Biobased thin films from thermosensitive and hydrophobic Passerini-modified polysaccharides

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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 materials offer environmentally acceptable solutions. However, their porous structure imparts them with poor barrier performances. Barrier effects are often achieved through petro-based and poorly biodegradable/recyclable coatings, prompting the search for alternatives conjugating efficiency and sustainability. The strong chemical affinity of some polysaccharides for cellulose make them good candidates for eco-friendly coatings1. However, their inherent hydrophilicity jeopardizes their long-lasting performances, especially under tropical storage conditions. Multicomponent reactions (MCRs) represent attractive routes to modify polysaccharides, consistent with some green chemistry's principles. Indeed, their atom-economy is good, they are conducted in 'one pot' conditions without catalysts or coupling reagents, and generate minimal waste. Among MCRs, the Passerini three component (P-3CR) reaction, achievable in mild aqueous medium, was shown well-suited for the dual modification of carboxymethyl cellulose (CMC) using model molecules2. The present work describes the extension of this approach to the dual functionalization of CMC and alginates (ALG) leading to more sophisticated structures by conjointly employing (i) hydrophobic moieties of various structures and rigidities3 and (ii) thermosensitive segments exhibiting a LCST (Jeffamine derivatives), to develop novel multifunctional coatings. A large series of CMC and ALG derivatives of different compositions was thus generated, and processed as thin films. Their wettability properties were analysed, emphasizing the impact of the modifications in various conditions. Their film-forming abilities on model substrates were examined by QCM-D (Quartz Cristal Microbalance) and AFM observations. This investigation gives insights into the effect of the double modification on adsorption onto solid surfaces, considering interactions between grafted hydrophobic moieties and thermo-induced conformational changes at LCST. Finally, dynamic vapor sorption experiments (DVS) on self-supported films, clarified the role of the chemical modifications on water sorption mechanisms.

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(2) a) Pettignano, A., Daunay, A., Moreau, C., Cathala, B., Charlot, A., Fleury, E., *ACS Sustain. Chem. & Eng.*, **2019**, 7, 17, 14685. b) Remy, L., Sudre G., Charlot A., Fleury E., *Carbohydrate Polymers*, **2023**, 320, 121228.

(3) Vuillet C., Charlot A., Fleury E., Article in progress.

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