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High-temperature stability of laser-joined silicon carbide components

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ABSTRACT

Silicon carbide is recommended for applications in energy technology due to its good high-temperature corrosion resistance, mechanical durability, and abrasion resistance. The prerequisite for use is often the availability of suitable technologies for joining or sealing the components. A laser-induced process using fillers and local heating of the components represents a possible low-cost option.

Investigations in which yttrium aluminosilicate glass was used for laser-induced brazing of SiC components of varying geometry are presented. A four-point bending strength of 112 MPa was found for these joints. In burst tests, laser-joined components were found to withstand internal pressures of up to 54 MPa.

Helium leak tests yielded leak rates of less than 10^{-8} mbar l s^{-1}, even after 300 h at 900 °C. In contrast, the assemblies showed an increased leak rate after annealing at 1050 °C.

The short process time of the laser technique – in the range of a few seconds to a few minutes – results in high temperature gradients and transients. SEM analysis showed that the filler in the seam predominantly solidifies in a glassy state. Crystallization occurred during later thermal loading of the joined components, with chemical equilibrium being established. Differences in seam structures yielded from different cooling rates in the laser process could not be equalized by annealing.

The results demonstrated the long-term stability of laser-brazed SiC assemblies to temperatures in the range of glass transformation (900 °C) of the yttrium aluminosilicate filler. In technological investigations, the suitability of the laser joining technique for sealing of SiC components with a geometry approximating that of a fuel element sleeve pin (pin) in a gas-cooled fast reactor was proven.

1. Introduction

Development of GEN IV reactors is aimed, among other things, at safe design [1]. To ensure this, the inclusion of the fuel in the form of TRISO particles in sleeve elements made of thermally stable and radiation-resistant material is foreseen for two reactor types, the (very)-high-temperature reactor (VHTR) and the gas-cooled fast breeder reactor (GFR).

The VHTR fuel element sleeves have a prismatic or spherical design. Depending on the design of the reactor type, different working temperatures are reported. The highest are given for the Frameatome design with a reactor outlet temperature of 1000 °C, a maximum fuel temperature of 1300 °C, and an accident peak fuel temperature of 1600 °C [2]. The pressure values for the primary coolant lie between 4 MPa [3] and 7 MPa [4].

The fast neutron spectrum of the GFR (CEA-design) requires a ceramic fuel concept with a good thermal conductivity. Fuel arrangements in pin- and plate-geometry are envisaged. For an average exit core temperature of 850 °C, the maximum fuel temperature is about 1200 °C. For accident conditions, a maximum fuel temperature of 1600 °C has been chosen. The nominal primary helium pressure is 7 MPa [5].

These requirements are particularly well met by ceramic materials, with silicon carbide (SiC) being the most preferred and investigated material. The characteristics of SiC include high thermal, mechanical, corrosion, and abrasion resistance as well as very good thermal shock resistance [6]. SiC is also thermally stable in conditions of high neutron activation [7]. On account of these properties, SiC is recommended for use in functional and structural components in high-temperature reactor applications. Implementation of the fuel concepts for VHTR and GFR requires a technology that ensures the gastight inclusion of the fuel in the specified conditions [8]. Use of glass–ceramic fillers can be suitable for ensuring the resistance of joined composites in these conditions [9].

The working temperatures of these two reactor types will be used as a basis for the thermal design of the cladding joints. To the best of the authors' knowledge, no requirements regarding the leak rates of ceramic claddings in nuclear reactors have been defined so far. The German KTA (Nuclear Technical Commission) fixed the leak rate for tube-to-tubes joints in components of the primary loop at 10^{-6} mbar l s^{-1} [10]. This value will be used as a target figure in consideration of the fact that the cladding is part of a multistage containment for the retention of fission products.

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A laser-based joining technology has been developed at the Technical University Dresden [11]. This technology enables the joining of the ceramic parts with a glass–ceramic filler, which is melted by a laser beam, as well as production of large parts, e.g., with electrically conductive joints [12]. One advantage of this technology is that the parts only need to be heated locally for joining. The laser joining process is also extremely fast (lasting a few seconds to a few minutes). The material to be joined must have a high thermal shock resistance to withstand the temperature gradients arising during the laser process without being damaged. Through its combination of high thermal conductivity and low thermal expansion coefficients [6], SiC is suitable for being heated via laser radiation.

Good wetting by the filler and suppression or minimization of mechanical stresses in the joint are the main prerequisites for the thermomechanical and chemical stability of the joint. Using glass–ceramic fillers, it is possible to match the thermal expansion coefficients of the filler and the ceramics [13]. SiO_2-based fillers exhibit very good wetting of SiC surfaces [14].

Y_2O_3–Al_2O_3–SiO_2 glasses are recommended due to their good mechanical strength, high Young's modulus, high glass transformation temperature, and good chemical resistance [15,16]. The thermal expansion coefficients of some of these glass compositions lie in the same range as that of SiC [17]. That is why these compositions are interesting as filler materials for the joining of SiC materials [18,19]. Smeacetto et al. used these glasses as coatings for carbon-bonded carbon fiber composites [20]. The NITE process for the joining of SiC_f/SiC components is also based on the use of the Y_2O_3–Al_2O_3–SiO_2 system; the addition of SiC-nanopowders leads to modification of the chemical system [21].

The suitability of such glasses for nuclear applications has been described by various authors. Microspheres made of yttrium aluminosilicate glasses were investigated by Hyatt and Day for radiotherapeutic purposes [17]. The suitability of such glasses for the immobilization of minor and long-lived actinides was studied by Gavarini et al. and Sadiki et al. [22,23]. Ferraris et al. pointed out the low neutron-induced radioactivity, resulting from the low activation rates of the elements yttrium, silicon, and oxygen [9]. Rohbeck et al. demonstrated the chemical resistance of a glass from this system to PuO_2 and UO_2 [24].

Decisive for the application of glass–ceramics as filler materials for applications in nuclear reactors is their resistance to neutron radiation, especially fast neutron radiation in the case of the GFR. The interaction between glasses and radiation has been published by different authors. For instance, Devine, Gavarini, and Sandhu studied the influence of neutron radiation on silicate glasses [22,25,26]. Boizot et al. described the effects of beta-irradiation on multicomponent aluminoborosilicate glasses used for nuclear waste storage [27]. Gedeon et al. has shown the changes in surface morphology of silicate glass induced by fast electron irradiation [28].

Coghlan et al. studied the impact of neutron radiation on glass–ceramics [29]. He used neutron fluxes up to 1×10^{23} n/m^2 for the treatment of MACOR and found changes in the swelling value that were not proportional to the neutron fluxes. This was explained by two simultaneous processes: the expansion of the crystalline mica phase and the contraction of the glassy phase. The differences in behavior of the crystalline phase (swelling) and the glassy phase (densification) lead to the conclusion that the application of glass–ceramic fillers is still debatable and needs further investigation [30]. The changes in the microstructure are influenced by the level of the neutron flux and the composition of the glass–ceramic.

Initial investigations on glass–ceramics from the system Y_2O_3–Al_2O_3–SiO_2 have been performed, and the results are available. Ferraris et al. used a glass–ceramic from this system for the joining of SiC_f/SiC-components for application in fusion and fission systems [18]. The activation of the glass–ceramic was limited through fixing the content of Al_2O_3 in the glass–ceramic at 18 wt% [9]. The radiation of SiC_f/SiC-joints made with Y_2O_3–Al_2O_3–SiO_2 glass–ceramic up to 820 °C and neutron fluxes $31–32 \times 10^{24}$ n/m^2 ($E > 1$ MeV) showed no influence of the neutron radiation on the crystalline phase composition or the microstructure. Swelling and/or shrinkage processes could not be observed [31]. For the neutron radiation of the joined samples, Ferraris et al. used a temperature below the glass transformation region of these glasses. Temperature-induced nucleation and crystallization is not expected at this temperature. Based on the fact that a neutron flux $31–32 \times 10^{24}$ n/m^2 did not affect the microstructure, it can be concluded that the neutron flux did not induce additional crystallization processes. These results support the authors' use of yttrium aluminosilicate fillers for laser-supported joining for nuclear applications.

The microstructure described by Ferraris et al. was developed using a furnace-induced crystallization process. The brief duration of the laser process is not conducive to the relaxation of the glass in the seam and the establishment of chemical equilibrium. Crystallization processes are inhibited, taking place later either in an additional thermal treatment step or during application of the joined components. Because the mechanical and thermal resilience of a joint is determined by the microstructure in the zone between the two joined components, the formation of a suitable seam structure is crucial to the process (for the resistance under application conditions). Information on the crystallization behavior of the Y_2O_3–Al_2O_3–SiO_2 glasses processed by the laser beam has been unavailable up to now. Thus comparison between the neutron irradiation behaviors of the laser joined samples and those reported in Ref. [31] cannot be straightforward. Additionally the reactivity of silica–allumina–yttria glasses/glass–ceramics silica-alumina-yttria glasses/glass-ceramics under reactor conditions (in particular humidity) should be assessed and also their creep behavior at the working conditions.

Nuclear fuels for future reactor designs are being developed within the framework of the EU-funded "F-Bridge" (Basic Research for Innovative Fuel Design for GEN IV Systems) project [32]. Development of joining technologies for components containing the fuel in the form of TRISO particles is included in this work. The goal of the work at TU Dresden was to demonstrate the suitability of laser-induced sealing of SiC components using yttrium aluminosilicate fillers through investigation of the thermal stability of the joints at temperatures similar to those that could occur during reactor operation and determination of the mechanical strength and the gas-tightness of the joined parts. The technological implementability of the process was also tested. A geometry approximating that of a possible pin geometry for GFR was used for investigations on the temperature distribution during the laser process and the reproducibility of the joining process.

2. Experimental work

For achieving a very low leak rate in the material to be joined, a pressureless-sintered silicon carbide (SSiC) material with no open porosity was chosen for the joining trials. The laser joining technology was tested on three different part geometries: pins, capsules, and rods.

A StarCeram® SSiC [33] material was used for the pins, which were composed of three separate parts, each having an outer diameter of 10 mm (Fig. 1a). A 100-mm-long tube with a wall thickness of 2 mm was sealed with 20-mm-long caps at each end, yielding a total length of 140 mm for the joined part. This part geometry was utilized for technological investigations of the laser process. Two