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First-principles investigations of the physical properties of binary uranium silicide alloys

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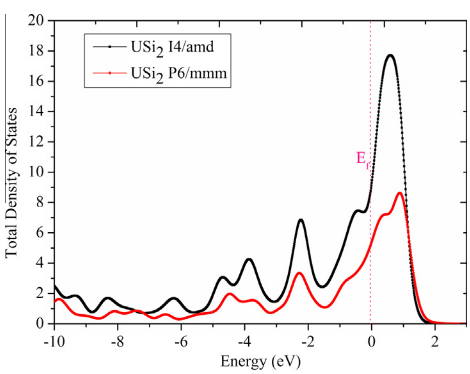
GRAPHICAL ABSTRACT

Total density of states for USi_2.



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ABSTRACT

The structural, elastic properties and the Debye temperature of binary Uranium Silicide (U-Si) alloys are investigated by using the first-principles plane-wave pseudopotential density function theory within the generalized gradient approximation (GGA). The ground states properties are found to agree with the available experimental data. The mechanical properties like shear modulus, Young's modulus, Poisson's ratio σ and ratio B/G are also calculated. Finally, The averaged sound velocity (v_m), the longitudinal sound velocity (v_l), transverse sound velocity (v_t) and the Debye temperature (θ_D) are obtained. However, the theoretical values are slightly different from few existed experiment data because the latter was obtained at room temperature while the former one at 0 K.

1. Introduction

The binary Uranium Silicide (U-Si) alloys have been studied in great detail due to their crystal structure, magnetic and electronic properties determined by susceptibility and calorimetry measurements at low temperature [1,2]. This binary system comprises such eight well-defined compounds as USi, USi_2, USi_3, U_3Si, U_3Si_2, U_5Si_4, U_3Si_5 and $USi_{1.88}$.

Remschnig et al. [1] described the magnetic behavior of the USi by a modified Curie–Weiss law. Yagoubi et al. [3] have studied the structural and electronic properties under high pressure using both theory and high pressure synchrotron X-ray diffraction experiments. The U_3Si_2, the state-of-the-art nuclear fuel material mostly used in modern research reactors, is made by melting metallic uranium and pure silicon in induction furnace. The compound U_5Si_4 has been elaborated and characterized by X-ray diffraction for

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the first time by Noel et al. [4,5] However, the temperature range of stability of this compound is unknown [6]. U_3Si_5 crystallized in hexagonal defect AlB_2-type. U_3Si_5 melted congruently at 2043 K and had a composition range o1-U_3Si_5 to o2-U_3Si_5 in the temperature range of 1273–1573 K [7]. $USi_{1.88}$ was the orthorhombic defect $GdSi_2$-type at its silicon poor phase boundary and was the tetragonal defect $ThSi_2$-type structure at its silicon rich phase boundary [1,8]. The stoichiometric and non-stoichiometric phases USi, U_3Si_2, U_5Si_4, U_3Si_5 and $USi_{1.88}$ have already been studied and were not further considered in the present study. To reveal the mechanical properties and Debye temperature, we calculated the structure, elastic properties and the Debye temperature of the USi_2, USi_3 and U_3Si alloys using the first-principles method in this work.

The rest of the paper is organized as follows: in Section 2, we described briefly the computational methods used in this work; Section 3 contains our results and discussions, involving structural and mechanical properties and Debye temperature of the U-Si system; finally, the conclusion is given in Section 4.

2. Calculation methods

The first-principle calculations were carried out by using the plane-wave pseudopotential method (PW-PP) within DFT, which was implemented in CASTEP [9]. As for the exchange and correlation terms, the PW91 [10] function was used within the generalized gradient approximation (GGA) [11]. Using the PW-PP method, $3s^23p^2$ of Si and $5f^36s^26p^66d^17s^2$ of U were treated explicitly as valence electrons. In this study, we employed 800 eV as the cutoff energy of plane-wave and a $9 \times 9 \times 9$ Monkhorst-Pack k-point mesh because they provided accurate enough energy for these compounds. The structural parameters of U-Si alloys were calculated by using the Brodyden-Fletcher-Goldfarb-Shanno (BFGS) method [12].

3. Results and discussion

3.1. Structure properties

The compound USi_2, with exact 1:2 stoichiometry, is stable below 723 K. It has two phases (Space group: *I4/amd*, $ThSi_2$-type, Fig. 1a; Space group: *P6/mmm*, AlB_2-type, Fig. 1b). For *I4/amd* phase [13], there are 12 atoms in the cell, with Si atoms at the 8e Wyckoff site (0, 0, 0.41) and U atoms the 4a Wyckoff site (0, 0, 0); for the *P6/mmm* phase [14], there are 3 atoms in the cell, with Si atoms at the 2d Wyckoff site (0.3333, 0.6667, 0.5) and U atoms the 1a Wyckoff site (0, 0, 0). The silicon-rich compound USi_3 has the cubic phase

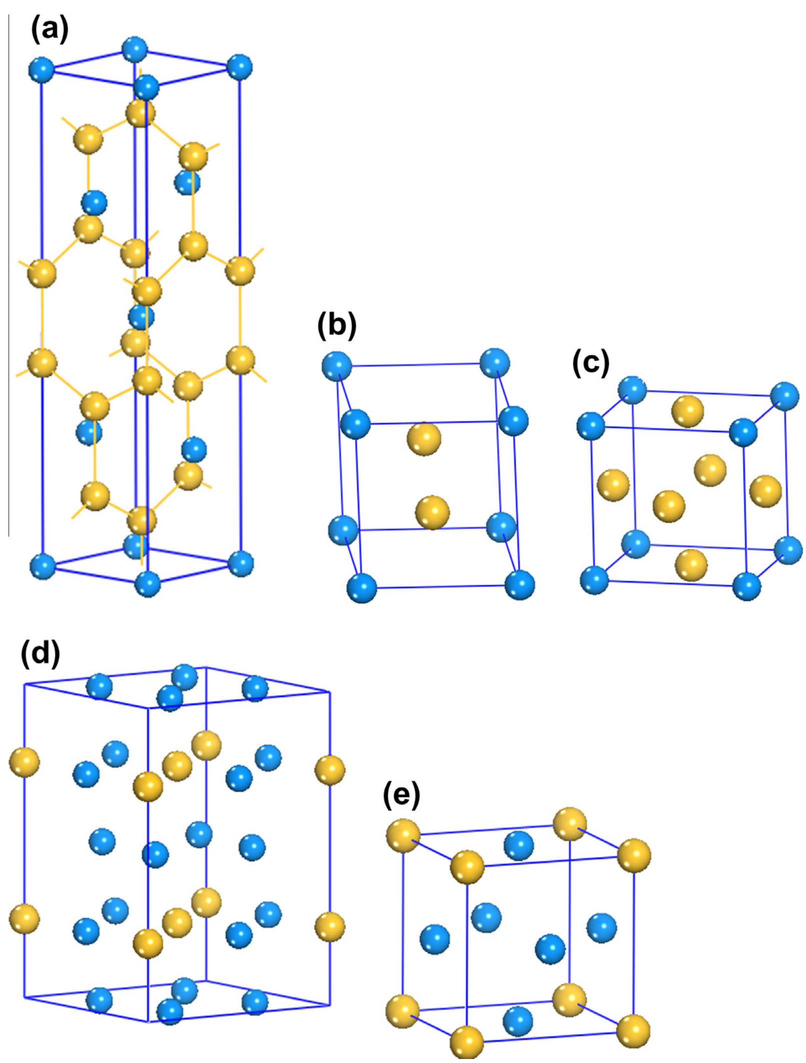

Fig. 1. Crystal structures of U-Si alloys. (a) Tetragonal phase of USi_2, (b) Hexagonal phase of USi_2, (c) Cubic phase of USi_3, (d) Tetragonal phase of U_3Si, and (e) Cubic phase of U_3Si. The blue and yellow spheres represent U and Si atoms, respectively. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)