Structural subtleties and catalytic activity of sodium aminophenolate complexes in polylactide degradation: towards sustainable waste management solutions

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Streszczenie

This study explores the intricate coordination chemistry of sodium aminophenolate species and their significant role in the depolymerization of polylactide (PLA), offering novel insights into catalytic degradation processes. By examining sodium coordination entities, including dimers and larger aggregates such as tetramers, we reveal how structural modifications, particularly the manipulation of steric hindrances, influence the formation and stability of these complexes. The dimers, characterized by a unique four-center core (Na–O–Na–O), serve as a foundational motif, which is further elaborated to obtain complexes with varied coordination environments through strategic ligand design. Our research delves into the lability of the amino arm in these complexes, a critical factor that facilitates the coordination of PLA to the sodium center, thereby initiating the depolymerization process. Moreover, DFT studies have been pivotal in identifying the most energetically favorable structures for catalysis, highlighting a distinct preference for an eight-membered ring motif stabilized by intramolecular hydrogen bonds. This motif not only enhances the catalyst's efficiency but also introduces a novel structural paradigm for sodium-based catalysis in PLA degradation. Experimental validation of the theoretical models was achieved through NMR spectroscopy, which confirmed the formation of the active catalyst forms and monitored the progress of PLA degradation. The study presents a comprehensive analysis of the influence of ligand structure on the catalytic activity, underscoring the importance of the eight-membered ring motif. Furthermore, we demonstrate how varying the steric bulk of substituents on the amino arm affects the catalyst's performance, with benzyl-substituted ligands exhibiting superior activity. Our findings offer a profound understanding of the structural factors governing the catalytic efficiency of sodium aminophenolate complexes in PLA degradation. This research not only advances the field of coordination chemistry but also presents a promising avenue for the development of efficient and environmentally friendly catalysts for polymer degradation.

Adres publiczny

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