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研究论文

N_2O 在 $Cu/t\text{-}ZrO_2(101)$ 表面的吸附与解离

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摘要 运用广义梯度密度泛函理论结合周期性平板模型方法研究了 N_2O 在完整及负载 Cu 的四方相 $ZrO_2(101)$ 表面的吸附与解离. 结果表明, N_2O 在完整 $ZrO_2(101)$ 表面的吸附均为物理吸附, Cu 在其完整表面的次表层第一氧位为最稳定吸附位, 且覆盖度为 0.25 ML 时的吸附最为稳定, 吸附能为 155.8 kJ/mol; N_2O 分子中 O 端弱物理吸附于 $Cu/ZrO_2(101)$ 表面, 其 N 端及平行吸附方式得到的稳定吸附能分别为 121.6 和 66.8 kJ/mol. 频率及电荷布居计算表明, 吸附后对称和反对称伸缩振动频率均发生红移, 电子由 Cu 负载底物表面转移给 N_2O 分子. 对 N_2O 分子的解离考虑了 N 端垂直吸附和平行吸附两种解离反应过程, 发现平行吸附过程的解离更易发生.

关键词: 密度泛函理论 一氧化二氮 四方相二氧化锆 铜 吸附 解离

Abstract: The density functional theory and slab models have been applied to investigate the adsorption and dissociation of N_2O on perfect t-ZrO_2 (101) and Cu/t-ZrO_2 (101) surfaces. The results indicated that N_2O adsorption on the $ZrO_2(101)$ surface is physical adsorption. The first of sub-surface oxygen site is the most stable adsorption site for the $Cu/ZrO_2(101)$ surface, and when the coverage is 0.25 ML, the most stable models were obtained with adsorption energy of 155.8 kJ/mol. The adsorption of N_2O on the Cu/t-$ZrO_2(101)$ surface by O-end is weak physical adsorption, and the N-end and parallel adsorption energy is 121.6 and 66.8 kJ/mol, respectively. Vibrational frequency and the Mulliken population were calculated, and the results indicated that the symmetric and antisymmetric vibrational frequencies are red-shifted and the charge transfers from Cu/t-$ZrO_2(101)$ to N_2O after adsorption. The N-end and parallel dissociation process were considered and the parallel dissociation process is more feasible.

Keywords: density functional theory, nitrous oxide tetragonal, zirconia, copper, adsorption, dissociation

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