Point Defects and Diffusion of Cesium in SiC Layer of TRISO coated fuel particle

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SiC Crystal Structure

- Tetrahedral sp^3 hyberdization
- Strong asymmetric valence charge
- Hexagonal crystalline structure

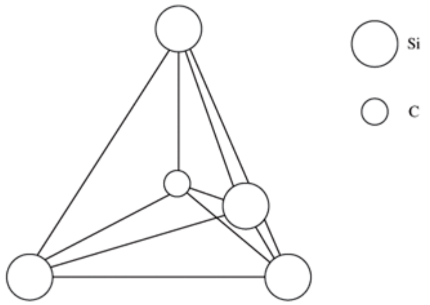
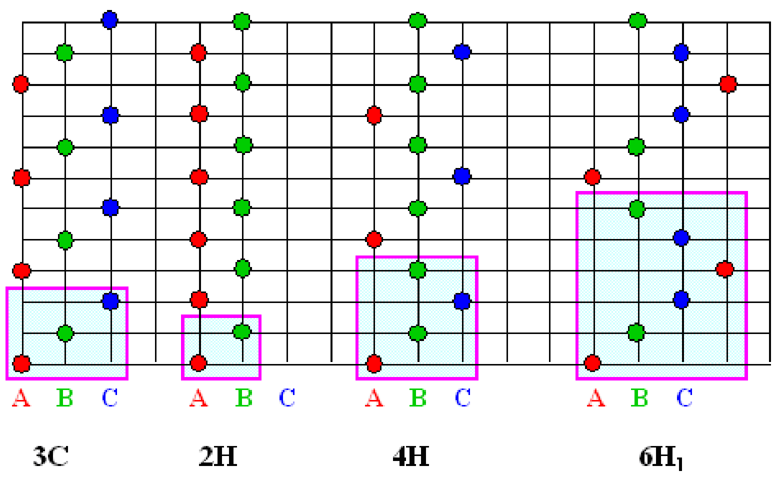


Figure 1. Representation of basic silicon carbide thetraedra.



SiC stacking blocks for different polytypes

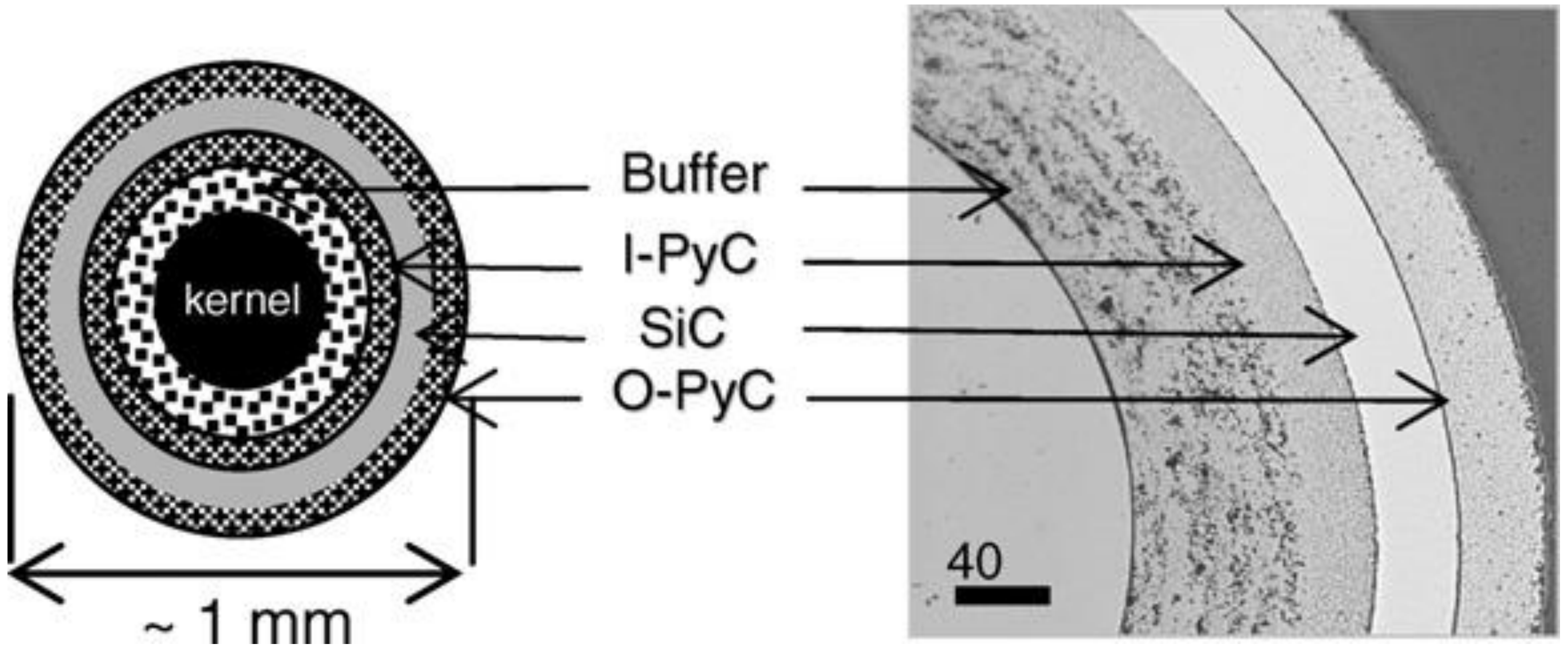
Industrial Usage of SiC

- Abrasive
- Electronics as semi-conductors
- Bearings and furnace parts
- Gemstone: synthetic moissanite
- Coating for TRISO fuel particles as the main diffusion barrier

SiC Material Property

- Wide band gap (4H: 3.26 eV)
- High sublimation temperature (2700 ^{o}C)
- Chemically inert (almost)
- High thermal conductivity (4H: 3.7 W/cm·K)
- Very low coefficient of thermal expansion (4.0 × 10^{-6} /K)

TRIstructure ISOtropic Fuel



- Fission Products are retained inside the TRISO fuel.
- 35 µm SiC layer is considered to be the main barrier to release of fission products.

TRISO Fuel

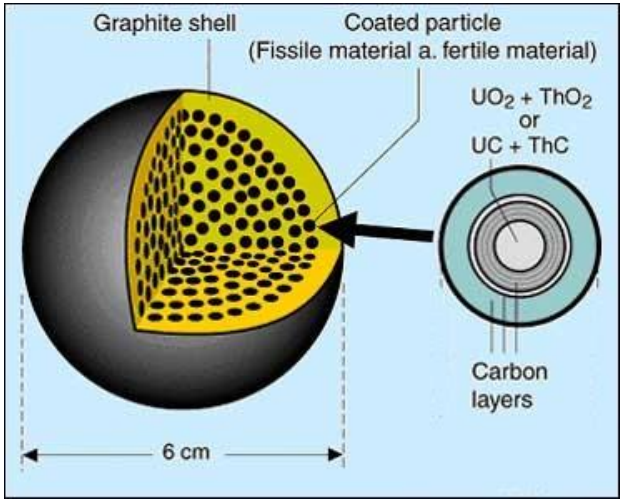
- TRIstructure ISOtropic 1mm micro spheres.
- Fuel kernel
  - stable oxides or carbides of uranium, plutonium or thorium such as UO_2
- Buffer layer
  - Void volume to contain fission gases
- High density pyrolytic carbon layer
  - Pressure barrier against fission products
- Silicon Carbide layer
  - Limited tensile strength
- Hight density pyrolytic carbon layer
  - Protects the SiC layer against damage

Very High Temperature Reactors (VHTR)

- Part of Generation IV reactors
- Outlet temperature of about 1000°C to enable applications such as process heat or hydrogen production.
- Graphite-moderated core with a once-through uranium fuel cycle.
- Prismatic block or a pebble-bed core composed of TRISO fuel particles.
- 400-600 MW_{th} reactor contains 10^9-10^{10} TRISO particles.

Reactors with TRISO Fuel

- AVR
  - Pebble bed prototype reactor
  - 15 MWe (40 MW_{th}) prototype reactor at Julich Research Center Operated from 1966 to 1988
  - Originally a breeder reactor (Thorium 232 to Uranium 233)
- THTR-300
  - Thorium High Temperature Reactor 300 MWe (750 MWth) Operated from 1985 to 1988
- HTR-10
  - Prototype pebble bed reactor in China
  - 10 MW_{th} reactor
- HTTR
  - Prototype prismatic core reactor in Japan
  - 40 MW_{th} reactor
- Gas Turbine-Modular Helium Reactor (GT-MHR)
  - General Atomics Prismatic (Block) Reactor
  - TRISO fuel compacts in Graphite Block



TRISO Particle failure Mechanisms

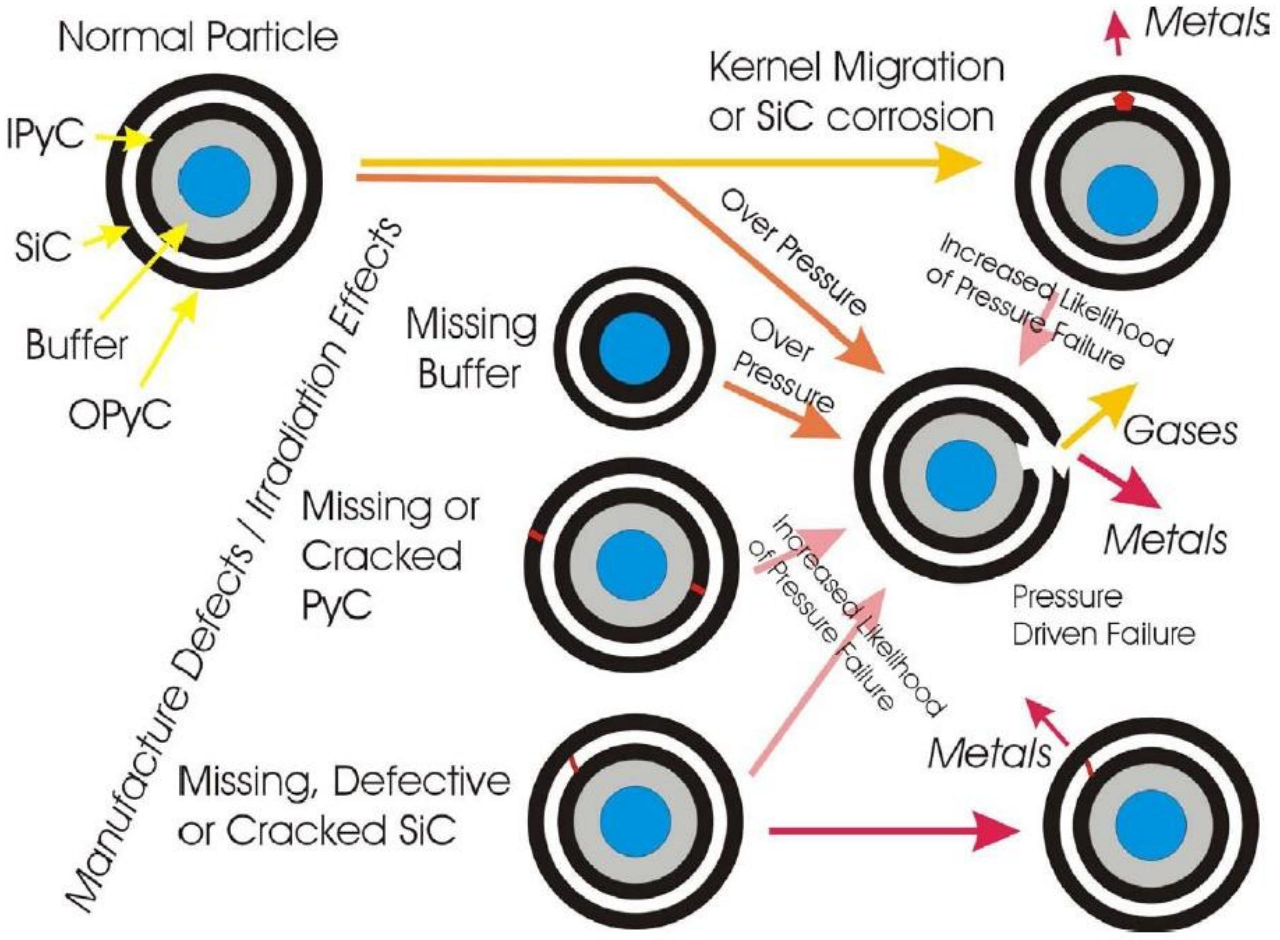


Figure 3-5. Particle Failure Mechanisms

Diffusion Concerns in SiC layer of TRISO Fuel

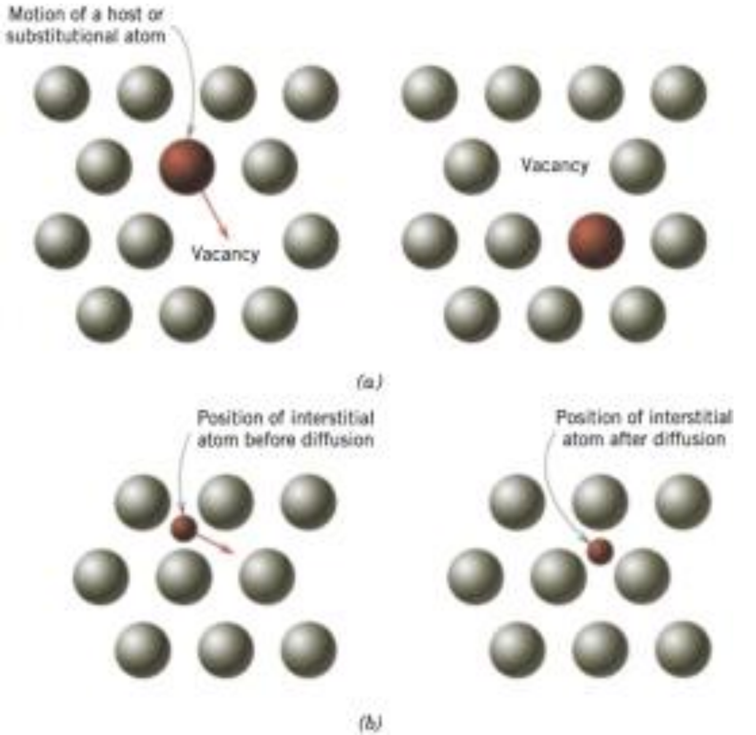
- Diffusion of fission products through SiC layer is one of the major concerns
- Research has been done on diffusion of Iodine, Silver, palladium and Cesium
- Data on cesium diffusion and mechanisms are limited.

cesium

- Relatively high fission yield (~13%)
  - about 6 atoms of Cs-137 and 7 atoms of Cs-135 are produced per 100 fission events.
- Cs-137
  - Major radionuclide in spent nuclear fuel, high level radioactive wastes
  - Half-life of 30.2 years
  - Decays by
    - beta decay with 416.3 keV average energy to stable barium-137.

Diffusion methods

- Vacancy Diffusion
  - Elevated temperatures
- Interstitial Diffusion
  - The diffusing atoms must be small enough to fit in the interstitial positions.



Factors that influence diffusion

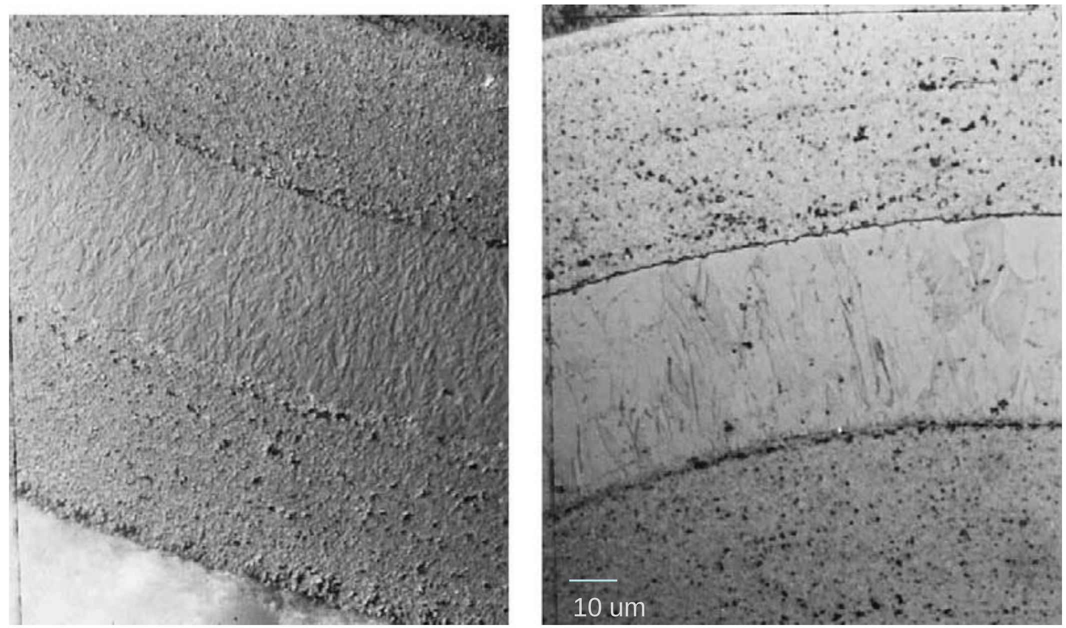
- Diffusing species
- Host material
  - Grain size
- Material defects
- Temperature

SiC Layer Microstructure

Following irradiation kernels heated to 1500 ^{o} C

- Columnar- large grained SiC
  Release of Ag 100%
  Release of Cs 24%

- Laminar- weaker SiC
  Release of Ag 82%
  Release of Cs 12%



•Smaller grained with higher tortuosity retains fission products better than large columnar SiC

Key differences in the fabrication, irradiation and high temperature of TRISO Fuel Particles. D.A. Petti, et al. (2003)

Migration of Fission Products

- Kernel type: MOX, UO,…
- A function of temperature
  - Thermal decomposition of SiC layer at temperatures > 2000 ^{o}C
- Burnup
  - Greater release of Cs when particles are exposed to higher burn up (14%) & higher fluence ($4.6 \times 10^{25} n/m^2$)
- Temperature gradient in the fuel
  - Function of power density and packing fraction

Research Objectives

- Develop an experimental model of diffusion of Cesium in the SiC layer of TRISO fuel.
- Determine the diffusion mechanisms of Cs through SiC crystal structure and grain boundaries.
- Measure Cs concentration profiles in SiC microstructures.

Experimental Approach

- Characterize SiC defects as a result of exposure to high temperature (up to 1800 ^{o}C) & mixed field radiation environment

- Implant Cs on SiC substrate, expose samples to neutron irradiation and high temperature
  - Study the resulting defects of SiC crystal structure
  - Verify the diffusion of Cs-137 through SiC layer
  - Determine the Cs-137 diffusion rate constant.

SiC & Diffusion Characterization

- X-Ray Diffraction
  - characterizing crystallographic structure
- SEM
  - surface characterization
- TEM
  - grain boundaries and cross sectional information
- EPR
  - Determining the number of the unpaired electrons in the speciment indicates the lattice vacancy by radiation.

Questions?