Title:	Strategic Design and Functionalization of an Amine-Decorated Luminescent Metal Organic Framework for Selective Gas/Vapor Sorption and Nanomolar Sensing of 2,4,6-Trinitrophenol in Water
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Abstract:	On the basis of the strategic design of a triazine-based dicarboxylate ligand with two primary amino groups and one secondary amino group, an amine-functionalized autofluorescent and polar three-dimensional metal organic framework (MOF) {[Cd(ATAIA)]·4H2O}n (1), where H2ATAIA = 5-((4,6-diamino-1,3,5-triazin-2-yl)amino)isophthalic acid, has been synthesized under two different solvothermal conditions and structurally characterized. Single-crystal X-ray analysis reveals that 1 crystallizes in the orthorhombic polar space group Fdd2, where each ATAIA ligand acts as a linear linker to connect four Cd(II) centers, resulting in the formation of a three-dimensional framework with a repeat of a double helical metal chain. It has been further characterized by elemental analysis, UV–vis and Fourier transform infrared spectroscopy, and thermogravimetric analysis. Its bulk phase purity and stability in aqueous acid and base solutions are confirmed by powder X-ray diffraction. Both field emission scanning electron microscopy and high resolution transmission electron microscopy images of 1 reflect the formation of microflowers by self-assembly of nanopetals. With the dehydrated framework of 1, sorption studies of different gases (N2, H2, and CO2) as well as polar and nonpolar solvents, such as water, benzene (Bz), and cyclohexane (Cy), have been performed. The CO2 sorption isotherm depicts type I isotherm at 298 and 273 K and type IV isotherm at 195 K. Furthermore, with an uptake of 129.2 cm3 g–1 (25.62 wt %) at 195 K, sorption of CO2 is selective over N2 (77 K) and H2 (77 K) because of the strong adsorbate–adsorbent interaction as clearly evident from an isosteric heat of adsorption (Qst) at zero coverage of 37.5 kJ mol–1, which is exceptionally higher than that of other functionalized MOFs. Using the ideal adsorption solution theory calculation for a CO2/N2 (15:85) mixture, selectivity values are found to be 54.08 (298 K) and 46.96 (273 K) at 100 kPa. For a major application, activated 1 has been utilized for selective and ultrafast detection of 2,4,6-trinitrophenol (TNP) in water with a limit of 0.94 nM (0.2 ppb), which supersedes any previous reported value. Excellent recyclability and stability of 1 for sensing experiments have been established. Time-resolved fluorescence studies and density functional theory calculations have been used to establish its mechanism of action. Furthermore, a prototype experiment for the real-time sensing of TNP in the vapor phase by fluorescence microscopy provides an easy colorimetric monitoring.
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