

A Study of Hydroxyl Nests in Zeolites

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Zeolites have been extensively utilised as catalysts in the petrochemical industry and are frequently modified to generate larger pores. However, the mechanism for the creation of mesopores is uncertain, although it is proposed that hydroxyl nests are generated as an intermediate step in reaction within the zeolites.¹

The present work uses computational techniques to examine the structure and stability of these defects. A hybrid QM/MM approach using the Chemshell software was employed to study defects in CHA, ZSM-5, FAU and Alpha Quartz. The model, shown in Figure (1) comprises a quantum mechanical description of a cluster of ~200 atoms around the reaction site near the porous channels, embedded in a much larger cluster modeled using a classical molecular mechanic's forcefield.^{2, 3}

We present results for the creation of hydroxyl nests as well as the aggregation of these defects to generate larger pores. We compare the generation of these defects from a point of origin outwards vs the removal of the key pentasil ring from the zeolitic framework. We aim to emulate and recreate cluster formation of various mesopores structures that have been proposed previously theoretically. Also observing the movement of protons around a potential hydroxyl nest site through the rotation of protons or the transfer of protons to adjacent oxygen atoms. As well as looking at the possibility of silanol inversion creating four O-H bonds but not removing silicon atoms.

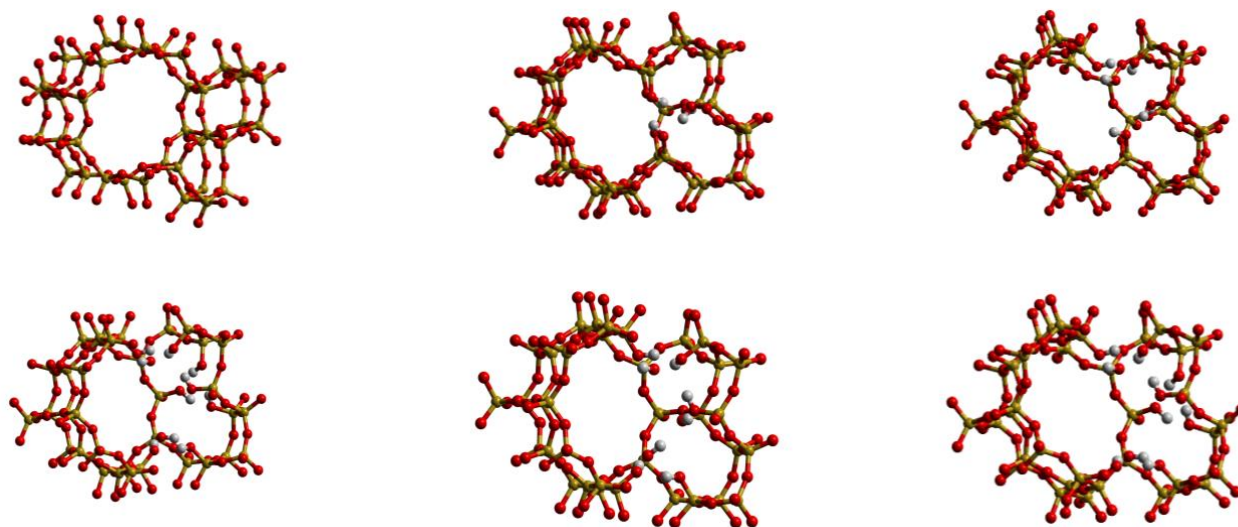


Figure 1: Formation of mesopore by removing 1 T site at each step in ZSM-5 cluster.

References

1. Barrer, R.M.M., M. B. Can., J. Chem, 1964. 42: p. 1481.
2. Sherwood, P et al. Faraday Discuss. 106 (1997): 79-92.
3. Lu, You, et al. J. Chem. Theory Comput 15.2 (2019): 1317-1328.