
Intrinsically Porous Crystalline Transition Metal Oxide Frameworks for Selective Noble Gas Recovery from Spent Nuclear Fuel Reprocessing Off-Gas Streams

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ABSTRACT:

The overall objective of the project is to develop readily synthesizable, thermally stable, and radiation-resistant solid adsorbents based on intrinsically microporous crystalline transition metal oxides (MCTMOs) for efficient and selective adsorption of noble gases (Xe and Kr) from off-gas streams of spent nuclear fuel reprocessing (SNFR) facilities. This project is driven by the hypothesis that MCTMOs with open channels of aperture diameters of 3.5 to 5.0 Å are potentially superior noble gas adsorbents to the current state-of-the-art systems. This hypothesis is rooted in the structural robustness of MCTMOs under extreme thermal and radiolytic environments, ease in scalable synthesis, and facile pore-size tuning through chemical and electrochemical redox processes, and is supported by the appealing preliminary results. To explore MCTMOs as selective Kr/Xe-adsorbing materials, we propose a systematic and collaborative research effort, combining computational simulations, experimental synthesis, and systematic performance evaluations. The **primary tasks** include 1) Molecular dynamic simulation and electronic structure calculations. 2) Synthesis of MCTMOs with varied compositions under different hydrothermal conditions. 3) Systematic exploration of MCTMOs under different calcination and chemical reduction conditions. 4) Thorough Structural characterizations of the resulting MCTMOs. 5) Performance evaluation of MCTMOs as Kr and Xe adsorbing media. And 6) Recyclability and recovery studies of end-of-life MCTMOs. The **aim** of the collaborative efforts is not only to develop novel MCTMOs as efficient Kr/Xe adsorbents with high selectivity and capacity but also to gain fundamental insights into the mechanistic formation pathways of hitherto still rare MCTMOs and MCTMOs-noble gas interactions. The proposed work will lead to the following **major deliverables**: 1) New MCTMOs that are computationally predicted to be highly Kr-selective adsorbents are identified. The nature and energies of MCTMOs/noble gas (Kr/Xe) interactions are elucidated. The effects of pore size and the chemical environment surrounding the pore on MCTMO's Kr/Xe adsorbing properties are quantitatively computed. 2) MCTMOs based on the M_3XO_x series and the $M_4X_3O_x$ series of different compositions and crystal forms are synthesized, and their crystal structures are determined. The correlation between hydrothermal conditions and the resulting MCTMO structures is established. 3) The effect of calcination conditions on the structure and porosity of MCTMOs is understood. 4) The effect of chemical reduction conditions (temperature, H_2 pressure, and time) and electrochemical reduction on the structures and porosity of MCTMOs is elucidated. 5) Adsorption isotherms of MCTMOs prepared under different conditions and with different extents of chemical reduction are systematically and comparatively measured. 6) Breakthrough experiments are performed on the most promising MCTMOs. At the conclusion of the project, the hypothesis that MCTMOs with precisely tuned pores and porous structures are superior Kr/Xe-capture materials from off-gas streams will be tested. If confirmed, several MCTMOs with unprecedentedly high Kr/Xe adsorption capacity and selectivity will be discovered.