

Short communication

Isorecticular interpenetrated pillared-layer microporous metal-organic framework as a highly effective catalyst for three-component synthesis of pyrano[2,3-*d*]pyrimidines

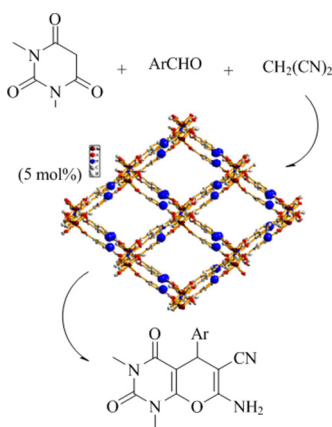
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GRAPHICAL ABSTRACT



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ABSTRACT

Two isorecticular microporous Zn(II)-MOFs, $[\text{Zn}_2(\text{NH}_2\text{-BDC})_2(4\text{-bpdh})]\cdot 3\text{DMF}$ (TMU-16-NH₂) and $[\text{Zn}_2(\text{BDC})_2(4\text{-bpdh})]\cdot 3\text{DMF}$ (TMU-16), (NH₂-BDC = amino-1,4-benzenedicarboxylate, BDC = 1,4-benzenedicarboxylate and 4-bpdh = 2,5-bis(4-pyridyl)-3,4-diaza-2,4-hexadiene) were used as heterogeneous catalysts. The TMU-16-NH₂ was used as an efficient heterogeneous base catalyst for the three-component cyclocondensation of 1,3-dimethylbarbituric acid, aryl aldehydes and malononitrile, giving rise to pyrano[2,3-*d*]pyrimidines. The ease of catalyst synthesis, excellent conversions and reusability of the catalyst for five consecutive cycles without a significant degradation in its catalytic activity, suggests significant future potential of this metal-organic framework for a wide range of base catalyzed reactions.

Metal-organic frameworks are crystalline compounds consisting of infinite lattices built up of the inorganic secondary building unit (SBU, metal ions, or clusters) and organic linkers, connected by coordination bonds of moderate strength. MOFs contain three well-differentiated

parts where the catalytic function can be allocated: the metallic component, the organic linker, and the pore space [1]. These materials have shown a variety of enormous potential applications including (but not limited to) gas adsorption [2], separations [3], luminescent [4], drug

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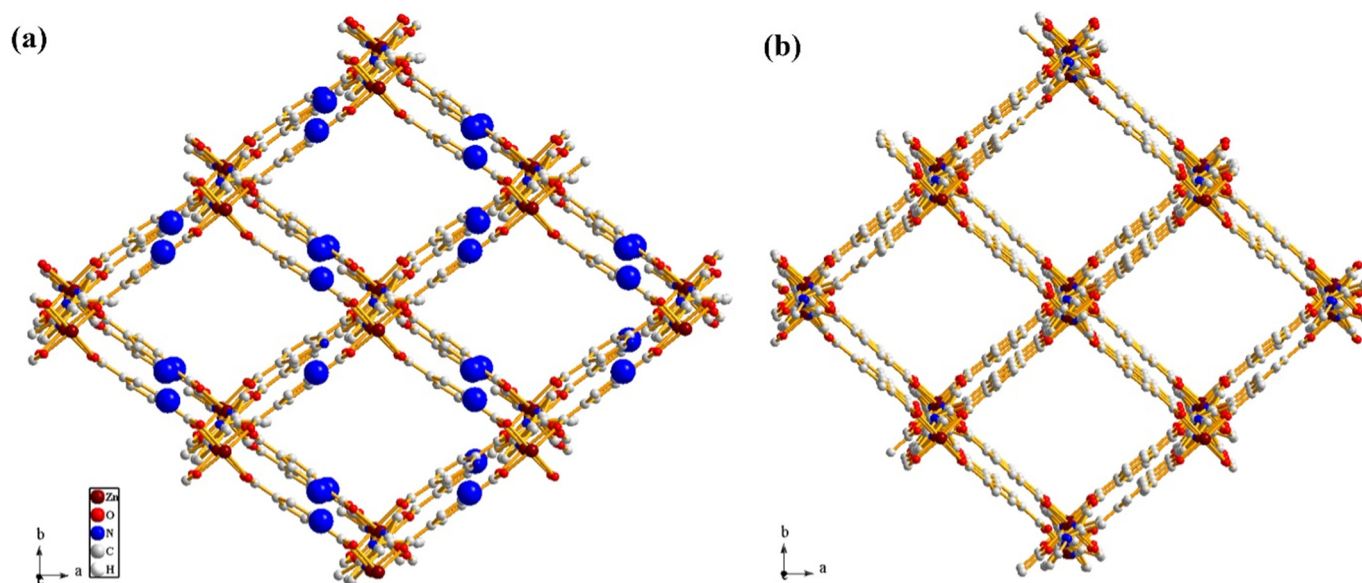


Fig. 1. Representations of the two-fold interpenetration (a) TMU-16-NH₂, highlighting the amine groups and (b) TMU-16, which contain 1D channels of 3 Å, viewed along the rectangular diagonal of the paddle-wheel clusters. All hydrogen atoms and the disordered guest molecules are omitted for clarity.

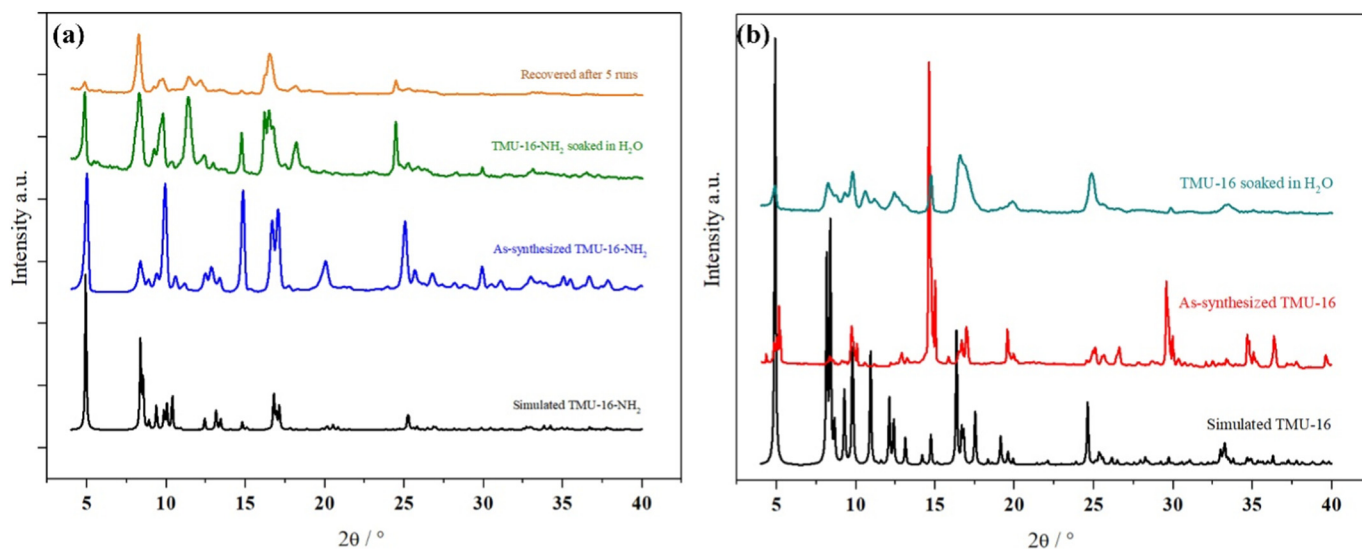


Fig. 2. PXRD of (a) TMU-16-NH₂: simulated (black), as-synthesized (blue), immersed in water for 2 h (green), and recovered after five runs, (b) TMU-16: simulated (black), as-synthesized (red), and immersed in water for 2 h (green). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

delivery [5] and sensing [6]. Over the last few years, abundant examples in literature have demonstrated the potential of MOFs in catalysis [7–11]. 4*H*-Pyrans and pyrimidinones are very important organic compounds with a wide range of biological activities. These compounds are reported to possess significant antibacterial, anticoagulant, anticancer, spasmolytic, diuretic, antianaphylactic, antihypertensive and anti-inflammatory activities [12–18]. On the other hand, pyrano[2,3-*d*]pyrimidines have received considerable attention over the past years due to their wide range of the diverse pharmacological action such as antitumor, cardiotoxic, hepatoprotective, antihypertensive and anti-bronchitic activity [19–23]. These compounds are generally synthesized via a one-pot three-component cyclocondensation of 1,3-dimethylbarbituric acid, aryl aldehydes and malononitrile in the presence of several catalysts such as 1,8-diazabicyclo[5.4.0]undec-7-ene [24], MgO [25], PEG-stabilized Ni nanoparticles [26], ZnFe₂O₄ nanoparticles [27], KF [28] and Mn/ZrO₂ [29]. Synthesis of these compounds using microwave irradiation [30] and electrocatalytic procedure in the

presence of sodium bromide as electrolyte [31] have been also reported. However, most of these methodologies suffer from disadvantages such as unsatisfactory yields, long reaction times and the use of relatively expensive catalysts. These finding prompted us toward further investigation in search for a new catalyst which will carry out the synthesis of these compounds under simpler experimental set up and eco-friendly conditions [32]. Continuing to our previous work [33], in this work we wish to report the utilization of isostructural two-fold interpenetrated microporous metal-organic framework, [Zn₂(NH₂-BDC)₂(4-bpdh)]·3DMF (TMU-16-NH₂) as an efficient heterogeneous catalyst for the condensation of 1,3-dimethylbarbituric acid, aryl aldehydes and malononitrile to achieve biologically interest Pyrano[2,3-*d*]pyrimidines. TMU-16-NH₂ is a pillared-layer MOFs like those was reported previously [34–37]. High catalytic activity was observed and catalyst could be reused without significant degradation in activity. Avoiding the utilization of harmful solvents in this environmentally friendly process is particularly appealing.

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