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Enhanced Stability for Propene Epoxidation with H_2 and O_2 on Au Nanocatalysts Supported on TS-1 nanosheets

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Highlights

- Titanosilicate MFI nanosheets of single-unit-cell thickness (NTS-1) were synthesized with a diquaternary ammonium surfactant.
- Au/NTS-1 catalyst exhibited notably catalytic stability with high epoxide selectivity for PO, much better than conventional Au/TS-1 catalyst.
- The intrinsic reason for the enhanced stability and high PO selectivity is elucidated.

1. Introduction

Since the discovery in 1998 of the propylene epoxidation activity of the Au-Ti system in the presence of hydrogen and oxygen[1], considerable efforts have been made to design Ti-based materials with enhanced diffusion of reactant/products, which can effectively increases the propylene epoxide selectivity and catalytic stability. In this work, titanosilicate MFI nanosheets (NTS-1) of single unit cell thickness were successfully synthesized with the diquaternary ammonium surfactant as the zeolite structure-directing agent. Compared with traditional TS-1, NTS-1 has shortened length of reactant/product and maximized molecular diffusion because of thinner thickness along the b-axis (2nm) and presence of mesopores.

2. Methods

2.1. Synthesis of diquaternary ammonium surfactant

The bifunctional structure-directing agent($C_{22\text{-}6\text{-}6}$) was synthesized following the procedures described in the literature[2]. The as-synthesized diammonium salt $C_{22\text{-}6\text{-}6}Br_2$ was converted into hydroxide form, $C_{22\text{-}6\text{-}6}(OH)_2$, through reaction with Ag_2O in distilled water.

2.2. Synthesis of NTS-1

In a typical synthesis of NTS-1, tetraethyl orthosilicate (TEOS) and tetrabutyl orthotitanate (TBOT) were hydrolyzed in an aqueous solution of $C_{22\text{-}6\text{-}6}(OH)_2$ with removal of ethanol, forming a synthetic gel with a molar composition of 1.0 SiO_2:0.02 TiO_2:0.02-0.2 $C_{22\text{-}6\text{-}6}(OH)_2$:200 H_2O. The resultant gel was transferred to a Teflon-lined stainless steel autoclave, which was heated at 413 K for 14 days under rotation (100 rpm). After crystallization, the solid product was filtered, washed with distilled water, and dried at 373 K. The product was further calcined to burn off the occluded organic species at 823 K for 6 h in air.

3. Results and discussion

This Au/NTS-1 catalyst exhibited notable catalytic stability with high epoxide selectivity for PO, much better than conventional Au/TS-1 catalyst. Moreover, the intrinsic reason of the enhanced performance is elucidated using HAADF-STEM, TGA, NMR, N_2 physisorption, XRD characterizations. Compared with traditional TS-1 (ca. 500 nm) supported Au catalyst, it is found that the much thinner thickness of NTS-1 (ca, 2 nm) facilitates the mass transfer ability. This work is of great essential to the design of highly efficient Au/Ti-based catalysts for propene epoxidation with hydrogen and oxygen.

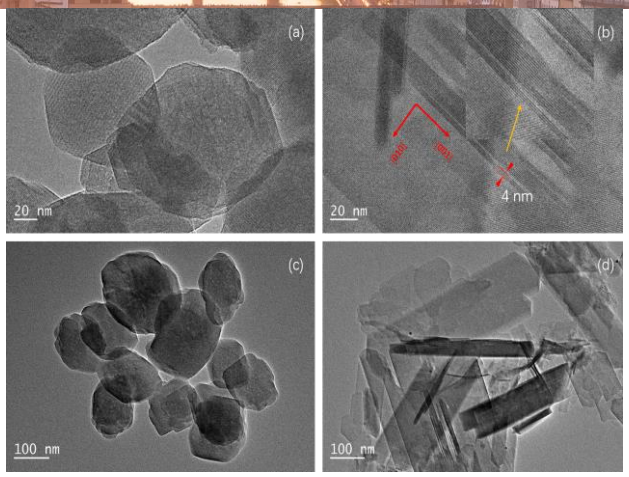

Figure 1. TEM images of conventional TS-1 (a and c) and NTS-1(b and d). The inserts show enlarged images.

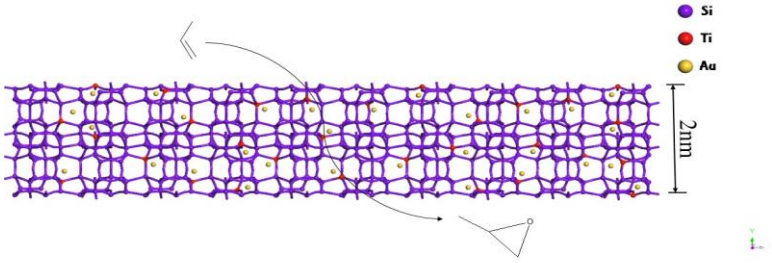

Figure 2. Propene epoxidation on Au/NTS-1.

4. Conclusions

In conclusion, TS-1 nanosheets with 2 nm thickness were synthesized to simultaneously improve the catalytic selectivity and stability of Au/Ti-containing catalysts for the green and simple propene epoxidation with H_2 and O_2 by enhancing the mass transfer ability of catalysts. The mechanism for the enhanced selectivity and stability is proposed by using multi-techniques. Compared with conventional Au/TS-1, the Au/NTS-1 catalyst has enhanced mass transfer ability by shortened reactant/product diffusion length and also better hydrophobicity. We conclude that the external surface of crystalline Ti-zeolite nanosheets are much better as epoxidation catalysts with H_2 and O_2.

References

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Keywords
TS-1; Nanosheets; Propene Epoxidation; Au