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Functional properties of cellulose derivatives to tailor a model sponge cake using rheology and cellular structure analysis

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ABSTRACT

The functional properties of cellulose derivatives were studied to understand their specific and respective roles in the structure formation at each step of the process of a model sponge cake composed of hydroxy propyl methyl cellulose (HPMC) and methylcellulose (MC) and native corn starch. During batter preparation, the shear-thinning behavior and the quantity of air in the final product increased with the HPMC/MC ratio. HPMC controlled the viscosity of the system and limited bubble loss during starch addition. However, the lower the batter density, the higher the inhomogeneity of the final structure. During baking, the MC governed the sol-gel transition but HPMC slightly impeded it, and the gelling temperature decreased along with the HPMC/MC ratio. Because of the cold water binding capacity of starch, the concentration of HPMC/MC increased in the continuous phase during the batter preparation, decreasing the gelling temperature. A compromise in the HPMC/MC ratio giving a gelling temperature of about 54 \degree C was necessary to have a homogeneous final structure. Image analysis of the cellular structure confirmed the results by a classification of the products. The higher the MC concentration, the smaller the cells and the thicker the cell walls. An analysis of the effect of the mixing time after starch addition showed that the density after baking increased along with it. It was possible to obtain different crumb structures from a same composition by only changing the mixing time. Finally, a hypothesis about the structuring mechanisms of a model sponge cake at each step of the process was proposed using a schematic representation of the structure at three different scales and along the process steps.

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1. Introduction

Cellulose derivatives are widely used in different industry fields such as pharmaceutics, polymers or food. The possibility to substitute different radicals on the cellulose chain allows a broad range of functions. Cellulose derivatives have been largely used to improve the quality of cereal products [\(Abdel-Aal, 2009; BeMiller,](#page--1-0) [2011; B](#page--1-0)á[rcenas](#page--1-0) & [Rosell, 2005](#page--1-0)) in order to ensure numerous specific functional properties such as texture improvement ([Armero](#page--1-0) & [Collar, 1996a, 1996b](#page--1-0)), increase in batter viscosity ([Shelke, Faubion,](#page--1-0) & [Hoseney, 1990](#page--1-0)), slowdown of amylopectin retrogradation ([Davidou, Le Meste, Debever,](#page--1-0) & [Bekaert, 1996](#page--1-0)), increase in water retention [\(Rojas, Rosell,](#page--1-0) & [Benedito de Barber, 1999](#page--1-0)) or replacement of gluten functionality [\(Haque, Morris,](#page--1-0) & [Richardson, 1994; Toufeili](#page--1-0)

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[et al., 1994](#page--1-0)). Methylcellulose (MC) and hydroxypropyl methyl cellulose (HPMC) were specifically used in bakery products for different functional properties such as gelation, water binding, coating, emulsifying, stabilizing or foaming ([Wüstenberg, 2015\)](#page--1-0).

The behavior of MC and HPMC in solution was commonly analyzed by rheological measurement. MC and HPMC go through a sol-gel transition at hot temperatures because of associations between the hydrophobic domains on the polymer chains ([Haque](#page--1-0) [et al., 1994; Li et al., 2001; Zhang et al., 2014\)](#page--1-0). These associations are possible only if the substitutions in the cellulose chains are heterogeneous [\(Haque et al., 1994; Hirrien, Chevillard, Desbri](#page--1-0)è[res,](#page--1-0) [Axelos,](#page--1-0) & [Rinaudo, 1998](#page--1-0)). Their gelling properties depend on the type and degree of the substitutions, their molecular mass and their concentrations (Bárcenas & [Rosell, 2005; Nasatto et al., 2015b,](#page--1-0) [2015a](#page--1-0)). In any case, tri-substituted units are compulsory for the gel formation ([Hirrien et al., 1998](#page--1-0)). At low MC concentration and low temperature, the system is described to go through the sol-gel torresponding author. AgroParisTech, 1 avenue des Olympiades, 91744, Massy transition because of association of the most hydrophobic groups.
Edex France of association of the most hydrophobic groups.

Cedex, France.

When the temperature increases hydrophobic interactions appear between the methyl substitutes which leads to an increase in viscosity and turbidity of the system and to the gel formation ([Hirrien](#page--1-0) [et al., 1998; Kato, Yokoyama,](#page--1-0) & [Takahashi, 1978; Nasatto et al.,](#page--1-0) [2015a\)](#page--1-0). This phenomenon is enhanced for higher MC concentrations. MC gels are inhomogeneous at microscopic scale. They are organized in micro-phase separation regions [\(Chevillard](#page--1-0) & [Axelos,](#page--1-0) [1997; Nasatto et al., 2015a; Tanaka, 1996\)](#page--1-0) forming a fibrillary network. HPMC behavior is slightly different from MC when the temperature increases. First, when a critical temperature is reached (generally between 60 \degree C and 70 \degree C) the viscosity suddenly decreases, followed 20 \degree C higher by a small increase. [Bodvik et al.](#page--1-0) [\(2010\)](#page--1-0) proposed that the decrease in viscosity can be related to the formation of compact objects. These objects are probably made of associated chains through hydrophobic interactions as proposed by Haque et al., in 1994. The subsequent viscosity increase at higher temperatures would be due to rearrangements of HPMC aggregates leading to the formation of fibrillary structures, which would be more linear and less entangled than for MC [\(Bodvik et al., 2010\)](#page--1-0) resulting in a softer gel.

However, used in a more complex system than an aqueous solution (for example in cereal products), the behavior of these cellulose derivatives may be extensively modified due to interactions with other ingredients (starch, wheat proteins, sugar and egg) at the different steps of the process. Understanding the mechanism of these cellulose derivatives in real cereal products is uneasy. Most of the studies use macroscopic techniques like image analysis, texture measurement or sensory aspect [\(Ashwini, Jyotsna,](#page--1-0) & [Indrani, 2009;](#page--1-0) [B](#page--1-0) a[rcenas](#page--1-0) & [Rosell, 2005; Hager](#page--1-0) & [Arendt, 2013; Jia et al., 2014\)](#page--1-0). However results were very descriptive, interpretations were specific to the components of the studied product, and it was impossible to follow the contributions of the cellulose derivatives at the different steps of the process. Generally speaking, it is possible to adjust dough rheology by the addition of cellulose derivatives with different structures (Correa, Añón, Pé[rez,](#page--1-0) & [Ferrero, 2010](#page--1-0)).

In order to study the cellulose derivatives behavior in more realistic conditions, particularly during the process, a model cake was developed (Bousquières, Bonazzi, & [Michon, 2017\)](#page--1-0). It is processed following the same production steps of a real cake: foaming, addition of the powder, cooking. Furthermore, it has a structure imitative of a sponge cake (crumb with a homogeneous cellular

Table 1

Characteristics of the cellulose derivatives MC type SGA7C and HPMC type K250M.

^a From [The Dow Chemical Company \(2002\).](#page--1-0)

b From [Bousqui](#page--1-0)è[res et al. \(2017\)](#page--1-0)

^c Determined at 20 °C using the Higgins equation ($[\eta - \eta_{solvent}]/[\eta_{solvent}^*C] = [\eta]+I$ $[\eta]^2 C$).

Table 2

Concentrations of cellulose derivatives for the 10 tested mixes.

structure and crust). However, its composition is simpler than for a sponge cake, as it contains only HPMC, MC, native corn starch and water.

The aim of the present study was to understand how the cellulose derivatives contribute to the cellular structure of the model sponge cake at the different steps of the process. Rheology measurements were performed to analyze the behavior of the cellulose derivatives during foaming and cooking. Image analysis was used to observe how the cellulose derivatives impact the final structure of the product. The effect of a modification of the powder addition step was studied as it appeared to have a great impact on the cellular structure of the crumb.

2. Materials and methods

2.1. Model product

2.1.1. Materials

The model sponge cake, as developed by Bousquières et al. [\(2017\)](#page--1-0), contained purified water, native corn starch (Cargill, Minneapolis, MN, US) with a water content of 12.4% w/w (measured according to NF norm V05 707), and two cellulose derivative (Dow Chemical, Midland, MI, USA): methylcellulose (MC) type SGA7C and hydroxypropyl methyl cellulose (HPMC) type K250M. Their characteristics are presented in Table 1. Viscosity grade and mean polymerization degree of cellulose derivatives were linked and followed a power law regression ([The Dow Chemical Company,](#page--1-0) [2002\)](#page--1-0). The step of a cellulose monomer was about 0.52 nm. Thus, the mean length of one HPMC type K250M molecule and one MC type SGA7C molecule were estimated to about 900 nm and 100 nm, respectively. The intrinsic viscosities at 20 \degree C were equal to 10.1 dL/ g and 4.3 dL/g for HPMC and MC, respectively. Comparing the mean lengths and the intrinsic viscosities, it seems that HPMC is more flexible than MC.

The water/starch ratio was kept constant at 2.1. The cellulose derivative quantities to be used were determined using equation (1) proposed by [Bousqui](#page--1-0)ères et al. (2017) , in order to have a constant batter viscosity of 1.4 Pa.s measured at 270 s^{-1} . The compositions of the 10 HPMC/MC mixes tested are given in Table 2, mixes 0 and 9 containing only HPMC and MC, respectively.

$$
[MC] = -1.74[HPMC] + 1.07
$$
\n(1)

2.1.2. Process

The production followed the process devised by Bousquières [et al. \(2017\).](#page--1-0) The main steps are summarized below. The cellulose derivative solutions were prepared on the day before each experiment by dispersion in hot purified water of HPMC and MC powders already mixed in the adequate proportions. The dispersions were refrigerated at $4 \degree C$ for 20 h to obtain the best solvation possible. The cellulose derivative solutions were then mixed for 10 min with a planetary mixer (Kitchen Aid model 5KSM150, Benton Harbor, MI, USA) equipped with a vertical whisk at speed 10. Then, starch was gradually added to the foam at speed 2 over 40 s, and the mixtures were further blended for 2 min 20 s also at speed 2. For each

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