

Abstract Submitted
for the MAR14 Meeting of
The American Physical Society

Rationalization of Hubbard U in CeO_x from first principles: Unveiling the role of local structure in screening¹ DEYU LU, PING LIU, Center for Functional Nanomaterials, Brookhaven National Laboratory — DFT+U method has been widely employed in theoretical studies on various ceria systems to correct the delocalization bias in local and semi-local DFT functionals with moderate computational cost. To rationalize the Hubbard U of Ce 4f, we employed the first principles linear response method to compute Hubbard U for Ce in ceria clusters, bulks, and surfaces. We found that in contrast to the commonly used approach treating U as a constant, the Hubbard U varies in a wide range from 4.1 eV to 6.7 eV, and exhibits a strong correlation with the Ce coordination numbers and Ce-O bond lengths, rather than the Ce 4f valence state. The variation of the Hubbard U can be explained by the changes in the strength of local screening due to O → Ce intersite transition. Our study represents a systematic, quantitative investigation of the relationship between the Hubbard U and the local atomic arrangement, enabling a DFT+environment-dependent U scheme that can have potential impact on catalysis research of strongly correlated systems.

¹This work is supported by the U.S. Department of Energy, Office of Basic Energy Sciences, under Contract No. DE-AC02-98CH10886.

Deyu Lu
Center for Functional Nanomaterials, Brookhaven National Laboratory

Date submitted: 15 Nov 2013

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