Development of Biodegradable, Bio-based Bioplastic-coated Paper with a Novel Spray Coating Method

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Understanding the end-of-life degradation challenges of petroleum-based plastic packaging is critical to developing eco-friendly alternatives. Paper and cellulose-based materials have emerged as sustainable options due to their biodegradability, recyclability, abundance, and low cost. However, their inherent hydrophilicity and porous structure, caused by high-density hydroxyl groups, lead to poor water and oxygen barrier properties, limiting their application in moist environments. To address these limitations, we explore the use of two bioplastics, Polyhydroxyalkanoates (PHAs) and biodegradable-polycarbonates (bio-PCs), known for their hydrophobicity, water insolubility, low gas permeability, and biodegradability under home-composting conditions. Despite these advantages, PHA is brittle with a narrow thermal processing window, while bio-PC exhibits poor mechanical strength and a low glass transition temperature. To enhance their performance, we blend bio-PC and PHA at different molar ratios and apply a novel spray-coating method to form triple-layer film structures on cellulose substrates. Cellulose nanofibrils (CNFs), at thin middle layer, effectively prevents penetration by organic solvents and bioplastics, enabling successful multilayer formation. A blend ratio of 5:1 (bio-PC: PHA) yields the smoothest surface. The blended films exhibit significantly improved thermal stability, with the decomposition temperature increasing from 165.5 °C (pure bio-PC) to 229.8 °C. Oxygen barrier properties were improved by two orders of magnitude compared to those of pure bio-PC or PHA-coated CNF paper. This study demonstrates a scalable strategy to enhance the thermal, physical, and barrier properties of bio-based coatings, offering a promising path for industrial applications in sustainable packaging.