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Stable Covalent Organic Frameworks for Exceptional Mercury Removal from Aqueous Solutions

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Supporting Information Placeholder

ABSTRACT: The predesignable porous structures found in covalent organic frameworks (COFs) render them attractive as a molecular platform for addressing environmental issues such as removal of toxic heavy metal ions from water. However, a rational structural design of COFs in this aspect has not been explored. Here we report the rational design of stable COFs for Hg(II) removal through elaborate structural design and control over skeletons, pore size, and pore walls. The resulting framework is stable under strong acid and base conditions, possesses high surface area, has large mesopores, and contains dense sulfide functional termini on the pore walls. These structural features work together in removing Hg(II) from water and achieve a benchmark system that combines capacity, efficiency, effectivity, applicability, selectivity, and reusability. These results suggest that COFs offer a powerful platform for tailor-made structural design to cope with various types of pollutions.

INTRODUCTION

Covalent organic frameworks (COFs) enable precise integration of organic building blocks into predesignable porous skeletons through topology design.¹ Progress over the last decade in the exploration of topology diagrams and synthetic reactions has developed a variety of COFs¹⁻⁹ with unique functions, such as semiconducting,⁴ emission,⁵ catalysis,⁶ proton conduction,⁷ and energy conversion and storage.⁸ Owing to the structural diversity of skeletons and pore walls, COFs offer a platform for designing high-performance materials that are promising for addressing environmental issues. However, exploring COFs to challenge environmental issues has been rarely developed.⁹

As a great threat to public health and environment, Hg(II) pollution causes serious damages to human beings, known as Minamata disease.¹⁰ In this context, to remove Hg(II) from polluted water to a concentration as low as possible is an important issue. Compared to traditional chemical reaction methods, adsorption using porous adsorbents is much superior because of its simplicity and cost efficiency.¹⁰ In addition to removal capacity, efficiency, and effectivity, suitable porous materials must be stable in water over a wide pH range (1–14) and must be reusable.^{10c} Traditional porous materials, including clays, activated carbons, and zeolites, usually have limited capacity.^{10d-f} Amorphous porous organic polymers are potential candidates but they lack principles for molecu-

lar design and methods for structural control.^{10g} Although metal-organic frameworks (MOFs) have been developed for Hg(II) removal based on their high surface areas, stable performance in aqueous solutions remains a challenging.^{11a,11b} A very first try using COFs is based on a long alkyl sulfide functionalized COF, i.e. COF-LZU8, which is useful for sensing Hg(II) owing to the fluorescence quenching of COF by Hg(II);^{11c} however, its limited capacity and stability preclude any implementation. Therefore, a porous material that meets the requirements of capacity, stability, and reusability remains a challenging in chemistry.

In response to the harsh requirements for Hg(II) removal from aqueous solutions, we rationally designed and synthesized a suitable COF structure. Firstly, for the stability, we have developed an extremely stable COF based on the imine-linked skeleton;^{6b} by integrating resonance effects (methyl sulfide units) to the phenyl edges, which is stable in aqueous solutions over a wide pH range. Secondly, for the capacity, efficiency, and effectivity, we introduced the shortest sulfide functional chains and developed two-dimensional (2D) COFs that enable the highest packing of sheets to achieve the highest sulfur content. The shortest sulfide chains on the pore walls suppress chain entanglement, help to expose the active sulfur sites to Hg(II), and endow the COFs with retained large pores, high surface area, and large

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