Organometallics for Sustainable Polymers and Copolymers

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Abstract: In recent years, a substantial amount of research has resulted in the genesis of new sustainable technologies towards the manufacture of polymers derived from renewable sources. The major reason for such a heuristic approach stems from the pollution problems associated with conventional plastics.[1] One such biodegradable material that has attracted increasing attention is poly(lactic acid) (PLA) which is commercially produced using the ring-opening polymerization (ROP) of lactide (LA).This methodology allows better control over molecular parameters (polydispersity index, molecular weight) under mild reaction conditions.[1] A study by Tolman et al. found that the stereoselectivity present in PLA is a result of thermodynamic preference of the initiating monomer molecule from a racemic mixture of LA enantiomers.[2] It is now beginning to unravel that an unsymmetrical transition state produced from LA and the catalyst during the initiation step plays a key role in deciding the stereospecificity of the propagation step, ultimately governing the final stereochemistry of the PLA, in addition to the thermodynamic preference for its molecular parameters. Jones et al. reported a Zr-*meso* salan complex, which is capable of producing an isotactic enriched PLA.[3] Recently, Kol et al. prepared an enantiopure salan magnesium complex which produces isotactic enriched PLA.[4] Earlier, we had reported benzotriazole phenoxide group (IV) complexes which are capable of producing highly heterotactic PLA**.**[5] However, the existing literature pays little attention towards the study of the symmetry of the transition state during initiation in LA polymerization. Thus, one of our objectives is an understanding towards the control of the molecular parameter as well as the tacticity of the PLA which is anticipated to depend on symmetry of the transition state. This was surmised to be solved either by using non-centrosymmetric ligand frameworks in the catalyst, or by using an enantiopure ligand. Our results proved the concept.[6, 7]

In addition, this lecture shall cover the important aspects of our research towards the synthesis of sustainable copolymers.[8-10]

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