200 by increasing the number of these small-sized bubbles, with a maximum of 50-70 per cross section, 201 beyond which the pastry crumb resembled coarse bread-like crumb.

202 3.2. Relation to the size of the widest bubbles after proving

Fig. 4 compares the number of bubbles per width class observed in the fat layers at the end of proving of Danish pastry prepared with 12 fat layers (Deligny et al., 2017) and that of eye-shaped bubbles in the finished product (data from Fig. 2a. interpolated between 8 and 16 fat layers).

The agreement was rather good. The higher number of 1-2 mm wide bubbles was attributed to the higher spatial resolution of the photographs (30 μ m) compared to MRI images. All bubbles are expected to increase in size, even slightly, during baking. This may explain the lower number of the smallest bubbles (> 2mm) and the higher number of the largest bubbles (> 20 mm) after baking; however, the extent of these variations was within the standard deviation. The results also suggest that the effect of the baking stage on the distribution in width of the eye-shaped bubbles was limited.

213 3.3. Relation to the size of fat fragments just after lamination

Fig. 2c presents the number of fat fragments as a function of their size, measured in Danish pastry with different numbers of fat layers just after lamination. For the purpose of comparison with Fig. 2b, these CLSM values are expressed for an equivalent section of Danish pastry (conversion from the width of CLSM images of about 3.78 mm to the width of the pastry, about 50 mm) and cumulated from the largest to the smallest sizes.

219 As expected, increasing the number of turns favored the fragmentation of fat: assumingly continuous 220 fat layers were first fragmented into large fragments, these then being fragmented into smaller ones, 221 and so on. For instance, the number of fat fragments whose width was more than 3 mm increased 222 steeply with 8 fat layers; and then decreased with 16 layers to the benefit of fragments 1-3 mm in 223 width (Supplementary Material 1). The maximum number of 1-2 mm wide fat fragments was reached 224 between 32 and 48 fat layers, given the high standard deviations. The number of smallest fragments 225 only increased monotonically, obviously at a much higher rate than the decreasing rate of the largest 226 sizes (loss of about 0.6 fat fragment per additional fat layer, versus a gain of 90 fat fragments per 227 additional fat layer).

The cumulative numbers of bubbles in the pastry after baking and of fat fragments after lamination 228 229 exhibited very similar trends (Fig. 2b-c). Especially the ranking of the number of fat layers was the 230 same: the number of large fragments/bubbles increased from 64 to 8 fat layers while the reverse was 231 the case for the number of small fragments/bubbles, with the crossing point at about 1 mm wide for 232 fat fragments/5 mm wide for bubbles. We thus attempted to find a correlation between the two and 233 to this end, 5 mm class intervals were used for bubbles. The results of the comparison are presented 234 in Fig. 5. The correlation was satisfactory. As previously mentioned, bubbles whose width was 235 between 10 and 15 mm did not differ much in number with the number of fat layers and explained the worse correlation coefficient R² (Fig. 5c). Very small fat fragments (< 0.2 mm) were not used in 236 237 Fig. 5a (otherwise the result would have been a very poor correlation); such small fragments were 238 also unlikely to host gas nuclei (see below).

239

240 Together with the observation of eye-shaped bubbles appearing in the fat layers (Deligny et al., 241 2017), the results in Fig. 5 suggest that the finite dimensions of fat fragments in some way 242 determined the final size of the eye-shaped bubbles. This result is discussed below. The slope in Fig. 5 also gave the average number of fat fragments required to produce a single bubble in the finished 243 244 product, a kind of "bubble yield". The smallest and largest fat fragments (less than 1 mm and more 245 than 3 mm in size) were not very "efficient" in forming eye-shaped bubbles (respectively 41 and 22 fat fragments for the gain of a single bubble in the finished product, Fig. 5a,d). The most "efficient" 246 247 fat fragments were 1-2 mm wide (about 6 fat fragments to gain a single bubble in the finished 248 product, Fig. 5b); the bubbles associated with these fragments in Fig. 5b were already shown to be 249 the best correlated with the total height of the Danish pastry (Fig. 3). This result is also discussed 250 below.

4. Discussion

Based on these results, the mechanisms governing the growth of eye-shaped bubbles in Danishpastry are now discussed. These mechanisms are also presented in schematic form in Fig. 6.

4.1. Gas nuclei in the fat layers and fat fragments

In puff pastry, water vapor is the main gas responsible for lift and large gas cavities in the fat layer 255 256 can be formed when the water in the dough boils (Cauvain & Telloke, 1993). In Danish pastry, the 257 temperature range at which gases are desolubilized or vaporized during the proving step requires the pre-existence of gas nuclei in the fat layers (Fig. 6). The layering fat used for the preparation of 258 259 Danish pastry is often supplied in the solid state, in the form of blocks. Some very tiny gas inclusions 260 may be present in the original block of fat and these could serve as gas nuclei. The fat extrusion step is also likely to incorporate air into the fat. Lastly, air pockets can also be entrapped between dough 261 262 and fat layers when the original "book" is formed. When the fat melts, most probably at the proving 263 temperature, air trapped at the dough/fat interface may move up into the fat layers by buoyancy, 264 while the liquid fat will flow downwards, lining the pocket of gas. It is plausible that both the former 265 and latter sources of nuclei were involved in the present study since the fat was not extruded, but 266 simply sliced from the block of fat and deposited onto the original dough layer (see "Materials and 267 Methods"). Tracking these gas inclusions in the fat or at the dough/fat interface would thus require 268 further investigations.

The probability of having a gas nucleus in the fat fragment is drastically reduced with a decrease in the size of the fat fragment, which may explain the low "yield" of the smallest fat fragments (smaller than 1 mm) in producing eye-shaped bubbles (Fig. 5a). In fact, the data in Fig. 5a did not correlate well when fat fragments less than 0.2 mm in width were included; the reason may be the more systematic absence of a nucleus in this size class of fat fragments.

4.2. Bubble growth during proving and baking

Bubble growth is a balance between gas production (inducing pressure forces) and mechanicalresistance to deformation of the dough surrounding the bubble.

At the proving temperature, CO_2 is known to be as soluble in fat as in water (Jacobsen et al., 1999). CO₂ produced by yeast in the base dough during proving and early baking solubilizes into fat and then diffuses towards gas nuclei incorporated in the fat (Fig. 6). Given that the fat layer is rather thin (5-6

times thinner than the dough layers, see Bousquières et al. (2014b)), resistance to diffusion is rathersmall.

282 On the other hand, fat deformability is a prerequisite for bubbles to grow. The consistency of fat is 283 usually optimized for the lamination step. If the fat is too hard, it will fragment and eventually tear 284 the dough sheet; if too soft, it will presumably soak into the dough, the dough layers will knit together and the layering effect will be lost. Fat deformability mainly depends on its crystal form -285 286 preferentially β' form for fat plasticity (Stauffer, 1996)– and its solid content. According to Telloke 287 (1991), solid fat content should be 40-50% for optimal fat plasticity at the lamination step; depending 288 on the fatty acid composition of the layering fat, it still ranges between 10 and 40% at the proving 289 step. In the present study, fat solid content was about 10% at the proving temperature and was 290 assumed to not resist bubble inflation too greatly. Hence, fat spreads at the surface of the bubble as 291 the latter increases in volume.

Local fat thinning is likely to continue when the dough is rising in the oven. A key element of interfacial fat related to gas retention is its solid fraction at temperatures higher than the proving temperature, since it is likely to supply additional matter for this interface by melting upon heating (Brooker, 1996). This should be taken into account when choosing the solid content profiles of the layering fat.

297

298 *Collapse of the largest bubbles* growing in the fat layers was frequently observed during MRI proving 299 (see films loaded as electronic elements in Deligny et al., 2017). The largest bubbles were associated 300 with large fat domains, most plausibly communicating with the ambient environment of the pastry, 301 and the escape of the gases to the outside probably explains this collapse. This would also explain 302 why the "bubble yield" of the largest fat fragments was low (Fig. 5d).

Further fragmentation of the fat with an increase in the number of turns (Supplementary Material 1) will break the connection to the outside and help retain gas inside the smaller fat fragments. Consistently with this view, no collapse was observed by MRI during the growth of medium to smallsized bubbles (Deligny et al., 2017) and the "bubble yield" of the smaller fat fragments in the finished product was consistently much higher, see Fig. 5b for 1-2 mm wide fat fragments.

The connection between the largest fat domains and the ambient environment in the oven might also be sealed as the edge of the pastry sets during early baking. Gas hosted in these domains may thus be retained and contribute to the lift. Consistently with this view, the number of the largest eyeshaped bubbles increased between the end of proving and the end of baking (Fig. 4). However, the

contribution of the largest eye-shaped bubbles to lift is limited because of their generally low
number per pastry section (Fig. 2a), but is crucial for pastries prepared with low numbers of fat layers
(see 8 fat layers in Fig. 1 and 4 fat layers in Fig. 6). Inflation of these large bubbles, whether successful
or not, is believed to be source of variability in lift between products.

316

The increase in size of a single bubble, assumed to be spherical, can be estimated from that observed 317 at the pastry scale: $\frac{D_b(t)}{D_h(0)} \approx \left(\frac{V_p(t)}{V_n(0)}\right)^{1/3}$ where D_b is the bubble diameter and V_p is the volume of the 318 pastry. The latter was multiplied by 3.4 during proving, and by 1.6 during baking, according to Deligny 319 320 and Lucas (2015) who used the same procedure to prepare pastry as in the present study. This 321 yielded an increase in bubble diameter of 1.7. The same calculation was applied to an eye-shaped 322 bubble whose width, length (in the depth direction in the pictures in Fig. 1) and height are noted: 323 $W_b(t), L_b(t), H_b(t)$, with the following assumptions: (i) the original nucleus was assumed to be spherical in shape: $W_b(0) = H_b(0) = L_b(0)$; (ii) the bubble was assumed to grow in the same 324 proportion in width and length: $W_b(t) = L_b(t)$; (iii) the final height of the bubble was shown to be 325 326 smaller than the width by a factor noted F, with F comprised between 2.5 and 5.0 (Fig. 1). Under these assumptions, the increase in bubble width was given by $\frac{W_b(t)}{W_b(0)} \approx \left(F \frac{V_p(t)}{V_p(0)}\right)^{1/3}$ and comprised 327 328 between 2.4 and 3.0. Although the calculation merits further refinement, these preliminary values

329 suggest that the growth of the bubble of gas inside fat fragments could be enhanced compared to330 that of a spherical bubble growing in the dough layer.

Likewise, the *5-10 mm wide bubbles* occupied 15% and 50% of the cross section of the finish product with 8 and 48 fat layers respectively (Fig. 1). This result was consistent with the above conclusion that eye-shaped bubbles contribute highly to the increase in lift.

Nevertheless, it does not agree with previous reports, namely: (i) the eye-shaped bubbles observed by MRI during proving contributed little to the cross section of the proven pastry, less than 10% (Deligny et al., 2017) and (ii) the baking stage did not much change the distribution in width of the bubbles at the end of proving (Fig. 4). We believe that these bubbles grew preferentially in the vertical (height) direction *during baking*, a dimension which was not taken into account in the elaboration of Fig. 4.

Better gas retention of the 5-10 mm wide bubbles (as mentioned in the discussion about collapse above) together with a preferentially upwards inflation mainly explains why this size class was best correlated with the total height of the pastry (Fig. 3). Let us remind that larger fat fragments do not

retain gas while smaller fat fragments are unlikely to host a gas nucleus. Pastry lift may be better
controlled through the number of 1-2mm wide fat fragments formed during the lamination step.
However, as Fig. 3 shows that a 10-fold increase in the number of 5-10 mm bubbles only produced a
1.4-fold increase in the height of the pastry, the effect remained small.

5. Conclusion

The results of the present study modify the common view of bubble growth and lift in Danish pastryin the following way:

with low numbers of turns, the gain in the lift capacity with increasing number of turns was 350 351 previously attributed to the increasing number of layers, assuming that they were still sufficiently continuous. The present study shows that increasing the number of layers 352 353 without fragmentation does not lead to the expected gain in lift but results in bubbles as 354 wide as the pastry and which hardly any retain gas. (Very) low numbers of fat layers (4-8) were included in the present study in order to obtain some points below the optimal range 355 356 (16-48 fat layers). Conversely, very low numbers of fat layers were often skipped in previous 357 studies, which mainly focused on puff pastry, since optimal lift in these products is associated to very high numbers of fat layers (130-250). This is the best explanation for why these 358 studies missed the role of fat fragmentation. Mc Gill (1975) mentioned however the 359 360 tendency of the layers to separate in puff pastry when the number of turns was too low (separation can be achieved if the bubbles are as wide as the product). 361

362 with high numbers of turns, the results of the present study confirm that fat fragmentation with increasing number of turns determines the loss of lift capacity. Past studies focused on 363 364 the void spaces between fat fragments that were thought to favor gas release and decrease gas retention in the dough layers. The results of the present study rather suggest focusing on 365 the size of fat fragments that contain eye-shaped bubbles, instead of on the void between fat 366 367 fragments. With high numbers of turns, most of the fat fragments do not host a gas nucleus 368 and are distorted from their initial function -to form domains for the growth of the eye-369 shaped bubbles. Likewise, the size of bubbles hosted in small fat fragments does not differ 370 much from that of bubbles growing in dough (between 1 and 5 mm in the present study). In the present study, such a case was encountered in pastries prepared with more than 64 fat 371 layers (6 turns) in which more than 100 of these bubbles were counted per pastry section. 372

In the literature, optimal lift is often linked to the number of fat layers or turns, which is one of the factors only affecting the size of fat fragments (the total amount of layering fat, the melting point of fat, the relative speed of rollers are other well-known factors). We propose to use instead the degree of fat fragmentation in order to work in a unique frame of interpretation, common to future studies.

Finally, the fat fragments that most affected both the regularity of Danish pastry and lift were 1 to 2 mm in width. Provided that in the future, this result is generalized to other dough recipes, other fat types, other ways of preparing the base dough, etc., this target size could be used for optimizing the crumb texture of Danish pastry from the lamination step. In this sense, the present study contributes to the reverse engineering of the Danish pastry-making process.

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421 **7. Figure captions**

422 Figure 1. Typical photographs of sections of pastry prepared with different numbers of fat layers: 8
423 (a), 16 (b), 32 (c), 48 (d), 64 (e), 81 (f), 128 (g).

Figure 2. Effect of the number of fat layers on the number of bubbles visible to the naked eye in cross sections of baked pastries as a function of their size (a; b in cumulated number), and on the number of fat fragments (c in cumulated number), as visible in samples observed by CLSM (adapted from Bousquieres et al. (2014a)).

Figure 3. Total height of the Danish pastry once baked (Deligny and Lucas, 2015) as a function of the total number of eye-shaped bubbles (more than 1 mm in width, see the Materials and Methods section) or only the number of eye-shaped bubbles 5-10 mm in width.

Figure 4. Number of bubbles in Danish pastry prepared with 12 fat layers as a function of their size; comparison with the number of bubbles at the end of proving (180 min) adapted from Deligny et al. (2017). During proving, bubble "collapse" was frequently observed. In order to take into account all the bubbles that could increase in size during baking, the maximum size they reached during the 180 minutes of proving was retained.

Figure 5. Number of bubbles as a function of the number of fat fragments. Each data point isaccompanied by a value corresponding to its number of fat layers.

Figure 6. Schematic representation of bubble growth inside the fat fragments under two fat fragmentation configurations. These configurations were encountered in the present study and varied the number of fat layers (on the left), and photographs of cross sections of pastries prepared with 4 and 16 fat layers (on the right) are used for the purpose of illustration.

Supplementary material 1. Number of fat fragments of different ranges of size as a function of the
theoretical numbers of fat layers (numbers of fat layers below 8, the numbers of fat fragments were
deduced from the number of fat layers assuming them to be continuous).

Figure 1. Typical photographs of sections of pastry prepared with different numbers of fat layers: 8 (a), 16 (b), 32 (c), 48 (d), 64 (e), 81 (f), 128 (g).





Figure 2. Effect of the number of fat layers on the number of bubbles visible to the naked eye from cross sections of baked pastries as a function of their size in (a); cumulated number, in (b), and on the number of fat fragments, cumulated number in (c), in samples observed by CLSM (adapted from Bousquieres et al. (2014a)).





Figure 3. Total height of the Danish pastry once baked (Deligny et al., 2015) as a function of the total number of eye-shaped bubbles (more than 1 mm in width, see Materials and Methods) or only the number of eye-shaped 5-10 mm wide bubbles.



Figure 4. Number of bubbles in Danish pastry prepared with 12 fat layers as a function of their size; comparison with the number of bubbles at the end of proving (180 min) adapted from Deligny et al. (2017). During proving, bubble "collapse" was frequently observed. In order to take into account all the bubbles that are candidates for growth during baking, their maximum size reached during the 180 minutes of proving was retained.







Figure 6. Schematic representation of bubble growth inside the fat fragments in two configurations of fat fragmentation. These configurations were encountered in the present study and varied the number of fat layers (on the left), and pictures of cross sections of pastries prepared with 4 and 16 fat layers (on the right) are used for the purpose of illustration.



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<u>Highlights</u>

- Fat fragments determined the size of the eye-shaped bubbles typical of Danish pastry
- The number of 5-10 mm wide bubbles was linearly correlated with lift during baking
- 10-20 mm bubbles typical of Danish pastry remained constant with increased folding
- 10-20 mm bubbles decreased in number with over-lamination
- 1-10 mm bubbles provided crumb regularity to Danish pastry.

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