Computational Chemistry-HW 1

Yujia Wang

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1. Mahyuddin MH, Staykov A, Shiota Y, Miyanishi M, Yoshizawa K. "Roles of Zeolite Confinement and Cu–O–Cu Angle on the Direct Conversion of Methane to Methanol by $[Cu_2(\mu\text{-}O)]^{2+}$-Exchanged AEI, CHA, AFX, and MFI Zeolites." *ACS Catalysis*, 7, (2017): 3741-3751. DOI: 10.1021/acscatal.7b00588.

2. Small-pore Cu-zeolites (Cu-CHA, Cu-AFX, and Cu-AEI) were reported to produce more methanol per copper atom than the medium-pore Cu-MFI and large-pore Cu-MOR zeolites do for methane hydroxylation. The question the authors tried to answer is the reason for the difference in catalytic activities between these small-pore Cu-zeolites and the medium-pore and large-pore Cu-zeolites. Specifically, they tried to explain the roles of zeolite confinement and Cu$-$O$-$Cu angle on direct conversion of methane to methanol by $[Cu_2(\mu\text{-}O)]^{2+}$-exchanged AEI, CHA, AFX, and MFI zeolites.

3. The answer the authors arrived at is, the difference in catalytic activities is related to the zeolite confinement and Cu$-$O$-$Cu angle, by effecting the stability of the transition state of C$-$H bond dissociation. AEI, CHA, AFX, and MFI zeolites exert similar confinement effects on methane in stabilizing transition state and increasing the orbital energies of methane; for Cu$-$O$-$Cu angle, the decrease in the Cu$-$O$-$Cu angle of the $[Cu_2(\mu\text{-}O)]^{2+}$ active site was found to lower the acceptor orbital energy of $[Cu_2(\mu\text{-}O)]^{2+}$-zeolite, which further stabilizes transition state.