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研究论文

金属Ir_4 Cluster催化丙烯加氢反应势能面的密度泛函理论研究

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摘要 用密度泛函理论(DFT)对金属Ir_4 cluster催化丙烯Propene加氢反应的反应机理进行了理论研究. 在B3LYP理论水平下优化了反应通道上反应物、中间体、过渡态和产物各驻点物种的几何构型, 构建了该反应的基态势能面. 计算结果表明, Ir_4 cluster催化丙烯加氢反应, 主要通过3条反应通道(c,d和e)进行. 主反应通道c 是H1原子先经过中间体1加成到丙烯的边端C上形成中间体3, 然后H2原子经过渡态TS3—5, 中间体5和过渡态TS5-P加成到中间C上生成产物P. c通道无论从动力学角度还是热力学角度都是最有利的; 反应通道d和e中的最高势垒和通道c上的相比差别不大, 具有一定的竞争性, 是次通道.

关键词 密度泛函理论(DFT) 反应通道 Ir_4 cluster 丙烯

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Density Functional Theoretical Study on the Potential Energy Surface of the Propene Hydrogenation Catalyzed by Metal Ir_4 Cluster

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Abstract Using density functional theory(DFT), the reaction mechanism of propene hydrogenation catalyzed by metal Ir_4 cluster were explored in detail theoretically. At B3LYP level, the geometries of stationary points(reactions, intermediates, transition states and product) were optimized and the ground state potential energy surface was ploted. The calculated results suggest that for the propene hydrogenation catalyzed by metal Ir_4 cluster, the reaction may follow three reaction channels, which is c, d and e. In the major reaction channel c, the H-atom at Ir_1 site first transforms to intermediate 1 after surmounting TSR-1, followed by the addition of H-atom to the side C of propene, leading to forming intermediate isomer 3. After that, the H-atom at Ir_2 site can add to the middle C, passing through transition state TS3—5, intermediate 5 and transition state TS5-P respectively. Channel c is the most feasible reaction channel on the PES on both kinetic and thermodynamic considerations. As the highest transition states in channel d and e are a little higher than that of in channel c, they are less competitive and belong to minor channels.

Key words Density functional theory Reaction channel Ir_4 cluster Propene

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