Roles of 6d states on the atmospheric corrosion mechanisms of uranium

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Abstract Surface hydrogenation, oxidation, and other corrosion-related reactions of uranium have attracted continuous concern for many years, because these processes are not only of great scientific interest, but also of significant practical importance for nuclear industries. Through several theoretical studies, we reveal that the 6d electronic states play important roles on the chemical properties of uranium. Our research progress can be summarized as follows: i) On γ-U surface, H_2 molecules dissociate barrierlessly due to s-d electronic hybridizations; ii) On α-U surface, there are also "s-d interaction" adsorption channels along which H_2 molecules dissociate barrierlessly; iii) Interactions between the three frontier molecular orbitals with surface d states lead to that H_2O dissociate spontaneously on the γ-U surface; iv) U-6d state is the only evolving state in different UnOm clusters, which hybridizes with U-5f states in U-rich clusters, and hybridizes with O-2p states in O-rich clusters.

Keywords hydrogenation, oxidation, uranium

Reference

[1] Y. Yang, P. Zhang, P. Shi, X.L. Wang, s-d Electronic Interaction Induced H_2 Dissociation on the γ-U(100) Surface and Influences of Niobium Doping, J. Phys. Chem. C 115 (2011) 23381.

[2] P. Shi, Y. Yang, B.Y. Ao, P. Zhang, X.L. Wang, Influences of Surface Substitutional Ti Atom on Hydrogen Adsorption, Dissociation, and Diffusion Behaviors on the α-U(001) Surface, J. Phys. Chem. C 118 (2014) 26634.

[3] Y. Yang, P. Zhang, First-principles molecular dynamics study of water dissociation on the γ-U(100) surface, J. Phys.: Condens. Matter 27 (2015) 175005.

[4] Y. Yang, H.T. Liu, P. Zhang, Structural and electronic properties of UnOm (n=1-3,m=1-3n) clusters: A theoretical study using screened hybrid density functional theory, J. Chem. Phys. 144 (2016) 184304.