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PII: S2095-9273(17)30369-9
DOI: http://dx.doi.org/10.1016/j.scib.2017.07.007
Reference: SCIB 178

To appear in: *Science Bulletin*

Received Date: 25 April 2017
Revised Date: 28 June 2017
Accepted Date: 4 July 2017

Please cite this article as: J-H. Lan, Z-F. Chai, W-Q. Shi, A combined DFT and molecular dynamics study of U(VI)/calcite interaction in aqueous solution, *Science Bulletin* (2017), doi: http://dx.doi.org/10.1016/j.scib.2017.07.007

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Received 25 April, 2017

Accepted 28 June, 2017

Accepted 4 July, 2017

A combined DFT and molecular dynamics study of U(VI)/calcite interaction in aqueous solution

Jian-Hui Lan,[1] Zhi-Fang Chai[1,2] and Wei-Qun Shi*[1]

[1]Laboratory of Nuclear Energy Chemistry, and Key Laboratory for Biomedical Effects of Nanomaterials and Nanosafety, Institute of High Energy Physics, Chinese Academy of Sciences, Beijing 100049, China

[2]School of Radiological and Interdisciplinary Sciences (RAD-X) and Collaborative Innovation Center of Radiation Medicine of Jiangsu Higher Education Institutions, Soochow University, Suzhou 215123, China

E-mail: shiwq@ihep.ac.cn

Abstract Here we present a combined DFT and molecular dynamics study of uranyl (U) interaction mechanisms with the calcite (104) surface in aqueous solution. The roles of three anion ligands (CO_3^{2-}, HCO_3^-, OH^-) and solvation effect in U(VI) interaction with calcite have been evaluated. According to our calculations, water adsorbed on the calcite (104) surface prefers to exist in molecular state rather than dissociative state. Energy analysis indicate that the positively charged uranyl species prefers to form surface complexes on the surface, while neutral uranyl species may bind with the surface via both surface complexing and ion exchange reactions of U(VI) → Ca(II). In contrast, the negatively charged uranyl species prefer to interact with the surface via ion exchange reactions of U(VI) → Ca(II), and the one with $UO_2(CO_3)_2(H_2O)^{2-}$ as the reactant becomes the most favorable one in energy. We also