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Evolution of thermo-physical properties and annealing of fast neutron irradiated boron carbide

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ABSTRACT

Boron carbide is widely used as a neutron absorber in most nuclear reactors, in particular in fast neutron ones. The irradiation leads to a large helium production (up to $10^{22}/cm^3$) together with a strong decrease of the thermal conductivity. In this paper, we have performed thermal diffusivity measurements and X-ray diffraction analyses on boron carbide samples coming from control rods of the French Phenix LMFBR reactor. The burnups range from 10^{21} to $8.10^{21}/cm^3$. We first confirm the strong decrease of the thermal conductivity at the low burnup, together with high microstructural modifications: swelling, large micro-strains, high defects density, and disordered-like material conductivity. We observe the microstructural parameters are highly anisotropic, with high micro-strains and flattened coherent diffracting domains along the (00l) direction of the hexagonal structure. Performing heat treatments up to high temperature (2200 °C) allows us to observe the material thermal conductivity and microstructure restoration. It then appears the thermal conductivity healing is correlated to the micro-strain relaxation. We then assume the defects responsible for most of the damage are the helium bubbles and the associated stress fields.

1. Introduction

Boron carbide is widely used as a neutron absorber for the control, safety and protection of nuclear reactors. This is due to the conjunction of a high melting temperature and abundance and a high ^{10}B neutron absorption cross section in the whole neutron energy spectrum. The main limitations arise from brittleness and the production of large quantities of helium and heat during the (n,α) neutron absorption reactions. As a matter, the (n,α) reaction is exothermal (about 2.8 MeV per capture). In a control rod, the burnup (neutron capture density) can be as high as $10^{22}/cm^3$ per year, this leading to a volume power about 150 W/cm^3. This generates high thermal gradients in the absorber material pellets leading to an extensive pellet cracking (Fig. 1).

It is of primary importance to have a good knowledge of the thermal conductivity of the material and its evolution during irradiation. It is now well known that the thermal conductivity degrades under neutron irradiation [1–5]. We present here results we have obtained from materials irradiated in experimental devices in the French LMFBR Phenix reactor: the materials are somewhat different from those in most of other studies. Some of those results, already published in Refs. [6–9] are here re-examined this allowing providing additional results.

Moreover, the thermal conductivity loss certainly arises from helium accumulation but also from structural damage. In order to estimate this contribution, we have performed X-ray diffraction analyses of the materials. The crystal structure of boron carbide is well known [10,11]. It is built by a close packing of nearly regular icosahedra (composition close to $B_{11}C$) located on the apex of a rhombohedral network. The main diagonal of the rhombohedra is occupied by a linear chain (most frequent composition CBC), this leading to the $B_{11}C$-CBC cell composition, i.e. B_4C. The rhombohedral structure is most often described in a hexagonal frame. Most of observations have shown that during neutron irradiations, helium

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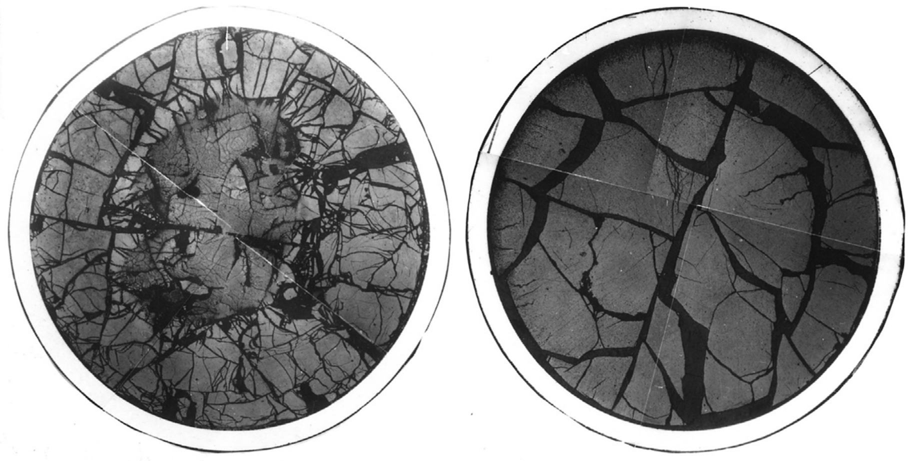

Fig. 1. Boron carbide pellet cracking in an experimental Phenix control rods. Left: burnup = $1.2.10^{22}/cm^3$. Right: burnup = $8.5.10^{21}/cm^3$. Scale: outer cladding diameter = 21 mm.

accumulate as flat bubbles perpendicular to the (00.l) direction, generating strong and anisotropic internal stresses [12].

At last, thermal treatments have been performed up to high temperature in order to determine the temperature ranges in which the thermal conductivity and the structure recover.

2. Experiment

The irradiations have been performed in the French Phenix Sodium cooled Fast Reactor in the frame of the development of the absorber rods of the Superphenix reactor. Two experiments are here considered:

- The first one (Hyperbare-1) aimed at testing advanced control rods designs. In this case, the boron carbide pellets are inserted in a metallic sheath aiming at reducing fragment relocation and the shroud carburization [13].This experiment has actually constituted 2 pins of a normal control rod. It has then been submitted to the normal displacements of such a component.
- The second one (Fracasse) aimed at analyzing the cracking of the material at the beginning of life as a function of the material manufacturing process and the pellets geometry and density. This experiment was a fixed capsule inserted in the center of the reactor.

Their main characteristics are reported on Table 1.

The boron carbide was synthetized with the magnesothermal process [10], the French reference process of the time. After acid washing, this leads to a micronic powder. As compared to the carbothermal synthesis process, the magnesothermal process leads directly to a material with a small, homogeneous grain size without cruching and sieving steps. Other differences stand in a higher specific surface of the powder, the impurity composition and content (Mg vs Fe, oxygen, free carbon), this leading to somewhat different hot pressing conditions (lower temperature for the magnesothermal process).The powder was then hot-pressed in a graphite die to obtain cylindrical pellets, the hot-pressing conditions (about 2000 °C, 50 MPa, 1 h) were adjusted to obtain a high density (circa 96%) and keep as a small grain size as possible. The pellets were then ground to remove the reaction surface and to obtain the required dimensions.

In both experiments, the boron carbide pellets were stacked in stainless steel shrouds and maintained by a spring to follow the deformations induced by swelling and fragments relocation. The lower and upper plugs of the pins are porous to allow sodium circulation inside the rod, between the B_4C pellets stacking and the clad. This allows first cooling the absorber and second evacuating the released helium arising from the (n,α) absorption reactions. In the case of control rods, this then allows rather large B_4C pellets diameter, since the surface temperature remains low, and short control rods, since no expansion chambers for released helium are required. The main drawback is an accelerated chemical interaction with the cladding, boronation and/or carburization, allowed by direct transfer through sodium.

After irradiation, the Hyperbare control rod and the Fracasse capsule were sodium drained, then submitted to non-destructive examinations (metrology, neutron and X-ray scanning). The pins were then cut to perform the destructive examinations: burnup, metallography, density, and so on. Before the analyses, selected sections are heated to 500 °C under a vacuum in order to evaporate residual sodium.

In the case of the Hyperbare-1 experiment, 2 control elements (here-under named 30A and 44C) have been analyzed. The burnup and boron carbide swelling distribution along the rod height are reported in Fig. 2. The exponential-like variation of the burnup results from the neutron distribution in the reactor (flux and energy spectrum) and the moving of the control rod in the core during its life. The similar distribution of the swelling confirms a quasi-linear variation as a function of burnup at least up to $10^{22}/cm^3$, about 0.15 vol % per 10^{20}capt./cm^3 [2,4,13].

In the case of the Fracasse experiment, short (40 cm) elements have been irradiated in fixed position, located at the mid-plane of

Table 1
Main characteristics of the Hyperbare-1 and Fracasse experiments (min-max burnups and volume power: from top of absorber column to reactor mid-plane).

Experiment	Hyperbare-1	Fracasse
Absorber	Magnesothermal B_4C	
Relative density	0.96	
^{10}B enrichment	0.48	
Pellets diameter (mm)	23.5	6.6
Grain size (μm)	2–5	2–5
Irradiation duration (efpd[a])	399	43
Min-max burnup ($10^{20}/cm^3$)	12–140	~25
Volumic power (n,α) reactions (W/cm^3)	15–165	~160
B_4C temperature, min – max (°C)	500–1100	500–600

[a] efpd: equivalent full power days (equivalent irradiation duration reported to a full power operation of the reactor).