A study of fission product migration paths in irradiated TRISO particle SiC

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Tristructural isotropic (TRISO) coated fuel particles used in high-temperature gas-cooled nuclear reactors (HTGRs) provide key advantages in terms of passive safety and reactor energy efficiencies. TRISO fuel consists of a number of special coating layers surrounding the fissile material in the kernel as shown in Fig. 1. The coating layers consist of a carbon buffer layer, inner pyrolytic carbon (PyC) layer, a silicon carbide (SiC) layer as well as an outer pyrolytic carbon layer. Together the layers act as a pressure vessel and barrier to the release of radioactive fission products into the reactor environment.

The cubic 3C-SiC layer of thickness approximately 35 m, acts as the primary barrier to the migration of metallic fission products out of the particle. The finding, more than three decades ago, that silver can be released by intact TRISO particles has led to significant research to determine possible transport mechanisms of certain fission products in SiC. Out-of-reactor experiments did not reveal any significant lattice diffusion of silver in SiC[1]. Many earlier findings alternatively proposed a dominating role of fast diffusion paths such as grain boundaries and dislocations in enhancing fission product migration in SiC. Recently the role of the fission product palladium in enhancing the transport rate of silver in SiC[2] was confirmed by transmission electron microscopy (TEM) of neutron-irradiated SiC from a TRISO coated particle from the US Department of Energy's Advanced Gas Reactor (AGR)-1 experiment[3] (Compact 4-1-1; average burnup of 19.38% fissions per initial metal atom; a time-averaged, volume-averaged temperature of 1072 °C; a time-averaged peak temperature of 1182 °C; and an average fast fluence of 4.13×10^{21} n/cm^2). Although other studies found that Ag was found separately (without Pd) in TRISO coated particles irradiated at 11.4 %, 16.7 and 18.6% FIMA, Pd could have been present at lower than measureable concentrations[4].

In this study, SiC focussed ion beam (FIB) sections obtained from Compact 4-1-1 of the AGR-1 experiment were analysed in a double Cs-corrected JEOL ARM 200F using atomically resolved scanning transmission electron microscopy (STEM). By using atom resolved STEM in combination with electron energy loss spectroscopy (EELS) and large angle energy dispersive spectroscopy (EDS) a much better understanding of the migration routes and mechanisms of fission products in irradiated polycrystalline 3C-SiC was achieved.

In general, the findings indicate that the transport mechanism of metallic fission products such as Pd and Ag in the SiC layer is based on a solid state diffusion process with a clear preference for fast diffusion paths such as grain boundaries (Fig. 2 (a)), sub domain boundaries and dislocation cores (Fig. 2 (b)). The diffusion mechanisms were found to have a strong association with Si exposed at the periphery of enhanced diffusion paths as determined by EELS fine structure analysis. The first evidence of intra-granular fission product transport along dislocation cores in SiC will be presented (Fig 2 (c)). The dislocation cores are present at the annihilation or termination points of stacking fault planes within the SiC, which allows for single-atom fission product migration as shown in Fig. 2 (d).

References
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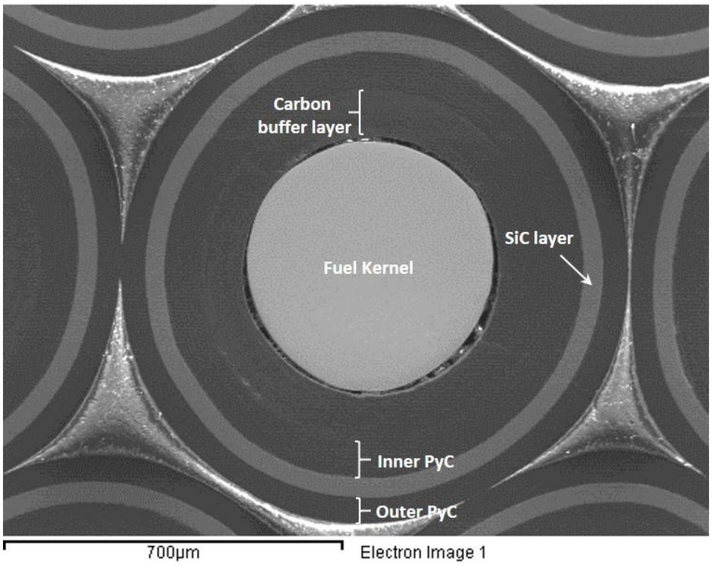

Fig 1. Secondary electron (SE) scanning electron microscope (SEM) image of a polished TRISO particle showing the various coating layers.

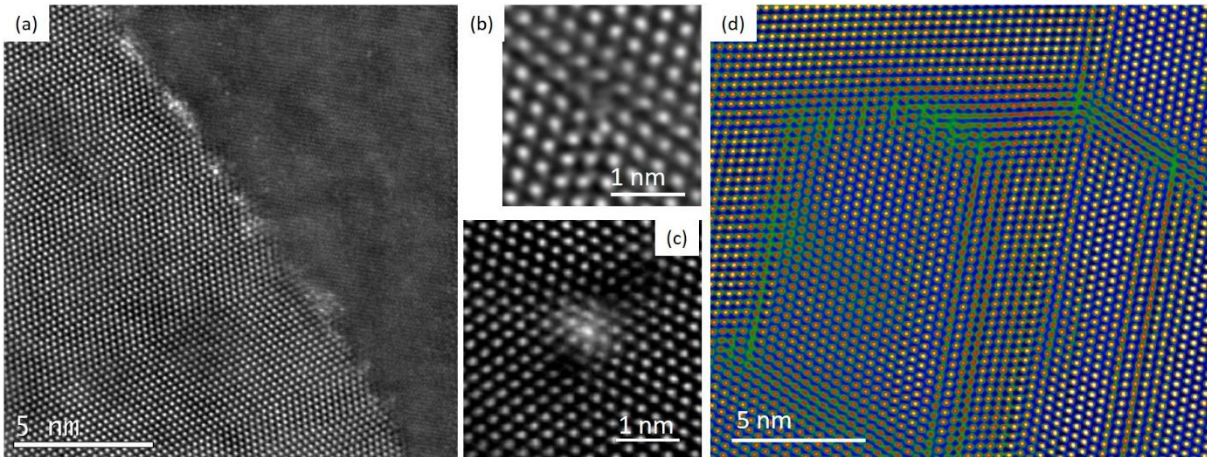

Fig. 2. (a) Annular dark field (ADF) STEM image of a SiC grain boundary showing the presence of isolated metallic fission product atoms along the boundary. (b) Atomic resolution image of an unoccupied Lomer-Cottrell dislocation in a SiC grain. (c) Atomic resolution image of a fission product atom present within a dislocation core in the SiC. (d) Atomic resolution false colour image showing the annihilation and termination points of stacking faults planes within a SiC grain.