Ab-Initio Simulation of Fission Product Diffusion in Graphene & Graphite

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Graphite has been used for neutron moderation from the beginning of the nuclear reactor era. While research activities have abated over the years, there is renewed interest in graphite motivated by its use in Very High Temperature Reactors (VHTRs) and Molten Salt Reactors (MSRs). Retention of the activated fission products is paramount during normal operating and accident conditions, and a mechanistic understanding of the bonding and diffusion properties of fission products is imperative for predicting the release rates and designing appropriate barriers.

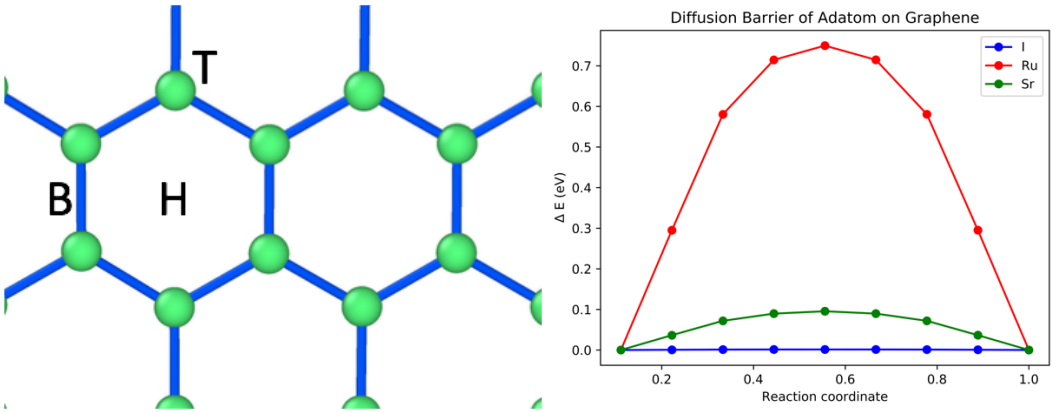

Figure 1: Preferential adsorption sites of adatom on graphene (left) and calculated diffusion barrier profiles using the vdW-DF-cx functional (right).

While the simulation, via Density Functional Theory, of the adsorptive properties of a number of elements on graphene has received some attention, to the best of our knowledge little work has been conducted on their bonding and diffusive properties. We have performed high-accuracy DFT simulations of nuclear fission products on graphene using LDA, GGA and van der Waals exchange-correlation functionals using the plane-wave Quantum Espresso software. Structural relaxation at symmetrical sites is performed and preferential sites are identified for a range of nuclear fission products (Ag, Ba, Ce, Cs, Eu, I, Kr, Ru, Sr). The bonding, structural and diffusive properties have been extracted using nudged elastic band (Figure 1) projected Density of States (Figure 2) and Bader charge analysis calculations, and the effect of the different EXC functionals is compared.

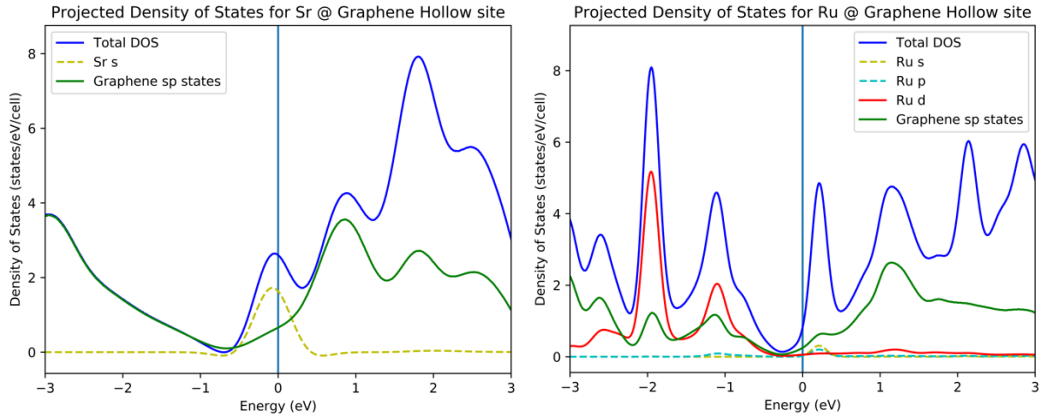

Figure 2: Sr (left) and Ru (right) spin-up projected Density of States at the energetically favourable Hollow adsorption site, calculated using the vdW-DF-cx functional. Shift in the Fermi level relative to Dirac point and partial occupation of the s-state are indicative of ionic bonding for the Sr adatom. Ru has a significant impact on the electronic structure, with evidence of strong hybridization due to the prominent peaks in the PDOS.

In addition, DFT calculations have been performed at LDA level, using HGH pseudopotentials, for these same fission products in bulk nuclear graphite using the AIMPRO code which uses Gaussian, filtrated basis sets to allow efficient simulation of large supercells. Formation energies have been calculated and preferential sites identified, and the structural, bonding and diffusive properties are extracted and compared to the results obtained on graphene sheets. These simulations form a basis for the understanding of the diffusion and retention of fission products in nuclear graphite.

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