Dual modifications of polysaccharides via Passerini reaction: toward biobased films with tunable properties

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Résumé

The ever-rising global awareness about plastic pollution push value chain key players of the packaging market to develop new alternatives. Cellulosic fiber-based packaging materials offer environmentally acceptable solutions. However, their porous structure imparts them with poor barrier performances, making them unfit for the food industry. Barrier effects can be achieved through petro-based and poorly biodegradable/recyclable coatings, which seeks to be replaced by alternatives conjugating efficiency and sustainability. The strong chemical affinity of some polysaccharides for cellulose make them good candidates to be used as eco-friendly physically deposited coatings (1). However, their inherent hydrophilicity jeopardizes the long-lasting performances of the coating, notably under tropical storage conditions. Multicomponent reactions (MCRs) represent a green route for the modification of polysaccharides. Indeed, they are conducted in 'one pot' mild conditions, in the absence of neither catalysts nor coupling reagent. Among them, Passerini-three component reaction (P-3CR) combines a carboxylic acid, an isocyanide and an aldehyde/ketone, to form an -acyloxycarboxamide function. Through P-3CR, previous work demonstrated the modification of carboxymethyl cellulose (CMC), a widely available ether derivative of cellulose (2). The present work follows on from this work and describes the dual functionalization of CMC and alginates (ALG) by various hydrophobic substituents differing in their structures and rigidity, via P-3CR (Fig. 1). The Passerini modification of ALG is successfully proved and leads to promising derivatives, never described in literature. The coating abilities onto model substrates of the as produced hydrophobized polysaccharides are examined by QCM-D, coupled with AFM analysis which emphasize the effect of the modification on their adsorption propensity. Rheological and light scattering analysis provide insights into the organisation of modified polysaccharide chains in water, explaining their observed peculiar mode of deposition onto substrates. Finally, dynamic vapor sorption experiments (DVS) carried out on self-supported films allow to investigate the impact of the hydrophobic grafted side-groups on mechanism of water sorption.

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