化学学报 2006 64 (1): 27-31 ISSN: 0567-7351 CN: 31-1320/06

研究论文

丙烯腈在**Cu(100)**表面化学吸附的密度泛函理论研究

夏树伟*,[1]，高林娜[1]，徐香[1]，孙雅丽[1]，夏少武[2]

([1]中国海洋大学化学化工学院 青岛 266003)
([2]青岛科技大学化学与分子工程学院 青岛 266042)

收稿日期 2005-4-20 修回日期 2005-9-20 网络版发布日期 接受日期

摘要 利用密度泛函方法, 模拟金属铜原子簇Cu_{14}(9,4,1)的(100)表面, 对丙烯腈(CH_2=CHCN)在Cu(100)面上不同吸附位的吸附状况进行了理论研究. 结果表明:
丙烯腈分子通过端位N原子垂直吸附在金属表面上为弱化学吸附, 部分电荷从丙烯腈分子转移至铜金属簇;
由N原子的孤对电子与金属铜形成弱σ共价键; 顶位是最佳吸附位, 吸附能为40.7391 kJ•mol^{-1},
N原子与金属表面间的平衡距离为0.2279 nm; 其次为桥位和穴位, 吸附能分别为36.2513和30.2158 kJ•mol^{-1},
平衡距离为0.2194和0.2886 nm; 吸附后C≡N键的强度降低, 活化了丙烯腈分子. 化学吸附使体系的熵减小,
是由于丙烯腈分子的平动和转动因吸附而被限制.

关键词 密度泛函 丙烯腈 Cu(100) 化学吸附
分类号

DFT Study of the Chemisorption of Acrylonitrile on the Cu(100) Surface

XIA Shu-Wei*,[1], GAO Lin-Na[1], XU Xiang[1], SUN Ya-Li[1], XIA Shao-Wu[2]

([1] *College of Chemistry and Chemical Engineering, Ocean University of China, Qingdao 266003*)
([2] *College of Chemistry and Molecular Engineering, Qingdao University of Science and Technology, Qingdao 266042*)

Abstract The adsorption of acrylonitrile (AN) on the different sites of Cu(100) surface has been studied theoretically by means of model copper cluster Cu_{14}(9,4,1) with density functional theory (DFT). AN standing up and adsorbed perpendicularly to the surface and bonded to the metal sites via a nitrogen-metal interaction took a weak chemisorption. Such chemisorption led to electron transfer from the AN molecule to the cluster. In the metal complexes of Cu_{14}-AN, the σ-bonding via the lone pair electrons on the N atom was observed. Chemisorption on top site was preferred with the adsorption energy of 40.7391 kJ•mol^{-1} and the equilibrium *N*-surface distance of 0.2279 nm, while both bridge and hole sites were less stable than the top one, with their adsorption energies of 36.2513 and 30.2158 kJ•mol^{-1}, *N*-surface distances of 0.2194 and 0.2886 nm respectively. AN was activated by the chemisorption, which made the decrease of the strength of C≡N. The major contributions to the entropy decrease came from rotations and translations of AN, since these motions were lost upon chemisorption.

Key words density functional theory acrylonitrile Cu(100) surface chemisorption

DOI:

通讯作者 夏树伟 shuweixia@hotmail.com