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Efficiently Dispersed MnOx on Mesoporous 3D Cubic Support for Cyclohexene Epoxidation

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Abstract : Epoxides constitute important intermediates for the production of fine and bulk chemicals as well as valuable building blocks for the synthesis of a variety of bioactive molecules. Manganese oxides are used as selective catalyst for various redox type reactions and also effectively used in the field of catalytic disposal of pollutants. Non-toxic, cost efficient factor and more over existence of wide range of oxidation state (+2 to +7) makes catalyst more interesting for both academic research and industrial applications. However, the serious drawback lying is the lower surface area. Exceedingly dispersed manganese oxide grafted over mesoporous solid material KIT-6 through ALD (Atomic Layer Deposition) technique effectively catalyze cyclohexene with H2O2 (30% in water) to corresponding epoxides. Highly selective epoxide >99% with 55.7% conversion of cyclohexene was achieved using huge dispersed active sites of MnOx species containing catalysts. Various weight percent such as (1, 3, 5, 7 & 10 wt %) of manganese (II) acetylacetonate complex was employed as Mn source to post-graft via active silanol groups of KIT-6 and are designated as (Mn-G-KIT-6). XRD, N2 sorption, HR-TEM, DRS-UV-VIS, EPR and H2-TPR were employed for structural and textural properties. Immense Mn species of about 95% proportion on silica matrix obtained was evident from ICP-OES.The resulting materials exhibited Type IV adsorption isotherms indiacting mesopore in nanorange. Si-KIT-6 and Mn-G-KIT-6 materials exhibited surface area of 519-289 m2/g and with decrease in pore volume of 0.96-0.49 cm3/g with pore diameter ranging 7.9- 7.2 with increase in wt%. DRS-UV-VIS spectroscopy and EPR studies reveal that manganese coexists as Mn2+/3+ species as extra-framework sites and frame-work sites that result in dispersion on surface of silica matrix of KIT-6 and incorporated manganese sites with silanol groups along with small sized MnO cluster, evident from HR-TEM which increase with Mn content. Conventional production of epoxides by the intramolecular etherification of chlorohydrins formed by the reaction of alkenes with hypochlorous acid is the major drawbacks obtained recently. The most efficient synthesis of oxiranes (epoxides) is obtained by mesoporous catalysts (Mn-G-KIT-6) are presented here and discussed.

Keywords : ALD, epoxidation, mesoporous, MnOx
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