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Author: Joshua Minwoo Kweun Chenzhe Li Yongping Zheng
Maenghyo Cho Yoon Young Kim Kyeongjae Cho



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Bulk-Surface relationship of an electronic structure for high-throughput screening of metal oxide catalysts

Joshua Minwoo Kweun^a, Chenzhe Li^a, Yongping Zheng^{a,b}, Maenghyo Cho^a, Yoon Young Kim^a, Kyeongjae Cho^{a,b}

^a Department of Mechanical and Aerospace Engineering, Seoul National University, Gwanak-gu, Seoul 08826, South Korea

^b Department of Materials Science and Engineering, The University of Texas at Dallas, Richardson, Texas 75080, United States

*kjcho@utdallas.edu, Tel.: +1 972 883 2845

*yykim@snu.ac.kr, Tel.: +82 2 880 7154

Highlights

- Bulk surface relationship was predicted by the ligand field nature of metal oxides.
- Antibonding and bonding d-bands occupancy clarified the bulk surface relationship.
- Different surface relaxations were explained by the bulk electronic structures.
- Transition from the bulk to the surface state was simulated by oxygen adsorption.

Abstract

Designing metal-oxides consisting of earth-abundant elements has been a crucial issue to replace precious metal catalysts. To achieve efficient screening of metal-oxide catalysts via bulk descriptors rather than surface descriptors, we investigated the relationship between the electronic structure of bulk and that of the surface for lanthanum-based perovskite oxides, LaMO_3 ($M = \text{Ti, V, Cr, Mn, Fe, Co, Ni, Cu}$). Through density functional theory calculations, we examined the d-band occupancy of the bulk and surface transition-metal atoms (n_{Bulk} and n_{Surf}) and the adsorption energy of an oxygen atom (E_{ads}) on (001), (110), and (111) surfaces. For the (001) surface, we observed strong correlation between the n_{Bulk} and n_{Surf} with an R-squared value over 94%, and the result was interpreted in terms of ligand field splitting and antibonding/bonding level splitting. Moreover, the E_{ads} on the surfaces was highly correlated with the n_{Bulk} with an R-squared value of more than 94%, and different surface relaxations could be explained by the bulk electronic structure (*e.g.*, LaMnO_3 vs. LaTiO_3). These results suggest that a bulk-derived descriptor such as n_{Bulk} can be used to screen metal-oxide catalysts.

Keywords

Bulk surface relationship (BSR), Density functional theory (DFT), Metal oxide catalyst, High-throughput screening (HTS), Ligand field, Correlation study.

1. Introduction

Much research in recent decades has focused on metal oxide due to its relative abundance, which can offer the opportunity to replace catalysts composed of precious platinum group metals (PGMs). Metal oxide is crystalline or amorphous materials and consists of metals and oxygen ligands. The materials provide a wide range of tunable compositions, mechanical strength, thermal stability [1], and selective oxidation property [2]. Metal oxide has many possible uses for catalytic applications such as the solid oxide fuel cell (SOFC) [3,4], environmental catalysts [2,5], water splitting [6,7], and thin film photovoltaic cell [8,9]. In most of the research fields, however, an inefficient trial-and-error approach has been faced with the difficulty to develop new metal oxide catalysts.

One way to accelerate the material design process is to conduct a study on density functional theory (DFT) for a better understanding of the atomic scale nature [5,7,10–21]. For example, high-throughput screening (HTS) was carried out for various applications such as intermetallic compounds, photovoltaic cells, thermoelectric materials, and heterogeneous catalysts and with different descriptors such as formation energy, band gap, and adsorption energy [10–13]. Especially, electronic structure based descriptors were developed for optimizing catalysts. For example, Vojvodic *et al.* demonstrated that metal oxides had the Brønsted-Evans-Polanyi (BEP) relationship, which is a linear relationship between molecule dissociation energy and its relative energy of transition states [14]. Similarly, Vojvodic *et al.* showed that metal carbides

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