The hydriding resistance of plutonium oxides and mononitride: A view from \textit{ab initio} molecular dynamics\textsuperscript{1} BO SUN, HAIFENG LIU, HAIFENG SONG, Institute of Applied Physics and Computational Mathematics, CAEP — Based on the non-local van der Waals density functional (vdW-DF)+\textit{U} scheme, we carry out the \textit{ab initio} molecular dynamics study of the interaction dynamics for H\textsubscript{2} molecules impingement against Pu-oxides and mononitride surfaces. We show that except for the weak physisorption, both PuO\textsubscript{2} and PuN surfaces are so difficult of access that almost all of H\textsubscript{2} molecules will bounce back to the vacuum when their initial kinetic energies are not sufficient. Although the dissociative adsorption of H\textsubscript{2} on PuO\textsubscript{2} surfaces is found to be exothermic, the collision-induced dissociation barriers of H\textsubscript{2} are very high (up to 2.2 eV). However, PuO\textsubscript{2} overlayer on Pu-metal can be reduced to $\alpha$-Pu2O\textsubscript{3} drived by oxygen-lean conditions, and H\textsubscript{2} can penetrate and diffuse in $\alpha$-Pu2O\textsubscript{3} easily. As a result, $\alpha$-Pu2O\textsubscript{3} can promote the hydriding process of Pu. Unlike PuO\textsubscript{2}, PuN is found to be one kind of stable and uniform passivation layer against Pu-hydriding. Specifically, the incorporation of PuN and H-atom is proven to be thermodynamically unstable. Overall, our current study reveals the mechanical and chemical resistances of Pu-oxide and Pu-nitride to hydrogen corrosion, which have strong implications to the understanding of the surface corrosion and passivation of Pu metal.

\textsuperscript{1}This work was supported by the FDST of CAEP under Grant No. 9090707.

Bo Sun
Institute of Applied Physics and Computational Mathematics, CAEP

Date submitted: 22 Jan 2015
Electronic form version 1.4