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Influence of boron carbide on core degradation during severe accidents in LWRs

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ABSTRACT

Boron carbide (B_4C) is widely used as neutron absorbing control rod material in light water reactors (LWRs). It was also applied in all units of the Fukushima Dai-ichi nuclear power plant. Although the melting temperature of B_4C is 2450 °C, it initiates local, but significant melt formation in the core at temperatures around 1250 °C due to eutectic interactions with the surrounding steel structures. The B_4C containing melt relocates and hence transports material and energy to lower parts of the fuel bundle. It is chemically aggressive and may attack other structure materials. Furthermore, the absorber melt is oxidized by steam very rapidly and thus contributes to the hydrogen source term in the early phase of a severe accident.

After failure of the control rod cladding B_4C reacts with the oxidizing atmosphere. This reaction produces CO, CO_2, boron oxide and boric acids, as well as significant amount of hydrogen. It is strongly exothermic, thus causing considerable release of energy. No or only insignificant formation of methane was observed in all experiments with boron carbide.

The paper will summarize the current knowledge on boron carbide behavior during severe accidents mainly based on experiments performed at KIT, and will try, also in the light of the Fukushima accidents, to draw some common conclusions on the behavior of B_4C during severe accidents with the main focus on the consequences for core degradation and hydrogen source term.

1. Introduction

The motivation for this paper came from considerations of the progress of the severe nuclear accidents in the Fukushima Dai-ichi NPP in March 2011. All affected units there were boiling water reactors (BWRs) using boron carbide as absorber material.

Worldwide, many light water reactors (LWRs) use boron carbide as neutron absorbing material for control units (Kawada, 1998); details will be given in the next section. Boron carbide may influence degradation of the fuel elements, and it is connected with release of energy and various gases. One big concern regarding gas release was the potential formation of methane CH_4 which could form volatile organic iodine compounds.

B_4C absorber behavior during severe accident scenarios was investigated in various CORA tests (Sepold et al., 2009). In these out-of-pile test series with prototypical materials it was found that the presence of B_4C causes the formation of low-temperature melt at around 1250 °C that attacks the adjacent channel box and fuel rod cladding. The absorber melt also interacts with UO_2 fuel and causes its dissolution at low temperatures. Large parts of the absorber material were relocated from the top to the bottom of the test bundles.

In the framework of the EC sponsored COLOSS program (Adroguer et al., 2005) various experiments on different scales were performed including CODEX at AEKI (Hózer et al., 2001) and QUENCH at KIT (Steinbrück et al., 2004a; Steinbrück, 2004b) bundle experiments and separate-effects tests in the VERDI (Cocuaud, 2003) and BOX (Steinbrück, 2005 and Steinbrück, 2010) test rigs at IRSN and KIT, respectively. One advantage of the QUENCH tests is the sophisticated off-gas analysis by mass spectrometry. Data of these experiments were used for development of advanced models (Seiler et al., 2008; Steiner, 2007; Steinbrück et al., 2007) and validation of severe fuel damage (SFD) codes (Drath et al., 2006 and Drath, 2007).

The in-pile bundle test Phebus FPT3 (Repetto et al., 2011; Payot, 2009), which was the last within the FPT series at CEA Cadarache, studied the impact of boron carbide control rod on fuel degradation, fission product transport and deposition in the circuit and the behavior in the containment using irradiated fuel. Substantial influence of B_4C on melt formation and relocation to the bottom of the test bundle was observed also in this test. In addition, transport of boron containing compounds to colder regions of the circuit with blockage formation was found (Haste et al., 2012). Complementary small-scale tests studying boron carbide interactions with

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the atmosphere and other materials were conducted in the framework of the International Source Term Program (ISTP) BECARRE at IRSN (Clément and Zeyen, 2005; Dominguez, 2012; de Luze, 2013). Extensive isothermal experiments with boron carbide – stainless steel mixtures up to 1527 °C in steam atmosphere revealed different kinetic regimes caused by the formation of liquid protective layers and melt splashing.

Recently, the impact of boric acid on volatile fission products specification was summarized by Kissane et al. (2011) showing that boron strongly affects the quantities and physico-chemical forms of these elements that reach the containment. Furthermore, boron carbide influences the in-vessel corium behavior in the lower RPV plenum. AREVA's EPICOR program (Fischer et al., 2012) performed at A.P. Alexandrov Research Institute of Technology, Sosnovy Bor, Russia, has clearly demonstrated that boron carbide may strongly reduce the density of metallic melts, thus affecting the inversion point in the metal-oxide corium system.

The risk of recriticality due to relocation of boron carbide from the fuel elements should be mentioned, but is beyond the scope of the paper.

This paper summarizes the main effects of boron carbide focused on bundle degradation as well as on the release of gaseous and condensable reaction products and chemical energy during severe nuclear accidents. It makes no claim to be complete and is mostly illustrated by examples of experimental results obtained at KIT. For more detailed information the reader is referred to the list of references.

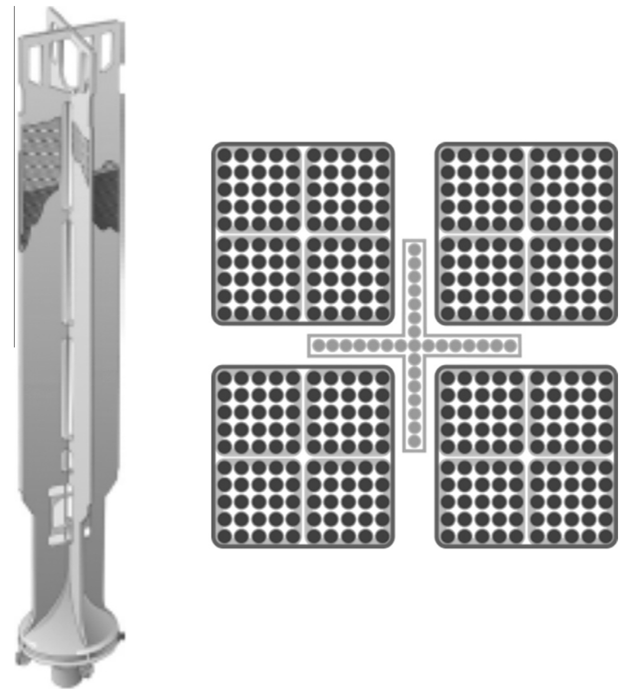

Fig. 1. Westinghouse BWR control rod with B_4C (left) and its typical arrangement between fuel elements (right).

2. B_4C inventory in LWRs

Boron carbide is used as neutron absorbing material in all boiling water reactors (BWRs), in later-generation pressurized water reactors (PWRs), and in Russian VVERs. It is applied in form of powder or pellets.

French 1300 and 1450 MR PWRs, the currently build European Pressurized Reactor (EPR), and VVERs use B_4C absorber rods, combined in control rod assemblies, with stainless steel (SS) cladding. Hybrid control rods with silver–indium–cadmium (SIC) alloy in the lower part of the rod are applied in the PWRs. The surrounding guide tube is made of Zircaloy-4 in PWRs and of stainless steel in VVERs.

In BWRs the boron carbide is embedded in a cruciform-shaped blade made of stainless steel and moving in a channel box made of Zircaloy. Fig. 1 gives an example of a Westinghouse type of BWR control rod (Westinghouse, 2013) and shows schematically the arrangement of the control blade between the fuel elements (Wikipedia, 2013).

The amount of B_4C per reactor varies between 300 and 2000 kg. Table 1 gives examples of the boron carbide inventory in various types of reactors. For instance, one control blade in the Fukushima Dai-ichi NPP units consists of 7 kg B_4C and 93 kg stainless steel.

Table 1
B_4C inventory of selected nuclear reactors.

Reactor	Mass B_4C, kg
German BWR line 69	ca. 1200
German BWR line 72	ca. 1700
GE BWR-3 (Fukushima Daiichi, unit 1)	680
GE BWR-4 (Fukushima Daiichi, unit 2–4)	960
VVER 1000	250
FRAMATOME PWR 1300 MW	320
FRAMAROM PWR 1450 MW	340
EPR	440

3. Degradation mechanism of B_4C absorber rods

Independently on the reactor type, the boron carbide absorber powder or pellets are encased in stainless steel structures (rods or blades). The B_4C–SS system is not thermodynamically stable. Chemical interactions start already at 800 °C and lead to complete failure of the control rod/blade at around 1250 °C by rapid liquefaction of the stainless steel approx. 200 K below its melting point (Steinbrück, 2010; Hofmann et al., 1990; Nagase et al., 1997; Belovsky, 1997). Fig. 2 illustrates this behavior showing results of separate-effects tests with one-pellet size control rod segments. These specimens were annealed in a horizontal tube furnace for 1 h at the specified temperature in steam atmosphere. After failure of the absorber rods, B_4C as well as the resulting absorber melt may interact with Zircaloy channel box and guide tubes, respectively, and the adjacent fuel rods. The B_4C-Zircaloy liquefaction occurs at much higher temperatures, i.e. at ca. 1650 °C.

The eutectic temperatures of the binary systems B–Fe, Cr, Ni are at 1174, 1630, and 1093 °C with the concentration of boron being 2–4 wt% (Massalski et al., 1990). The minimum of the liquidus surface in the ternary phase diagram Fe–B–C is at 1029 °C in the iron-rich part of the system (Rogl, 2008). Experimentally, it was shown that in the more complex system B_4C–SS only 1 wt% boron carbide is necessary to completely liquefy the stainless at about 1250 °C (Steinbrück et al., 2004c). Fig. 3a reveals the complete melting a SS cylinder filled with 1 wt% B_4C after 1 h at 1250 °C. More recent experiments allowed the direct observation of the B_4C–SS reaction by video camera. Approx. 1-cm sized stainless steel samples completely liquefied at ca. 1250 °C within less than one minute only by having loose contact with a boron carbide surface.[1]

Having in mind that a control blade in the Fukushima Daiichi NPPs consists of 93 kg stainless steel and 7 kg boron carbide, it is very likely that these assemblies were completely destroyed very early in the accidents.

[1] The video can be obtained from the author on request.