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Radiation-tolerant joining technologies for silicon carbide ceramics and composites

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ABSTRACT

Silicon carbide (SiC) for nuclear structural applications, whether in the monolithic ceramic or composite form, will require a robust joining technology capable of withstanding the harsh nuclear environment. This paper presents significant progress made towards identifying and processing irradiation-tolerant joining methods for nuclear-grade SiC. In doing so, a standardized methodology for carrying out joint testing has been established consistent with the small volume samples mandated by neutron irradiation testing. Candidate joining technologies were limited to those that provide low induced radioactivity and included titanium diffusion bonding, Ti–Si–C MAX-phase joining, calcia–alumina glass–ceramic joining, and transient eutectic-phase SiC joining. Samples of these joints were irradiated in the Oak Ridge National Laboratory High Flux Isotope Reactor at 500 or 800 °C, and their microstructure and mechanical properties were compared to pre-irradiation conditions. Within the limitations of statistics, all joining methodologies presented retained their joint mechanical strength to ~3 dpa at 500 °C, thus indicating the first results obtained on irradiation-stable SiC joints. Under the more aggressive irradiation conditions (800 °C, ~5 dpa), some joint materials exhibited significant irradiation-induced microstructural evolution; however, the effect of irradiation on joint strength appeared rather limited.

1. Introduction

The use of silicon carbide (SiC) composites in nuclear power applications, primarily fusion reactor structural elements or light-water reactor (LWR) fuel cladding and core constituents, has received increasing attention over the past decade and has been discussed as a critical need over the past two decades [1–3] for conceptual fusion reactor designs such as the European TAURO design [3], the US ARIES-AT and ARIES-ACT designs [4,5], and the Japanese DREAM [6]. Fig. 1 gives a schematic representation of the TAURO design, essentially consisting of nested plates made from SiC composite that form a simple compartmentalized box design in which the first wall and blanket internals are separately cooled by Pb-17Li. The ARIES-AT concept is of a similar design, again utilizing SiC composite and Pb-17Li coolant, in fairly simple geometry for ease of fabrication and joining. At the time of these fusion reactor designs, joining technologies were not specified.

For fission power applications, two generic design types employing SiC composites for LWR cladding are under discussion: fully ceramic SiC cladding and ceramic/metal cladding. For the fully ceramic cladding, some combination of fibrous composite is utilized to achieve fracture resistance and high-temperature performance with a single layer or multiple layers of monolithic SiC ceramic used to seal the inherently porous and microcracked composite. Using the metal/ceramic design, sealing is achieved either by an inner metallic bladder (liner) or outer metallic seal. For the fully ceramic system, an end-cap seal is required, which is a critical technology development still awaiting a solution.

As previously stated, the ability of joining SiC to itself or other materials is a critical, unresolved technological need regarding the application of SiC ceramics and composites in nuclear systems. This is in contrast to the ability to join SiC or SiC/SiC for non-nuclear applications, which can be accomplished in a reliable and rugged manner through a number of conventional and advanced techniques. The suite of nominally conventional techniques that has been successfully demonstrated includes pre-ceramic polymer joining, glass–ceramics, reaction bonding, active metal/pre-ceramic polymers, and active metal solid state displacement reactions. Additional details of these methods are provided in Section 2 of this paper. Although comparison of results from different studies on the strength of various bonding techniques is confounded by

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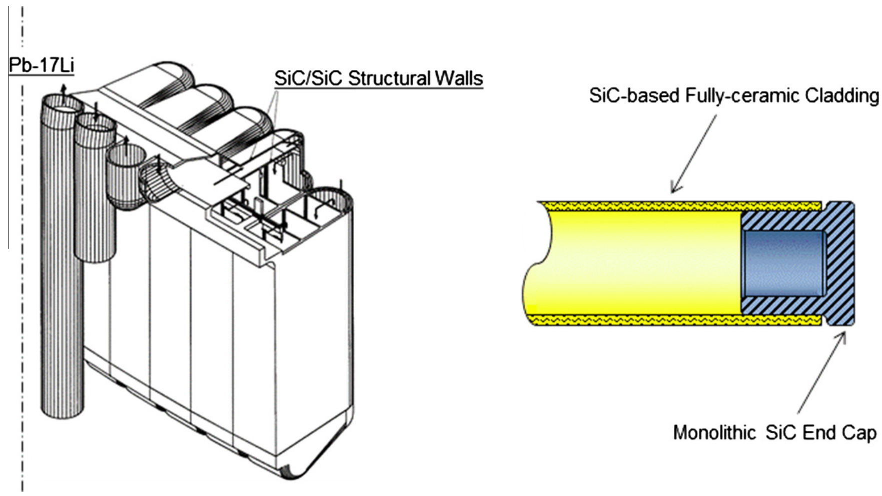

Fig. 1. Conceptual schematic of TAURO blanket design (left) and end plug section of fully ceramic LWR fuel rod (right).

a lack of standards for testing and the variety of test types used (e.g., bend vs tensile, etc.), the strength of the joints produced appears adequate for fusion and nuclear applications.

A key issue is that the methods studied to date, and the limited data available on the irradiation of joints and the materials making up those joints, have demonstrated poor irradiation stability. As discussed previously, a reliable joining technique for SiC still remains to be developed for nuclear applications. This is why mechanical component designs for near-term applications (such as control rod sleeves for high-temperature gas-cooled reactors) avoid the issue of joining entirely, preferring instead to use mechanical joints [7,8]. However, this option is not possible for fusion blanket systems, which require leak-tightness to PbLi or in some cases helium, or in the case of LWR cladding, as hermeticity (the retention of fission products and helium) is a functional requirement.

Another issue related to joining of SiC is the lack of a widely accepted standard test method: a reliable test not prohibitively difficult and/or expensive to prepare and capable of yielding reproducible shear strength is still unavailable [9]. An additional concern for nuclear applications is the use of miniaturized specimens necessary for testing irradiated samples. Torsion of solid specimens seems to be the appropriate test method for post-irradiation shear properties of joints of SiC-based materials [10]. The optimization of torsional specimen size and geometry was performed through the collaborative efforts of Oak Ridge National Laboratory (USA), Kyoto University (Japan), and Politecnico di Torino (Italy). Based on this development, a reliable comparison among different joints before and after neutron irradiation is presented in this work.

2. Joining technologies for silicon carbide

Various methods have either already been established or are currently considered promising for joining SiC to SiC, either monolithic or composite, for non-nuclear applications. These methods include solid state sintering of SiC ceramics, diffusion bonding using various active fillers [11], transient eutectic-phase routes [12–15], glass–ceramic joining [9,10,16], metallic braze-based joining [17–20], SiC reaction forming [21–23], MAX-phase bonding [24], pre-ceramic polymer routes [25–27], and selective area chemical vapor deposition (CVD) [28], to name a few. Joining processes that utilize solid state diffusion or transient liquid phases may often be assisted by additional local/internal heating schemes such as laser, electron beam, and pulsed electrical field (as used for spark plasma sintering) applications. When these techniques are discussed for application in nuclear environments, consideration must be given to issues related with neutron irradiation, in addition to those common to non-nuclear applications: strength and reliability during mechanical loading, required pressure for processing, potential process-induced damages to the base materials, chemical compatibility of the joint with the specific operating environment, and the ability to satisfy the design-specific hermeticity requirement. Specific to fusion applications is the strong bias towards materials with low induced radioactivity to ensure the structure remains a "low-activation" component. Below, representative joining methods for SiC ceramics and composites are briefly reviewed and discussed in terms of potential critical issues for applications in fusion and nuclear energy systems.

2.1. Solid state diffusion bonding

SiC to SiC can be achieved through a purely solid state self-diffusion process at very high temperatures and high pressure. This is obviously the ideal method, along with the selected area CVD, for producing a robust permanent joint in terms of chemical purity and structural continuity. However, considering that self-diffusion bonding typically requires high pressure at a temperature of at least $0.75T_m$, its utilization in practical applications will be severely limited, although the field-assisted technique achieves adequate bonding at a "low" temperature of $\sim$2000 °C.

Far more common methods to join SiC to SiC are metal diffusion bonding techniques, which are accomplished by solid-state diffusive conversion of a metal insert into compounds with the host elements, silicide and/or carbide with SiC, to produce strong joints [29,30]. Metal diffusion bonding of SiC has been successfully achieved with various refractory metals, such as titanium, molybdenum, tantalum, niobium, and tungsten, which form carbides that are thermodynamically more stable than SiC [11]:

$$(xy)\mathrm{SiC} + (x+y)\mathrm{M} \rightarrow (y)\mathrm{MC}_x + (x)\mathrm{MSi}_y. \quad (1)$$

Solid state diffusion for these metals is generally fast enough at temperatures higher than $\sim$1200 °C to result in full conversion of a thin metal foil (tens of microns) insert or powder slurry into the ceramic phases. Titanium makes Ti–Si–C ternary MAX phases in addition to the binary conversion products under certain conditions, and these are specifically classified as MAX-phase joining. The strength of the metal diffusion bonding is generally determined