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Exploring the Rheological, Crystallization, and Mechanical Property Behaviour of Stereocomplexed Poly(lactide) Blends, and Their Crystalline Network Structures

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Abstract

Poly(L-lactide) (PLLA) is a bio-based and compostable polymer with significant potential for replacing petroleum-derived polymers. However, its poor crystallization kinetics, melt strength and low thermal resistance limit PLLA's processability and application in high-performance components. Nevertheless, these issues can simultaneously be overcome by melt-blending PLLA with poly(D-lactide) (PDLA) to form stereocomplex crystallites with a melting temperature about 50°C above that of neat homopolymers. These stereocomplex crystallites have a strong nucleating effect and remarkably enhance PLLA's melt strength, while imparting enhanced thermal resistance. Moreover, if drawn from the melt-state, the stereocomplex crystallites can be transformed into *in situ* nanofibers. The incorporation of stereocomplex nanofibers in PLLA has been linked to noticeable improvements to crystallization behaviour and a reduction in boiling water shrinkage, relative to conventional spherulitic stereocomplex crystallites. Synthesizing from previous work, the aim of this current work is to tune the properties of *in situ* fibrillated PLLA/PDLA blends to demonstrate the applicability of PLA as a high-performance polymer. This is done through studying the *in situ* fibrillation of PLLA blends containing varying concentrations of PDLA, with comparison to neat PLLA and non-stretched blend analogues. A comprehensive comparison between stretched and non-stretched blends reveals the development of concentration-dependent network structures of polymorphic nature. Below the critical concentration for network formation, differences between the properties of stretched and non-stretched blends differ greatly. Above this concentration, blend properties are rather similar. The effect of blend composition, multi-tiered morphology, and polycrystallinity, are systematically investigated to identify structure-property relationships.