Thermophysical and Atomic Transport Properties of Mixed-Oxide Fuel: Interatomic Empirical Potential and *Ab Initio* Calculations

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Mixed-oxide (MOX) fuel, commonly used in Light Water Reactors (LWR), is the reference fuel for fast breeder reactors and is envisaged in France to implement plutonium multi-recycling in LWR. For these purposes, it is necessary to determine accurately its properties. Among them, thermodynamic and atomic transport properties can be computed by empirical potentials and first-principles calculations. In the present study, we show the necessity to use these two methods to determine rigorously the properties of $U_{1-y}Pu_yO_2$ MOX fuel.

Concerning thermodynamic properties, we focus on the heat capacity over the entire range of Pu content and a large range of temperature (from 300 K to the melting temperature) using classical (CMD) and *ab initio* (AIMD) molecular dynamics. CMD with CRG interatomic potential [1] predicts a full Bredig transition [2] at high temperature (T > 1800 K) and an effect of the Pu content solely on this transition. For UO_2, the occurrence of the complete Bredig transition is in good agreement with several available experimental studies [3]–[6]. We performed AIMD calculations in the cases of UO_2 and PuO_2, which show a satisfactory agreement and thus confirm the CMD results.

Concerning atomic transport properties, we investigate the influence of the chemical disorder on the defect formation energies (DFE) of bound Schottky defects (BSD) and oxygen vacancies in $(U,Pu)O_2$ fuel. This disorder is characterized by the random distribution of uranium and plutonium on the cation crystallographic sublattice leading to multiple possible configurations around a point defect. CRG potential calculations show that the effect of the disorder on DFE is significant up to 6.5 Angströms around a defect and that DFE deviate by 1.2 eV depending on the multiple neighboring configurations around BSD. First-principles calculations have been performed on selected configurations to confirm this result.

References

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