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研究论文

CH_2ClO与NO反应机理的理论研究

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摘要 采用B3LYP, MP2方法在6-31＋(d,p)和6-311＋＋G(d,p)水平研究了CH_2ClO自由基与NO反应的微观机理, 找到了三个可能的反应通道. 并得到了各反应通道的反应物、中间体、过渡态和产物的优化构型、谐振频率. 成功地解释了Wu等的实验结论. 从电子密度拓扑分析的角度, 讨论了化学反应过程中化学键的变化规律, 为实验研究大气化学反应提供理论依据. 找到了该反应的结构过渡态(结构过渡区)和能量过渡态, 发现了反应热与结构过渡区之间的关系.

关键词 甲基氯氧自由基 反应机理 过渡态 电子密度拓扑分析

分类号

Theoretical Studies on the Reaction Mechanism of CH_2ClO with NO

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Abstract The reaction mechanism of CH_2ClO radical with NO was investigated by using B3LYP and MP2 methods with the 6-31＋G(d,p) and 6-311＋＋G(d,p) basis sets, and three reaction channels were found. Geometries of the reactants, intermediates, transition states and products were optimized and IRC calculations were carried out. The calculated results successfully explained the conclusion of Wu's experimental study. From the view of topological analysis of electron density, the changes of chemical bonds in the reactions were discussed, which will be helpful for the experimental research of the atmospheric chemistry. The "energy transition state" and the "structure transition state" in the reaction have been found. The calculated results revealed the relationship between the reaction enthalpy and the "structure transition region".

Key words CH_2ClO radical reaction mechanism transition state topological analysis of electronic density

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