Hexatriene Cyclization in Acid Zeolites Investigated Using DFT Methods

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Aromatic compounds are an important segment of the chemical industry that impacts many aspects of everyday life. Although the majority of aromatic compounds are made from petroleum, there is renewed interest in converting shale-gas derived light alkanes into aromatics over acid zeolite catalysts. It is known that these catalysts are good at converting light hydrocarbons into higher olefins, which can then form aromatic compounds. However, there is very little mechanistic understanding about the elementary steps and intermediates between linear olefins and cyclic precursors to aromatics.

Hexatriene was used as a model olefin to gain new insights on the cyclization mechanisms that happen in acid zeolites. With the aid of DFT methods, mechanisms involving Diels-Alder cycloaddition, pericyclic electrocyclization, and carbocation mediated cyclization were investigated in the gas phase. In the process, new and more complete mechanisms were developed to connect hexatriene to cyclic products via carbocation intermediates. The effect of zeolite acid sites on these reaction pathways was then investigated using quantum cluster models. This work paves a new path towards understanding aromatization in acid zeolites by shedding light on the underlying fundamental chemistry.