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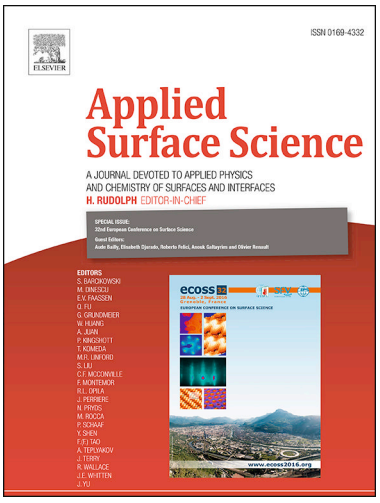

Full Length Article

DFT study on water oxidation on nitrogen-doped ceria oxide

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DFT study on water oxidation on nitrogen-doped ceria oxide

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Abstract: Heteroatom doping is effective to regulate electronic structure and improve the performance of photocatalyst. Herein, water oxidation on nitrogen-doped CeO_2(110) was investigated by using density functional theory calculation to understand the role of nitrogen doping. The results show that the optimal pathway of water oxidation on perfect and nitrogen-doped CeO_2(110) is $H_2O \rightarrow OH+H \rightarrow O+2H$; $O+2H+H_2O \rightarrow OOH+3H \rightarrow O_2+4H$, and the rate-limiting step of whole water oxidation process is $O+2H+H_2O \rightarrow OOH+3H$. The presence of nitrogen dopant in CeO_2(110) induces new characteristic peak of impurity level, and reduces the activation energies of water oxidation, especially for the rate-limiting step of O-O bond formation. The reverse reaction of O and H to OH that is easily occur on perfect CeO_2(110) is effectively prohibited on nitrogen-doped counterpart. Additionally, nitrogen dopant makes the rate-limiting step easier than that on oxygen-defective CeO_2(110) from the kinetic perspective. Therefore, nitrogen-doped CeO_2 is expected to be a promising photocatalyst for water oxidation.

Keywords: water oxidation, ceria oxide, nitrogen doping, density functional theory

1. Introduction

Photocatalytic water oxidation, the bottleneck for water splitting to produce hydrogen, has attracted a wide range of research interests to alleviate energy crisis and solve environmental problems [1-7]. Highly efficient and low-cost artificial catalyst is the key to overcome the slow kinetics of four sequential electron transfer steps in