An Investigation of Light Alkane Conversion Reactions on Zeolites with A Cluster Approach

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Introduction

The conversion of hydrocarbons by zeolite acid catalysts is essential for the modern oil and chemical industries. In this work, density functional theory methods were used to investigate light alkane catalytic conversion reactions on zeolites, including methane, ethane, propane and *iso*-butane. The zeolite acidity plays a very important role in the reactions. The deprotonation energy, which varies over a range of 20 to 50 kcal/mol among different zeolite structures, is often used as a theoretical measurement of zeolite acidity. In this work, we also investigated the effect of zeolite acidity on the light alkane conversion reactions.

Results and Discussions

An aluminosilicate cluster model containing three tetrahedral (Si, Al) atoms (T3 cluster), $H_3SiOAlH_2(OH)SiH_3$, was used to study the reaction pathways. The stationary points and transition state structures of the reactions were calculated at the B3LYP/6-31g* level and the energies were obtained using CBS-QB3, a complete basis set composite energy method. The calculated activation energies had good agreement with the available experimental data and the absolute errors were within a few kcal/mol. The activation barriers were the largest for dehydrogenation reactions, second highest for cracking reactions, and lowest for hydrogen exchange reactions, indicating the hydrogen exchange reactions were most energetically favorable.

The zeolite acidity effect was mimicked by changing the terminating Si-H bond lengths of the zeolite cluster. Linear relationships were found between the activation energies and the deprotonation energies. Applying the correlations, activation energies can be predicted for different zeolite catalysts as long as their deprotonation energies are first acquired.

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