

Silica islands regulated external Ti-site environment of TS-1 for enhanced performance of 1-hexene epoxidation

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Abstract

Rational regulation of local environment of Ti-sites in TS-1 harbours tremendous industrial and scientific significance to epoxidation reactions. Herein, we report a facile and environment-friendly strategy to boost catalytic selectivity by covering the Ti-sites on external surface of TS-1 (ITS-1) without sacrificing activity of internal Ti sites. By quantitative analysis of d3-acetonitrile and quinoline-DRIFTS, 1H MAS NMR and XPS, it is found that percentage of external Ti-sites decreased from 11% to 6% after depositing surface silica islands. This successfully inhibits ring-opening reaction of 1,2-epoxyhexane on catalyst surface, as demonstrated by slower kinetic decomposition rate of 1,2-epoxyhexane. Compared with conventional TS-1 catalyst, selectivity of 1,2-epoxyhexane over ITS-1 catalyst significantly increased from 83.5% to 98.5% while maintaining high 1-hexene conversion. Furthermore, overmuch surface silica coverage only leads to extremely low conversion (2%) due to inhibition of mass transfer. This work paves the way for rational construction of Ti-containing catalysts for 1-hexene epoxidation

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