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First principles calculation of the Al_3U–Si_3U pseudobinary fcc phase equilibrium diagram

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ABSTRACT

In the frame of RERTR Program (Reduced Enrichment for Research and Test Reactors) it is being developed a high density uranium based fuel that could remain stable in the body cubic centered (bcc) phase during fabrication and irradiation in the reactor. Research is focused in a U–Mo alloy dispersed fuel in aluminum matrix. The main problem focuses in an undesirable growth of the interface between fuel and Al matrix. This problem could be reduced with the addition of Si to the matrix and the subsequent stabilization of UAl_3 in the interface. We investigated this possibility in this work using first principles theory and methods. We demonstrated that $U(Al,Si)_3$ phase is stable in the UAl_3–USi_3 pseudobinary system. Moreover, calculations were able to justify the existence of an experimentally reported phase with $U_4Al_9Si_3$ stoichiometry and a wide composition range.

1. Introduction

Several uranium alloys are being studied in the frame of the RERTR (Reduced Enrichment for Research and Test Reactors) program. The objective is that the fuel could remain stable in the body centered cubic phase (γU bcc solid solution) during fabrication ($\sim$500 °C) and irradiation ($\sim$250 °C), i.e., at temperatures at which αU is the equilibrium phase [1]. Fuel powder is dispersed in aluminum and co-laminated to form the fuel element. Under irradiation diffusion takes place and an interlayer (IL) grows around the fuel particles. As a consequence, swelling of the fuel element and porosities are observed [2]. To prevent a failure caused by this behavior, the IL nature must be characterized and optimized.

The IL composition has been experimentally measured in a diffusion couple U-7 wt% Mo/Al, and classified in two layers of 77 and 82 at% Al [2]. These authors reported these compositions as $(U,Mo)Al_3$ and $(U,Mo)Al_{4.4}$ since the content ratio U/Mo in both layers was not altered by irradiation. Besides, Mirandou et al. [3] found that the IL constituents in the out of pile heat treated diffusion couples at 580 °C U-7wt%Mo/Al, were $(U,Mo)Al_4$ (oI20, space group 74), $(U,Mo)Al_3$ (cP4, space group 221) and also UMo_2Al_{20} (cF184, space group 227), in agreement with previous experiments.

Research was then focused on the identification of the reason for the undesired behavior of these or some of these compounds. It was suggested [4] that the IL is amorphized during irradiation and its instability is due to amorphous character. The same authors pointed out that an IL with a greater Al content tends to amorphized more easily so the key to avoid massive pore formation could be to obtain an IL constituent between U–Mo alloy and Al with a low Al content such as $(U–Mo)Al_3$. Their proposal was to stabilize $(U,Mo)Al_3$ compound against the formation of a compound with a greater Al content such as UAl_4. UAl_4 was also identified as an undesired product in the IL with other arguments. Gan et al. [5] commented that the UAl_4 compound remains crystalline under irradiation, but its fragility could be responsible for the swelling and following breaking of the fuel element inside the reactor.

Among the possible solutions to the problem, modifications of the fuel composition by the addition of Si have been successful. The objective was to inhibit the formation of UAl_4 compound during the fabrication process at high temperature of a fuel element based on the dispersion of UAl_3 compound in Al matrix [6–9]. Other researchers showed that in the case of U_3Si_2 dispersions in Al the IL composition after irradiation was $U(Al_{0.75},Si_{0.25})_3$ [10]. More recently, the undesired behavior of the irradiation products in the IL was not observed in diffusion couples U_3Si/Al [11] and U_3Si_2/Al [12].

For the same purpose experiments in dispersed U–Mo fuel in a Si-modified Al matrix were performed [13]. Out of pile studies showed that Si is accumulated in the IL in between the U–Mo alloy and Al–Si [14,15]. On the other hand, in pile experiments

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also show the beneficial effect of Si addition: IL thickness was smaller than the one observed in fuel elements based in a pure Al matrix and no porosity was formed [16,17].

The USi_3–UAl_3 pseudobinary system was evaluated by Dwight [18] through measurements in binary or ternary fields of the U–Al–Si phase equilibrium diagram. He suggested a complete miscibility between UAl_3 and USi_3 at 900 °C but commented that a gap in the solid solution $U(Al,Si)_3$ could exist at lower temperatures. Another proposal [19] is based on the identification of an intermetallic compound by x-ray diffraction techniques that is stable till 1340 °C, crystallographically belonging to the 223 space group, with 256 atoms in the unit cell and the stoichiometry $Al_{144}Si_{48}U_{64}$ but exhibiting a wide solubility range at room temperature (between 0.07 and 0.5 at% USi_3 in the pseudobinary UAl_3–USi_3).

We present in this work the results of our investigation of the disordered phase $U(Al,Si)_3$. We evaluated the formation energy of the perfectly disordered solid solution in the pseudobinary UAl_3–USi_3 using first principles calculated total energies in a cluster expansion method. We also show the phase equilibrium diagram resulting from the Monte Carlo simulations of thermodynamic properties at finite temperatures.

2. Calculation methods

2.1. First principles calculations of properties of ordered structures

The total energies of the superstructures and the pure elements were calculated using DFT together with the full-potential linearized augmented-plane wave (FP-LAPW) method in the generalized gradient approximation (GGA) [20], including scalar relativistic corrections [21,22], and implemented in the WIEN2K code [23]. We have neglected here the correction introduced by the spin-orbit term taking into account that the aim of the work is to evaluate relative stability of compounds. As it has already been noticed [24], changes in total energies are canceled at a great extent when differences in total energies are made to obtain formation energies. All parameters were optimized so as to enhance accuracy of calculations. We searched for the best value for the kinetic energy cut-off for the plane-wave basis that met the requirements of convergence and acceptable computer time by testing the parameters RKmax and muffin-tin radii (rmt). They were set as rmt[U]=2.5, rmt[Al]=2.2, rmt[Si]=2.2 and RKmax=10. The core configurations [Xe] $4f^{14}$ $5d^{10}$, [He] $2s^2$ $2p^6$ and [He] $2s^2$ $2p^6$ were considered for U, Al and Si respectively. The magnitude of largest vector in charge density expansion (Gmax) was set to 18. The calculations used a number of k points such that the total energy of the structures changed less than 0.1 mRy/atom (approx. 0.13 kJ/mol). The number of k points in the calculation was 256 in an irreducible wedge of the Brillouin zone. The criterion to obtain the total energy was to minimize it as a function of lattice parameter.

2.2. Cluster expansion method applied to disordered structure properties

The cluster expansion is a generalization of the well-known Ising Hamiltonian [25]. Over the last twenty years, several publications have taken care of the formalism for obtaining solid state properties of an alloy from quantum mechanical energy calculations involving a small number of ordered structures [26–29].

The expectation value of a function of the configuration of the system $<P>$, such as the thermodynamic average of the configurational energy, can be expressed easily in terms of the multisite correlation function ξ_α:

$$\langle P\rangle = \sum_{\alpha} \quad V_\alpha \quad \xi_\alpha \tag{1}$$

where α denotes a cluster of $|\alpha|$ sites. The expansion coefficients V_α are known in the literature as the effective cluster interactions (ECI) for the physical property P. The complete cluster expansion of Eq. (1) is formally exact, however, the utility of this rests in the possibility of identifying a hierarchy of a small number of clusters whose contributions V_α to the physical property P dominate those of the remaining clusters.

The formation energy ΔE_F of any ordered or disordered alloy lattice may be now described with a truncated expression of the bilinear form of Eq. (1). The unknown parameters ECIs of the cluster expansion can then be determined by fitting this expression to a set of formation energies of ordered compounds, for which the corresponding correlation functions are known. These energies can be obtained, for instance, through first-principles electronic calculations. Because those calculations demand time, we have access to a finite number of structural energies. The number of V_α parameters to compute can be equal or smaller than the number of known structural energies. In case they are equal, ECIs can be obtained by the direct inversion method of Connolly–Williams [30]. However, it has been determined that a better cluster expansion is obtained if the system of Eq. (1) is over determined [28]. The choice of the number and hierarchy of the clusters is determined using a Cross-Validation (CV) score defined as

$$CV = \left(\frac{\sum_{n=1}^{N} (\Delta E_F^n - \Delta \hat{E}_F^n)^2}{N} \right)^{1/2} \tag{2}$$

where ΔE_F^n is the first-principles electronic calculated formation energy of the ordered compound n, whereas $\Delta \hat{E}_F^n$ is the value predicted by the cluster expansion with ECI's obtained with a least-squares fit to the $(N-1)$ other structural energies. For a finite number of structural energies the CV score goes through a minimum when the number of clusters included in the expansion increases.

The right clusters and number of known structures that will enter in the cluster expansion must fulfill the following three criteria:

i) The ground state phase diagram should exhibit known stable phases and the predicted energies for structures other than the ones included in the cluster expansion should lie above the ground state tie lines.
ii) The predicted cluster-expanded energies of ordered compounds in the expansion should agree with the corresponding formation energy calculation via first principles (the CV score is small).
iii) The magnitude of the ECI decays as a function of the diameter of the corresponding cluster and as a function of the number of sites it contains.

We used the MAPS program within the ATAT package [31] in conjunction with the WIEN2k code [32] to evaluate the needed structures, calculate their total energies, calculate the CV score and construct the set of ECIs to reproduce the system energetic.

2.3. Finite temperature thermodynamic properties. Phase diagram

The equilibrium states at constant volume and temperature were obtained by calculating the Gibbs free energies for all compounds in the ground state and for the solid solution at the same constant temperatures by means of a Monte Carlo simulation [33]. With this purpose, the cluster expansion obtained as described above was used as input for the Easy Monte Carlo Code (EMC2) program [34]

within the ATAT package [31]. Simulations were made within a semi-grand canonical ensemble with a fixed number of atoms where energy and concentration are allowed to fluctuate and both temperature and the difference μ between chemical potentials of the two species are imposed.

The starting point for calculation is chosen as a value of chemical potential and temperature where the grand canonical potential can be computed analytically. This is, a low temperature for ordered phases and a sufficiently high temperature for disordered solid solution [35]. Then, by scanning over temperature and chemical potential range related values for potential and composition can be obtained for the whole phase diagram. Finally, the equilibrium state emerges from intercepting curves in grand canonical potential vs. μ plots drawn for all metastable phases at the same fixed temperature. This method has been chosen instead of the common tangent method because it proved to be simpler to implement. Details of calculations are given below in the results section.

3. Results and discussion

3.1. UAl_3–USi_3 pseudobinary system ground state

In order to apply the cluster expansion model to the pseudobinary system UAl_3–USi_3 we had to adopt a model for the sites occupation. In the $L1_2$ base structure (space group 221), we consider fixed the U atoms in the Wyckoff positions 1a, and we consider the atoms Al and Si interchangeable species en the Wyckoff positions 3c. We assumed sustitutional occupation of Al and Si in 3c positions. As a consequence, this choice introduces a low mismatch between atom sizes of interchangeable species leading to a relative difference of 5% in Wigner Seitz ratio between the extreme compounds of the expansion UAl_3 and USi_3. Based on this fact, and though it has been proven that local displacements should be allowed when relaxing ordered structures to be entered in the expansion [36,27], we decided not to perform internal relaxation of atomic positions since it is not a demanding ingredient for low atom size mismatches [37,38].

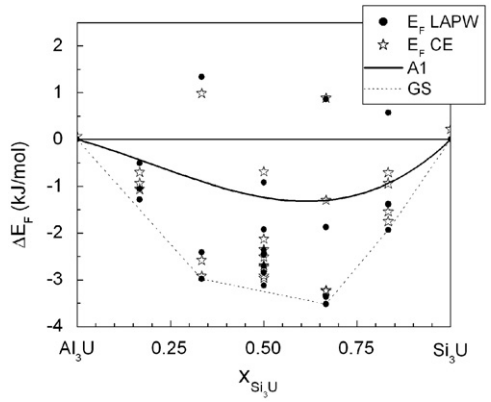

Fig. 1. fcc ground state phase diagram of the Al_3U–Si_3U system. Circles and stars stand for first principles formation energy and cluster expansion ones respectively. Solid and dotted lines represent the formation energy of the disordered solid solution (A1) and the ground state (GS) respectively.

Total energy calculations were performed for 24 fcc ordered structures. UAl_3 and USi_3 in the $L1_2$ structure were taken as references for the calculation of formation energies. The UAl_3–USi_3 pseudobinary system ground state diagram is shown in Fig. 1.

The minimum clusters set that best fit the three convergence criteria contains pairs till sixth order and triplets including pairs of first and second order:

i) The formation energies calculated through the cluster expansion for structures containing up to 16 atoms and not included in the calculation of ECIs are shown in Fig. 2a. The ground state obtained through the expansion agrees with the first principles calculated one and no energies are predicted below the ground state.
ii) The acceptable low value for CV (0.854 kJ/mol) reveals that a good agreement was found between first principles calculated energies and cluster expansion obtained energies for the 24 ordered structures included in the calculation of ECIs.
iii) ECIs intensity diminishes as a function of size and number of atoms (Fig. 2b).

3.2. 0.25 at% USi_3 composition

In order to account for the finding of a stable intermetallic compound $U_4Al_9Si_3$ [19], we calculated its formation energy through the cluster expansion and we obtained a positive value. Stabilization can then take place by the presence of defects, in agreement with the wide solubility range experimentally reported; or in the other hand, the equilibrium atoms positions could experience a slight displacement from the symmetry positions, as the authors report [19]. We have neglected in this work internal relaxation of atom positions based on the low mismatch between Al and Si atoms, but the result for the $U_4Al_9Si_3$ compound may indicate that some electronic effect other than atom size may render local relaxations meaningful. Possibly, local charge distribution could be strongly influenced by the Al and Si open p shells [39]. Calculations including local relaxation of atomic positions during the ab initio calculations of the ordered structures entering the cluster expansion will be the subject of a forthcoming report. We also analyzed the possible existence of other structures with a size greater than 16 atoms that could stabilize in the ground state at 0.25 at% USi_3 composition. Cluster expansion predicted negative values for several structures in that composition with cell sizes up to 48 atoms, but none of them stabilized below both the calculated and the predicted ground states.

3.3. UAl_3–USi_3 pseudobinary system phase equilibrium diagram

Simulations by Monte Carlo method were performed within a $31 \times 31 \times 31$ cell (in units of the unit cell), at each fixed temperature, and with a step in chemical potential of 0.0001 Ry/at (approx. 0.13 kJ/mol). Calculations for the disordered

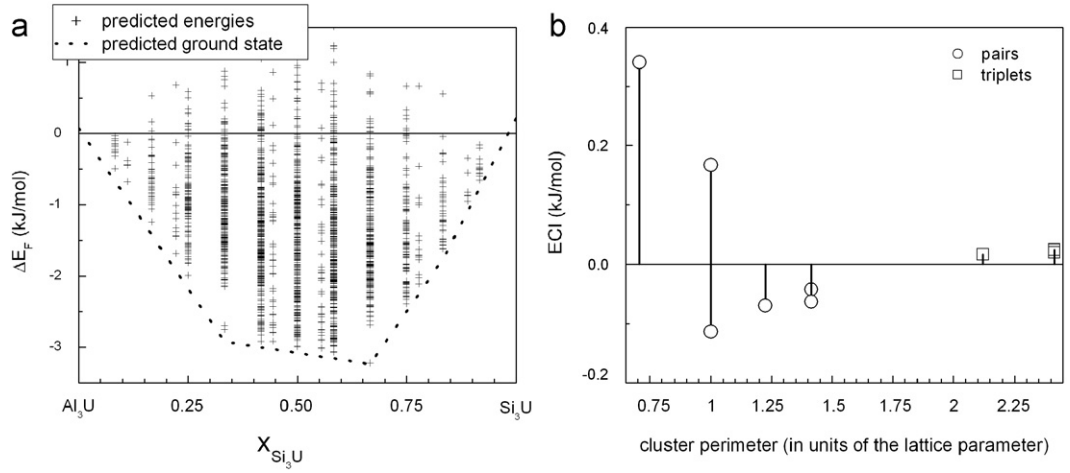

Fig. 2. (a) Predicted energies for structures not included in the calculation of ECIs; (b) Effective Cluster Interactions (ECIs) for fcc pseudobinary alloys Al_3U–Si_3U. The triplet perimeter is taken as parameter of the cluster size for ECIs corresponding to triplets.

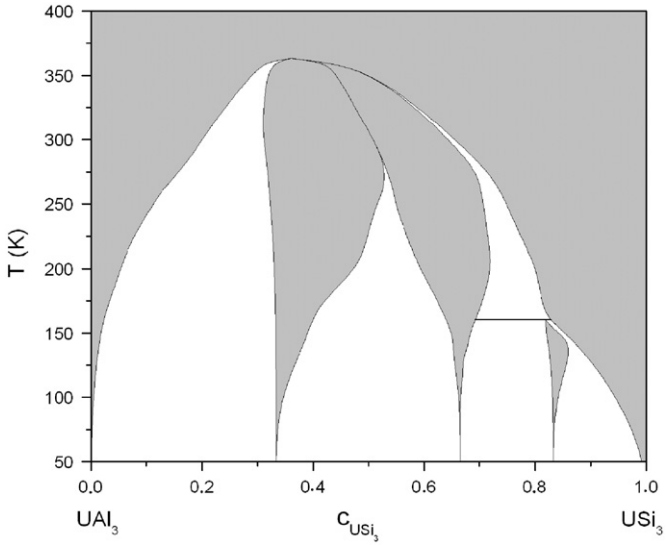

Fig. 3. fcc calculated equilibrium phase diagram for the pseudobinary system UAl_3–USi_3.

structure began at 30000 K to ensure the randomness of the configuration, and at $T=0$ K for the ordered phases. Convergence was attained by imposing a precision in composition of 10^{-3} at% for all phases. Equilibria were found from the intersection between grand potential vs. chemical potential graphs for all phases. We show in Fig. 3 the resulting phase equilibrium diagram. No evidence was found to justify a miscibility gap in the solid solution at low temperatures as had been previously suggested [18]. On the other hand, we found two ordered phases stable at low temperatures with a wide solubility range that undergo a transformation to a sole partially ordered phase above 260 K. These results agree with the experimental finding of the $U_4Al_9Si_3$ phase [19]. Above 360 K we only found the solid solution with the evidence of a short length ordering for compositions between 25 and 82 at%, with a maximum at approximately 60 at%. The solid solution is completely disordered above 1600 K. It is worth to note that this fact also agrees with the experimental report in Ref. [19] where it is stated that the partially ordered $U_4Al_9Si_3$ phase forms from the disordered solid solution at 1340 °C.

4. Conclusions

We successfully investigated a relevant feature concerning the development of a nuclear fuel based on the dispersion of a bcc γU based alloy in an Al based matrix applying theory and methods of first principles. Facing the problem of the stability of the interaction layer between a U based alloy and an Al based matrix, we demonstrated that $U(Al,Si)_3$ phase is stable in the UAl_3–USi_3 pseudobinary system. The calculations also gave a basis for understanding the existence of the $U_4Al_9Si_3$ phase with a wide solubility range experimentally reported.

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Appendix A. Supplementary information

Supplementary data associated with this article can be found in the online version at http://dx.doi.org/10.1016/j.calphad.2012.04.004

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