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Investigation of Mesoporous TS-1 for the Catalytic Formation of N-oxides

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Titanium silicalite-1 (TS-1) is a microporous zeolite with MFI structure. The tetrahedral coordinated titanium atoms incorporated in the silica framework make TS-1 into a remarkable heterogeneous catalyst for oxidation of several organic compounds with hydrogen peroxide [1-2]. Unfortunately, the microporous zeolite often suffers from severe mass transfer limitations, which results in low overall activity. Another obstacle is that the micropores are too small to be accessed by bulky molecules, which hinders its use. Different methods to overcome these obstacles have been proposed over the years. One method is to use zeolite with frameworks containing larger pores e.g. BEA and MWW. However, evidence has been found that these catalysts are not as active as TS-1 [3]. Another method to overcome the diffusion limitations is to decrease the mean diffusion path, either by synthesizing mesoporous zeolites (using templating or demetallation) or by synthesizing small zeolite crystals (*e.g.* confined space synthesis).

Here, we present the investigations of the catalytic oxidation of N-heterocycles over mesoporous TS-1. The mesoporous zeolites are synthesized by a carbon templating method (cTS-1) adapted from [4], desilication with NaOH (dTS-1), and a mixture of both procedures (cdTS-1).

All TS-1 samples were tested for the oxidation of N-heterocycles to their corresponding N-oxides using aqueous H_2O_2 as the terminal oxidant. Initially, the oxidation of pyridine to pyridine N-oxide (PNO) as a model reaction was used. The reaction was also performed with several other substrates to demonstrate the versatility of the catalyst. PNO is a valuable synthetic intermediate in numerous organic reactions and for the manufacturing of pharmaceuticals and agrochemicals [5].

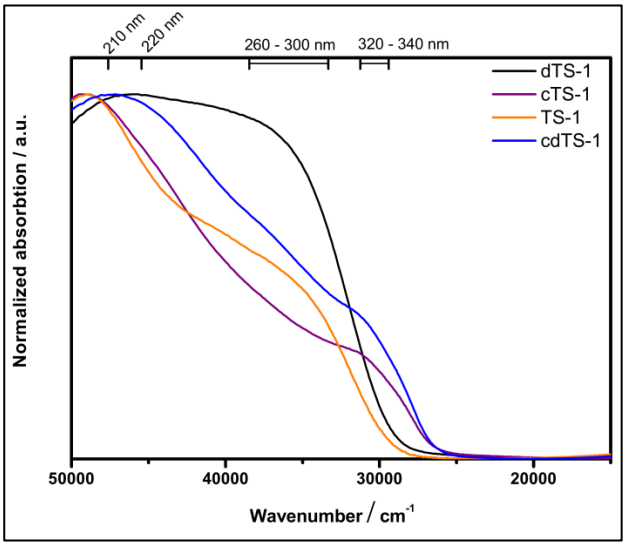

Figure 1. UV-Vis spectra of TS-1 and its mesoporous derivates

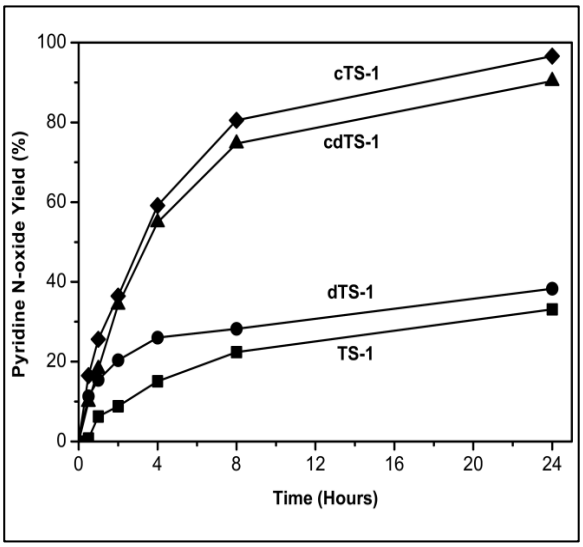

Figure 2. Yield of pyridine N-oxide over time

References

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