165g Effects of Cation Migration upon Energetics of Fe/Zsm-5 Catalyzed Reactions

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Isolated iron cations that are ion-exchanged into ZSM-5 can migrate to different locations on the walls of the zeolite channel. The preferred location can change depending upon the identity of the species adsorbed on the cation and upon the proximity of additional cations. The energetic consequences of this cation migration upon the thermochemistry of catalytic N_2O decomposition were studied using density functional theory. The results were combined with experimental measurements of the reaction kinetics, in-situ gravimetric characterization of catalyst surface coverage, and infrared spectroscopy leading to a consistent mechanistic model for the reaction. The promotional effect of gas phase NO was also studied experimentally and computationally. It can be explained in terms of the reduction of a surface oxo species generating gas phase NO_2, followed by its gas phase decomposition, regenerating NO.