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研究论文

3-硝基-1,2,4-三唑-5-酮与NH_3及H_2O分子间相互作用的理论研究

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摘要 在DFT-B3LYP/6-311＋＋G**水平上, 求得3-硝基-1,2,4-三唑-5-酮(NTO)/NH_3和NTO/H_2O两种超分子体系势能面上5种全优化构型. 经基组叠加误差(BSSE)和零点能(ZPE)校正, 求得NTO与NH_3和H_2O的分子间最大相互作用能依次为－37.58和－30.14 kJ/mol, 表明NTO与NH_3的分子间相互作用强于与H_2O的作用. 超分子体系中电子均由NH_3或H_2O向NTO转移, 相互作用能主要由强氢键所贡献, 由自然键轨道分析揭示了相互作用的本质. 对优化构型进行振动分析, 并基于统计热力学求得200.0～800.0 K温度范围从单体形成超分子的热力学性质变化. 发现由NTO和NH_3形成超分子II和III在常温下可自发进行; 而NTO和H_2O只在低温下才能自发形成IV, V和VI超分子.

关键词 3-硝基-1,2,4-三唑-5-酮(NTO) NH_3 H_2O 分子间相互作用 密度泛函理论 自然键轨道分析 热力学性质

分类号

Theoretical Study on Intermolecular Interactions of 3-Nitro-1,2,4-triazol-5-one with NH_3 and H_2O

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Abstract Five fully optimized geometries of 3-nitro-1,2,4-triazol-5-one (NTO)/NH_3 and NTO/H_2O supermolecules have been obtained with density functional theory method at the B3LYP/6-311＋＋G** level. The intermolecular interaction energy was calculated with basis set superposition error correction and zero point energy correction. The greatest corrected intermolecular interaction energies of the NTO/NH_3 and NTO/H_2O supermolecules are －37.58 and －30.14 kJ/mol respectively, indicating that the intensity of interaction between NTO and NH_3 is stronger than between NTO and H_2O. Electrons in supermolecular systems transfer from NH_3 or H_2O to NTO. The strong hydrogen bonds contribute to the interaction energies dominantly. Natural bond orbital (NBO) analysis was performed to reveal the origin of the interaction. Based on the vibrational analysis, the changes of thermodynamic properties from the monomer to supermolecules with the temperature ranging from 200.0 to 800.0 K have been obtained using the statistical thermodynamic method. It was found that structures II and III can be produced spontaneously from NTO and NH_3 at room temperature, while structures IV, V and VI can only be produced spontaneously from NTO and H_2O at lower temperature.

Key words 3-nitro-1 2 4-triazol-5-one ammonia water intermolecular interaction density functional theory natural bond orbital analysis thermodynamic property

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