CHARACTERIZATION OF SILICON CARBIDE AND PYROCARBON COATINGS FOR FUEL PARTICLES FOR HIGH TEMPERATURE REACTORS (HTR)

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AGENDA

• Background
• SiC microstructure
• PyC microstructure
• PyC/SiC interfaces
• Concluding remarks
BACKGROUND

• Reviving interest worldwide for HTR technology (co-generation, safety, rentability)
• Fuel design = key element: 1st confinement barrier for fission products

SiC/PyC multilayer coating

Buffer (100 µm, d~1)
I-PyC (35 µm, d~1,9)
SiC (35 µm, d~3,2)
O-PyC (35 µm, d~1,9)

• Production and qualification of fuel particles are conducted by the CEA in collaboration with AREVA
**BACKGROUND**

**PyC and SiC material challenges:**

- **PyC**: gas-tight and perfect isotropic texture to improve performance under irradiation

- **SiC**: β-SiC (cubic structure) to compensate the swelling effect due to irradiation and tight layer: fine-grained and strong grain boundaries to reduce the migration of fission products, less flaws as possible to sustain strain

- **PyC/SiC interfaces**: strong for mechanical synergy

**NEED FOR:**

- Developing analytical procedures
- Characterising as-processed particles to improve process parameters
- Comparing with analytic know-how developed in the 1970’s
SiC MICROSTRUCTURE

SEM micrograph of SiC in cross-section after Murakami etching

**Columnar grains:**
- Thickness ~ 1 µm
- Length ~ 4-5 µm

**Finer grains** in the beginning of the coating

Exhibition of preferred orientation during growth (Electron Back-Scattering Diffraction)

- <111> pole parallel to the growth direction
- XO normal to the particle
SiC MICROSTRUCTURE

Crystallographic nature: $\beta$-SiC (3C)

No excess free carbon and silicon

**SiC stoichiometric**
Si/C=1.02 (Electron Probe Micro Analysis)

Measuring of chlorine:
- At the extreme surface concentration: 0.43 % at. (X-ray Photoelectron Spectrometry)
- In the core layer: **Cl<15 ppm** (EPMA)
SiC CHEMICAL ANALYSIS

SIMS DEPTH PROFILE : semi quantitative results

Profil sur bille

XPS analysis at:
C 83.24 %,
O 6.61 %,
Si 9.72 %,
Cl 0.43 %

SiC surface

e (nm)

C
O
37Cl
SiC
243D
SiC MICROSTRUCTURE

SEM fractography

Predominantly transgranular cleavage

Strong grain boundaries
no free C, Si and Cl detected

Bright field TEM image

No intergranular porosity
SiC MICROSTRUCTURE

Evidence of flaws by short etching on polished surface (reducing the SiC elastic limit)

Microcavities = Potential cracks initiators

Prevision on the cracks apparition and propagation in the layer
Evidence of crystallographic Stacking Faults (SF) by TEM Observations along the <110> β-SiC crystal direction

Bright field TEM image

HRTEM image

Very few α-SiC
Rather “one-dimensionally-disordered” polytypes
PyC texture: competition of 2 growth mechanisms

Reactions in the gas-phase: Isotropic PyC

Direct deposition onto the surface: Anisotropic PyC

J.L. Kaae, *Carbon*, 23, 6, 665-673, 1985
PyC MICROSTRUCTURE

SEM images

Buffer fractography

I-PyC fractography

I-PyC polished and etched section

Agglomerated spheroids formed in the gas phase (Diameter = 300-400 nm)

Heterogeneous distribution of opened porosity
Agglomerated spheroids
(mean diameter=500 nm)
surrounded by PyC deposited
directly on the substrate
PyC MICROSTRUCTURE

SEM images

Buffer fractography  I-PyC fractography  I-PyC polished and etched section

Isotropic PyC fraction
Anisotropic PyC fraction

ARWIF 2005
OAK RIDGE
PyC MICROSTRUCTURE

HRTEM images

Good agreement with CEA (Pelissier & Lombard)

PyC classification (1976)

Buffer

“Tangled fiber”

I-PyC

“Layered”

Flat substrate

“Mosaic”

“Layered”

“Mosaic”

“Layered”

“Mosaic”
PyC/SiC INTERFACES

Raman mapping on polished cross-sections

Smooth O-PyC/SiC interface
Strong interface

Weak interface
CONCLUDING REMARKS

- Re-appropriation of analytical know-how developed in the 1970’s

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CONCLUDING REMARKS

- New analytical procedures for qualifying the microstructure of fuel particles

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CONCLUDING REMARKS

• Re-appropriation of analytical know-how developed in the 1970’s

• New analytical procedures for qualifying the microstructure of fuel particles

• Next challenge:
  correlation growth mechanisms / microstructure