



# Matrix effects on the crystallization behaviour of butter and roll-in shortening in laminated bakery products



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## ABSTRACT

Two hydrogenated roll-in shortenings (A & B), one non-hydrogenated roll-in shortening and butter were used to prepare croissants. The impact of the laminated dough matrix on fat crystallization was then investigated using powder X-ray diffraction (XRD), pulsed nuclear magnetic resonance (p-NMR) and differential scanning calorimetry (DSC). The fat contained within a croissant matrix has never before been analyzed using these techniques. In each case, XRD revealed that the polymorphism of a roll-in fat will be different when baked within the dough matrix than when simply heated and cooled on its own. Both hydrogenated roll-in shortenings and butter experienced only minor changes, largely retaining their  $\beta'$  polymorphs, but the non-hydrogenated shortening experienced significant conversion from  $\beta'$  to the  $\beta$  form. However, this conversion did not take place immediately upon cooling, but after approximately 24 h of storage time. The fat contained within the croissants exhibited a significantly lower SFC than the same fats in bulk. Further, DSC results demonstrated that a greater temperature was required to completely melt all of the fat in a croissant than the same fat in bulk, observed visually as broader peaks in the melting endotherms. Analysis of croissant firmness over storage time, measured as the maximum force required to cut a croissant was used as an indication of potential sensory consequences. Results suggested that only croissants prepared with non-hydrogenated shortening experienced significant changes in firmness over one week of storage. These results indicate that there is an interaction between the shortenings and the ingredients of the croissant matrix, and given the differences observed between roll-in fats used, the extent of interaction is potentially influenced by the composition of the roll-in fat itself.

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## 1. Introduction

Laminated doughs are high fat bakery products, including croissants, Danishes and puff pastry. Croissants are the type of laminated dough selected for this study, known for their distinct shape. These products consist of many thin, alternating layers of fat and dough formed by repeated rolling and folding. It is for this reason that fats used in laminated dough production are often referred to as “roll-in fats.” Upon baking, the layering causes each individual dough layer to bake separately, creating the characteristic visual separation of layers and flaky texture. Croissants often contain approximately 30% fat by weight, therefore the properties of the fat have the ability to impact the overall quality of the products (Lai & Lin, 2006). The fat serves many purposes in these products, contributing functionality as well as taste and lubrication.

The properties of a roll-in fat which achieve optimum functionality have been well established. First, roll-in fats require optimum plasticity at the specific temperature at which the croissants will be prepared due

to the rolling that will take place. The fats must be soft and spreadable to facilitate layering without tearing the dough layers, but not so soft to leak out under the pressure. The solid fat content (SFC) is one factor that gives a good indication of the hardness and rolling ability of a fat at a given temperature. Roll-in shortenings are produced with a SFC ranging from 10 to 40% over a range of 33.3–10.0 °C, making them plastic and workable in a wider range than other bakery fats (Baldwin, Baldry, & Johansen, 1972). A minimum SFC is also required to entrap and stabilize of small pockets of air and water. Once laminated, these pockets are dispersed throughout the product, expanding during baking, and resulting in a visible rise.

However, SFC is not the sole factor controlling the functionality of a fat (Marangoni & Rousseau, 1998). Another factor involved in the plasticity and functionality of roll-in fats is polymorphism. The three major polymorphic forms occurring in fats are:  $\alpha$  (hexagonal),  $\beta'$  (orthorhombic), and  $\beta$  (triclinic), listed in increasing order of melting point, density and stability, and classified according to their subcell structure. The polymorphic form present will directly affect the melting point of a fat (Marangoni & Wesdorp, 2013). Fat polymorphism is often used as an indicator for fat functionality in the food industry, and in the case of laminated doughs, optimum plasticity has been correlated to the presence

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of  $\beta'$  crystals. These crystals tend to be smaller in size with needle-like shapes and create strong, plastic shortenings (Macias-Rodriguez & Marangoni, 2016). Conversion to  $\beta$  polymorphs, the most stable crystal form, can occur but these crystals are often larger and associated with hard and brittle textures making them undesirable for rolling (Baldwin et al., 1972; DeMan, DeMan, & Blackman, 1991). The presence of  $\beta'$  polymorphism has become standard for laminating or roll-in shortenings.

While butter is often thought of as the benchmark for laminated dough preparation, it is not favourable for large scale production due to its high cost and narrow plastic range. In the industry, roll-in shortenings manufactured by partial hydrogenation from different oils were first utilized, and produced highly accepted laminated dough products. However, these shortenings contain high proportions of *trans* fatty acids (TFA), which have a known association with heart disease and other chronic diseases (Keys, Anderson, & Grande, 1965; Lichtenstein, 2014; Mensink & Katan, 1992; Mensink, Zock, Kester, & Katan, 2003; Thompson, Minihane, & Williams, 2011). This has led to the removal of partially hydrogenated shortenings in favour of non-hydrogenated shortenings produced via interestification or by blending oils with high melting fractions (Gibon & Kellens, 2014; McNeill, 2014). These shortenings, however, have not provided the same sensorial acceptability as the partially hydrogenated shortenings, leaving many consumers unsatisfied with the alternative options (Wang, Gravelle, Blake, & Marangoni, 2016). Continuing research has uncovered that the consumption of unsaturated fatty acids (UFA) is correlated with positive health effects (Hu, Manson, & Willett, 2001; Lunn & Theobald, 2006). This has led to new efforts to produce a roll-in shortening containing primarily UFA, however these shortenings also have limitations when it comes to functionality (Acevedo & Marangoni, 2014; Blake & Marangoni, 2015; Garcia-Macias, Gordon, Frazier, Smith, & Gambelli, 2011). At this point, no roll-in shortening developed meets both the health and functionality criteria.

The focus now must be to understand how the hydrogenated roll-in shortenings behave once incorporated into these products and ultimately develop a TFA free, and low SFA roll-in shortening that behaves in the same way. It cannot be assumed that the properties of a roll-in shortening prior to incorporation remain after baking. First, the high temperatures required to bake laminated doughs are sufficient to completely melt any existing fat crystals, erasing their prior crystalline form and allowing for changes in crystallization behaviour upon cooling within the matrix. Second, melting and cooling the fat in the presence of the other ingredients creates opportunity for ingredient interactions. Notable interactions between lipid materials and starch, as well as lipid materials and proteins have been documented, meaning there is great potential for the croissant components (wheat starch and gluten) to interact with the roll-in fats and alter the crystallization behaviour (Chiming Tang & Copeland, 2007; Eliasson, 1994; Karel, 1973; Pareyt, Finnie, Putseys, & Delcour, 2011). Storage time will also be introduced as a variable given the known changes that occur in bakery products, including starch retrogradation and the migration and loss of moisture, which could impact the fat crystallization behaviour over the croissants' week long shelf life. To complete the investigation, samples of each fat will also be baked at the same time and temperature conditions used in baking the croissants to ensure that any noted differences between bulk fat and fat within a croissant are not simply caused by melting and recrystallization.

Until now, the crystallization properties of a fat contained within a baked matrix have never been investigated. In this work, the impact of baking a commonly used roll-in fat into a laminated dough matrix on a fat's crystallization behaviour was investigated, including polymorphism, melting behaviour, SFC, and product firmness. Four different fats were used to prepare four different types of croissants: butter, two different hydrogenated shortenings (termed A & B) and one non-hydrogenated shortening. The shortenings used are all specialized for lamination. The inclusion of both hydrogenated and non-hydrogenated shortenings will allow for speculative assessment of why the products produced vary significantly in perceived acceptability. The information obtained from this research has the potential to be used in the creation of guidelines for developing alternative roll-in shortenings, those which are more in line with current health concerns, but which match the behaviour of the most acceptable shortenings.

## 2. Materials and methods

### 2.1. Materials

Butter and all non-fat ingredients were purchased at a local supermarket. Three different shortenings were obtained from Bunge® Canada. The composition of each shortening used, as stated by the supplier, is listed in Table 1. To maintain the fat crystal memory, the shortenings and butter were stored at 5 °C prior to measurement. Wheat starch was obtained at a local supermarket, and pre-gelatinized wheat starch was obtained from ADM Food Ingredients. Prepared croissants were stored at room temperature for one week. One week was the determined shelf life as croissants became mouldy after this length of time.

### 2.2. Fatty acid (FA) composition

Milk fat was obtained from butter by heating gently on hot plate until bubbling ceased. Foam was skimmed off the surface and the solids were discarded, retaining only the fat portion for analysis. Each of the roll-in fats were converted into fatty acid methyl esters (FAMES) using the protocol described by Christie (Christie, 1982). An Agilent 6890 series GC (Agilent Technologies Inc., Wilmington DE, USE) equipped with a CP-Sil 88 capillary column (100 m × 0.25 mm × 0.20  $\mu$ m), flame ionization detector (FID), split/splitless injection port and a 7683-series auto-sampler was used for the analysis of prepared FAMES. 1–2  $\mu$ g/ $\mu$ L of total FAMES were dissolved into hexane and 1  $\mu$ L was then injected. Hydrogen was used as the carrier gas at a flow rate of 1 mL/min. The oven began at a temperature of 110 °C, increasing to 230 °C at a rate of 4 °C/min and holding here for 10 min. The injector temperature was 240 °C, and the detector temperature was 280 °C. Triplicate analysis was performed and the average reported. Fatty acid (FA) composition was determined with the use of an internal standard. Quantification of FAs was done by integration of the relative peak area.

### 2.3. Triacylglycerol (TAG) compositions

Determination of TAG composition of milk fat (obtained from butter using the same method described above) and each shortening was carried out by high performance liquid chromatography (Agilent HPLC model 110, Agilent Tech, Palo Alto, CA), equipped with a quaternary pump, autosampler and Hewlett-Packard Chem Station software

**Table 1**  
Roll-in fat compositions (as stated by manufacturer).

Roll-in fat type	Composition
Unsalted butter	Butter
Hydrogenated shortening A	Hydrogenated vegetable and hydrogenated modified palm oil
Hydrogenated shortening B	Hydrogenated soybean and cotton seed oils
Non-hydrogenated shortening	Canola oil, modified palm oil and palm kernel oil

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