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Title:	Topologically Driven Pore/Surface Engineering in a Recyclable Microporous Metal–Organic Vessel Decorated with Hydrogen-Bond Acceptors for Solvent-Free Heterogeneous Catalysis
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Keywords:	Topologically Driven Pore Recyclable Microporous Metal–Organic Hydrogen-Bond Acceptors Solvent-Free Heterogeneous Catalysis
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Abstract:	<p>The use of metal–organic frameworks (MOFs) comprising custom-designed linkers/ligands as efficient and recyclable heterogeneous catalysts is on the rise. However, the topologically driven bifunctional porous MOFs for showcasing a synergistic effect of two distinct activation pathways of substrates (e.g., involving hydrogen bonding and a Lewis acid) in multicomponent organic transformations are very challenging. In particular, the novelty of such studies lies in the proper pore and/or surface engineering in MOFs for bringing the substrates in close proximity to understand the mechanistic aspects at the molecular level. This work represents the topological design, solid-state structural characterization, and catalytic behavior of an oxadiazole tetracarboxylate-based microporous three-dimensional (3D) metal–organic framework (MOF), {[Zn2(oxdia)(4,4'-bpy)2]·8.5H2O}_n (1), where the tetrapodal (4-connected) 5,5'-(1,3,4-oxadiazole-2,5-diyl)diisophthalate (oxdia4-), the tetrahedral metal vertex (Zn(II)), and a 2-connected pillar linker 4,4'-bipyridine (4,4'-bpy) are unique in their roles for the formation, stability, and function. As a proof of concept, the efficient utilization of both the oxadiazole moiety with an ability to provide H-bond acceptors and the coordinatively unsaturated Zn(II) centers in 1 is demonstrated for the catalytic process of the one-pot multicomponent Biginelli reaction under mild conditions and without a solvent. The key steps of substrate binding with the oxadiazole moiety are ascertained by a fluorescence experiment, demonstrating a decrease or increase in the emission intensity upon interaction with the substrates. Furthermore, the inherent polarizability of the oxadiazole moiety is exploited for CO₂ capture and its size-selective chemical fixation to cyclic carbonates at room temperature and under solvent-free conditions. For both catalytic processes, the chemical stability, structural integrity, heterogeneity, versatility in terms of substrate scope, and mechanistic insights are discussed. Interestingly, the first catalytic process occurs on the surface, while the second reaction occurs inside the pore. This study opens new ways to catalyze different organic transformation reactions by utilizing this docking strategy to bring the multiple components close together by a microporous MOF.</p>
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