

對觸媒科學研究者的應用實例

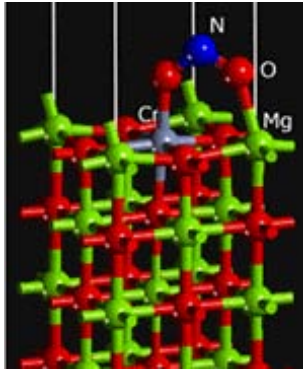
稀土元素、過渡金屬、以及混合金屬氧化物的脫氮化物和脫硫化物之活性：經由軌域-能帶相互作用研究所作的觸媒改良之系統設計

CASTEP—the plane wave total energy code to carry out detailed investigations of the interaction of the above pollutant molecules with the surfaces of MgO, TiO₂, Cr₂O₃, ZnO, and CeO₂. Also studied were the electronic properties of mixed-metal oxides and pure and doped metal surfaces. Much of the above work also investigated the effects of structural defects (steps, kinks, corners, O-vacancies) and doping with a second metal.

The close coupling between theory and experiment is making possible a fundamental understanding of many phenomena associated with the chemistry of molecules on oxide surfaces. In particular, the importance of band-orbital interactions for the reactivity of oxide surfaces has become clear, and a correlation between the electronic and chemical properties of mixed and doped oxides has been established. This has opened the way for using simple models based on band-orbital mixing to provide a conceptual framework for modifying or controlling the chemical activity of pure oxides, and for better engineering of mixed-metal oxides.

DeNOx and DeSOx activity of rare-earth, transition-metal and mixed-metal oxides: systematic design of better catalysts through orbital-band interaction studies

Reference: A. Maiti et al, *J. Phys. Chem.* 104, 3630 (2000); *J. Chem. Phys.* 112, 9929 (2000). *J. Am. Chem. Soc.* 122, 12362 (2000); *J. Phys. Chem. B* 105, 5497 (2001); *J. Am. Chem. Soc.* 123, 9597 (2001).

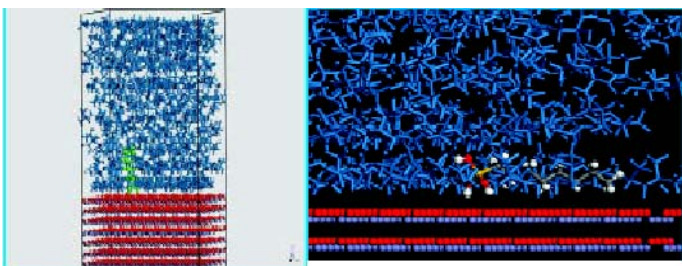


鋅腐蝕防止劑的改良 - ZnO 上的矽烷化合物的作用

This is to study the building of silane molecules on the surface of zinc oxide. Amorphous cell was used to create solvent-silane cells which are placed on the oxide substrate. The behavior of three different silane molecules, on the surface was studied using molecular dynamics simulation code DISCOVER and COMPASS was used as a force field. These silanes were studied as the degree of polarity on the tail varies and hence can be used to understand the role of the tail on the dominant configurations found on the surface. The results show that there exists two types of stable configurations (1) where only the silane head group binds on the surface and the other is the parallel configuration.

Toward improved zinc corrosion inhibitors-understanding the role of silanes on ZnO

Reference: A. Kornherr et al *J. Chem. Phys.* 119 (18), 9719 (2003); *Chem. Phys. Lett.* 393, 107 (2004)

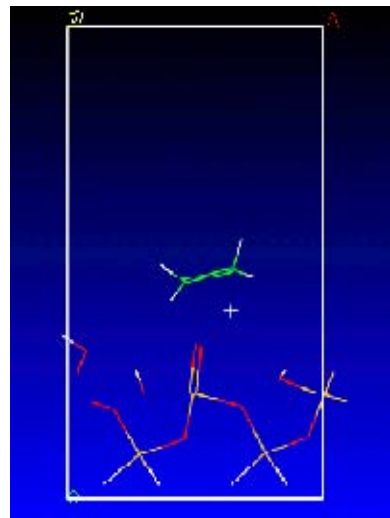


表面的磷對從乙烷到乙烯之氧化脫氫作用的影響

DMol3—the DFT code, was used to carry out an extensive investigation of the full ODH cycle of ethane to ethylene on a SiO₂ surface. A large number of accurate calculations were made possible by: (1) the representation of the catalytic surface with a periodically repeated slab; (2) fast structural relaxation in delocalized internal coordinates even in the presence of cartesian constraints¹; and (3) automatic search and refinement of transition states using periodic super cells. Scientists from Accelrys, in collaboration with experimental groups from Akzo-Nobel and Sandia National Laboratories, obtained important results from this study: (1) ethane cannot insert into a defect-free oxide surface. It needs to be pre-enriched with atmospheric O₂; (2) the rate-limiting steps involve the insertion of O₂ into the defect-free oxide surface, and the release of ethylene into the atmosphere; (3) activation barriers are lowered by ~10 kcal/mol in the presence of surface

Effect of surface phosphorus on the oxidative dehydrogenation of ethane to ethylene

Reference: 1. J. Andzelm, R. D. King-Smith, and G. Fitzgerald, *Chem. Phys. Lett.* 335, 321 (2001); *J. Chem. Phys.* 117, 8080 (2002).



在沸石中二氧化碳的吸附機制

Sorption modules was used to study Carbon dioxide adsorption in faujasite. A proposed mechanism for the carbon dioxide adsorption was validated by the trends observed for the enthalpies of adsorption. The top picture shows that the result obtained from the Monte Carlo calculation represents many local energy minima to choice between the best adsorption behavior. This is followed by adsorption isotherm calculation. Both the results of isotherm and enthalpies of adsorption were in good agreement with the data obtained from micro calorimetric experiments. This also provide an atomistic view.

Adsorption mechanism of carbon-dioxide in zeolites

Reference: G. Mourin et al *J. Phys. Chem. B.* 109, 16084 (2005); A. Goj et al *J. Phys. Chem. B* 106, 8367 (2002).

