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Title: Luminescent 2D Pillared-Bilayer Metal-Organic Coordination Networks for Selective Sensing of

ReO4- in Water

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Keywords: nuclear energy

perrhenate ion sensing

Issue Date: 2021

Publisher: ACS Publications

Citation: ACS Applied Materials & Interfaces, 13(38), 45465–45474.

Abstract:

For the preference in nuclear energy, one of the high-level liquid waste materials in the form of pertechnetate anion (TcO4-) has become an environmental hazard due to its mobility into groundwater and soil. For its sequestration, numerous efforts have been reported in recent years. However, its selective sensing, even using its nonradioactive surrogate oxidizing perrhenate ion (ReO4-), in aqueous media is very limited. To develop novel materials for such a purpose, we have designed an amino acid-functionalized bent dicarboxylic acid, 4-(((4-((carboxymethyl)carbamoyl)phenyl)amino)methyl)benzoic acid (H2hipamifba), for the strategic room-temperature synthesis of two isostructural and highly luminescent two-dimensional (2D) metal-organic coordination networks (MOCNs), {[Cu(hipamifba)(4,4'-azbpy)]·2CH3OH·2H2O}n (1) and {[Zn(hipamifba)(4,4'-azbpy)]·2CH3OH·2H2O}n (2), where 4,4'-azobipyridine (4,4'-azbpy) as a pillar linker imparts luminescent properties in the architectures. The single-crystal X-ray structural analysis demonstrates that 1 and 2 have pillared-bilayer 2D networks with the sq1/Shubnikov tetragonal plane net topology. These multiresponsive luminescent materials were gainfully employed for the selective sensing of ReO4- in water with a detection limit of 3.4 and 5.4 ppm for 1 and 2, respectively. It is noteworthy to point out that these are the first neutral sensors for such study as the only other two sensors reported in the literature are cationic in nature. Their suitability (selectivity, stability, and recyclability) as excellent water-stable sensors was established through the competitive analyte test and a comparison of pristine and spent samples by powder X-ray diffraction (PXRD) and scanning electron microscopy (SEM). Further, the mechanism of selective detection is explained by the time-resolved studies and density functional theory (DFT) calculations

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