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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 wastes materials make them a scourge for the environmental. Waste storage in landfills and waste incineration are not sustainable solutions, as well as the mechanical recycling which gradually degrades the physical properties of plastic polymers. 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, the reductive depolymerization processes have appeared as alternative approaches to access new value added products from plastics. The goal is to develop catalytic systems 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).

Such reductive deconstruction methods are scarce and are all based on the use of H₂ 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 La[N(SiMe₃)₂]₃ as a 4f-catalyst and the pinacolborane as a hydride donor, for the reductive depolymerization of a wide range of polyesters and polycarbonates.⁶



¹ *Plastic -The Facts*, PlasticsEurope, Belgium, 2020.

² C. Jehanno, M. M. Pérez-Madrigal, J. Demarteau, H. Sardon and A.P. Dove, *Polym. Chem.*, 2019, **10**, 172–186.

³ E. Feghali, L. Tauk, P. Ortiz, K. Vanbroekhoven and W. Eevers, *Polymer Degradation and Stability*, 2020, **179**, 109241.

⁴ A. C. Fernandes, *Green Chem.*, 2021, **23**, 7330–7360.

⁵ C. J. Barger, A. Motta, V. L. Weidner, T. L. Lohr and T. J. Marks, *ACS Catal.*, 2019, **9**, 9015–9024.

⁶ *Chem. Commun.*, 2022, Accepted Manuscript.

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