

**Acetylene hydrogenation catalyzed by bare and Ni doped CeO₂(110): The role of frustrated
Lewis pairs**

Shulan Zhou,^{1,2}* Qiang Wan,³ Sen Lin,^{3,*} and Hua Guo²

*¹School of Chemistry and Chemical Engineering, Shandong University of Technology, Zibo
255000, China*

*²Department of Chemistry and Chemical Biology, University of New Mexico, Albuquerque, New
Mexico 87131, USA*

*³State Key Laboratory of Photocatalysis on Energy and Environment, College of Chemistry,
Fuzhou University, Fuzhou 350002, China*

*Corresponding authors. Email: zhoushulan2009@126.com, slin@fzu.edu.cn

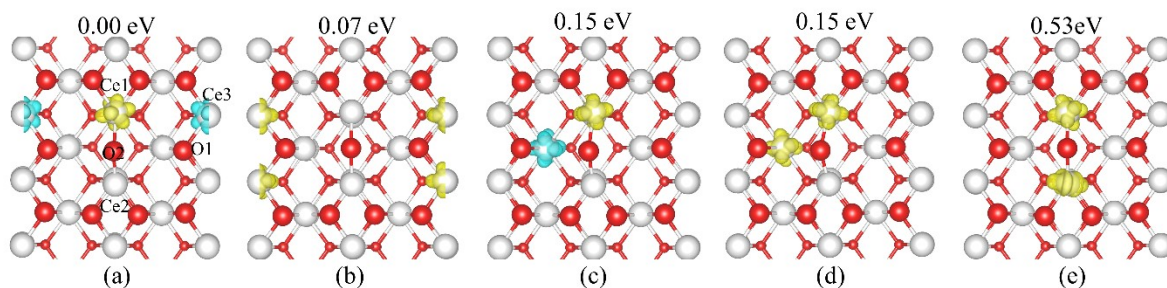


Figure S1 Calculated spin density with Ce^{3+} located at different positions for $\text{CeO}_2(110)\text{-O}_v$. The data is the energy of each configuration relative to the most stable one (a). Color scheme: Ce, white; oxygen, red; spin density, cyan or yellow.

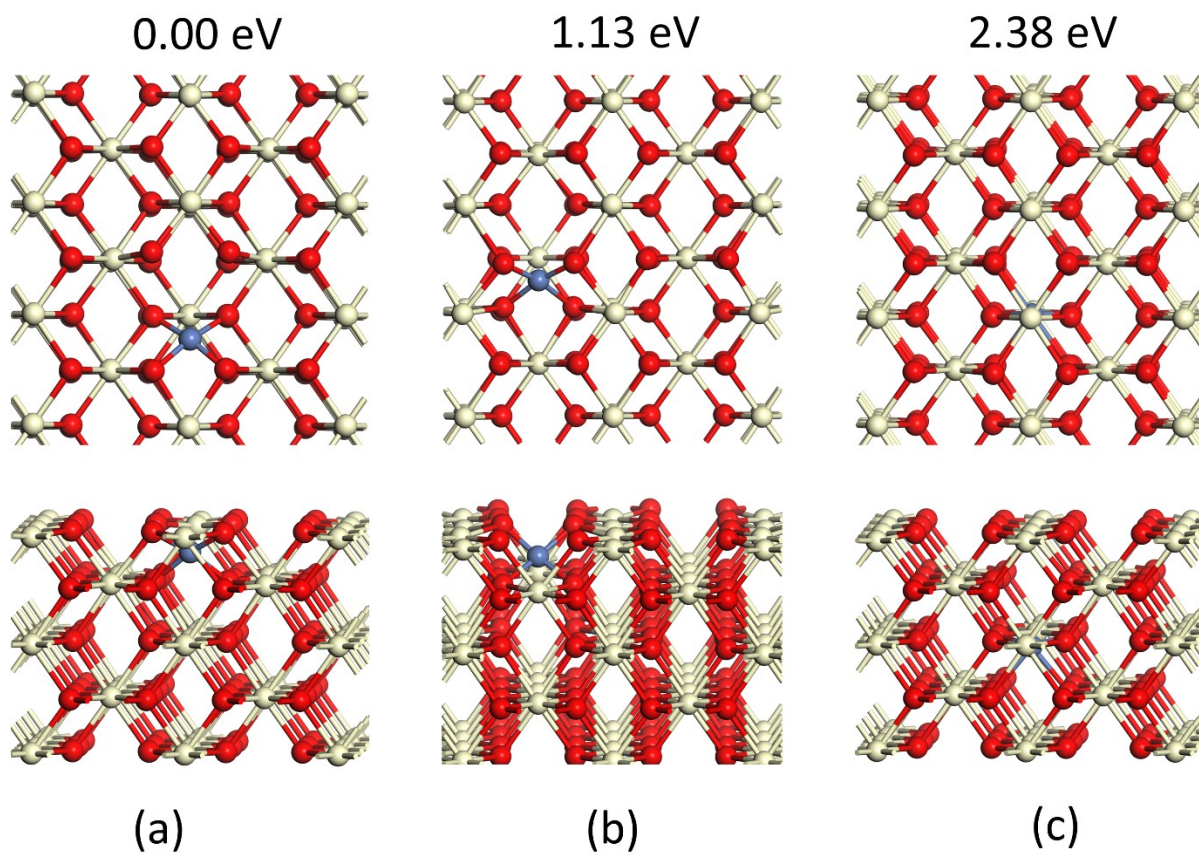


Figure S2 Top and side views of geometries of Ni doped $\text{CeO}_2(110)$ with substituted Ce located at different layers. (a) Configuration 1 which a surface Ce (first layer) replaced by Ni; (b) Configuration 2 which a second layer Ce replaced by Ni; (c) Configuration 3 which a third layer Ce replaced by Ni. The values in the figure indicated the energies relative to that of configuration 1. Color scheme: Ni, blue; Ce, yellow; oxygen, red.

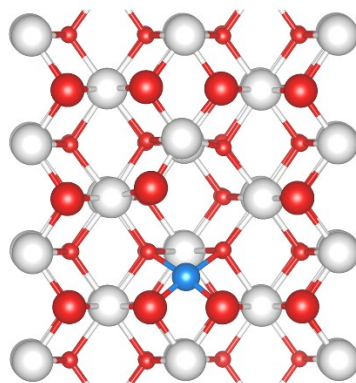


Figure S3 Calculated spin density of Ni-CeO₂(110)-O_v. Color scheme: Ni, blue; Ce, white; oxygen, red; spin density, cyan or yellow.

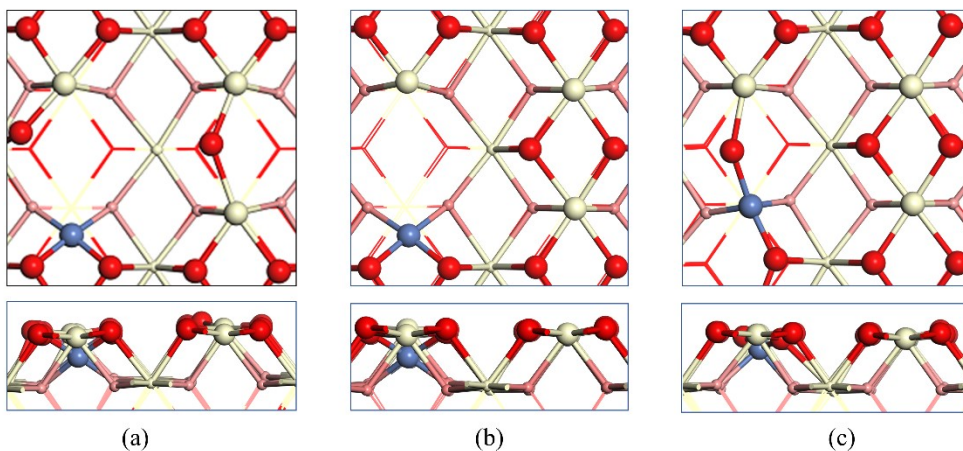


Figure S4 Top and side views of geometries of Ni-CeO₂(110) with two O_vs. (a) second O_v located at O1, (b) second O_v located at O2, (c) second O_v located at O3. O1, O2 and O3 are indicated in Figure 1 (d). Color scheme: Ni, blue; Ce, yellow; oxygen, red; subsurface oxygen, light red.

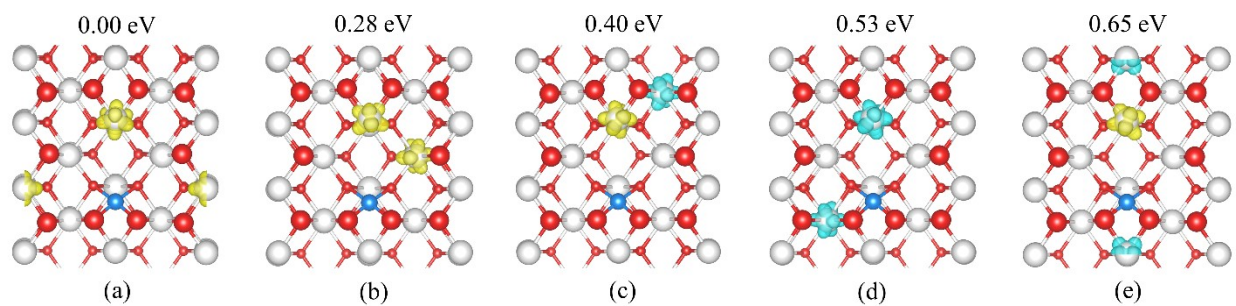


Figure S5 Calculated spin density with Ce^{3+} located at different positions for $\text{Ni-CeO}_2(110)\text{-}2\text{O}_v$. The data of each image is the energy of each configuration relative to the most stable one. Color scheme: Ni, blue; Ce, white; oxygen, red; spin density, cyan or yellow.

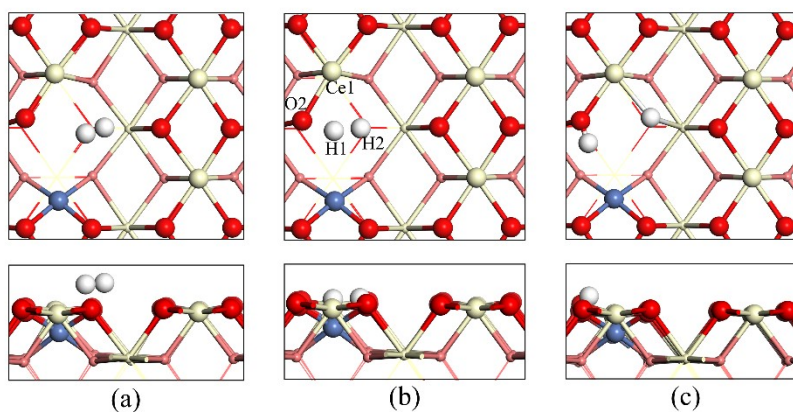


Figure S6 Top and side views of geometries related to H₂ dissociation on the Ce/O CLP at Ni-CeO₂(110)-O_v. (a) H₂ adsorption (H₂*), (b) TS of the H₂ dissociation, (c) heterolytic products (H*-Ce + H*-O). Color scheme: Ni, blue; Ce, yellow; oxygen, red; subsurface oxygen, light red; H, white.

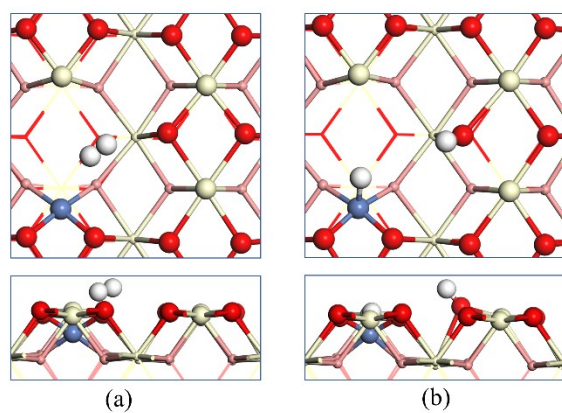


Figure S7 Top and side views of geometries of adsorbed H₂ (a) and dissociated H₂ (H*-Ni+ H*-O) (b) on Ni-CeO₂(110)-2O_v. Color scheme: Ni, blue; Ce, yellow; oxygen, red; subsurface oxygen, light red; H, white.