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一氧化碳在原子簇**Zn_4O_4**上吸附的从头算研究

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摘要 以原子簇Zn_4O_4模型，用量子化学的从头算方法与补偿方法（counterpoise ）相结合研究了一氧化碳在氧化锌极化表面Zn-ZnO(0001)和O-ZnO(000-1)及在非极 化（10-10）表面的吸附态和吸附键能。研究表明，无论锌离子以何种方式出现在 晶体表面，锌离子都是较强的活化吸附中心，CO的碳原子向内的吸附键最强。这一 结论与宏观实验测试的结果相一致，尽管宏观实验测试的结果在上述不同晶体表在相近的CO脱附热。这种不同氧化锌晶体表面有相近的CO脱附热的现象是由于晶体表 面存在固有的晶体缺陷-表面层阶梯造成的。补偿方法，主要用于计算不同活化吸 附点的吸附质与吸附剂的弱作用。以Zn_4O_4为模型，以氧离子为活化吸附位，对 CO在氧化锌表面的计算结果表明，当CO分子垂直于晶体表面显排斥作用，当CO分子 平行于晶体表面仅有弱的吸附键。

关键词 一氧化碳 氧化锌 晶体 吸附 从头计算法

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Ab initio Study of the Adsorption of CO on a Zn_4O_4 Cluster

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Abstract Using the Zn_4O_4 clusters as model and combining quantum chemical calculations with counterpoise method, we study the states of CO absorbed on polar (0001), (000-1) and nonpolar (10-10) surfaces of ZnO and evaluate binding energies of the states. It can be infered that the cations Zn~(2+) on any surface of ZnO are the strongest active sites and the bond which carbon atom coordinated to a surface Zn is the strongest one, which are in accordance with experimental results. Although the similar binding energies for all surfaces of ZnO were found in experiments by some researchers, which might be attributed to that initially CO populates preferentially defect sites. A weak interaction between absorbent and adsorbate can be described well by counterpoise-method. No binding for either end of the CO molecule coordinates to a surface oxygen atom of ZnO. When the CO molecule is parallel to the surface of ZnO and coordinates to the surface oxygen atom, there occurs a weak binding only.

Key words CARBON MONOXIDE ZINC OXIDE CRYSTALS ADSORPTION AB INITIO CALCULATION

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