**8.3 FIRST-PRINCIPLES CALCULATIONS OF INTRINSIC DEFECTS AND Mg TRANSMUTANTS IN 3C-SiC** — S. Y. Hu, W. Setyawan, R. M. Van Ginhoven, W. Jiang, C. H. Henager, Jr., and R. J. Kurtz (Pacific Northwest National Laboratory)

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Silicon carbide (SiC) possesses many desirable attributes for applications in high-temperature and neutron radiation environments. These attributes include excellent dimensional and thermodynamic stability, low activation, high strength, and high thermal conductivity. Therefore, SiC based materials draw broad attention as structural materials for the first wall (FW) and blanket in fusion power plants. Under the severe high-energy neutron environment of D-T fusion systems, SiC suffers significant transmutation resulting in both gaseous and metallic transmutants. Recent calculations by Sawan, *et al*. [2] predict that at a fast neutron dose of ~100 dpa, there will be about 0.5 at% Mg generated in SiC through nuclear transmutation. Other transmutation products, including 0.15 at% Al, 0.2 at% Be and 2.2 at% He, also emerge. Formation and migration energies of point defects in 3C-SiC have been widely investigated using density functional theory (DFT). However, the properties of defects associated with transmutants are currently not well understood. Fundamental understanding of where the transmutation products go and how they affect microstructure evolution of SiC composites will help to predict property evolution and performance of SiC-based materials in fusion reactors.

In this work, the Vienna Ab-initio Simulation Package (VASP) is used to calculate the formation energies of different defects in 3C-SiC-Mg. The convergence and accuracy in terms of pseudopotentials, energy cutoff, supercell size, and k-point mesh used in DFT calculations are first examined by comparing the thermodynamic properties from our calculations with experimental data and existing DFT results, including cohesive energy, lattice constants and elastic constants of perfect crystals {Si(diamond), Mg(fcc), 3C-SiC, $Mg_2Si$, C(diamond), and Mg(hcp)}, and formation energies of defects and defect clusters in 3C-SiC. Different defect configurations including substitutional, interstitial, and dumbbell are considered in the calculations. The substitutional and interstitial are described as $A_B$. A denotes the species and the subscript B denotes the lattice site. There are two interstitial sites in 3C-SiC. One is the tetrahedral center of Si, and the other is the tetrahedral center of C. The two interstitial sites are described as TSi and TC, respectively. A dumbbell is described as $A_1^+$-$A_2$<**a**>. $A_1$ and $A_2$ denote the two species in the dumbbell. Subscript "+" of $A_1$ denotes that the dumbbell locates at $A_1$ lattice. <**a**> denotes the direction of the dumbbell. The formation energies of defects and defect complexes in SiC-Mg are

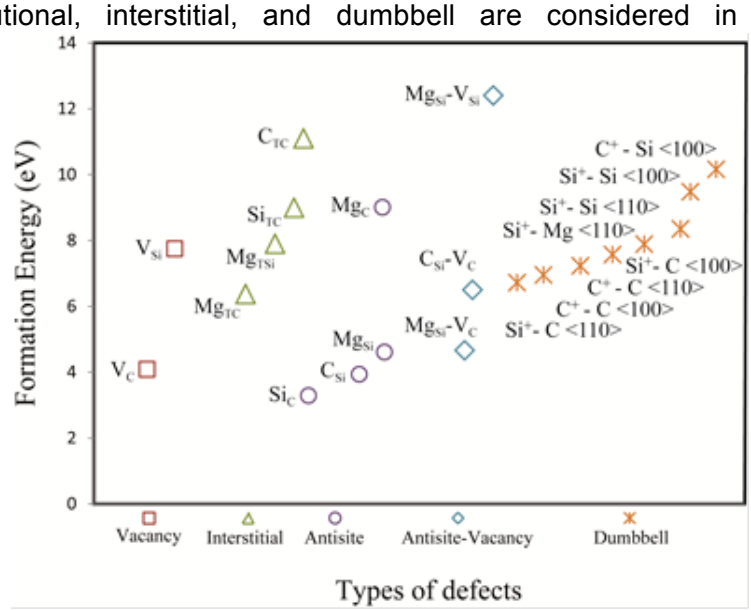


Figure 1. Summary of formation energies of different point defects and defect complexes in 3C-SiC.

presented graphically in Figure 1. The results show that substitutional $Mg_{Si}$ is the most stable defect of Mg, with formation energy 4.61 eV. The complex $Mg_{Si}$-$V_C$ has a little higher formation energy (4.66 eV). The defect complexes: $Mg_C$-$V_{Si}$, $Mg_{TSi}$-$V_C$, $Mg_{TSi}$-$V_{Si}$ are unstable. They will transform to $Mg_{Si}$-$V_C$, $Mg_C$, and $Mg_{Si}$ without overcoming energy barriers, respectively. The stability of $C^+$-Mg<100>, $C^+$-Mg<110>, $Si^+$-Mg<100> and $Si^+$-Mg<110> dumbbells were carefully checked by relaxing the dumbbell configuration with different initial distances from 0.84 Å to 1.4 Å. It is interesting to find that only $Si^+$-Mg<110> dumbbell is stable. The remaining three dumbbells are unstable, and dissociate to form Mg interstitials. The interstitial $Mg_{TC}$ has lower formation energy than that of the interstitial $Mg_{TSi}$.

The minimum energy paths (MEP) of defect migration and small cluster evolution are determined with a generalized solid-state elastic band (G-SSNEB) method. In the simulations, the first step is to relax the defect configurations of the initial and final states, after that three images are inserted between the initial and final states, and then a search for the MEP using the G-SSNEB method is performed with both atomic motion and the relaxation of the supercell. The MEPs of C, Si, $Mg_{Si}$ migrations to their nearest neighbor vacancies show that the activation energy of Si to $V_{si}$ is 3.51 eV which is in good agreement with the DFT result 3.4 eV [3]. The activation energy of C to $V_{si}$ is about 3.33 eV compared to 4.58 eV for $C_{Si}$ to $V_C$. It is found that the activation energy of $Mg_{Si}$ to $V_{Si}$ (0.97eV) is much smaller than that of Si to $V_{si}$ (3.51 eV). The migrations of $Mg_{Si}$ to $V_C$, $Mg_{Si}$ to $V_{Tsi}$, and $Mg_C$ to $V_{TC}$ are energetically not favored because the process causes an energy increase. Figure 1 shows that the $Si^+$-Mg<110> dumbbell is a metastable. It may transform to more stable defect $Mg_{TC}$ via two different processes. One is $Si^+$-Mg<110> to $Mg_{TC}$. The other has two steps, i.e., $Si^+$-Mg<110> to $Mg_{TSi}$ and then $Mg_{TSi}$ to $Mg_{TC}$. The MEPs for these two processes are calculated. The results show that the activation energy of $Si^+$-Mg<110> to $Mg_{TC}$ is 0.23 eV. The activation energies of $Si^+$-Mg<110> to $Mg_{TSi}$ are 0.3 eV while it is 0.91 eV for $Mg_{TSi}$ to $Mg_{TC}$ migration. The first process $Si^+$-Mg<110> to $Mg_{TC}$ has lower activation energy than that of the second process. Interstitial $Mg_{TC}$ has a lower formation energy than that of interstitial $Mg_{TSi}$. The activation energy from $Mg_{TC}$ to $Mg_{TSi}$ is 2.44 eV. It is small compared with the activation energies of Si and C self-diffusions (3.5 eV). Therefore, the transition from $Mg_{Si}$ to $V_{Si}$, $Si^+$-Mg<110> to $Mg_{TC}$, and $Mg_{TC}$ to $Mg_{TSi}$ could be Mg diffusion mechanisms during the formation of Mg-related clusters and $Mg_2Si$ nucleus.

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