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Thermoplastic starch hybrid biocomposite films with improved strength and flexibility produced through crosslinking via carboxylic acid

Abstract

Thermoplastic starch (TPS) suffers from its intrinsic low mechanical strength and high brittleness due to its strong hydrogen bonding and low chain mobility. The conventional way to crosslink the TPS film can improve the strength and stiffness of the films, but usually reduces the flexibility of the film, and increases its brittleness. In this study, the incorporation of the hybrid nanofiller [1 wt% nanocellulose (C) and 4 wt% nano bentonite (B)] into the TPS proved to improve greatly the films' strength and flexibility. The hybrid nanofillers with ratio 4B:1C was incorporated into the crosslinked thermoplastic corn starch (CR-TPCS) film to increase the its flexibility and toughness and produced a high mechanical strength fully biodegradable film. Two different aqueous carboxylic acids: citric acid (CA) and tartaric acid (TA) with different pH values (2,4,6) as the green crosslinker were employed. Substantial increase of tensile strength (3.98 to 9.17 MPa), Young's modulus (9.10 to 46.30 MPa) and elongation at break (55.2 to 135.7%) was observed for the CA- 4B1C/pH2 films compared to the CR-TPCS films. The melting temperature (Tm) of the CA-4B1C/pH2 improved compared to the TPCS/4B1C (un-crosslinked) film due to its crosslinking effect. Meanwhile, the CA-4B1C films exhibited the highest degree of substitution and di-esterification with the lowest swelling and water solubility properties due to the formation of a special "bridge" structure between the CA, nanocellulose and plasticizer. The "bridge" structure developed between the TPCS chains serves as the toughener to motivate higher chain stress relaxation and load endurance. The crosslinked "bridge structure" also proved to effectively reduce the retrogradation phenomenal in the TPCS films. This combination method of hybridization and crosslinking is an efficient, low cost, and environmentally friendly technique to overcome the low flexibility and brittleness problem of the TPS based packaging film.

Keywords

Crosslinking carboxylic acid; Nano-biocomposite; Nanocellulose; Nanoclay; thermoplastic starch