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研究论文

金属Pt表面水蒸汽分子吸附的量子力学计算

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摘要 基于电子与振动近似方法和密度泛函B3LYP理论, 氧和氢原子选择6-311G**基函数, Pt选择赝势基组LanL2DZ, 优化得到Pt-OH_2结构和微观性质, 稳态结构Pt-H_2O分子中, Pt与H_2O不在同一平面, Pt倾向于与O原子结合. 计算了100～898.15 K温度下, 水蒸汽分子在Pt表面吸附反应的热力学函数值和平衡压力, 拟合得到ΔS^0,ΔH^0, ΔG^0, ln p与温度的函数关系. 室温以上$\Delta G^0>0$ kJ•mol^{-1}, 水蒸汽分子在Pt表面不能稳定吸附; 200 K以下, $\Delta G^0<0$ kJ•mol^{-1}, 能够稳定吸附. 计算了不同温度下水蒸汽分子在Pt表面发生解离反应的ΔG^0和平衡压力, 室温以上$\Delta G^0>0$ kJ•mol^{-1}. 100～898.15 K温度下, 水蒸汽分子在Pt表面不容易发生解离, 实际反应过程中以完整分子形式参与反应.

关键词 热力学函数 密度泛函理论 Pt 水蒸汽 吸附 解离

分类号

Adsorption Study of Water Vapor Molecule on Metal Platinum Sur-face by Quantum Mechanism Computation

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Abstract Based on electron and vibration approximate means and the density functional theory B3LYP, with LanL2DZ basis sets for Pt and 6-311G** basis sets for hydrogen and oxygen, the different structures of Pt-OH_2 have been optimized. Pt and H_2O of stable Pt-OH_2 molecule are not in the same plane, and Pt atom is in connection with O atom directly. The thermodynamic functions and the equilibrium pressures of adsorption reactions of water vapor have been calculated, and their relationships with temperature were obtained. Above 298.15 K, $\Delta G^0>0$ kJ•mol^{-1} in 100～898.15 K, and the adsorption of water vapor molecule was not stable on Pt surface. Under 200 K, $\Delta G^0<0$ kJ•mol^{-1}, and the water vapor molecule could adsorb on Pt surface steadily. ΔG^0 and the equilibrium pressures of dissociation reactions have also been calculated. Above 298.15 K, $\Delta G^0>0$ kJ•mol^{-1}. In 100～898.15 K, the water vapor molecules have no tendency to dissociate on Pt surface, and they participate in most reactions in intact molecule.

Key words thermodynamic function density functional theory Pt water vapor adsorption dissociation

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