A materials approach to site-isolation of Grubbs catalysts from incompatible solvents and m-chloroperoxybenzoic acid.

Mwangi MT, Runge MB, Hoak KM, Schulz MD, Bowden NB.

Source

Department of Chemistry, University of Iowa, Iowa City, IA 52242, USA.

Abstract

The development of a method for site-isolation of Grubbs second-generation catalyst from MCPBA is described. In these reactions, Grubbs catalyst was dissolved in a solvent consisting of a mixture (1:1 v/v) of 1-butyl-3-methylimidazolium hexafluorophosphate and methylene chloride and completely encapsulated within a thimble fabricated from polydimethylsiloxane (PDMS). A series of molecules that react by cross metathesis or ring-closing metathesis were added to the interior of the thimble and allowed to react. In the last step, m-chloroperoxybenzoic acid (MCPBA) dissolved in MeOH/H₂O (1:1 v/v) was added to the exterior of the PDMS thimble. Small organic molecules diffused through the PDMS to react with MCPBA to form epoxides, but the Grubbs catalyst remained encapsulated. This result is important because Grubbs catalyst catalytically decomposes MCPBA at ratios of MCPBA to Grubbs of 3000 to 1. The yields for this two-step cascade sequence ranged from 67 to 83 %. The concept behind this sequence is that small organic molecules have high flux through PDMS but large molecules--such as Grubbs catalyst--and ionic reagents--such as MCPBA--have much lower flux through PDMS. Small molecules can thus react both outside and inside PDMS thimbles, whereas incompatible catalysts and reagents remain site-isolated from each other. This method does not require alteration of structures of the catalysts or reagents, so it may be applied to a wide range of homogeneous catalysts and reagents. To demonstrate further that the catalyst was encapsulated, the Grubbs catalyst was successfully recycled within the cascade sequence.