Supplemental Material

Migration and aggregation of fission products and their impacts on physical properties in UO_2: Deep potential molecular dynamics simulations

Zhihong Chen[a], Yunfei Hong[a], Junkai Deng[a,*], Zhibin Gao[a], Ronghua Chen[b], Rui Tang[c], Hongxing Xiao[c,*], Xiangdong Ding[a], Jun Sun[a]

[a] State Key Laboratory for Mechanical Behavior of Materials, Xi'an Jiaotong University, Xi'an 710049, China

[b] School of Nuclear Science and Technology, Xi'an Jiaotong University, Shaanxi, Xi'an, 710049, China

[c] National Key Laboratory of Nuclear Reactor Technology, Nuclear Power Institute of China, Chengdu, 610213, China

[*]Corresponding authors: junkai.deng@mail.xjtu.edu.cn (J. Deng); xiaohongxing2003@163.com (H. Xiao)

In this work, for the defect formation energy calculations (including vacancy and interstitial defects), the reference state is the perfect UO_2 crystal. For the incorporation energy calculations, the reference structure is a UO_2 crystal containing a pre-existing vacancy. Specifically, the formation energies of vacancies and interstitials were calculated using the following formulas:

$$E_{vacancy} = E_X^{N-1} + E_X - E^N \quad (1)$$

$$E_{interstitial} = E_X^{N+1} - E^N - E_X \quad (2)$$

where E^N is the energy of the perfect supercell without defects, E_X^{N-1} is the energy of the supercell with a single vacancy of type X, and E_X^{N+1} is the energy of the supercell with a single interstitial of type X. E_X is the corresponding chemical potential of a uranium or oxygen atom in the reference state (α-Uranium and oxygen molecule, respectively).

The incorporation energy $E_{Incorporation}$ of each FP atom at sites X, including a uranium vacancy (V_U) or an oxygen vacancy (V_O), is estimated using the following formula:

$$E_{Incorporation} = E_X^N - E_X^{N-1} - E_{fp} \quad (3)$$

where E_X^N is the energy of the supercell containing the FP atom at site X, and E_{fp} is the energy of the FP atom in isolation.

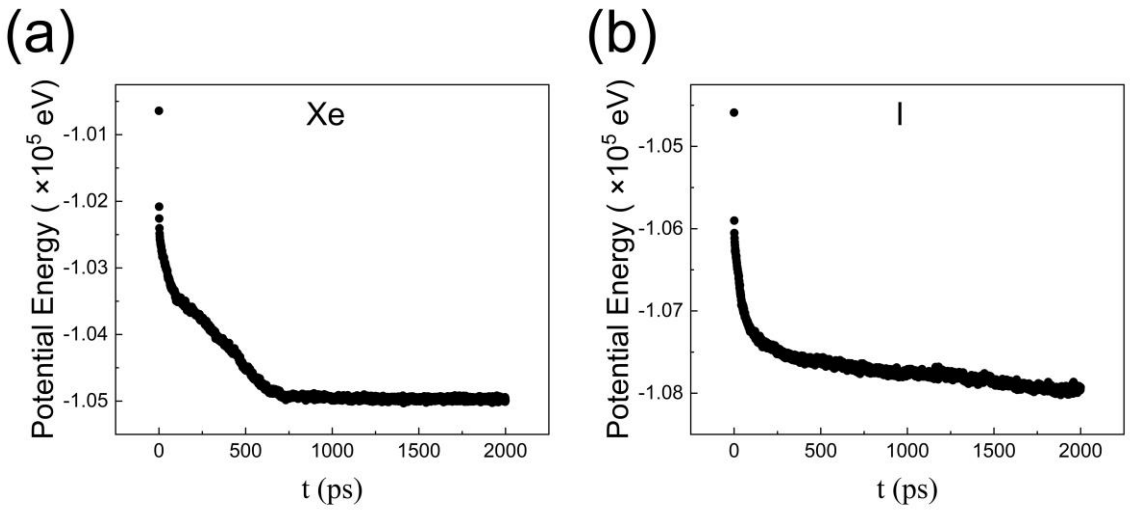

FIG. S1. Potential energy variation in UO_2-Xe (a) and UO_2-I (b) systems in MD&MC simulations.

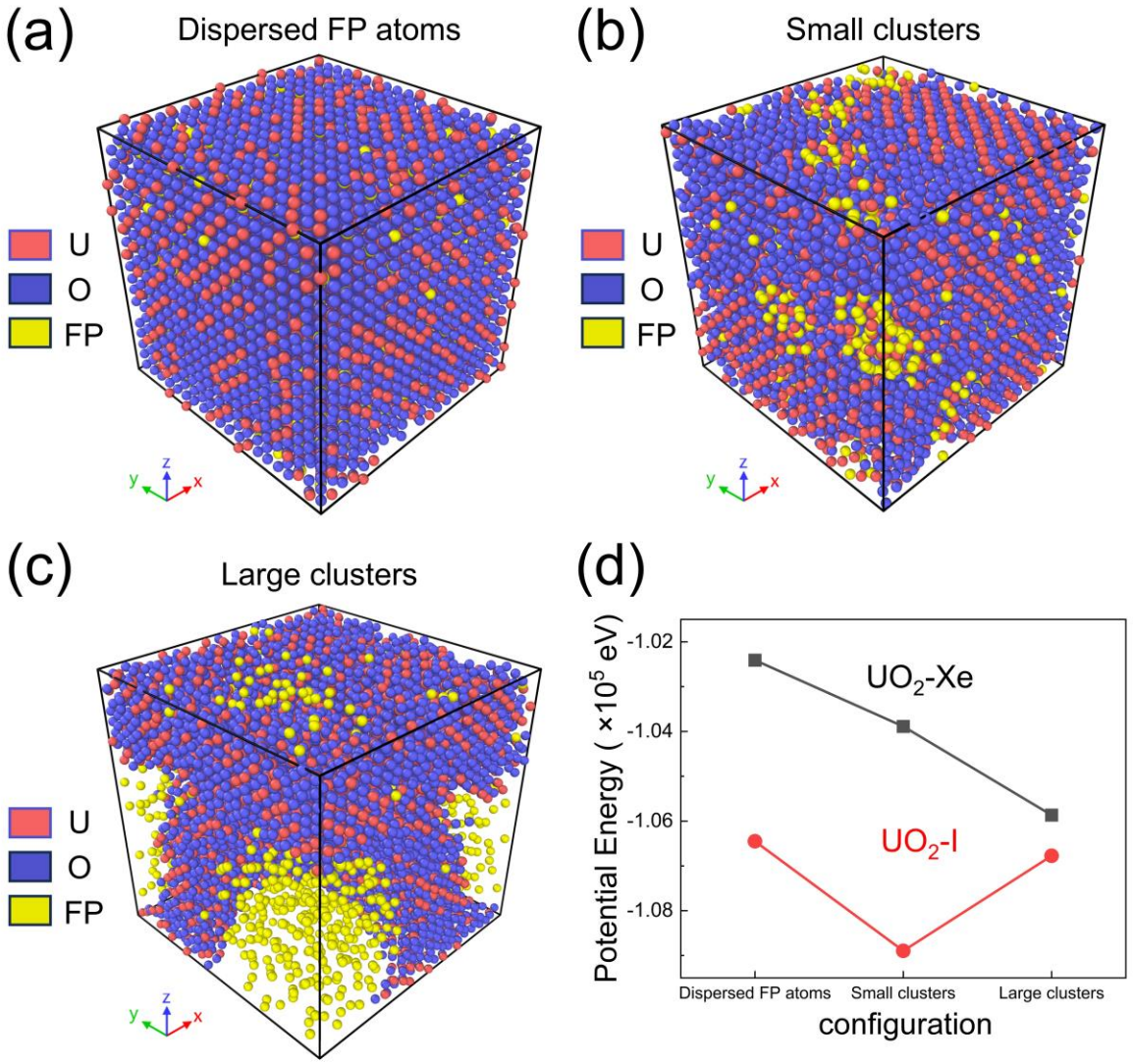

FIG. S2. Different configurations: (a) dispersed FP atoms, (b) small clusters, (c) large clusters for UO_2-Xe and UO_2-I systems, and (d) comparison between their potential energies for these different configurations.

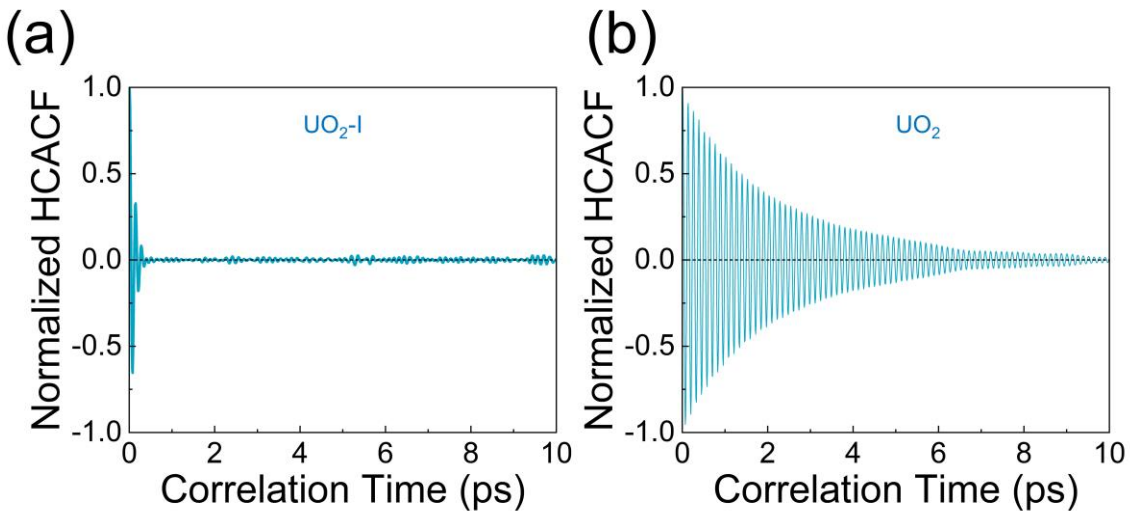

FIG. S3. The heat current autocorrelation function for UO_2-I system and pristine UO_2 at 300 K.

TABLE S1. UO_2 properties calculated by $DP_{U\text{-}O\text{-}Xe}$ and $DP_{U\text{-}O\text{-}I}$ models.

properties	$DP_{U\text{-}O\text{-}Xe}$	$DP_{U\text{-}O\text{-}I}$
Bulk modulus (GPa)	217.54	206.72
Hill's Shear modulus (GPa)	88.54	94.66
Young's modulus (GPa)	233.89	246.36
Poisson's ratio	0.32	0.30

TABLE S2. Comparison of UO_2 thermal conductivity in different sizes calculated by DP models with other MD and experimental results. The values in parentheses were converted according to a correction equation [1], accounting for 95% of the theoretical UO_2 density to facilitate comparison with experimental data.

	thermal conductivity ($W \cdot m^{-1} \cdot K^{-1}$)
4 × 4 × 4 supercell by DP	15.08 (13.23)
5 × 5 × 5 supercell by DP	15.07 (13.22)
10 × 10 × 10 supercell by DP	14.85 (13.03)
12 × 12 × 12 supercell by DP	14.82 (13.00)
Cooper (MD)	about 16.86
Arima (MD)	about 15.17
Maxwell (MD)	about 13.52
Weisensee (experiment)	10.2 [2]

REFERENCE

[1] R. Brandt and G. Neuer, Thermal Conductivity and Thermal Radiation Properties of UO_2, J. Non-Equilib. Thermodyn. **1**, 3 (1976).

[2] P. B. Weisensee, J. P. Feser, and D. G. Cahill, Effect of ion irradiation on the thermal conductivity of UO_2 and U_3O_8 epitaxial layers, J. Nucl. Mater. **443**, 212 (2013).