Ultraporous Covalent Organic Frameworks (COFs) in Seconds *via* Organic Terracotta Process

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Covalent organic frameworks (COFs) have concerned substantial deal of recognition to the researchers because of its summoned inordinate scientific attention owing to its diverse applications in catalysis, gas storage, sensing and optoelectronics. The conventional synthetic route for obtaining such ordered covalent networks is the connection of symmetrical building blocks through covalent bonds. However, tedious synthetic procedures, longer reaction time combined with the usage of toxic solvents and poor yield appear to be the bottleneck towards their greener fabrication, thereby limiting their possible potential applications. In this regard, we have introduced the simplest route *via* molecular organization approach towards the exceptionally rapid synthesis of highly crystalline, ultraporous COFs in seconds. The as-synthesized COFs ranked at the top among all reported two-dimensional COFs in terms of BET surface area till date. Accounting crystallography, we have detailed the molecular level fundamentals of COF crystallization and control over layered planarity and interlayer stacking. In addition, processing such porous, crystalline materials into various geometric shapes of industrial importance faces inadequacy due to the intricate problems like insolubility, grain boundaries as well as the difficulties in bulk scale synthesis. In this regard, we have further explored the possibility of industrial-scale (kg/h) synthesis of COFs

and *in situ* processing them into different shapes (beads, cylinders, tube etc.) and sculpture mimicking the ancient Terracotta Process by keeping their potentiality intact. The shapes outperformed the conventional zeolites and other MOFs in dehumidification performance.