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A new methodology for studying neutron absorber materials: First results with boron carbide

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ABSTRACT

This paper presents a new methodology used to study transmutation damage in boron carbide (B_4C). B_4C samples were set in a specially designed sample holder and irradiated in a neutron thermal flux in the ILL reactor. After describing the sample holder and the irradiation conditions, this paper discusses the results of the first post-irradiation examinations by secondary-ion mass spectrometry (SIMS), X-ray diffraction (XRD) and scanning electron microscopy (SEM). The results obtained for a B_4C disc with natural boron composition prove the efficiency of the methodology.

1. Introduction

In the nuclear field, there is an increasing demand for more physically based modelling of material properties. The reason for this is twofold: to limit the number of validation irradiations required in real reactor conditions, and to provide more reliable predictions outside the validation domain. This tendency largely applies to nuclear fuel, with numerous papers moving from first-principle calculations to fuel microstructure modelling [1]. This demand for physically based modelling is also relevant for neutron absorber materials, though less work has been done in this field. Boron carbide is one of the most frequently used materials for neutron absorbers and its phenomenology under irradiation has been long documented [2,3]. In actual reactor conditions, it is subjected to both atomic collision with fast neutrons and transmutation reactions due to neutron capture by ^{10}B isotopes. Neutron atomic collisions produce collision cascades that result in point or extended defects. ^{10}B transmutation reactions (up to $10^{22}\,cm^{-3}$, i.e. about 10% of the total boron in current fast reactor conditions) produce He and Li recoils that induce defects during their stopping and chemical modification of the pristine boron carbide. Helium clusters in over-pressurized small bubbles could lead to inter- and intragranular cracking [4]. The fate of lithium is less clear since this element is not visible using classical methods such as electron probe micro analysis (EMPA) or X-ray photoelectron spectroscopy (XPS).

Physically based modelling requires the identification of each physical mechanism that is needed to describe the material. In the case of boron carbide under irradiation, the effects of atomic collisions with fast neutrons and transmutation reactions cannot be easily separated from the available experimental results obtained by irradiation in nuclear reactors or by ion implantation experiments. During irradiation in a reactor, the material is subjected to a broad neutron energy spectrum leading to a complex weighting of the two interactions, i.e. absorption and ballistic damage. Thus, ballistic and transmutation effects are intricately linked. With ion implantation, a specific irradiation defect can be reproduced by choosing a suitable ion and energy level, but this type of experiment cannot be used to reproduce the transmutation phenomenon as a whole.

Each mechanism also has to be characterised as a function of temperature but samples are required to do this. They must be irradiated at a constant temperature and they must be large enough so post-irradiation characterisation tests can be performed. In reactor irradiation conditions, the temperature follows a complex history as a function of the reactor power and varies spatially as a function of the neutron flux. In ion implantation experiments, the temperature may be known with better accuracy but the resulting irradiated area is usually only a layer less than 1 μm thick. In this case, only a few experimental methods (e.g. TEM) can provide valuable information; X-ray diffraction cannot be used in this instance.

In order to overcome these difficulties, we developed a new methodology for studying neutron absorber materials in which only transmutation defects are produced within bulk samples at a practically constant temperature. This methodology was used for the irradiation of

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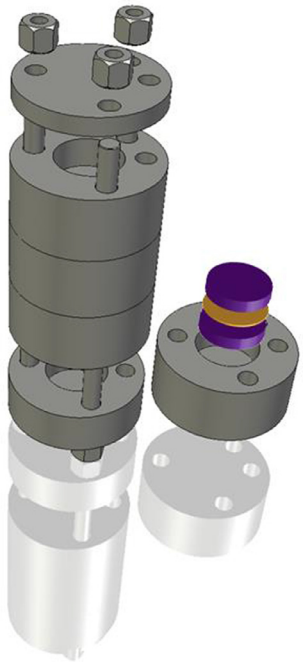

Fig. 1. B_4C sample (orange) sandwiched between two SiC discs (purple) positioned in their sample holder (grey). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

B_4C discs in a new sample holder in the high flux reactor at the Institut Laue Langevin (ILL). This paper first describes the sample holder, before detailing the irradiation experiment and the preliminary characterisation results. These results are discussed in the conclusion, showing how the new methodology met our requirements.

2. Sample holder

The samples were first settled in an aluminium shuttle so they could fit into the T4 beam tube. A new sample holder was designed not only to meet the above irradiation objectives presented, but also to fit into this shuttle. A schematic diagram of this sample holder is given in Fig. 1.

The B_4C discs were 0.3 mm thick and 10 mm in diameter. They were prepared from a high-density (> 98% of theoretical density = 2.52) pellet with a small grain size (5–10 μm) and mirror-polished surfaces. Two β-SiC discs – 2 mm thick and 10 mm in diameter – were placed on each side of the B_4C discs. These β-SiC (theoretical density = 3.51) discs were prepared from a CVD-made ceramic pellet and were placed in contact with the mirror-polished surface of the B_4C samples. Each SiC-B_4C-SiC sandwich was placed inside a hole drilled in an aluminium disc. Five aluminium discs were fabricated and stacked on top of each other to build the sample holder. The ^{10}B enrichment of the samples was natural (19.9%) for the three bottom B_4C discs, 48% for the fourth disc and 90% for the top disc. The aluminium discs were held together by three screws. An additional hole was drilled through all the discs to let the air flow out when the sample holder was placed in the capsule. Small zirconium foils were inserted in this hole at different heights to determine the neutron flux using post-irradiation gamma spectrometry. Fig. 2 shows how the sample holder was assembled (Fig. 2a) and its installation in the capsule (Fig. 2b).

This design also guarantees the safe irradiation of the sample holder inside the capsule in the T4 tube. The high thermal conductivity of SiC and the level of contact between the mirror-polished surfaces of B_4C and SiC ensures that the heat produced by the transmutation reactions is removed efficiently. The irradiation time was also set to 5 days, not only to produce a sufficient level of transmutation for further characterisation, but also to minimise the helium production and maintain the pressure increase in the capsule at less than one atmosphere, assuming the total release of helium.

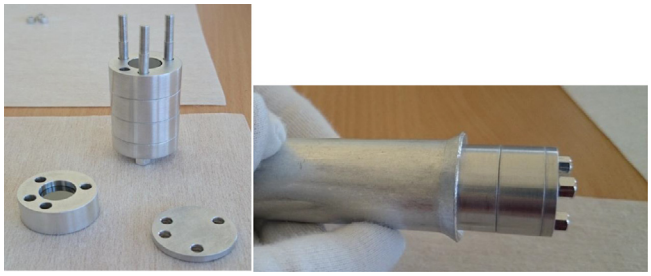

Fig. 2. Sample holder fabrication and installation in the aluminium capsule.

3. Irradiation

The T4 irradiation tube was positioned in the reflecting light water vessel, very close to the heavy water tank in the ILL reactor. This location leads to a neutron flux with a Maxwellian distribution at a temperature about 20 °C (calculated in reference [5]).

The shuttle was positioned vertically in the T4 tube so that the B_4C discs were parallel to the neutron flux from the core. Unfortunately, the position of the sample holder inside T4 could not be known. As a consequence, the relative position of the zirconium foils (ϕ1) and the SIMS quantitative line scan (ϕ2, discussed below) were not known relative the neutron flux. This neutron flux is known for its large local variations because of nuclear reactions with ^{10}B that reduce the flux in the vicinity of the B4C disc. Not knowing ϕ1 and ϕ2 induces large uncertainties in the neutron flux at the positions of the zirconium foils and SIMS line scan. Consequently, we did not attempt to model the neutron flux theoretically but instead chose to rely on its experimental determination.

The penetration depth of 0.025 eV thermal neutrons in natural B_4C is about 130 μm ($\sigma\mathbf{N} = 76\,cm^{-1}$). Thus, the irradiation damage was more or less homogeneous in our 300 μm thick B_4C discs that had natural ^{10}B enrichment, despite the local neutron flux changes discussed above.

The capsule containing the sample holder and the five SiC-B_4C-SiC sandwiches was irradiated for five days in June 2015. Because of the radiation level of the capsule after irradiation, it was only removed from the hot cell in September 2015. First of all, the two ends of the capsule were cut and the small zirconium foils were removed from the hole drilled for air flow. Gamma spectrometry measurements were used to determine their accumulated dose and the neutron flux during irradiation at different heights inside the sample holder (see Table 1). The average measured flux was consistent with the nominal flux in T4 [6].

Secondly, the capsule was sent to the CEA/Cadarache centre. Its dose rate measured at a distance of 5 cm was 1500 μSv/h after a period of 40 days following the end of the irradiation. This was mainly due to the activation of chromium, which is used as alloying element in the aluminium sample holder and capsule. The capsule was dismantled in a controlled area so the SiC-B_4C-SiC sandwiches could be removed. The radioactivity of the SiC and B_4C discs was measured. The dose rate for each sample was less than 0.1 μSv/h at a distance of 5 cm and the labile contamination was less than $0.4\,Bq\,cm^{-2}$ for alpha activity and

Table 1
Calculated neutron flux at the different zirconium foil positions.

Foil number	❶	❷	❸	❹	Average
Foil mass (mg)	1.59	1.63	1.71	1.35	
Calculated flux in 10^{13} n/cm^2/s	1.8	1.73	1.39	2.18	1.7 ± 0.3