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Changes in gelatinisation and pasting properties of various starches (wheat, maize and waxy maize) by the addition of bacterial cellulose fibrils

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ABSTRACT

The aim of this work was to analyse the effect of bacterial cellulose fibrils (BCF) on the gelatinisation profile and pasting properties of starches from different sources (wheat, maize and waxy maize) and amylose contents. Blends of 8% starch with different BCF levels (0, 0.5, 2, 6 and 10% based on the dry weight of starch) were prepared and tested by Rapid Visco-Analysis (RVA), Differential Scanning Calorimetry (DSC) and both Optical and Polarized Light Microscopy. Results showed that BCF produce a significant modification of pasting properties. The pasting temperature was reduced but viscosities (peak, final, trough, breakdown and final) increased. The reduction in pasting temperature at the highest BCF addition was 20 °C higher for maize and wheat starches but only 2 °C higher for waxy maize starch. In contrast to the pasting temperature, the gelatinisation temperature by DSC for all three starches slightly varied upon BCF addition, but the gelatinisation enthalpy was reduced to a greater extent than values reported for the addition of other hydrocolloids to starch blends. Optical and polarized light microscopy showed the presence of domains rich in starch and highly aggregated BCF in all three starches evaluated. The increase in viscosity and decrease in pasting temperature are discussed in terms of changes in starch concentrations in the starch rich domain. These results open interesting perspectives in the use of bacterial cellulose and plant cell walls to design novel bio-composites to structure foods.

1. Introduction

Cellulose is the most abundant biopolymer in nature. It is mainly produced by plants, trees and bacteria by condensation of glucose units during the photosynthesis process in plants and trees. Long chains of anhydro-glucose units, joined via β-1,4-glycosidic linkages (C—O—C), are formed during this process (Eichhorn, 2011). Cellulosic materials consist of both crystalline and amorphous domains, in different proportions depending of cellulose source (Ciolacu, Ciolacu, & Popa, 2011). The presence of para-crystalline or amorphous cellulose is often significant, although it varies from species to species (Eichhorn, 2011). The physical properties of cellulose, as well as their chemical behaviour and reactivity, are strongly influenced by the arrangement of the cellulose molecules with respect to each other and to the fibre axis (Ciolacu et al., 2011). One type of cellulosic material that has received much recent attention as potential new functional material for industrial applications is bacterial cellulose.

Bacterial cellulose is a biopolymer formed by nanofibrils, which is synthetized mainly by *Acetobacter xylinus* and *Acetobacter hansenii* (Shah et al., 2013). These microorganisms are able to create in their extracellular matrix a complex network of cellulose fibres by a highly regular intra- and inter-molecular hydrogen bonds network resulting in a weak gel structure. This is the basis of Nata de Coco a traditional sweet candy desert originating in the Philippines (Tabuchi, 2007). Bacterial cellulose has a unique structure, composed by nanofibrils forming a nanostructured network characterized by high purity (free of components such as lignin and hemicellulose) Because of its nanostructure bacterial cellulose shows a high mechanical stability, high water absorption capacity in the wet state and full biocompatibility making this material feasible to be used in wide variety of applications (Lee, Buldum,

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Mantalaris, & Bismarck, 2014; Picheth et al., 2017; Shah, Ul-Islam, Khattak, & Park, 2013). More recently the production, structure and applications of bacterial cellulose has been reviewed in the context of food use (Grishkewich, Mohammed, Tang, & Tam, 2017; Shi, Zhang, Phillips, & Yang, 2014; Ullah, Santos, & Khan, 2016). Incorporation of hydrocolloids, particularly pectin and mannans during the synthesis of bacterial cellulose has frequently been used to prepare models of the plant cell wall structure (Lopez-sanchez et al., 2017; Whitney, Brigham, Darke, Reid, & Gidley, 1998). Remnants of plant cell walls can be used to replace soluble hydrocolloids in structuring foods and may give health benefits (Foster, 2011; Padayachee, Day, Howell, & Gidley, 2017).

For many years there has been extensive interest in hydrocolloid:starch blends because of their inclusion in a wide range of food products. A review by Bemiller (2011) identified a large number of starch hydrocolloid blends, however we are not aware of any studies where bacterial cellulose has been added to starch to modify pasting behaviour. This paper describes a preliminary study to determine how bacterial cellulose fibrils modify the gelatinisation profile and pasting properties of starch. Starch gelatinisation is a physical transition that takes place in a starch granule and modifies the functional properties (e.g. solubility, viscosity, water holding capacity) as a response to high temperature and water. Although there is not a formal definition, gelatinisation has been described as "the collapse of molecular order inside the starch granule which produce irreversible structural changes related with an increase in granule volume, melting of crystalline form, loss of birefringence and increasing in starch solubility due to effect of temperature in an environment of high moisture" (Belitz, Grosch, & Schieberle, 2009; J. N.; BeMiller & Huber, 2008). Normally the gelatinisation is measured by microscopy, differential calorimetry, X-Ray diffraction among other techniques. The modification of the gelatinisation profile of starches by other biostructures is important for a number of reasons, including its potential effect on the extent of retrogradation on cooling and presumably on generation of low-digestive and resistant starch (Mishra, Hardacre, & Monro, 2012). Appelqvist, Brown, and Norton (1995) also described an application of freeze-thaw resistance in starch sauces when mixed with hydrocolloids. In the case of starch pasting, it is regarded as a consequence of gelatinisation and is generally followed by viscosity changes. Indeed, as a result of starch gelatinisation, a viscoelastic mass is obtained (called paste), which consists of a continuous phase that is a molecular dispersion of suspended starch polymer molecules forming a network and a discontinuous phase of swollen granules, granules ghosts and granule fragments (Bemiller, 2011). A common technique used to follow starch pasting is the Rapid-Visco Analysis (RVA) which was developed from the well-known bravender curves of starch viscosity used in the industry.

Studies looking at starch hydrocolloid interactions have generally involved only one starch source and several hydrocolloids. In this study maize, wheat and waxy maize starches were selected because of their industrial importance, but also due to some structural differences between them. For instance, the waxy maize starch contains only traces of amylose whereas the amylose content of maize and wheat starches is ~25–29% but this could vary with source and extraction method (Bertoft, 2017). Swelling of granules on heating will be influenced by the presence of amylose-lipid complexes, which could be more present in high amylose cereal starches than do normal and waxy starches (Debet & Gidley, 2006; Pérez, Baldwin, & Gallant, 2009). In terms of starch granule size, they have been well characterized. 5–20 μm (diameter) in maize and waxy maize and 2–36 μm (diameter) in wheat. However, wheat starch shows a bimodal distribution in size. Considering the typical X Ray diffraction pattern all these starches correspond to type-A starch (Buléon et al., 1998; Jane, 2009).

The objective of this work was to determine how the addition of bacterial cellulose fibrils modify the gelatinisation profile and pasting properties of starch from different sources (wheat, maize and waxy maize).

2. Materials and methods

2.1. Materials

Native wheat, maize and waxy maize starches were purchased from Sigma Aldrich (Germany) in powder form. Dried sheets of bacterial cellulose fibrils (BCF) were kindly provided by Membracel (Brazil). The starches and bacterial cellulose were used as received without further purification and stored at room temperature until further use.

2.2. Preparation of starch-BCF suspensions

BCF was added to each starch in a concentration of 0, 0.5, 2, 6 and 10% w/weight dry starch (Equation (1)), using distilled water as solvent. BCF dried sheets were processed prior mixing following the protocol proposed by Quero et al. (2015). In the first step a well defined amount of BCF was held overnight in excess of distilled water in order to promote full hydration. In the next step, the BCF suspension was homogenized using a high power kitchen blender (Thomas "Premium", Germany) for 20 min, then followed by vacuum filtration using 8 μm diameter filter papers (Whatman 541, USA). At the same time, starch water suspensions at 8% (w/v) were prepared for each starch type. In the final step, the filtered BCF was added to each starch suspension and stirred for 15 min at room temperature in order to get homogeneous suspensions.

$$BCF\ concentration\left(\%, \frac{w}{w}\right) = \left(\frac{BCF\ weight}{BCF\ weight + Starch\ weight}\right) x100 \quad (1)$$

A control sample, in this case BCF in the absence of starch, was prepared following the same protocol.

2.3. Measurement viscoelasticity of BC suspensions

A preliminary characterization of the viscoelasticity of BCF suspensions in water in the absence of starch was carried out using a rheometer (Physica MCR 301, Anton Paar, Germany) equipped with parallel plate geometry. BCF suspensions were prepared at concentration of 0.05, 0.1 and 0.2% (w/v). Measurements were made in the linear viscoelastic region at a frequency of 1Hz and strain of 0.5%. The temperature was scanned from 10 °C to 40 °C at a rate of 5 °C/min.

2.4. Measurements of pasting properties

Pasting properties of starch-BCF blend were analysed by Rapid-Visco-Analysis (RVA super 4, Newport Scientific, Australia) in accordance with the methodology proposed by Sullo and Foster (2010) with minor modifications. 25–28 g of each suspension was weighed in aluminium canisters and inserted into the instrument. Pasting profiles were obtained as a function of temperature as follow: holding at 25 °C during 5 min, heating between 25 and 95 °C at 5 °C/min, holding at 95 °C during 5 min, cooling to 25 °C at 5 °C/min and holding at 25 °C during 5 min. The analysis was performed under constant stirring (160 RPM). The pasting properties measured were: 1) pasting temperature (temperature at which starch granules begin to swell and gelatinise due to water uptake, which is recorded from the onset of the viscosity peak); 2) peak