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Reductive Depolymerization of Polyesters and Polycarbonates with Hydroboranes by Using a Lanthanum(III) Tris(amide) Catalyst

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The worldwide massive production of plastics (368 Mt per year) [1] and the poor recycling rate of waste materials make them a scourge for the environment. Waste storage in landfills, their incineration or the mechanical recycling which gradually degrades the physical properties of plastic polymers, are not sustainable solutions. Chemical recycling, which is the depolymerization of materials into valuable monomers or chemicals, seems to be an appealing route for the future.

Currently, well-known solvolysis processes catalyzed by bases, acids and ionic liquids enable the depolymerization of polymers by hydrolysis, aminolysis or transesterification reactions.[2] They offer the recovery of pure monomers useful for the production of new virgin plastics.

Recently, reductive depolymerization processes of oxygenated and nitrogenated materials have appeared. These methods offer alternative approaches to access new value added products from plastics. The routes need to develop catalytic systems (catalyst and reducing agent) that would be able to selectively cut and reduce polarized bonds (carbon-oxygen and/or carbon-nitrogen) of the polymers to obtain the corresponding monomers (alcohols, amines or hydrocarbons).

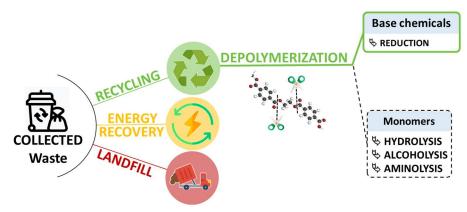


Figure 1. Reductive depolymerization, an alternative approach in plastic recycling

Such reductive deconstruction methods are scarce and are all based on the use of H_2 or hydrosilanes as reductants. The catalytic hydrogenolysis of oxygenated and nitrogenated polymers (polyesters, polyamines and polyurethanes) was reported with Ru(II) and Ir(III) catalysts under high pressures and temperatures, whereas their catalytic hydrosilylations could take place under milder conditions with metal-based catalysts (Ir(III), Zn(II), Mo(VI)) or organocatalysts ([Ph₃C][B(C₆F₅)₄] and B(C₆F₅)₃).[3,4] Hydroboranes as reductants could offer distinct reactivity and selectivity due to their higher hydride donor ability than hydrosilanes and an additional pronounced Lewis acidity.

Inspired by the work of T. J. Marks *et al.*,[5] on the reduction of esters, we considered the trisamide complex La[N(SiMe₃)₂]₃ as a 4f-catalyst and the pinalcolborane as a hydride donor, for the reductive depolymerization of a wide range of polyesters and polycarbonates.[6]



Figure 2. Our catalytic system for the reductive depolymerizaiton of polyesters and polycarbonates

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