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Legume enriched cereal products: A generic approach derived from material science to predict their structuring by the process and their final properties

A.F. Monnet^{1*}, K. Laleg^{2*}, C. Michon¹, V. Micard^{2§}

¹UMR Ingénierie Procédés Aliments, AgroParisTech, Inra, Université Paris-Saclay, Massy, France

²UMR IATE, Montpellier SupAgro, INRA, CIRAD, University of Montpellier, Montpellier, France

* Co-first authors

§ Corresponding author: Valérie Micard, Montpellier SupAgro 2 place Pierre Viala, 34060 Montpellier cedex 01, France, valerie.micard@supagro.fr

Highlights

- Composition and behavioral differences in the components of wheat and legume flours
- Texture, sensory and nutritional properties of legume enriched cereal products
- Impact of adding legume flour on the multi-scale structure of cereal products
- Modifying processing to control the quality of legume enriched cereal products

Abstract

Enriching cereal products with legume flour has nutritional advantages, notably that of improving the amino acid balance, but also modifies their processability and properties. Being able to predict such modifications is a major issue for industry.

In this review, we focus on the properties of the main components of cereal and legume flour (proteins and starch), and their functionality during processing of five typical cereal products (snacks, pasta, biscuits, cakes and bread), and pay particular attention to their role in structuring the product throughout processing. The nutritional and other properties (color, texture, structure, and sensory acceptability) of the legume-enriched cereal products are described.

Adding legume flour changes the structure of the product and results in a different texture. As the structure is usually designed during processing, a good knowledge of the product-process interactions is required. State diagrams of starch and protein in both cereals and legumes are compared. The gelatinization and the denaturation temperatures of starch and protein in legumes are slightly higher than those of cereals. These differences may affect the structuring dynamics and the final structure of the products. Process modifications are proposed to limit such effects and are discussed in light of several results in the literature.

Applying an integrative approach enabled us to demonstrate the expression of the functional properties throughout processing, from the composition of the different flours to the end products. We propose a generic representation that can be used by all those who wish to rationally formulate legume enriched cereal products.

Key words: Food; chemical-physics; functional properties; protein; starch; state diagram; structure

38 Introduction

39 Meat is a major source of protein in the human diet and in 2013, accounted for 40% of the world's
40 total protein supply (FAO, 2016). Meat consumption reached 34 kg/capita/year in the 2013-2015
41 period (OECD/FAO, 2016) and is projected to increase by ~70% by 2050 (FAO, 2011). However, a
42 decrease in meat consumption is recommended due to growing concerns about the environmental
43 impact of livestock and the health risks associated with high meat intake. This leads to the need for
44 more plant based diets and more energy efficient processing.

45 Cereal and legume grains are ancestral staple foods that are rich in carbohydrates, fibers and
46 micronutrients. They also provide a fair amount of proteins (9-18 and 20-40 g/ 100 g of grains wb,
47 respectively). When combined, protein efficiency is improved thanks to complementary essential
48 amino acid profiles (Young & Pellett, 1994). Both kinds of whole grain also play a key role in the
49 prevention of cardiovascular disease, type 2 diabetes, some cancers, digestive tract diseases,
50 overweight and obesity (Duranti, 2006; Mellen, Walsh, & Herrington, 2008; Ye, Chacko, Chou,
51 Kugizaki, & Liu, 2012).

52 In another connection, agricultural research has shown the potential advantages of cereal-legume
53 intercropping that makes better use of natural resources at crop or rotation scale (Pelzer *et al.*,
54 2012). Considering that wheat based baked products are widely consumed and accepted (more than
55 4 million metric tons consumed in Europe in 2015 (CAOBISCO, 2016)), improving the formulation of
56 pulse enriched products could be a step towards more sustainable food chains.

57 In the literature, a wide variety of studies focus on the formulation of cereal products with legume
58 flours or isolates (Noorfarahzilah, Lee, Sharifufin, Mohd Fadzelly, & Hasmadi, 2014). The first boom in
59 this research started in the late 1970s driven by the quest for more competitive and high quality
60 protein sources in many parts of the world. Special programs were conceived to promote the use of
61 composite flours, i.e. binary or ternary mixtures of flours and fractions (starch, proteins) of different

botanical origin that would enable the formulation of fortified products, especially valuable for developing countries. This led to studies of the nutritional, structural and sensory properties of fortified products for example by (D'Appolonia, 1977; Fleming & Sosulski, 1977a, 1977b; Mcwatters, 1978).

A second boom began in 2000 due to the strong resurgence of concerns about protein in a context of forced nutritional transitions at global scale. In line with pioneer works, recent studies have focused on the impact of replacing wheat flour with legume flour on nutritional, structural and sensory properties of enriched products (Noorfarahzilah *et al.*, 2014). Such experiments are underway worldwide. However, among studies on the use of legumes in cereal or cereal-like products, only a few have focused on the flours and on the effect of partial or total replacement of cereal flour by legume flour on the product structure at different spatial scales and throughout processing (Laleg, Barron, Santé-Lhoutellier, Walrand, & Micard, 2016; Laleg, Cassan, Barron, Prabhasankar, & Micard, 2016; Petitot, Barron, Morel, & Micard, 2010).

Processing such products raises technological questions due to differences in functionality between wheat and legume flours, at first sight the most obvious being the type and functionality of proteins. Foschia, Horstmann, Arendt, & Zannini (2017) recently reviewed the uses of legumes (mainly as concentrate and isolate) as an ingredient in different gluten free bakery and pasta products. These authors described the resulting changes in the properties of the end product in an attempt to link them with the functional properties of proteins, and underlined the difficulty in drawing any generic conclusions concerning the effect of legume ingredients on product quality due to the variation in their formulas and in processing conditions. To be able to properly monitor processing and to achieve constant product quality, changes in the structure of the product need to be better assessed at each stage of processing.

The present review investigates the impact of adding varying amounts of legume flour to wheat flour on the multi-scale structure of five standard cereal products: bread, pasta, biscuits, soft cakes and

87 extruded snacks. In line with the above mentioned review by Foschia *et al.* (2017) of gluten free
88 products enriched in legumes, particular attention is paid to the functional properties of the legume
89 components, particularly starch and proteins. Their influence on the resulting texture, color, flavor
90 and nutrition of final food products is described. The structure-properties relationships are
91 highlighted in an original way. Finally, for the first time, a generic approach is proposed to monitor
92 the transformation of material during processing of legume enriched cereal products.

93

Formulation of classical wheat products likely to be complemented with legume flours

Formulation offers the opportunity to tailor the structure of a food by controlling the processing conditions (Aguilera & Stanley, 1999). Table 1 provides a simplified comparison of the formulas of the five products based on their major ingredients. Flour content varied homogeneously over the range of formulas. Three cases can be distinguished: (i) products rich in flour, whose second ingredient is mainly water (snacks, pasta, bread); ii) biscuits, with an intermediate amount of flour and equal proportions of sugar and fat as second ingredients; iii) soft cakes, with a small amount of flour, the same amount of sugar, and smaller amounts of fat, egg, and water. Additional ingredients are yeast, raising agents or emulsifiers in bread, biscuit and cake formulas. These are minor ingredients as far as their concentration is concerned but they contribute to the formation of a gas phase and/or influence the distribution of air and fat inside the product structure.

Beyond the nature of the ingredients, their components, i.e. starch, proteins of various origins, water, lipids and fibers, also require attention. The balance between these components and the way they come into contact (mixing order, intensity and duration) define the way they interact to form one or several phases that, in turn, determine the product structure or matrix. For an overview of the different balances between components, data from the National Nutrient Database of USDA (2016) for the composition of an average wheat flour (11.5% protein content) and whole eggs was used to deduce the approximate starch, protein, lipid, fiber composition, and total water content of all formulas (Table 1). The result highlights the importance of starch and water, followed by proteins, which accounted for around 10% or less. Starch and proteins are essential components in the process of structure building of these products, and their action is closely linked to water availability.

[TABLE 1]

Comparative composition of wheat and legume flours

Replacing wheat flour with legume flour, which differs in composition, has an impact on the balance of the components in the formula. Legume flours contain less starch, more protein and more fiber than wheat flour and semolina, at an equal or higher lipid content.

Starch content accounts for between 60% and 80% db of soft and durum wheat, whereas it ranges from 40% to 65% for legumes except for lupin (less than 1% db) (Ahmed, 2014; Feillet, 2000; Laleg, Barron, Santé-Lhoutellier, *et al.*, 2016; Osorio-Díaz, Agama-Acevedo, Mendoza-Vinalay, Tovar, & Bello-Pérez, 2008; Petitot, Boyer, Minier, & Micard, 2010; Sosulski & Mc Curdy, 1987; N. Wang, Hatcher, Toews, & Gawalko, 2009).

Protein contents ($N \times 6.25$) vary between 20% and 30% db in legume flours (D'Appolonia, 1977; N. Wang *et al.*, 2009) except in faba and lupin flours in which protein can reach almost 40% db (Abou-Zaid, Ramadan, & Al-Askany, 2011; Ahmed, 2014). In wheat flour and semolina, protein content usually varies between 9% and 18% db (Feillet, 2000).

Fiber contents are around 2% db in wheat flour and semolina while it can reach 10% db in pea and faba flours, and even 20% to 40% db in chickpea, lentil and lupin flours (Abou-Zaid *et al.*, 2011; Ahmed, 2014; Osorio-Díaz *et al.*, 2008; Petitot, Boyer, *et al.*, 2010; Previtali *et al.*, 2014; Sosulski & Mc Curdy, 1987). In the latter species, the flours are made from grains that have not been dehulled, and the result is a higher proportion of fiber. Lipid content varies between 1% and 3% db in wheat and legume flours except for chickpea and lupin flours in which it can reach 10% to 13% db (Ahmed, 2014; Laleg, Cassan, Barron, *et al.*, 2016; Petitot, Boyer, *et al.*, 2010; Sosulski & Mc Curdy, 1987; Zhao, Manthey, Chang, Hou, & Yuan, 2005).

Comparative composition and structure of proteins and starches

In addition to varying in proportions, starch and proteins in wheat and legume grains also vary in nature.

The proportion and size of the main proteins contained in wheat and legume grains vary. In wheat, gluten proteins i.e. prolamins, are the dominant group, accounting for 80% of total protein (Shewry, Tatham, Forde, Kreis, & Mifflin, 1986). Prolamins are storage proteins and are classified as gliadins and glutenins depending on their structure and size (Shewry *et al.*, 1986). Glutenins are high molecular mass polymeric proteins (> 100 kDa) composed of subunits linked by disulfide bonds. Gliadins are lower mass monomeric polypeptides (25-75 kDa) that can form intra-polypeptide disulfide bonds (Feillet, 2000; Shewry *et al.*, 1986). In legumes, globulins are the dominant group, accounting for 50% to 70% of total protein (Boye, Zare, & Pletch, 2010). Globulins are also storage proteins that can be divided into two classes according to their Svedberg sedimentation coefficients, 7S and 11S. Vicilins are 7S trimeric proteins with three subunits linked by weak bonds (hydrogen, hydrophobic interactions) for a total molecular mass of ~175 kDa (Boye, Zare, *et al.*, 2010). Legumins are 11S hexameric proteins with six subunits held together by weak bonds for a total molecular mass ranging from 270 to 420 kDa (Duranti & Gius, 1997). Wheat gliadins and glutenins contain a higher concentration of sulfur amino acids than legume globulins meaning they have more reactive cysteine residues (Boye, Zare, *et al.*, 2010; Mann, Schiedt, Baumann, Conde-Petit, & Vilgis, 2014). Finally, low molecular mass metabolic albumins account for the remaining fraction in both cereal and legume grains, reaching respectively, 15% and 15-40% of total protein contents (Boye, Zare, *et al.*, 2010; Feillet, 2000). In legumes, they are subdivided into a major fraction PA1 (dimeric, 50 kDa), a minor fraction PA2 (dimeric, 11 kDa), lectins (tetrameric, 50 kDa) and enzyme inhibitors (Perrot, 1995).

Starch in both wheat and legumes is composed of starch granules containing linear amylose and branched amylopectin organized in semi-crystalline and amorphous structures. However they differ in their shape and amylose/amylopectin ratio (Buléon, Colonna, Planchot, & Ball, 1998). Wheat

starch presents a bimodal distribution of large lenticular starch granules (25-40 μm) and small spherical starch granules (5-10 μm) (Buléon *et al.*, 1998). The amylose/amylopectin ratio in wheat granules varies from 11/89 to 26/74 depending on the variety (Kumar *et al.* 2016). Legume starches are generally composed of oval or irregularly shaped starch granules. Their size is similar, ranging from 8 to 50 μm in pea, 6 to 37 μm in lentil, and 14 to 30 μm in chickpea (Hoover, Hughes, Chung, & Liu, 2010). Legume starches have a higher proportion of amylose than wheat starch: their amylose/amylopectin ratios range from 24/76 to 40/60 for pea and lentil starches and from 23/77 to 35/65 for chickpea starch (Hoover *et al.* 2010).

Comparison of the functional properties of proteins and starches

During the formation of the structure, proteins and starch express their functional properties in response to different environmental parameters: water availability, temperature, mechanical treatment, and potential chemical or physical interactions with other components. Fundamental properties concern the ability to form specific structures (networks, interfaces) and to undergo thermal transitions that are all closely related to the process of structure building. First, we discuss the usual functional properties of proteins from both origins. We then provide insights into the thermal transitions of protein and starch based on available data, as shown in figures 1 and 2.

Most wheat proteins are insoluble in water at neutral pH, which corresponds to standard processing conditions (Shewry *et al.*, 1986). This almost makes them unsuitable for several functional properties requiring solvation, i.e. emulsification and foaming. However, under specific hydration and shear conditions, wheat proteins have the ability to form dough with viscoelastic properties. These mechanical properties are enabled by the formation of a gluten network made of “fibers” composed of glutenins and gliadins linked by inter- and intra-disulfide bonds. This is made possible by the large amount of reactive cysteine residues in these proteins (Mann *et al.*, 2014). Conversely, legume protein isolates exhibit a satisfactory degree of solubility in water at pH 7, and the mean values for pea, lentil and chickpea are ~70% (Boye, Aksay, *et al.*, 2010; Karaca, Low, & Nickerson, 2011).

Concerning interfacial properties, legume protein isolates have low emulsifying properties (~20 m² of oil covered/g of isolate) and stability (~10 to 20 min) compared to animal protein isolates (Fenn, Lukow, Humphreys, Fields, & Boye, 2010; Karaca *et al.*, 2011; Withana-Gamage, Wanasundara, Pietrasik, & Shand, 2011). However, they show better foaming activities (~100% to 200%) and stabilities (~60% to 90% at 30 min) (Boye, Aksay, *et al.*, 2010; Toews & Wang, 2013). This ability is related to their low molecular mass that enables them to rapidly diffuse to the air-water interface. In any case, their interfacial properties are better than those of wheat gluten (only ~5 m² of oil covered/g of isolate and 9 min of emulsifying stability, and foaming activity and stability indexes of 29% and 40% (J. Wang, Zhao, Yang, & Jiang, 2006)).

As polymers, protein and starch are subject to thermal transitions. Their transition temperature depends on the available water content that is expected to vary during the course of the process.

Figure 1 summarizes thermal transitions in the form of state diagrams for proteins and figure 2 for starch.

[FIGURE 1]

Proteins are subject to two kinds of thermal transition: glass transition and thermosetting. Glass transition corresponds to an increase in molecular mobility enabled by an increase in temperature and/or in water content. This change in organization at a molecular scale is accompanied by major changes in the mechanical properties of the material, which changes from a solid brittle state or “glassy state” to a soft viscoelastic state or “rubbery state” (Cuq, Abecassis, & Guilbert, 2003). Glass transition is well described in wheat proteins. Changes in the glass transition temperature (T_g) of wheat gluten proteins as a function of water content, as reviewed by Cuq *et al.* (2003) and in supplementary data by Bengoechea, Arrachid, Guerrero, Hill, & Mitchell (2007) are shown in figure 1. Only a few studies have focused on glass transition in legume proteins, considering either isolates (Bengoechea *et al.*, 2007; Pelgrom, Schutyser, & Boom, 2013) or purified fractions (Mizuno, Mitsuiki, & Motoki, 2000; Morales & Kokini, 1997). Changes in the T_g of pea and soy protein isolates as a

function of water content reported by Pelgrom *et al.* (2013) and Bengoechea *et al.* (2007) are included in figure 1. This comparison shows that glass transitions in wheat gluten and legume protein isolates take place in the same ranges of temperature and water content. Thermosetting refers to the fact that above a certain temperature, there is a change in protein structure. This phenomenon is described as the succession of protein unfolding or denaturation and protein aggregation or crosslinking, both occurring in a relatively narrow temperature range. The beginning of thermosetting is usually estimated by the denaturation temperature (T_d) measured by differential scanning calorimetry. As a result of protein crosslinking by hydrogen, hydrophobic and disulfide bonds, a three-dimensional network is formed that confers new rheological properties to the matrix (Mann *et al.*, 2014). Figure 1 shows the changes in the thermosetting temperature of wheat gluten proteins as reported by Kokini, Cocero, & Madeka (1995). The T_d of wheat gluten proteins varies from ~90 °C for water contents below 20% water db to ~70 °C for water contents above 20% water db, assuming that it remains constant above 43% water db (Fig. 1). Many studies have documented the denaturation of proteins in different legume species and protein fractions at water contents of between 80% and 94% wb and at different DSC heating rates (2, 5 or 10 °C/min). To get an idea of the variation in T_d in these parameters, we chose to calculate an average T_d for 50 values from five studies, considering 11 different legume species and three heating rates after having checked that the difference in the heating rate had very low impact on the values. Averaging the legume species was possible given the low standard deviation with regard to the positioning of the T_d curve in the state diagram (Fig. 1). The result was $88\text{ °C} \pm 7\text{ °C}$ over the 80-94% moisture range, indicating a higher T_d for legume proteins than for wheat gluten proteins.

[FIGURE 2]

As a semi-crystalline polymer, starch is subject to glass transition of its amorphous zones and melting of its crystalline zones. Like for wheat proteins, data are available on the glass transition of wheat starch (Cuq *et al.*, 2003). Figure 2 shows changes in the glass transition temperature (T_g) of wheat

237 starch as a function of water content, as reviewed by Cuq *et al.* (2003). In contrast, very few data can
238 be found on the Tg of legume starches. Pelgrom *et al.* (2013) documented changes in the Tg of pea
239 starch between 10% and 16% water wb (Fig. 2). On this basis, the comparison of Tg in wheat and pea
240 starches suggests that glass transition occurs in the same ranges of temperature and water content.
241 As a semi-crystalline polymer, starch is subject to the melting of its crystalline zones at the
242 temperature Tm. Crystallites melt in a three-stage process composed of the rapid melting of
243 amylopectin crystallites (Tm1), slow melting of remaining ones (Tm2) and melting of amylose-lipid
244 complexes (Tm3) (Roos & Drusch, 2015). The occurrence of Tm1 and Tm2 depends on water content;
245 usually only Tm2 is observed at low water content (< 35% wb), both Tm1 and Tm2 at intermediate
246 water contents (35% to 60% wb) and only Tm1 at high water contents (> 60% wb) (Roos & Drusch,
247 2015). Theoretically, it is only in excess water (i.e. > 60% wb) conditions that gelatinization *stricto*
248 *sensu* can occur. Gelatinization is a complex process including melting of the crystallites at Tm1 but
249 also a group of other structural changes leading to hydration and swelling of the granules, leaching
250 and potential loss of granular integrity. In the case of wheat starch, Vanin, Lucas, & Trystram (2009)
251 summarized data from three studies on Tm1, Tm2 and Tm3; the corresponding Tm ranges are shown
252 in figure 2. In the figure, data from the study by Eliasson (1980) show the occurrence of Tm2 above
253 60% water wb; however, this is not standard behavior. Like in the case of legumes, the study of the
254 starch thermal transition has mostly focused on gelatinization properties with little research devoted
255 to the different melting transitions. Consequently, available data usually provide information about a
256 single transition endotherm in the 70-80% moisture range that we believe to be Tm1. For the same
257 reason as for protein denaturation, we calculated an average Tm1 for legume starches based on 46
258 values taken from four studies concerning 11 legume species. In this case, all the studies used the
259 same heating rate of 10 °C/min. The result was an average temperature of 70 °C ± 5 °C over the 70-
260 82% moisture range (Fig. 2). This suggests a higher Tm1 for legume starches than for wheat starch,
261 whose Tm1 lies around 55 °C ± 5 °C. Foschia *et al.* linked this to the higher amylose/amylopectin ratio
262 of legume starches (Foschia *et al.*, 2017).

To conclude, it appears that wheat and legume components have contrasted properties. While they exhibit distinct behaviors in terms of solvation and structure building abilities (e.g. the formation of a network versus foam), their thermal properties appear to be similar. However in legumes, higher protein denaturation and starch gelatinization temperatures (T_{m1}) may point to a tendency to a shift in these reactions as a function of temperature during thermal treatment.

Current knowledge about the impact of adding legume flour on the end properties of the product

A good number of studies are available concerning the impact of replacing wheat flour on the textural, sensory and nutritional properties of enriched products. They are summarized in Table 2 according to the trends shared by the five cereal products.

[TABLE 2]

Modifications in color and texture caused by adding legume flour

Concerning color, a decrease in lightness values (L) with an increase in the proportion of legume flour is regularly reported. The same decrease was observed in all five kinds of product, both in the crust and crumb in the case of bread and cakes. This could be related to the color of the legume flour (Zhao *et al.*, 2005), and to the thermal induced protein-carbohydrate Maillard reactions which involve reducing sugar and the terminal amino group of a free amino acid, in particular lysine. These reactions could be more pronounced in the case of legume-enriched products, resulting in a larger decrease in the lysine content in comparison with that in wheat products (-14% and -2% decrease in lysine between low and very high temperature treatment of 35% faba bean pasta and wheat pasta respectively) (Laleg, Barron, Santé-Lhoutellier, *et al.*, 2016). In dried pasta, lightness decreased by 16% in 35% split pea enriched pasta (Petitot, Boyer, *et al.*, 2010). With 100% enrichment, the extent of pasta darkening has been shown to depend directly on the type of legume. The lightness decreased by 43%, 60% and 26% respectively with 100% faba, lentil and black gram pasta (Laleg,

287 Cassan, Barron, *et al.*, 2016; Petitot, Boyer, *et al.*, 2010). In biscuits, lightness decreased by 10% when
 288 50% of lupin flour was added (Jayasena & Nasar-Abbas, 2011). In snacks, lightness decreased by 13%
 289 when 45% of bean flour was introduced compared to 15% of added bean flour (Anton, Gary Fulcher,
 290 & Arntfield, 2009). In cake, crumb lightness decreased by 3% and crust lightness by 10% when 50% of
 291 chickpea flour was added (Gómez, Oliete, Rosell, Pando, & Fernández, 2008). In bread crumb,
 292 lightness decreased by 7% and crust lightness by 24% when 30% of chickpea flour was added
 293 (Mohammed, Ahmed, & Senge, 2012). A tendency to increased yellowness-blueness (b^*) was also
 294 observed in snacks, biscuits, bread crumb and crust, and cake crumb. In snacks b^* increased by 22%
 295 when 45% of bean flour was added compared to 15% of added bean flour (Anton *et al.*, 2009). In
 296 biscuits, b^* increased by 51% when 50% of lupin flour was added (Jayasena & Nasar-Abbas, 2011). In
 297 bread, crumb b^* increased by 1,230% and crust b^* increased by 160% when 30% of chickpea flour
 298 was added (Mohammed *et al.*, 2012). In cake crumb b^* increased by 72% when 50% of chickpea flour
 299 was added (Gómez *et al.*, 2008). Variations in redness-greenness (a^*) depended on each case; they
 300 generally depended on type of legume and the proportion of flour.

301 In terms of texture, incorporating legume flour always increases the hardness, firmness or stiffness of
 302 the product, measured by TPA, other compression tests, or bending tests. For example, in snacks
 303 Anton *et al.* (2009) measured a +5% and +15% increase in the breaking strength index (peak breaking
 304 force over surface area measured with a three point bending test) in snacks containing 30% and 45%
 305 of legume flour compared to snacks containing 15% of legume flour. A +38% and +48% increase in
 306 pasta firmness has been observed in 35% faba or split pea pasta, and in 100% faba bean pasta,
 307 respectively (Petitot, Boyer, *et al.*, 2010; Rosa-Sibakov *et al.*, 2016). However, the firmness of gluten
 308 free 100% faba bean, lentil and black gram pasta was shown to still be 2.3, 1.7 and 1.5 fold lower
 309 than a cereal gluten free pasta (Laleg, Cassan, Barron, *et al.*, 2016). In biscuits Jayasena and Nasar-
 310 Abbas (2011) observed increases in hardness and brittleness (evaluated with a three point bending
 311 test) ranging between +51% and +342% for hardness and +96% and +223% for brittleness in biscuits
 312 containing 10% to 50% of lupin flour. In bread, using a compression test, Previtali *et al.* (2014)

showed an increase in firmness of + 26% and +82% in breads containing 20% and 25% of lentil flour, respectively. De la Hera, Ruiz-París, Oliete, & Gómez (2012) reported increases in hardness (TPA test) ranging from +18% to +80% in layer cakes containing 50% of lentil flour (different cultivars and particle sizes) and between +32% and +145% in cakes containing 100% lentil flour.

In pasta and biscuits, the increase in hardness can only be attributed to the structure and composition of the dense matrix in contrast to snacks, bread and cake (aerated products) where cellular structure may also play a role. In the case of aerated products, an increase in firmness can be attributed either to the cell wall material itself or to the distribution of air within the structure (size, number and distribution of the cells, thickness of the cell walls). Some of these aspects have been shown to be impacted by the incorporation of legume flour in some products. First, product expansion and density decreased and increased, respectively, with an increasing proportion of legume flour: reaching -20% of expansion in snacks containing 45% of legume flour compared to 15% in snacks (Anton *et al.*, 2009); +33% density in layer cakes containing 100% lentil flour (De la Hera *et al.*, 2012); up to +31% of density in breads containing 40% of soy flour (Shogren, Mohamed, & Carriere, 2003). Secondly, in the case of enriched snacks and breads, thickening of the cell walls and an increase in porosity (meaning more holes in the walls) have been reported (Fleming & Sosulski, 1978; Lazou, Michailidis, Thymi, Krokida, & Bisharat, 2007; Lazou & Krokida, 2010), concomitant with a decrease in the number of cells in enriched snacks.

Sensory perception and acceptability of legume-enriched products

Sensory results provide qualitative information on variations resulting from descriptive profiles that take color, flavor, texture and structure characteristics of products into account. The few results available in descriptive profiles appear to confirm instrumental observations of volume and textural properties. Indeed, Shogren *et al.* (2003) reported an increase in the perceived denseness and firmness of breads when the proportion of soy flour was increased from 0% to 40%. Petitot, Boyer, *et al.* (2010) and Rosa-Sibakov *et al.* (2016) also reported an increase in perceived hardness in pasta

containing 35% faba bean and split pea and 100% faba bean flour, respectively, in comparison with wheat pasta. These descriptive profiles also made it possible to detect the emergence of specific flavors or aftertastes, with the mention of a “beany” attribute. In the same study by Shogren *et al.* (2003), the increase in the proportion of soy from 0% to 40% caused an increasing tendency to a beany flavor as well as an increase in bitter flavor that is described as “mildly bitter aftertaste associated with soy flour”. In pasta, Wojtowicz & Moscicki (2014) reported the “poor taste” related to a beany flavor in pasta containing more than 20% of legume flour.

Acceptability tests have also been performed. The majority of these studies are based on acceptability scores based on a scale of 0 to 10. The results show a general tendency to decreasing values of acceptability of certain attributes with an increase in the amount of legume flour incorporated. This may concern overall acceptability as well as general appearance, color or texture. Specific attributes like taste, aroma, flavor and associated persistence are also concerned and linked to unusual “beany” perceptions. Such studies usually investigate the maximum amount of legume flour that can be used and still ensure a product is of acceptable sensory quality. To our knowledge, no study has been conducted of the effect of the incorporation of legume flour in the acceptability of snacks. The 15% and 30% enriched pasta were scored as equally acceptable as the durum control, with no difference in color, flavor or overall acceptability (Wood, 2009). At higher levels of enrichment, the overall acceptability of pasta decreased due to a beany flavor (Wójtowicz & Mościcki, 2014), and to changes in major textural attributes, e.g. an increase in hardness, elasticity and fracturability (by 60%, 30–40% and 170%, respectively, for 35% legume pasta (Petitot, Boyer, *et al.*, 2010). However, the acceptability of pasta enriched with 70-100% legume flour was still equivalent to that of their high fiber or gluten free counterparts. Indeed, acceptability of 70% faba bean pasta was close to or better than whole wheat or gluten free pasta, respectively, and 100% faba bean pasta tended to be better appreciated than commercial gluten free pasta (Laleg, Barron, Cordelle, *et al.*, 2017). For biscuits, Noor Aziah, Mohamad Noor, & Ho (2012) reported an improvement in flavor, after taste, crispness and overall acceptability with the addition of 35% of

mung bean or chickpea flour, but a decrease in aroma acceptability. Conversely, Jayasena and Nasar-
Abbas (2011) and Mc Watters (1978) reported a decrease in flavor, taste, crispness and overall
acceptability of biscuits with 20% and 50% of added lupin, soy or pea flours. In cakes, Gómez,
Doyagüe, & de la Hera (2012) reported a decrease in acceptability in the appearance, texture, odor,
taste, persistency and overall acceptability of cakes containing 35% of pea flour. However scores for
the enriched cake were higher than 5 (neither like nor dislike), indicating relatively good
acceptability. Finally, a decrease in the acceptability of breads containing more than 10% to 20% of
chickpea, soy, pea and lentil flour was observed (Alam, Talukder, Rahman, Prodhan, & Huq, 2013;
Mohammed *et al.*, 2012; Previtali *et al.*, 2014; Raidl & Klein, 1983).

Nutritional impact of the incorporation of legume flour

The optimization of the composition of all cereal products by incorporating legumes results in
beneficial nutritional properties (table 2) starting with an increase in protein, fiber and mineral
content. The increase in protein content is associated with an improvement in the essential amino
acid score, notably that of lysine, which depends on the type or on the amount of legume
incorporated (Laleg, Barron, Cordelle, *et al.*, 2017; Sparvoli *et al.*, 2016). Even if lysine essential amino
acid is reduced by the Maillard reactions that take place during the thermal treatment, it is still
higher in legume enriched products than in wheat products (21 mg/g of protein in low temperature
wheat pasta and 33 mg/g of protein in very high temperature mixed faba bean pasta) (Laleg, Barron,
Santé-Lhoutellier, *et al.*, 2016). *In vitro* protein hydrolysis is also impacted; increases of 11% in 15%
green pea snacks (Patil, Brennan, Mason, & Brennan, 2016) and of 24% in 100% faba bean pasta
(Laleg, Barron, Cordelle, *et al.*, 2017) have been reported. In the case of pasta, this could be
explained by the molecular weakening of the protein network structure that facilitates accessibility
of proteolytic enzymes to specific protein cleavage sites (Laleg, Barron, Cordelle, *et al.*, 2017). Only a
few studies of the effect of legume enrichment on *in vivo* protein digestion of foods can be found in
the literature (Fleming & Sosulski, 1977b; Torres, Frias, Granito, Guerra, & Vidal-Valverde, 2007;

Torres, Frias, Granito, & Vidal-Valverde, 2007). No impact of the incorporation of legume flour on protein true digestibility was reported in these studies that focused on pasta and bread. However increases in the protein efficiency ratio (amount of weight gain per unit of protein consumed) have been reported, from +53% for bread (Fleming & Sosulski, 1977b) to +64% to +86% for pasta (Torres, Frias, Granito, Guerra, *et al.*, 2007; Torres, Frias, Granito, & Vidal-Valverde, 2007).

The digestive behavior of the starch fraction is more widely documented. Generally, the incorporation of legume flours reduces the *in vitro* glycemic response in any product (Gularte, Gómez, & Rosell, 2012; Laleg, Cassan, Barron, *et al.*, 2016; Sparvoli *et al.*, 2016). However, the same difference measured *in vivo* is not statistically significant in many wheat products (Fujiwara, Hall, & Jenkins, 2017; Greffeuille *et al.*, 2015). Only studies on pasta by Goñi and Valentín-Gamazo (2003) and on bread by Zafar *et al.* (2015) reported a significant decrease in the *in vivo* glycemic index (bread and pasta), although enriched products generally still have low *in vivo* glycemic and insulin indexes (Greffeuille *et al.*, 2015).

Legume flour enrichment is also an effective way to provide micronutrients and bioactive compounds. Some of these bioactive compounds, including protease inhibitors, phytic acid and α -galactosides, are considered as “anti-nutritional factors” because of their ability to reduce protein digestibility, nutrient absorption and to cause intestinal discomfort, respectively (Duranti, 2006; N. Wang *et al.*, 2009). The amount of bioactive compounds in legumes can be significantly reduced by preprocessing (soaking, fermentation and germination) or during standard processing. Indeed, the inhibitory activity of trypsin is almost completely inactivated in cooked pasta. Total α -galactosides (44 to 57 mg/g db, in lentil, faba and black gram flours) are also remarkably reduced (-41% to -71%) (Laleg, Cassan, Barron, *et al.*, 2016). However, no effect of these processing steps has been reported for phytic acid. Germinating *Cajanus cajan* seeds led to a 58% reduction in phytic acid (Torres, Frias, Granito, & Vidal-Valverde, 2007). Finally, interest has recently focused on the nutritional function of the bioactive compounds supplied by legumes as intestinal, metabolic or probiotic/prebiotic factors.

In conclusion, the incorporation of legume flour in cereal products has a positive impact on their nutritional properties. The change in color does not appear to be a problem. The off-flavor reduces the acceptability score, but could be limited by the use of flavoring agents. Finally the change in texture, which is linked to the structure, needs to be controlled. A good knowledge of transformations that occur during the process will help control the final structure and texture.

From the raw materials to the end product: structure building by the action of the process

Simplified state diagram for protein and starch of legumes and cereal and process paths of five main cereal products

In the following review, we refer to a simplified state diagram summarizing the thermal transitions of both wheat and legume components (Fig. 3a) to propose hypotheses concerning the effect of incorporating legume flour on the structuring and the resulting structure of each type of product. The four zones in the diagram correspond to different physical states of the material. Zones 1 and 2 refer to the glassy and rubbery states separated by the average glass transition lines. The shift from the lower part of the diagram (zone 2) to the upper part (zones 3 and 4) corresponds to the general softening of the material toward the liquid state that is related to the melting of starch crystallites (Cuq et al., 2003), and may be emphasized by the presence of shear. Zones 3 and 4 differ in the viscosity of the liquid as a function of the variation in water content. When thermosetting of the proteins occurs, leading to the formation of a network, the product in the upper part of the state diagram (zones 3 and 4) can be qualified as soft solid. It should be noted that dependence of transition temperatures on water content is taken into consideration for each component separately as if it had free access to total water in the product with no competition from other components or ingredients. This is a simplified approach to the distribution of water in the product (Lazou & Krokida, 2011). Furthermore, the effect of sugar on the increase in the starch melting temperatures and the decrease in the starch and protein glass transition temperatures has been well described (Roos &

Drusch, 2015). Such an effect should be taken into account in the case of biscuits and soft cakes that contain 20% and 30% sugar wb, respectively (table 1).

[FIGURE 3]

[TABLE 3]

Table 3 compares the water content and temperature of the products during the course of processing. Processing is summarized in four steps: raw material, mixing before thermal treatment, hot product and end product. For each step, the link with the appropriate zone in the state diagram (Fig. 3a) is defined based on water content and temperature. This comparison illustrates the wide range of water contents (from 15% to 43% wb) of the flour or semolina during the processing of five main cereal products, in the maximum product temperature (from 100 °C to 250 °C) and in the final water content (from 1% to 69% wb, with a difference between the core and the surface of the product, or not).

Based on these data, the path of each product in the simplified state diagram can be drawn (Fig. 3). Each process leads to the formation of a final structure that is schematically represented in figure 4. We now describe the path of each product to show how closely it is related to structure building during the course of processing. The potential impact of incorporating legume flour, together with the above mentioned implications are also discussed. In a material science approach, the changes in the products depending on variations in temperature and moisture during processing are described in the order of increasing final water content of the product.

[FIGURE 4]

The temperature-hydration history and its effect on the final structure of snacks

In snacks (Fig. 3b) the initial semolina is rather poorly hydrated (15% wb). In the extruder barrel, it is highly sheared (SME of 300-900 kJ/kg) resulting in a rapid and large increase in pressure and

461 temperature (Della Valle, Kozlowski, Colonna, & Tayeb, 1989). The product goes through all the
462 transition temperatures (T_g , T_m , T_d) leading to the formation of a very viscous liquid called “melt”
463 with the disruption of starch granules but also the depolymerization of proteins, amylose and
464 amylopectin (Colonna, Doublier, Melcion, De Monredon, & Mercier, 1984). The melt is pushed
465 through the extruder die (zone 4 on Fig. 3b) and returns to room pressure while still very hot. Flash
466 vaporization of water occurs accompanied by rapid expansion of the product corresponding to the
467 formation of numerous large bubbles (Kokini, Chang, & Lai, 1992). Melt viscosity is a key parameter
468 in snack expansion and is influenced by the size and structure of starch and protein macromolecules,
469 even if these are depolymerized. In the case of legume enriched melts, viscosity could be reduced by
470 the lower molecular mass of legume proteins, and by the higher amylose/amylopectin ratio of
471 legume starches, even though the impact of the latter factor is still the subject of debate in the
472 literature (Moraru & Kokini, 2003). A lower melt viscosity could increase the expansion of the matrix.
473 Upon bubble growth, stress is exerted on the cell walls that open and holes appear (Fig. 4).
474 Moreover, depending on their resistance to expansion stress, some walls may completely disappear.
475 Water vapor evacuates thereby accelerating cooling and causing the stiffening of the walls as
476 proteins and starch return to a glassy state (Kokini *et al.*, 1992) (Fig. 3b). Glass transition should occur
477 at a sufficiently high temperature to prevent the bubbles collapse. Because legume enriched snacks
478 contain significantly more proteins than 100% wheat snacks, one can hypothesize that the glass
479 transition temperature of the mixed protein + starch matrix would decrease towards the T_g of
480 proteins (Fig. 1). This would delay stiffening after the opening of the structure, resulting in shrinking
481 of the snack and consequently a lower final volume. In this case, the final water content of the snack
482 is low (1-5% wb); its texture is crispy. At macroscopic scale, the snack is an open solid foam with large
483 interconnected cells (Fig. 4). At microscopic scale, the cell walls are composed of a continuous phase
484 made of a mix of proteins, amylose and amylopectin in the amorphous state.

The temperature-hydration history and its effect on the final structure of biscuits

In biscuits, the dough is poorly hydrated at the mixing step (16% wb). The presence of sugar in the dough reduces the glass transition temperatures of both proteins and starch (Roos & Drusch, 2015). Thus, at the mixing step, proteins are nearly able to undergo glass transition whereas starch is not (Fig. 3c). Biscuit dough can be qualified as a homogeneous mix of swollen flour particles and fat globules in an aqueous phase with high concentrations of sugar (Chevallier, Colonna, Buléon, & Della Valle, 2000). During baking, two paths can be distinguished depending on the changes in temperature. The surface of the product rapidly reaches high temperatures (~200 °C) that cause glass transition in proteins and in starch with no denaturation or gelatinization phenomena because of the limited water content. The core of the product reaches a lower temperature (~125 °C) that only allows glass transition of proteins. Given that biscuits are not subject to either protein thermosetting or to starch gelatinization (Chevallier, Colonna, Della Valle, & Lourdin, 2000), replacing wheat flour with legume flour would be expected to have little impact on structuring. During baking, the reaction of raising agents can lead to the formation of small air bubbles. After baking, the surface and core of the product return to the glassy state. The product has low water content (1-5% wb) and a crunchy texture. At macroscopic scale, the biscuit is a dense matrix that is homogeneous between the core and the surface. At microscopic scale, the matrix contains unpasted starch granules, fat globules, and small air cells entrapped in a continuous, highly concentrated phase of sugar and proteins (Fig. 4).

The temperature-hydration history and its effect on the final structure of pasta

In pasta (Fig. 3d), the combination of 32% hydration and low shear stress during the mixing step transform the semolina into a sandy dough, in which both starch and proteins are in the rubbery state and no gluten network is detectable (Petitot, Abecassis, & Micard, 2009) (Fig. 3d). The extrusion step (SME from 70-120 kJ/kg and pressure up to 17.6 MPa in a single screw extruder) forms a dough with a partial gluten network and sufficient compactness to survive cooking. Several thermal treatments are possible to dry the pasta (Fig. 3d). A low temperature treatment (40-60 °C/70-80%

relative humidity/18-28 h) returns the protein and starch to the glassy state without causing protein thermosetting. Conversely, a high temperature treatment (> 84 °C/74-90% relative humidity/2-5 h) allows gluten proteins to undergo thermosetting before reaching the glassy state. In a 0% to 100% legume-enriched pasta dried at low temperature, the percentage of proteins linked by weak bonds (hydrogen, hydrophobic) and covalent bonds (notably by disulfide bonds) linearly increased and decreased, respectively (from 70% to 94% and 30% to 5% respectively in faba bean-wheat pasta) inherent to the smaller quantity of cysteine residues in legume proteins (Laleg, Barron, Cordelle, *et al.*, 2017). When dried at high temperature, these differences between legume enriched and wheat pasta due to composition persisted. In addition, the proportions of covalently linked proteins in all the pasta increased when the drying temperature reached or was above the Td of wheat protein, and higher than the higher Td of legume protein. Only partial gelatinization of starch (20%) may occur (Güler, Köksel, & Ng, 2002) (Fig. 3d). The complete gelatinization of starch and protein thermosetting will occur during the final cooking step (mentioned in table 3 but not in Fig. 3d) in boiling water, and at higher temperatures when legume flour is incorporated. Ready to eat pasta is soft and viscoelastic. Its microscopic structure (Fig. 4) is characterized by swollen starch granules held in a compact and poorly porous protein network.

The temperature-hydration history and its effect on the final structure of soft cakes

In soft cakes, at the mixing step, the batter is relatively highly hydrated (30% wb), which allows proteins and starch to go into the rubbery state (Fig. 3e). Due to the low proportion of flour in the formula (table 1) and to the design of the mixer, no gluten network is formed (Dewaest, Villemejeane, Berland, Michon, *et al.*, 2017). Nevertheless, mixing incorporates air in the form of small bubbles that are stabilized in the batter thanks to surface active compounds (Sahi & Alava, 2003). The paths at the surface of the product and in the core differ during baking. The surface dries rapidly, leading to the formation of the crust (Fig. 3e) that limits water transfers between the core and the oven atmosphere (Lostie, Peczalski, Andrieu, & Laurent, 2002). The surface temperature reaches high temperatures (180-210 °C) and the proteins thermoset but the starch granules do not gelatinize (Fig.

3e). The water content of the product core remains constant while the temperature slowly increases up to 100 °C. Bubbles grow thanks to water vaporization, carbon dioxide formation, and gas dilatation. Like for snacks, bubble growth is associated with structure opening due to the formation of holes in the cell walls and to the disappearance of the cell walls (Dewaest, Villemejane, Berland, Neron, *et al.*, 2017; Lostie *et al.* (2002)) (Fig. 4). However, no effect on expansion has been proven, as opening occurs when temperature and expansion pressure are still high. Cake expansion is limited by the formation of the crust and further stopped by protein thermosetting and starch gelatinization in the core that set the structure (Fig. 3e). In cakes, pasted starch granules called “bricks” are considered to be the first structuring element of the cell walls, with denatured proteins forming the “mortar” between the bricks (Donovan, 1977) (Fig. 4). In legume-enriched cakes, starch content is lower which means the proportion of structuring elements in the cell walls is lower. There is also an increase in the thermosetting temperature that could delay the increase in viscosity and consequently promote cake expansion before setting. The result depends on the balance between expansion and stiffening kinetics. After baking, the product surface (crust) changes to the glassy state while the core (crumb) remains at the rubbery state. Usually in industry, the product is rapidly placed in hermetic packaging, which leads to water redistribution within the product (last arrows in Fig. 3e). At macroscopic scale, the crumb can be qualified as a soft open solid foam made of small interconnected cells with holes in their walls (Dewaest, Villemejane, Berland, Neron, *et al.*, 2017) (Fig. 4).

The temperature-hydration history and its effect on the final structure of bread

In bread, the phenomena are the same as in cakes with two paths (crust and crumb) in the state diagram. However they differ in two important points. First, thanks to the supply of mechanical energy (10-30 kJ/kg) and to the design of the mixer, gluten proteins are able to form a 3D network that gives the dough higher elastic properties (Feillet, 2000). When legume flour is incorporated, the presence of lower molecular mass proteins that are unable to form a network by themselves and might thus affect structuring (Boye, Zare, *et al.*, 2010). Second, during the fermentation step, the

562 bubbles start increasing in size at room temperature because yeast produces carbon dioxide. During
563 baking, the bubbles continue to expand thanks to water and carbon dioxide vaporization and gas
564 dilatation. Holes appear in the cell walls but in bread, the cell walls are more resistant to deformation
565 than in cake thanks to their viscoelastic properties (Fig. 4). Loaf expansion is stopped by the
566 formation of a crust that is more rigid than that of cake, and by the setting of the core (Fig. 3f). The
567 crust goes into the glassy state and becomes brittle when it returns to room temperature. The core
568 remains in a rubbery state and becomes a soft viscoelastic crumb with relatively high water content
569 (40-50% wb). At macroscopic scale, the crumb can be qualified as a soft open solid foam, made of
570 rather large cells with holes in their walls (Fig. 4). At microscopic scale, the cell walls are made of
571 pasted starch granules and a denatured gluten network that may be diluted by legume proteins
572 (Fleming & Sosulski, 1978; Mohammed *et al.*, 2012).

573 In conclusion, this material science generic approach allows the formulation of hypotheses
574 concerning the effects of incorporating legume flour on the structure of cereal products. However,
575 analyzing such effects is no trivial task, due to the combination of various structuring phenomena at
576 different length scales. To illustrate this complexity, in the following section, we include a
577 supplementary analysis of the incorporation of legume flour in order to describe the differences in
578 behavior we observed during processing, to discuss the validity of some of the previously proposed
579 hypotheses, and to propose changes in the process to insure the quality of such products. It aims at
580 giving an additional highlight to the generic approach developed so far, for two of the five cereal
581 products.

Incorporating legume flour in wheat foods requires modifying food processing: examples of low hydrated (pasta) and hydrated (cakes) products

The examples below illustrate the effects of product-process interactions on the structure of pasta and cake during dough formation and thermal treatment at different length scales.

During dough formation

Legumes incorporated in the pasta dough act as a diluting agent for the gluten network and slightly weakened the protein interactions (from 11% in wheat to 7% in 35% legume enriched pasta) at molecular scale (Petitot, Barron, *et al.*, 2010). At macroscopic scale, a formation of large sticky heterogeneous lumps of dough (particle size > 1-4 mm) was observed upon hydration and mixing, rendering the extrusion step difficult especially when the proportion of legume flour was greater than 35% (Petitot, Boyer, *et al.*, 2010; Wood, 2009). This can be attributed to the difference in particle size of legume flour and semolina that can affect their hydration properties and increase dough heterogeneity. Moreover, the higher proportion of protein and enzyme in legume flour can modify its granulation properties (Laleg, Cassan, Abecassis, & Micard, 2016). An increase in mixing speed from 60 to 120 rpm and a decrease in dough hydration from 47% db in wheat pasta to 45-40% db in pasta containing at least 35% of legume have been suggested as a way of limiting the formation of aggregates and lumpy dough (Petitot, Boyer, *et al.*, 2010). New alternatives are proposed under patent WO 2016097328 A1 (Laleg, Cassan, Abecassis, *et al.*, 2016).

In the processing of industrial soft cakes (e.g. layer cakes or pound cakes), batter can be formed by multi-stage mixing of dry ingredients (flour, sugar, emulsifiers) with egg and water, and with fat in the form of liquid oil. During the first step, the flour is hydrated and a foam is created. At macroscopic scale, this leads to the formation of a continuous phase of hydrated proteins in which air bubbles, fat droplets, and starch granules are dispersed. Proteins and starch undergo glass transition. At this level, the ability of wheat or legume proteins to solvate or hydrate determines batter viscosity, which is a fundamental criterion for cake expansion (Sahi & Alava, 2003). Concerning

foaming at mesoscopic scale, the presence of 20% to 40% of pea flour in a mix with wheat flour reduced batter density (-12% and -16% of batter density respectively compared to 100% wheat batter in a pound cake recipe, Monnet *et al.*, unpublished results). The better interfacial properties of legume proteins enabled more efficient stabilization of the bubbles than wheat proteins. One way to incorporate the same amount of air at this step would be to reduce the duration of mixing when pea flour is used. During the second step of mixing, an oil-in-water emulsion is formed. Usually a partial loss of air can be observed at this step due to the antifoaming properties of oil. After oil is incorporated, the batter densities are equivalent (-1% or -2% with respectively, 20% or 40% of pea flour). Thus, the higher surface activity of pea proteins does not seem to prevent the antifoaming action of oil. However, in layer cakes made with a single-stage mixing method, Gómez *et al.* (2008, 2012) and De la Hera *et al.* (2012) reported significant decreases in batter density with the incorporation of 50% to 100% of chickpea, pea or lentil flour (-3% to -10% of density depending on the proportion, the cultivar and particle size of the legume concerned). Beyond the quantity of air at macroscopic scale, incorporating legume flour may affect the size and distribution of air bubbles in the batter. Gómez *et al.* (2012) showed that the distribution of air in batters containing 50% or 100% of pea flour was more stable with smaller bubbles. In turn, this could impact cell structure in the cake and hence its global structural and textural properties.

During thermal treatment

In standard processing of wheat pasta, drying promotes the aggregation of protein polymers through disulfide bonds, leading to the formation of larger protein aggregates (Petitot, Barron, *et al.*, 2010). The extent of protein aggregation depends on the drying temperature (55 °C vs. 90 °C). At a low drying temperature (55 °C), as there is no thermosetting of either wheat or legume proteins, incorporating legume flour has no other impact than gluten dilution. The low drying program does not provide the right conditions for legume proteins to interact with each other covalently or with gluten via disulfide bonds. At a higher drying temperature (90 °C) additional covalent linkages (mainly disulfide, isopeptide or Maillard products) involving both wheat and legume proteins can be created

633 (Laleg, Barron, Cordelle, *et al.*, 2017). A proportion of between 35% and 100% faba bean pasta dried
634 at a very high temperature was subject to fewer covalent protein interactions than wheat pasta.
635 However, the increase in covalently linked proteins at very high temperature versus low temperature
636 drying conditions was greater when the amount of faba bean proteins was increased from 0% to
637 100%, showing that very high drying temperature enables additional reactivity of faba proteins.

638 Cooking results in extensive protein aggregation through covalent bonds in both wheat and in
639 legume enriched pasta. This reduces the difference in network structuring between pasta with and
640 without legume flour even if one difference can still be observed: a significant increase in losses
641 during cooking, depending on the proportion of legume flour added, has been reported for pasta
642 enriched with different types of legumes (lentil, pea, black gram, white bean and faba bean) (Laleg,
643 Barron, Cordelle, *et al.*, 2017; Laleg, Cassan, Barron, *et al.*, 2016; Rosa-Sibakov *et al.*, 2016; Wójtowicz
644 & Mościcki, 2014). The losses were linked to several phenomena, including the weaker final protein
645 structure due to the reduced capacity of legume proteins to covalently interlink, to the disruption of
646 gluten matrix by the higher quantity of fiber provided by legume flour (Petitot, Barron, *et al.*, 2010),
647 and to the increased proportions of salt-soluble proteins resulting from the incorporation of legume
648 flour (Petitot, Boyer, *et al.*, 2010). Drying pasta at a high temperature can help counteract cooking
649 losses notably by increasing the proportion of proteins linked by covalent linkages in the pasta
650 protein network (Laleg, Barron, Cordelle, *et al.*, 2017).

651 When soft cakes are baked, wheat glutenins and gliadins bind to each other through disulfide bonds
652 and egg proteins do the same separately. It is only at the very end of baking that both kinds of
653 proteins interact through disulfide bonds (Dewaest, Villemejane, Berland, Michon, *et al.*, 2017). This
654 interaction reduces the proportion of non-extractible proteins due to steric hindrance (Dewaest,
655 Villemejane, Berland, Michon, *et al.*, 2017). When 20% to 40% of pea flour was incorporated in a
656 pound cake recipe, an almost linear increase in the proportion of proteins linked by weak
657 interactions was observed (+ 43% and + 81% respectively, Monnet *et al.*, unpublished results). This

658 increase was offset by a decrease in the proportion of covalently linked proteins (-7% and -13%,
659 respectively) however, with little variation in the proportion of proteins linked by disulfide bonds (-
660 3% and +1%). This is inherent to the structure of legume proteins. Thus, introducing legume proteins
661 may weaken the force of interaction between proteins, especially their capacity to strongly bind to
662 wheat and egg proteins. At macroscopic scale, incorporating pea flour increased the density of the
663 cake (+3% for 20% or 40% compared to the control). At microscopic scale, the distribution of air
664 among the cell walls appeared to be more homogeneous, and the cell walls appeared to be thicker.
665 This suggests later and/or less efficient stiffening of the structure (due to a delay in thermosetting
666 and/or reduced starch content) that may have caused shrinking of the cells and thickening of the cell
667 walls during baking.

668 These two examples show how, by changing the nature, proportion, and functionalities of the
669 components (especially the binding abilities of proteins), incorporating legume flour can result in
670 differences in the structure building process, thereby elucidating the product-process interactions
671 that occur from the raw material to the end product.

672

Conclusion

Mixing legume and cereal flour leads to process different nature and amounts of their two main components, starch and proteins. Even though our approach, which consists in considering their distinct thermal transitions independently (one from another, and from other components and ingredients) may limit our overall understanding, the theoretical differences in their transition temperatures enabled us to propose preliminary hypotheses concerning the multi-scale structure building during baking or drying in connection with expansion properties in the case of aerated products. Comparing these hypotheses with results in the literature and with our own experience provided information on the effects of incorporating legume flour in standard cereal products at macroscopic, mesoscopic and macromolecular scales. Yet it is clear that establishing relationships between phenomena at different length scales is far from simple. Considering the information collected at different length scales, it can be assumed that incorporating legume flour has a significant effect on product structuring during processing. Knowledge about how to adapt process parameters to counteract such effects is already partially available. For snacks, barrel temperature, screw speed, feed rate and moisture content can be adjusted to modulate melt viscosity and expansion with respect to stiffening. For pasta, an increase in mixing speed and a decrease in hydration of the dough are recommended to limit the formation of lumps, and more drastic drying conditions to compensate for the weakening of protein interactions and the dilution of the gluten network. For cake, adjustments in mixing speed or in the order of incorporating the ingredients during batter formation could make it possible to adjust the amount and the distribution of air, and improvements could be made in structure setting by changing baking parameters (temperature, air speed). Finally for bread, dilution of the gluten network could be counteracted by sequential mixing to enable the formation of the protein network before the legume flour is added. The use of a structuring agent, such as a gum or animal proteins (ovalbumins) to imitate gluten structure is a possible alternative. In this way, wheat and legume flours could be successfully combined in a wide variety of cereal products. Aside from structural aspects, the nutritional quality of legume-enriched

products is significantly improved. Adverse sensory effects are still an obstacle to the wider use of these products, but it is believed that such effects could be counteracted by proper aromatic formulation, and should thus not be considered as limiting. Finally, we propose an approach that could help predict the effect of replacing wheat flour with legume flour and make it possible to control the structure and texture of the product at each stage of processing, depending on the path taken by the components in the state diagram. This approach should be completed by studies of thermal transitions in real formula conditions, as well as by detailed studies on the structural changes that occur at microscopic and macromolecular length scales in all cereal products.

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1031 **FIGURE CAPTIONS**

1032 **Fig. 1** Summary of data on glass transition temperatures (T_g) and denaturation temperatures (T_d) of wheat and
1033 legume proteins from different sources in the literature.

1034 * T_g range of wheat gluten adapted from Cuq *et al.* (2003)

1035 ** Average T_d value for legume proteins calculated from data from five studies on protein isolates or 7S and
1036 11S globulins between 80% and 94% water (wb), for 11 different legume species and 3 heating rates (2, 5 and
1037 10 °C/min). The error bars show the standard deviation associated with the calculation (T_d = 88 °C ± 7 °C).

1038 Sources: Kitabatake, Tahara, & Doi (1990); L'hocine, Boye, & Arcand (2006); Shevkani, Singh, Kaur, & Rana
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1040 *** Extrapolation of T_d ranges for wheat and legume proteins at high water contents (40-100% water wb)
1041 *All temperatures correspond to peak temperatures of endotherms measured by differential scanning*
1042 *calorimetry.*

1043 **Fig. 2** Summary of data on glass transition temperatures (T_g) and melting temperatures (T_m) of wheat and
1044 legume starches from different sources in the literature.

1045 * T_g range of glass transition temperatures in wheat starch, adapted from Cuq *et al.* (2003)

1046 ** Ranges of T_{m1}, T_{m2}, T_{m3} in wheat starch, adapted from Vanin *et al.* (2009)

1047 *** Average T_{m1} value for legume starches calculated from data from four studies between 70% and 82%
1048 water on a wet basis (wb), for 11 different legume species and 1 heating rate (10 °C/min). The error bars show
1049 the standard deviation associated with the calculation (T_m = 70 °C ± 5 °C). Sources: Chung, Liu, Hoover,
1050 Warkentin, & Vandenberg (2008); Hoover and Ratnayake (2002); Singh, Sandhu, Kaur, Singh Sandhu, & Kaur
1051 (2004); Zhou, Hoover, & Liu (2004).

1052 *All temperatures correspond to peak temperatures of endotherms measured by differential scanning*
1053 *calorimetry.*

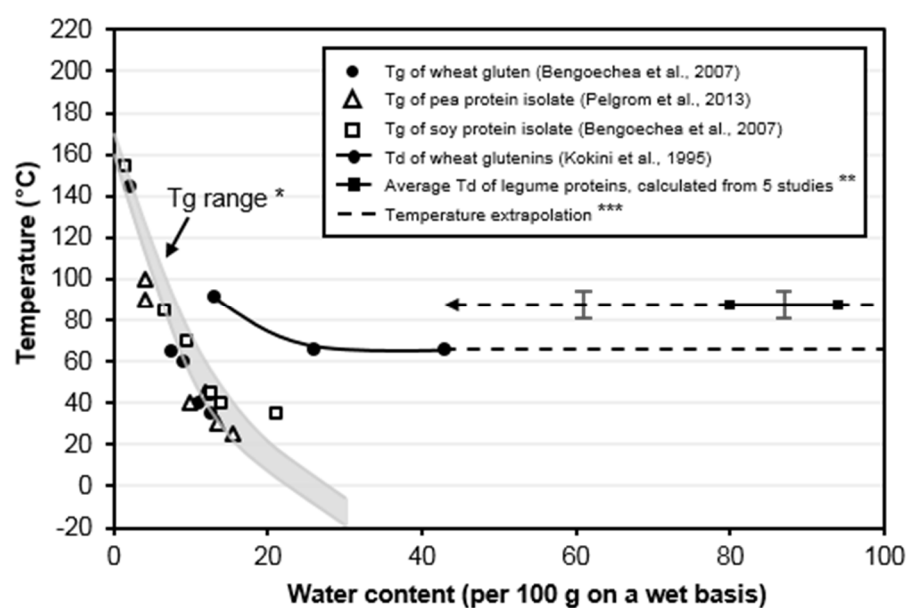
1054 **Fig. 3** Simplified state diagram for starch and proteins with four zones (a) and paths of the products: snacks (b),
1055 biscuits (d), pasta (d), soft cakes (e), bread (f).

1056 *Standard deviation bars in (a) show the ranges of variation of transition temperatures shown in figures 1 and 2.*

1057 **Fig. 4** Schematic representation of all structures at two length scales. Distribution of the products according to
1058 water content and type of structure (presence of air in the form of cells interconnected to varying degrees, or
1059 absence of air).

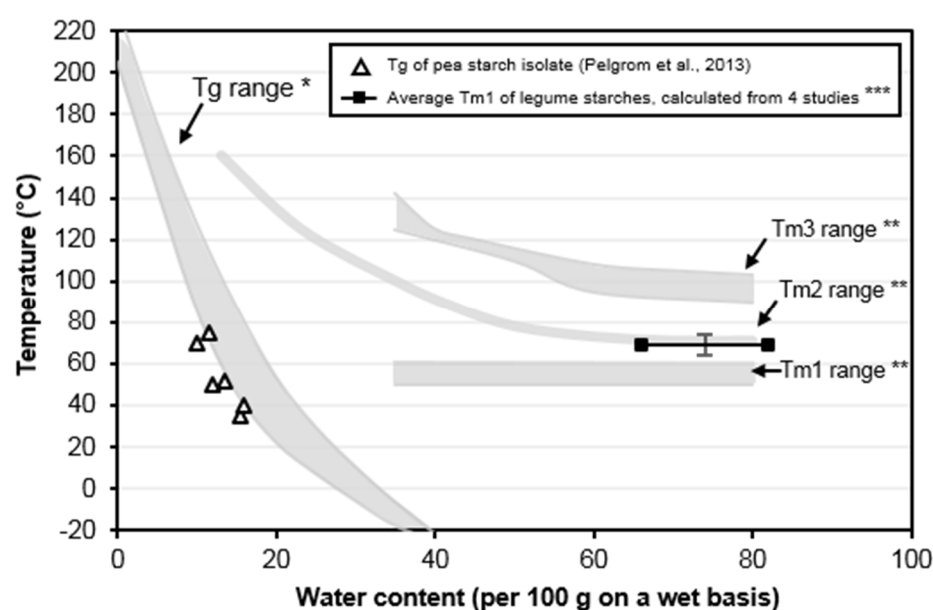
1060

1061 **FIGURE 1**



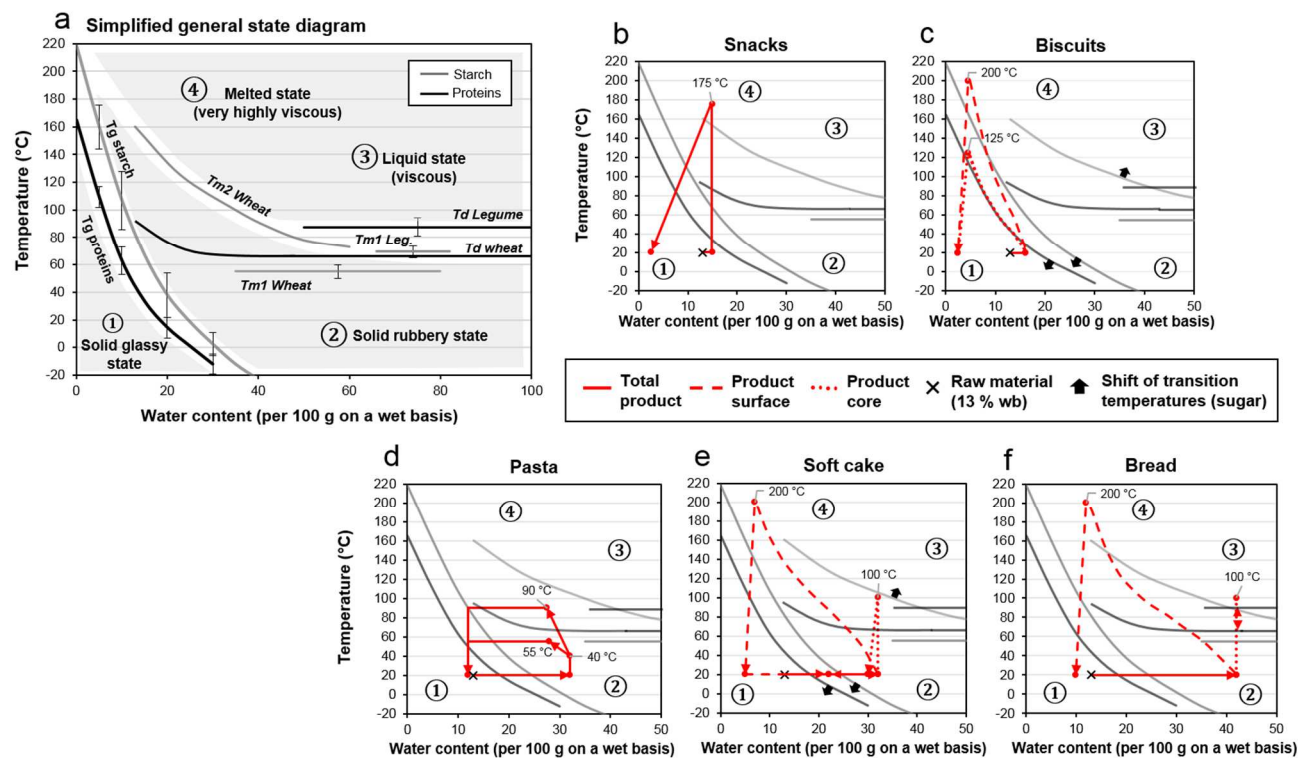
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1063 **FIGURE 2**



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1065 **FIGURE 3**



1066 **FIGURE 4**

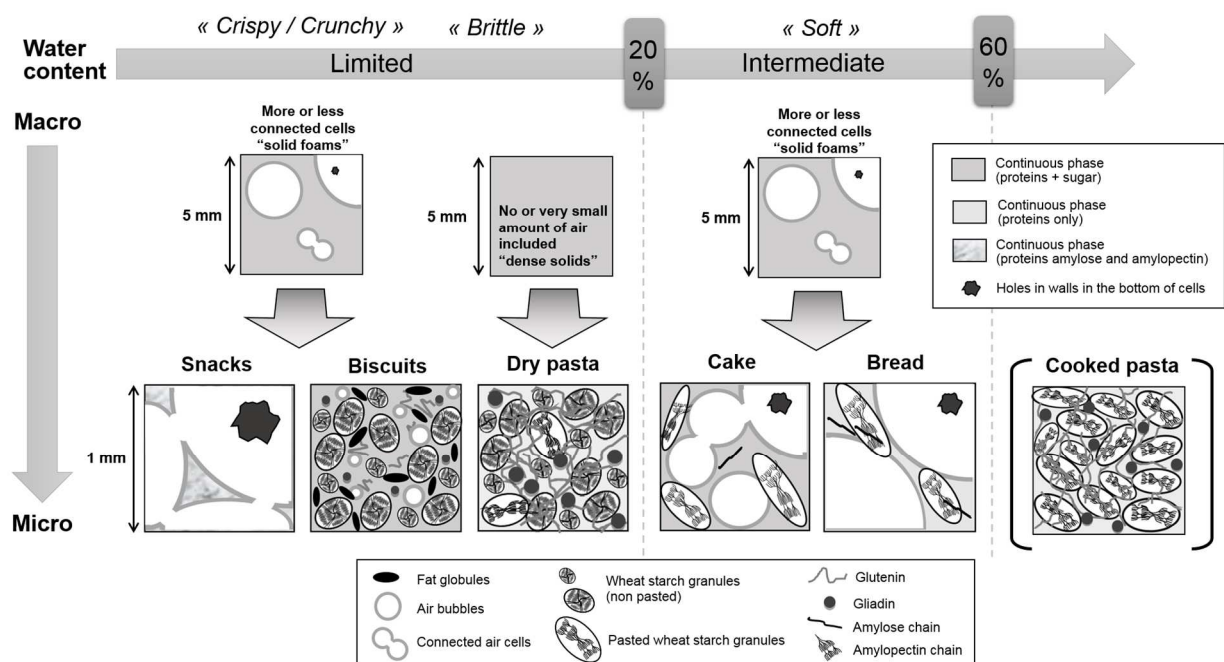


Table 1. Main characteristics of wheat product formulas before processing						
		Snacks ^a	Pasta ^b	Bread ^c	Biscuits ^d	Soft cakes ^e
Major ingredients (g/ 100 g of total formula)	Wheat flour or semolina	97	78	60	50	29
	Sugar	-	-	*	20	30
	Fat	-	-	*	20	9
	Egg	-	-	-	-	15
	Water	3	22	35	10	17
Other ingredients				Yeast, Salt	Raising agents, Emulsifiers*,Salt	
Components (g/ 100 g of total formula)	Total water	15	32	43	16	32
	Starch	61	49	38	31	18
	Proteins	11	9	7	6	3 (wheat) 2 (eggs)
	Lipids	1	1	1	21	11
	Total dietary fibers	2	2	2	1	1
^a Kaletunç and Breslauer (1996) ^b Petitot, Brossard, <i>et al.</i> (2009) ^c Alam <i>et al.</i> (2013); Heenan, Hamid, Dufour, Harvey, & Delahunty (2009); Shogren <i>et al.</i> (2003) ^d Short type; Heenan <i>et al.</i> (2009); Rababah, Al-Mahasneh, & Ereifej (2006); Zucco, Borsuk, & Arntfield (2011) ^e Layer type; De la Hera <i>et al.</i> (2012); Gularte <i>et al.</i> (2012) * Optional ingredients <i>Composition of the wheat flour chosen for calculations: 11.5% proteins, 63% starch, 2% fibers, 1.5% lipids, 13% water (USDA, 2016).</i>						

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Table 2. Impact of replacing cereals with different legumes on textural & structural, color, sensory and nutritional properties of the food products

Legume-enriched food	% Legume flour	Food properties			
		Texture & Structure	Color	Sensory attributes	Composition & Nutrition
Snacks ^a	0-90%	<ul style="list-style-type: none"> - ↓ expansion ratio - ↑ density and porosity - ↑ breaking strength index (N/mm²) and stiffness (Young's modulus) - ↓ crispness (distance of travel to F max during compression and percent deformation) - Scanning electron microscopy: ↓ number of cells, thickening of cells and cell walls 	<ul style="list-style-type: none"> - ↓ L - ↑ a* and b* 	<ul style="list-style-type: none"> - ↓ crunchiness 	<ul style="list-style-type: none"> - ↑ protein content (up to 12 fold) - ↑ antioxidant activity - ↑ phytic acid and trypsin inhibitors - ↑ <i>in-vitro</i> protein hydrolysis
Cooked pasta ^b	0-100%	<ul style="list-style-type: none"> - ↓ breaking energy, cohesiveness and resilience - ↑ stickiness - ↑ firmness and chewiness - Confocal laser scanning microscopy: no change in protein network at 35% - Brightfield microscopy: denser protein film, at 100% 	<ul style="list-style-type: none"> - ↓ L - = a* and b* 	<ul style="list-style-type: none"> - ↓ general appearance - ↑ fracturability, hardness and stickiness, and ↓ in bulkiness, elasticity, surface smoothness - ↑ adhesiveness and grittiness - ↓ taste acceptability - ↑ intensity of "beany" flavor - ↓ color acceptability - ↓ overall acceptability 	<ul style="list-style-type: none"> - ↑ protein content - ↑ amino acid score - = or ↓ glycemic index - = or ↓ rapidly available glucose - ↑ <i>in-vitro</i> protein hydrolysis - ↑ <i>in-vivo</i> protein digestibility and efficiency
Bread ^c	0-40%	<ul style="list-style-type: none"> - ↓ volume - ↑ density (↓ specific volume) - ↑ firmness or hardness - Scanning electron microscopy: disruption of protein-starch complex, thickening of cell walls, presence of holes in the walls 	<ul style="list-style-type: none"> - Crumb and crust: ↓ L, ↑ a* and b* 	<ul style="list-style-type: none"> - ↓ overall appearance acceptability - ↑ crumb denseness - ↑ size of the cells - ↑ crumb hardness, firmness - ↑ crust color - ↑ flavor intensity (e.g. "beany") - ↓ taste and odor acceptability - ↓ overall acceptability 	<ul style="list-style-type: none"> - ↑ protein, fiber and mineral contents - ↑ amino acid score - ↓ blood glucose response (glycemic index) - ↓ satiety - Rat growth assay : ↑ feed consumption, weight gain, net protein ratio and protein efficiency ratio
Cake ^d	0-100%	<ul style="list-style-type: none"> - ↓ volume - ↑ density (↓ specific volume) - ↓ symmetry - ↑ firmness or hardness (TPA) 	<ul style="list-style-type: none"> - Crust : ↓ L - Crumb : ↓ L, ↑ a* and b* 	<ul style="list-style-type: none"> - ↓ general appearance - ↓ flavor, aroma, persistency - ↓ texture acceptability - ↓ overall acceptability 	<ul style="list-style-type: none"> - ↑ total and available protein, mineral and fiber contents - ↓ rapidly available glucose
Biscuits ^e	0-100%	<ul style="list-style-type: none"> - Diameter, thickness, spread ratio: variable effect depending on type and % of legume incorporated - ↑ hardness (F max) and fracturability 	<ul style="list-style-type: none"> - ↓ L and ↑ b* above 20% - ↓ a* 	<ul style="list-style-type: none"> - ↑ color acceptability - flavor, aroma, overall acceptability: variable effect depending on type and % of legume incorporated - ↑ aftertaste (e.g. "beany") 	<ul style="list-style-type: none"> - ↑ protein, fiber and mineral contents - ↑ amino acid score - ↑ antioxidant activity - ↓ predicted glycemic index

^a Anton *et al.* (2009); Lazou *et al.* (2007); Lazou and Krokida (2010); Patil *et al.* (2016)

^b Goñi and Valentín-Gamazo (2003); Greffeuille *et al.* (2015); Laleg, Barron, Cordelle, *et al.* (2017); Laleg, Barron, Santé-Lhoutellier, *et al.* (2016); Laleg, Cassan, Barron, *et al.* (2016); Rosa-Sibakov *et al.* (2016); Wójtowicz and Mościcki (2014); Wood (2009)

^c Alam *et al.* (2013); Fleming & Sosulski (1978, 1977a, 1977b); Mohammed *et al.* (2012); Previtali *et al.* (2014); Raidl and Klein (1983); Shogren *et al.* (2003); Villarino *et al.* (2015); Zafar *et al.* (2015)

^d Campbell, Euston, & Ahmed (2016); De la Hera *et al.* (2012); Gómez *et al.* (2008); Gómez *et al.* (2012); Gularte *et al.* (2012)

^e Jayasena and Nasar-Abbas (2011); Mcwatters (1978); Noor Aziah *et al.* (2012); Patel and Rao (1995); Sparvoli *et al.* (2016); Zucco *et al.* (2011)

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Table 3. Changes in water content and in the temperature of the five products and the corresponding zone in the state diagram (Fig. 3)					
	Snacks ^a	Biscuits ^b	Pasta ^c	Soft cakes ^d	Bread ^e
Flour or Semolina	① 13% wb Room temp.	① 13% wb Room temp.	① 13% wb Room temp.	① 13% wb Room temp.	① 13% wb Room temp.
After mixing/kneading or during extrusion: Dough, Batter or Mix	② 15% wb Room temp. 300-900 kJ/kg	① 16% wb Room temp. 5-10 kJ/kg	② 32% wb Mixing: Room temp. Extrusion: 40 °C single screw 70-120 kJ/kg twin-screw 900 kJ/kg	② 32% wb Room temp.	② 43% wb Room temp. 10-30 kJ/kg
During baking or drying : Hot product	④ Surface & core: 150-200 °C at the die	② Surface: 180-220 °C Core: 100-150 °C	② ③ [Drying] Surface & core: 55-90 °C	② Surface: 180-210 °C Core: 100 °C	④ ③ Surface: 200-250 °C Core: 90-100 °C
			③ [Cooking] 100 °C		
After cooling and equilibration: End product	① 1-5% wb Room temp.	① 1-5% wb Room temp.	① [Dried] 12% wb Room temp.	① 22% wb Room temp.	① ② Crust: 5-15% wb Crumb: 40-50% wb Room temp.
			② [Cooked] 69% wb Consumption temp. (~ 50 °C)		

^a Della Valle *et al.* (1989); Kaletunç and Breslauer (1996)

^b Feillet (2000); Wade and Davis (1964)

^c Abecassis, Abbou, Chaurand, & Morel (1994); Petitot, Brossard, *et al.* (2009)

^d De la Hera *et al.* (2012); Dewaest, Villemejeane, Berland, Michon, *et al.* (2017); Lostie *et al.* (2002)

^e Cuq *et al.* (2003); Feillet (2000); Vanin *et al.* (2009)

In some cases, the specific mechanical energy (SME) (kJ/kg) is given.

A standard moisture loss of 2% was used for the cooling step based on data on bread from Feillet (2000). Moisture loss during snack cooling was 12.5%. Moisture loss during pasta cooling and bread/cake crumbs during baking were considered negligible. Moisture loss during baking or drying in all other cases was calculated to obtain a water content after cooling of + 2%.