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First principle calculation of helium in $La_2Zr_2O_7$: effects on structural, electronic properties and radiation tolerance

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Abstract:

First principle calculations based on density functional theory have been employed to study structural effects of trapping helium in $La_2Zr_2O_7$ pyrochlore. Lattice swelling and the distortion of unit cell have been found in He-$La_2Zr_2O_7$ systems. By analyzing the electronic structures and chemical bonding of He-$La_2Zr_2O_7$ systems, weak repulsive and attractive chemical interactions of helium in $La_2Zr_2O_7$ pyrochlore have been observed. The formation energies have been calculated to assess the relative stability of various helium interstitial configurations and the results show that the octahedral interstitial site is the most stable structure. The cation antisite defect formation energies and the *x* positional parameter for 48*f*-site oxygen are calculated to predict the radiation resistence of He-$La_2Zr_2O_7$ systems. The results indicate that the presence of low concentration of He interstitials may increase the radiation resistance of $La_2Zr_2O_7$ pyrochlore.

Keywords:

Density functional theory; $La_2Zr_2O_7$ pyrochlore; Helium interstitial; Formation energy;

1. Introduction

Nuclear energy gets extensive development worldwide as a clean energy, which can solve the ecological environment problems caused by the over exploitation of fossil fuels, but it also creates challenges. The most serious one is dealing with high-level nuclear waste in reasonable ways [1-5]. $La_2Zr_2O_7$ pyrochlore has been proposed as a potential superior host phase for the long-lived transuranic (TRU) elements in nuclear waste, because it possesses high chemical durability, good thermal stability, low leach rate and the enhanced radiation tolerance after TRU elements incorporated [6-9]. In recent decades, many of experimental and theoretical investigations involving $La_2Zr_2O_7$ pyrochlore have been carried out to evaluate the radiation stability and the solubility of actinides in it [9-17]. For example, Lian *et al.* [16] argued that there was a close correlation between the radiation resistance of $La_2Zr_2O_7$ and its deviation from the ideal fluorite structure. Besides, the cation antisite formation energy has been introduced to predict the disordering extent and then assess the radiation tolerance of $La_2Zr_2O_7$ [16, 18, 19]. Cerium has been used as a nonradioactive surrogate for plutonium to investigate

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