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Group IV complexes of tridentate salicylaldiminato ligands and their catalytic activity in ring-opening polymerizations of ε-caprolactone, lactides and epoxides

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Biodegradable polymers have various biomedical and environmental applications and the preferred method for producing these polymers is ring-opening polymerizations (ROP) of ε-caprolactone (ε-CL) and lactides. Particularly group IV complexes are found to be most efficient and stereoselective as catalysts for the ROP. New Ti(IV), Zr(IV) and Hf(IV) complexes containing tridentate [NNO]-type salicylaldiminato ligands [L1H {2,4-di-tert-butyl-6-((quinol-8-ylimino)methyl)phenol} and L2H{2,4-di-chloro-6-((quinol-8-ylimino)methyl)phenol)}] have been synthesized by reacting metal alkoxides with the ligands via alcohol elimination. In the solid state, Zr and Hf complexes are monomeric whereas, the Ti complexes are dimeric, where both Ti atoms are connected through mono-μ-oxo bridge formation due to controlled hydrolysis. During the complexation with L1H *in situ* intramolecular Meerwein–Ponndorf–Verley (MPV) type reduction of the imine moiety happened and the amine got deprotonated. In contrast, the ligand L2H has produced imine complexes under similar conditions. DFT calculations were carried out to investigate the activation energies of MPV type reduction. These complexes are found to be active catalysts for the ROP of ε-CL and *rac*-LA with narrow MWDs of the produced polymers in a living manner through coordination-insertion mechanism. Also, these complexes initiate the polymerization in a stereoselective manner to produce heterotactic-rich PLA from *rac*-LA with P_r value upto 0.84. In addition, these complexes are quite active for the catalytic ROP of epoxides such as *rac*-cyclohexene oxide, *rac*-styrene oxide and *rac*-propylene oxide.

Biography

Dipa Mandal has completed her MSc in Chemistry from Indian School of Mines, Dhanbad in 2010 and doing PhD in Organometallic Chemistry/Polymer Chemistry in IIT Madras and having interest in catalyst synthesis for the ring-opening polymerization processes and copolymerization of CO2 with epoxides.

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