Poster Presentation

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Supramolecular construction of an aldehyde-functionalized cyclobutane in the solid state

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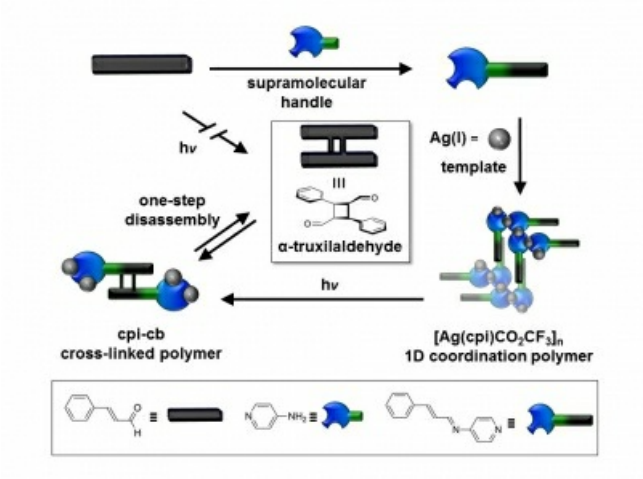
A goal of supramolecular chemistry which pursues the understanding and subsequent control of molecular packing in the solid state has led to the advent of rational design of crystalline structures. Employing non-covalent forces to direct assembly of reactant molecules, namely olefins, has guided development in the field of solid state reactivity for regio- and stereoselectivly controlled [2+2] photocycloadditions. A main focus of the field has employed hydrogen bonding and coordination chemistry to direct photodimerizations and subsequent synthesis of molecules challenging to generate through solution methods, such as ladderanes and paracyclophanes. A general restriction is the presence of supramolecular recognition sites 'irreversibly' attached (i.e. pyridyl) to a desired cyclobutane photoproduct. Here, we describe a method to attain an aldehyde functionalized cyclobutane by employing reversible imine chemistry combined with metal-organic coordination. First, a pyridine-based 'supramolecular handle' was installed utilizing imine chemistry to an aldehyde-functionalized olefin. Modification of the aldehyde to an imine generated a recognition site for subsequent supramolecular assembly. Second, olefins were self-assembled using metal-organic coordination in the form of a Ag(I) template. The result was a 1D coordination polymer that undergoes a regiocontrolled, UV-induced [2+2] photocycloaddition in the solid state. The photoreaction proceeds stereoselectively and in quantitative yield. One-step removal of the Ag(I) template and 'supramolecular handle' yields the aldehyde-functionalized cyclobutane α-truxilaldehyde.

[1] MacGillivray, L. R. et. al. (2008). Acc. Chem. Res. 41, 280-291.



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