In situ Growth of ZIF-8 on TEMPO-Oxidized Cellulose Nanofibril Film and Coated with Pectin for pH and Enzyme Dual-Responsive Controlled Release Active Packaging

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Abstract : The growth and reproduction of microorganisms in food packaging can cause food decay and foodborne diseases, which pose a serious threat to the health of consumers and even cause serious economic losses. Active food packaging containing antibacterial bioactive compounds is a promising strategy for extending the shelf life of products and maintaining the food quality, as well as reducing the food waste. However, most active packaging can only act as slow-release effect for antimicrobials, which causes the release rate of antimicrobials not match the growth rate of microorganisms. Stimuli-responsive active packaging materials based on biopolymeric substrates and bioactive substances that respond to some biological and non-biological trigger factors provide more opportunities for fresh food preservation. The biological stimuli factors such as relative humidity, pH and enzyme existed in the exudate secreted by microorganisms have been expected to design food packaging materials. These stimuli-responsive materials achieved accurate release or delivery of bioactive substances at specific time and appropriate dose. Recently, metal-organic-frameworks (MOFs) nanoparticles become attractive carriers to enhance the efficiency of bioactive compounds or drugs. Cellulose nanofibrils have been widely applied for film substrates due to their biodegradability and biocompatibility. The abundant hydroxyl groups in cellulose can be oxidized to carboxyl groups by TEMPO, making it easier to anchoring MOFs and to be further modification. In this study, a pH and enzyme dual-responsive CAR@ZIF-8/TOCNF/PE film was fabricated by in-situ growth of ZIF-8 nanoparticles onto TEMPO-oxidized cellulose (TOCNF) film and further coated with pectin (PE) for stabilization and controlled release of carvacrol (CAR). The enzyme triggered release of CAR was achieved owing to the degradation of pectin by pectinase secreted by microorganisms. Similarly, the pH-responsive release of CAR was attributed to the unique skeleton degradation of ZIF-8, further accelerating the release of CAR from the topological structure of ZIF-8. The composite film performed excellent crystallinity and adsorb ability confirmed by X-ray diffraction and BET analysis, and the inhibition efficiency against Escherichia coli, Staphylococcus aureus and Aspergillus niger reached more than 99%. The composite film was capable of releasing CAR when exposure to dose-dependent enzyme (0.1, 0.2, and 0.3 mg/mL) and acidic condition (pH = 5). When inoculated 10 μL of Aspergillus niger spore suspension on the equatorial position of mango and raspberries, this composite film acted as packaging pads effectively inhibited the mycelial growth and prolonged the shelf life of mango and raspberries to 7 days. Such MOF-TOCNF based film provided a targeted, controlled and sustained release of bioactive compounds for long-term antibacterial activity and preservation effect, which can also avoid the cross-contamination of fruits.

Keywords : active food packaging, controlled release, fruit preservation, in-situ growth, stimuli-responsive
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