

Novel Mesoporous Uranium-Oxide Based Catalysts for Removal of Volatile Hydrocarbons and Chloro-Organic Compounds: Synthesis and Reactivity Studies

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1. Introduction

Catalytic combustion of volatile hydrocarbons and chloro-organic compounds at low temperatures provides an environmentally benign as well as an energy efficient route for their removal. The removal of chloro-organic compounds has been reported to occur at moderate temperatures (<673 K) with the use of supported and unsupported uranium-oxide-based catalysts that exhibit exceptionally high activity (1). The highly active uranium-oxide-based catalysts, synthesized by conventional catalyst preparation techniques, are nonporous materials where the bulk of the uranium-oxide catalysts may not be involved in the reaction. The use of molecular assemblies of surfactants or related substances as structure directors has led to the synthesis of highly ordered mesoporous aluminosilicates and various transition metal oxides with very high surface areas (2-4). These methods have been used in the present study for the synthesis of novel mesoporous supported and unsupported uranium-oxide-based catalysts that should exhibit exceptionally high activity for the thermal oxidation of volatile hydrocarbons and chloro-organic compounds. The synthesis and use of ordered porous uranium-oxide-based catalysts would not only greatly enhance the catalyst efficiency for the destruction of volatile hydrocarbon and chloro-organic compounds but also provide an effective approach for the utilization of large amounts of depleted uranium from nuclear fuel production.

2. Catalyst Syntheses

Two types of catalysts have been synthesized in the present study: (1) U_3O_8 supported by mesoporous SiO_2 (U_3O_8/SiO_2) and (2) mesoporous uranium oxide (U_3O_8). U_3O_8/SiO_2 was synthesized by preparing a mixture of 0.12 cetyltrimethylammonium bromide (CTAB): 0.1 uranium nitrate: 1.0 tetramethylorthosilicate (TMOS): 1.0 methanol:0.02 HNO_3 : $4H_2O$. The resulting solid was heated in air at 873 K to obtain the mesoporous U_3O_8/SiO_2 . The U_3O_8 catalyst was prepared by mixing CTAB and UCl_4 (CTAB/ UCl_4 =0.16) in water. NH_4OH was added to adjust the pH to 10. The solution

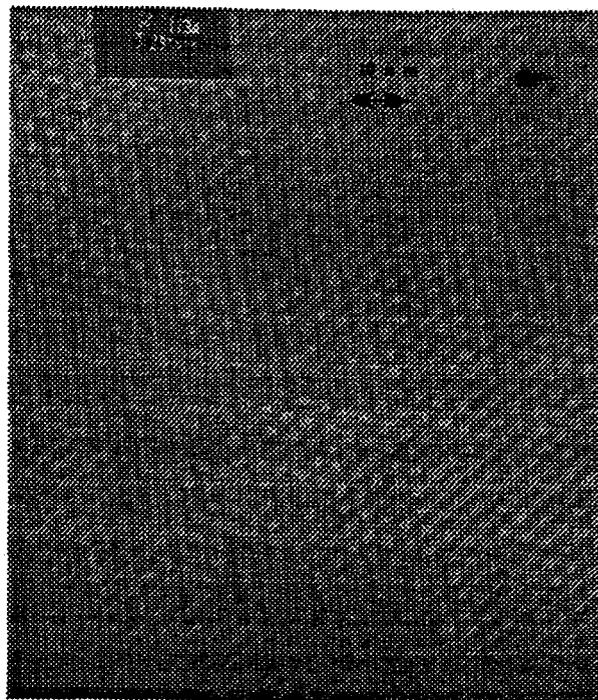


Figure 1 Transmission electron micrograph of mesoporous SiO_2 showing hexagonally packed mesopores.

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was filtered and the solid filtrate calcined in air at 723 K to obtain mesoporous U₃O₈.

3. Results

The transmission electron micrograph (TEM) of mesoporous SiO₂ prepared without the addition of the uranium precursor is shown in Figure 1. The synthesis produced a highly ordered SiO₂ with hexagonally packed pores. Brunauer Emmitt Teller (BET) analysis performed on U₃O₈/SiO₂ revealed a surface area of 310 m²/g with a maximum pore diameter of 72 Å. In addition, N₂-adsorption/desorption isotherms indicate that the pores are mesoporous with a fairly narrow distribution. The surface area, pore sizes, and pore size distribution for the mesoporous U₃O₈ catalysts were also measured and are listed in Table 1. The surface area of U₃O₈ obtained from the present study is significantly greater (650:1) than that of a commercially available U₃O₈.

Table 1 Comparison of Surface Area and Pore Diameter of Mesoporous Uranium oxide Catalysts

Catalyst	Surface Area (m ² /g)	Pore Diameter (Å)
Mesoporous U ₃ O ₈ /SiO ₂	310	72.0
Mesoporous U ₃ O ₈	65	114.7
U ₃ O ₈ (commercial)	0.1	-

The determination of catalytic activity and selectivity of these catalysts for the removal of toluene and chlorobenzene is currently underway. Future studies will aim at understanding the adsorption/desorption of reaction intermediates and the role that these intermediates play in the catalytic oxidation of volatile organic and chloro-organic compounds.

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4. References

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