An Ab-initio Calculation of the Actinides and the Lanthanides for a Simulated Uranium Metal Fuel

Chang Je Park, Kweon Ho Kang, Sang Ho Na, Young Hee Kim and Kee Chan Song
Korea Atomic Energy Research Institute, P.O. Box 105, Yuseong, Daejeon, Korea, 305-600
cjpark@kaeri.re.kr

1. Introduction

Nuclear metal fuel is widely used in a fast reactor due to its high thermal conductivity. Especially in the sodium-cooled fast reactor, uranium and zirconium based metal fuel is considered including transuranic elements (TRU) and rare earth (RE) elements. Thus, it is required to obtain basic material properties to evaluate a fuel performance in a reactor as well as a transportation and deposition. However, because the active TRU fuel contains various sensitive elements such as Pu which is deeply connected with a proliferation problem, it is very difficult to measure the properties of such U-based metal fuels. It is one of the solutions to use surrogates and to make a simulated fuel with them. For example, CeO_2 is used to replace PuO_2 due to its similar characteristics including its thermo-dynamical properties. An ab-initio calculation is used in this study to provide basic data for TRU, especially the distribution of electron which is fundamental data to determine an element's properties. Additionally, various lanthanides are also used to compare the elementary properties with TRUs.

2. Method and results

The ab-initio calculation is used with VASP code which approximates the Born-Oppenheimer theory.[1] Thus, the motions of electrons are separated from the ion's motions. It composes a multiple electrons problem from a multiple ions and electrons problem. The VASP also uses a density functional theory (DFT), which expresses the total energy as a function of the density of the electrons. Furthermore, a local density approximation is used to express the coherent relations between electrons. The pseudo-potential approximation is used to elucidate the forces between ion and electron. This results in a very fast convergence in the calculation. A projector-augmented wave potential (PAW) is used in the VASP calculation in this study. The k-points are given as 11X11X11. Due to the lack of potential data from VASP, as TRU elements, U, Pu, Np are chosen. Most Lanthanides such as La, Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, and Er are used. Table 1 shows some results of the calculations. Total free energy, Fermi energy, magnetization, and charge are obtained from the ab-initio calculation. Fig. 1 shows the density of the state for various elements such as U, Pu, Np, La, and Ce. The non-symmetry of the TRU elements comes from a polarization property,[2] which shows high magnetization values in the table. Especially, the TRU elements have 5f valence electrons which exhibit a quite different behavior compared with other families. From Table 1, the distributions of electrons show differences between the TRUs and Lanthanides. The crystal structures of the absolute temperature and ion radius are also compared in the table. However, because the crystal structure changes as a function of the temperature and impurities, more detail conditions should be followed to obtain reliable data. The crystal structures of the Lanthanides are almost HCP which is a typical metal structure. The ion radii of all the metal elements are distributed from 1.75 to 1.95 angstrom. The total free energies of the TRU elements are slightly lower than the lanthanides, which means they are more stable in the steady states. The Fermi energy shows an irregular distribution due to the electron behaviors. The uranium exhibits the highest values for the Fermi energy about 8.8 eV. In the Lanthanides, the cerium provides a large Fermi energy such as 6.0 eV.

From the figure, the uranium and lanthanides show a similar behavior for the density of a state. However, the Pu and Np show a quite different behavior due to magnetization characteristics. In the future, more TRU elements should be calculated with reliable pseudo potential data.

3. Conclusion

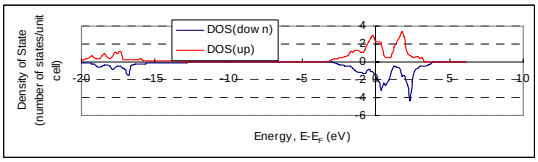
To grasp an electronic behavior of the TRU elements, an ab-initio calculation was performed and compared with the Lanthanides. There are quite different behaviors in the DOS, and total energies due to different valence electrons. However, to find a surrogate for the metal fuel, for example, Pu, Np, Am, and Cm, more data should be analyzed not only a calculation but also literature and experimental data. It should also be noted that the ab-initio calculations do provide basic data in terms of the electronic behavior. However, this approach will be a useful tool to apply to evaluating an active fuel element and to obtain a fundamental data.

Acknowledgements

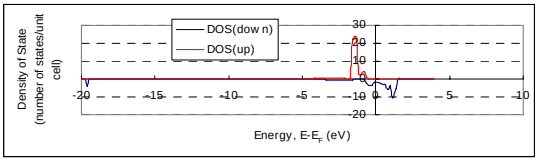
This work has been carried out under the Nuclear Research and Development program of the Korea Ministry of Science and Technology.

REFERENCES

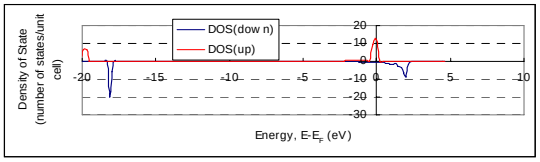
[1] G. Kresse and J. Furthmuller, VASP the Guide, Vienna University, 2007.
[2] L.R. Morss, N.M. Edelstein, J. Fuger, J.J. Katz, The Chemistry of the Actinide and Transactinide elements, Third Edition, Vol.2, 2006.



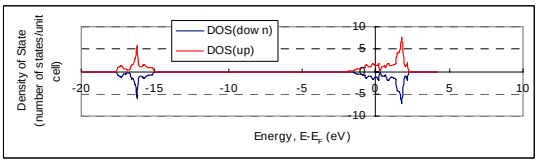
(a) U



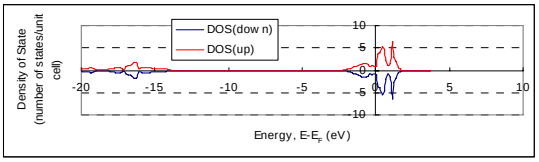
(b) Pu



(c) Np



(d) La



(e) Ce

Fig. 1 Density of states for various elements.

Table 1. Results of the Ab-initio Calculations for Various Elements

Element	Electron distribution	Crystal structure	Ion radius(A)	Free energy(eV)	Fermi energy(eV)	Magnetization (μ_B)	Charge (C)
U	5f36d17s2	Ortho	1.75	-10.859	8.776	1.373	13.082
Pu	5f66d2	Mono	1.75	-10.430	0.727	5.924	13.957
Np	5f46d17s2	Ortho	1.75	-8.742	1.753	4.691	12.681
La	5d16s2	HCP	1.95	-4.228	0.586	-0.007	8.768
Ce	4f26s2	HCP	1.85	-6.057	5.993	-0.001	9.921
Pr	4f36s2	HCP	1.85	-3.657	1.027	0.187	8.864
Nd	4f46s2	HCP	1.85	-3.575	0.892	0.211	8.954
Sm	4f66s2	Trig	1.85	-3.446	0.821	0.199	9.144
Eu	4f76s2	BCC	1.85	-1.113	-0.079	0.001	7.013
Gd	4f75d16s2	HCP	1.80	-3.346	0.905	0.030	7.772
Tb	4f96s2	HCP	1.75	-3.294	0.835	0.019	7.836
Dy	4f106s2	HCP	1.75	-3.242	0.749	0.006	7.918
Ho	4f116s2	HCP	1.75	-3.191	0.722	0.011	8.023
Er	4f126s2	HCP	1.75	-3.159	0.607	0.037	8.033