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Joining of machined SiC/SiC composites for thermonuclear fusion reactors

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Abstract

A low-activation glass–ceramic based on silica, alumina and yttria has been designed and tested as joining material for 2D fusion grade SiC/SiC. Neutron-induced radioactivity of elements present in the glass has been simulated by European Activation System EASY-2007 code package. The mechanical strength of the joined SiC/SiC has been tested by 4-point bending on three different kinds of joined samples. Bending strength higher than 120 MPa has been measured at room temperature, with composite failure in most cases.

1. Introduction

Ceramic fibre reinforced ceramic matrix composites (CMC) are attractive materials for many applications that require extreme high temperature stability and damage tolerance, especially for space, terrestrial and nuclear applications. Among CMC, silicon carbide fibre reinforced silicon carbide matrix composites (SiC/SiC) show a much greater resistance to high temperatures and aggressive environments than metals or other conventional engineering materials, thermal shock resistance, radiation stability and low neutron-induced activation [1–3], which make them suitable candidate materials for nuclear applications.

Since the manufacturing of complex and large parts cannot be performed, applications for SiC/SiC will depend on the capacity to join simple components into more complex structures. SiC/SiC composites have no melting phase, therefore it is impossible to join them by conventional fusion welding and diffusion bonding does not seem suitable as the inter-diffusion of SiC is very low, even at high temperature. A well known method for joining SiC ceramic parts is the co-sintering of ceramic components [4] but this method needs high pressure and high processing temperature.

Furthermore, several indirect joining techniques suitable for SiC/SiC or SiC ceramics have been investigated: laser assisted joining of SiC/SiC with oxide based solders [5], joining by pre-ceramic polymers [6,7], joining by glass and glass–ceramic [8,9], reaction forming [10], metallic brazing [11,12] and microwave assisted joining [13].

In the field of nuclear energy production (fusion and fission), requirements for SiC/SiC joining are extremely severe: a joining material must be compatible with a neutron environment and the joining technique must comply with the fusion nuclear reactor design where SiC/SiC components several meters long and 3 mm thick must be joined in a reliable and feasible way [14,15].

The very few available solutions for SiC/SiC joining in the field of nuclear applications are reported or reviewed in [6,8,11,12,16–18]. None of them have been tested in a nuclear environment up to now.

The aim of this paper is twofold: to design a low-activation glass–ceramic as joining material for SiC/SiC, and to use it to join machined SiC/SiC, as requested in [14], in order to obtain a mechanically reliable, low-activation

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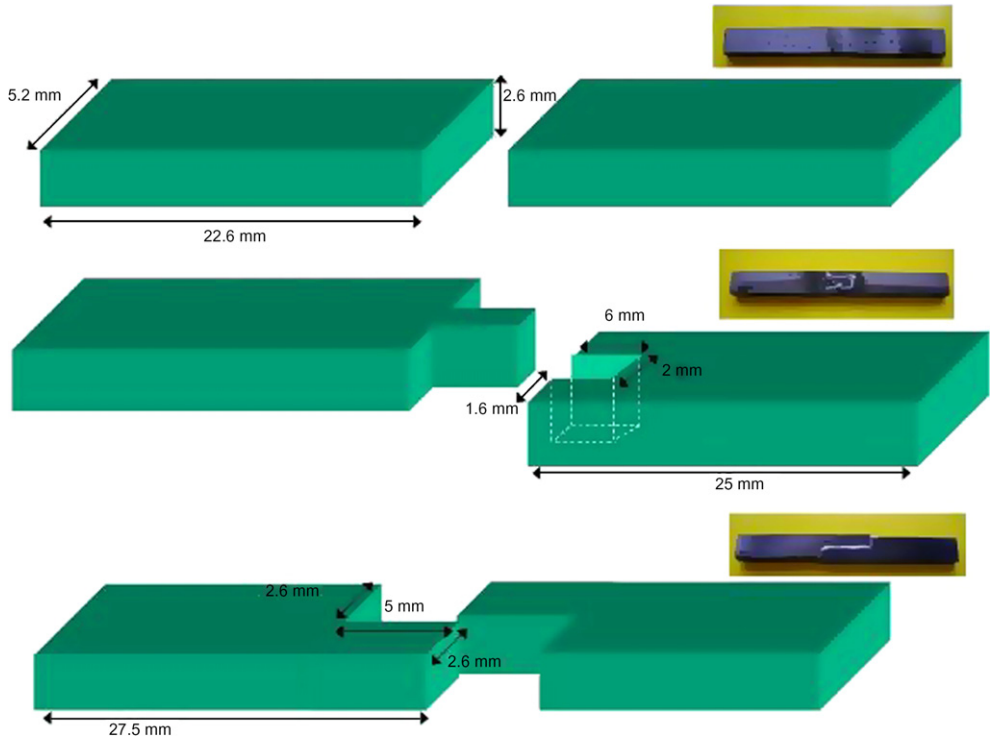

Fig. 1. Three different joint configurations of joined SiC/SiC composites tested by four-point bending test. (SiC/SiC thickness = 2.6 mm, width = 5.2 mm, length = 45 mm).

and hermetic joint for SiC/SiC to be used in a nuclear environment.

2. Experimental

The SiC/SiC composites used for this study are fusion-grade 2D composites provided by MT Aerospace, Germany, with a CVD–SiC coating of about 60 μm. The fibre type in the composite is SiC Tyranno Type S, the matrix is crystalline SiC, via CVI process with a carbon fibre/matrix interface. The SiC/SiC composite has a 0°/90° plain weave fabric parallel to mechanical loads used to characterize the joined samples.

The composition (wt%) of the glass (a SiO_2–Al_2O_3–Y_2O_3 based glass, referred to as SAY) used as joining material was designed according to

1. neutron-induced radioactivity of elements present in the glass, simulated by European Activation System EASY-2007 code package,[1]
2. SiC/SiC thermomechanical characteristics: glass transition, Tg, and coefficient of thermal expansion, CTE have been calculated by Sciglass 6.6 (ScienceServe GmbH, Germany) for several glass compositions,
3. wettability on CVD–SiC coated SiC/SiC, measured by hot stage microscopy (Leitz GmbH AII) equipped with a Leica DBP (Ernst Leitz GMBH, Wetzlar, Germany) camera.

[1] See website: http://www.fusion.org.uk/easy2007/index.html.

The glass was synthesised by melting/quenching: the powdered raw products have been melted in a platinum–rhodium crucible at 1750 °C for 5 h in air; then the glass was poured on a brass plate and subsequently powdered and sieved.

Its thermal expansion coefficient was measured in a Perkin–Elmer TMA on cubic samples ($5 \times 5 \times 5\ mm^3$) according to standard [19]. Differential thermal analysis (Perkin–Elmer DTA 7) was used to measure the glass transition and the crystallization temperatures. Wettability tests of glass on composite substrate were performed in Ar flow in a hot stage microscopy.

Sandwich-like joined samples were produced by depositing the glass slurry at room temperature on the surface of the as received CVD–SiC coated SiC/SiC. The slurry is a mixture of glass powder (grain size 38 μm) dispersed in ethanol or in PVA. The optimized joining heat treatment was performed in a tubular oven in Ar flow at 1375 °C for 20 min and at 1235 °C for 1 h, heating rate of 1000 °C/h, resulting in a white glass–ceramic joining material.

Polished cross-sections of the joined samples were characterized by optical microscopy and scanning electron microscopy SEM equipped with EDS analyzer. The phase composition of the glass–ceramic was determined by X-ray diffraction analysis (X'Pert Philips diffractometer) using the Bragg Brentano camera geometry and the Cu Kα incident radiation.

SiC/SiC joint mechanical strength was measured at room temperature by a mechanical test machine (SINTEC D/10); four-point bending strength was calculated using

2.6 mm × 5.2 mm × 45 mm samples (sample size adapted from ASTM C1341-00). Three different kinds of SiC/SiC joined samples were produced and tested, as in Fig. 1.

3. Results and discussion

SAY (Silica–Alumina–Yttria based) glass composition was selected in order to obtain a glass material with the following properties:

- low neutron-induced radioactivity,
- CTE close to that of SiC/SiC (4×10^{-6} °C^{-1}),
- good wettability to SiC/SiC,
- feasibility of the SiC/SiC joining process.

Irradiation has been simulated by means of the European Activation System EASY-2007 code package [20], and contact dose rates have been evaluated. Irradiation conditions are those in the TAURO fusion reactor [21] first wall.

The low-activation criteria used here deal with the possibility of easily performing the recycling operations of the material after its service in the reactor. A first, more restrictive condition deals with the possibility of allowing hands-on recycling (contact dose rate lower than 10 μSv/h) of the component after an adequate cooling period (less than 100 years as a maximum). A second, less demanding condition permits remote handling recycling operations with the use of simple shielding apparatuses: in that case, the contact dose rate must be lower than 2 mSv/h, again after less than 100 years of cooling [22].

Table 1
Results of the irradiation simulation

Nuclide	Dose rate @ 50 years mSv/h	Dose rate @ 100 years mSv/h	Time to reach hands-on limit	Comments
Al	11.3	11.3	>200 years	'Condition 2' LAM[a]
B	0	0	<1 years	'Condition 1' LAM[b]
Ba	2569	291	>200 years	
C	0	0	<1 year	'Condition 1' LAM
Ca	0.045	0.035	100 years (30% conc.)	'Condition 1' LAM
Co	1.5 10^6	2100	>200 years	
Mg	0.00003	<0.00001	50 years	'Condition 1' LAM
Ni	6450	10.2	>200 years	'Condition 2' LAM[c]
Pd	1960	1800	>200 years	
Si	0.0018	0.0018	<1 year	'Condition 1' LAM
Ti	0.23	0.081	100 years (12% conc.)	'Condition 1' LAM
Y	0.0036	0.00025	40 years	'Condition 1' LAM

LAM = low-activation material. Condition 1: $D < 10$ μSv/h. Condition 2: $D < 2$ mSv/h.

[a] Maximum allowable concentration: 18%.

[b] A lot of He is generated as transmutation product.

[c] Maximum allowable concentration: 20%.

Results of the irradiation simulation are available in Table 1. According to the first condition, C, Mg, O, Si, and Y can be considered fully low-activation elements: a material with a 100% concentration of those elements reaches the hands-on limit after a reasonably short decay time, as required; therefore, there is no limit to the concentration of those elements in the material. Ca and Ti are low-activation elements too, provided that their concentration is below 30% and 12%, respectively. All the previously quoted elements can be used as oxides to form glasses. B could be considered a low-activation element too, since it reaches the hands-on limit in less than 1 year. However, under neutron irradiation, B generates a lot of He as a transmutation product, then its use must be carefully verified. On the other hand, Al, Ba, Co, Ni, and Pd and their oxides are not low-activation materials according to the first condition, and their use is restricted (see Table 1).

The first low-activation condition, however, can be hardly applicable to a plasma-facing joining material, since all the first wall and blanket materials of TAURO can reach – as a maximum result – the second condition, that is, simple remote handling recycling (2 mSv/h after less than 100 years). Therefore, it has no sense to apply the first condition low-activation requirement to the joining material only. On the other hand, the second condition low-activation requirement must be applied, to avoid compromising the low-activation properties of the whole plasma-facing component.

If we apply the second condition low-activation requirement to the joining material, we have that (besides C, Mg, O, Si, Y, Ca and Ti) also other elements can be seen as low-activation ones: in particular, Al and Ni. In fact their contact dose rate after 100 years of cooling is around 10–11 mSv/h, if 100% concentration of each element is assumed. Being the dose limit equal to 2 mSv/h, it turns out that a concentration around 18–20% in weight permits to comply with the limit: therefore, Al and Ni can be classified as low-activation elements provided that their concentration is not superior to 18–20% in weight.

According to the low-activation requirements specified in the second condition, we have selected a glass composition able to fulfill such requirements. The selected composition of the glass is: SiO_2 54 wt%, Al_2O_3 18.07 wt%, Y_2O_3 27.93 wt% (SiO_2 74.92 mol%, Al_2O_3 14.77 mol%, Y_2O_3 10.31 mol%); details on this glass are reported in Ref. [23]. CTE of the glass before the joining process, measured by TMA between 400 °C and 700 °C with heating rate of 20 °C/min, is $3{,}83 \times 10^{-6}$ °C^{-1}, close to the SiC/SiC one.

Wettability tests were performed in the hot stage microscopy to measure the contact angle and adhesion of the glass on the SiC/SiC surface. The tests showed a very good wettability of SAY on SiC/SiC (contact angle near to zero was measured at 1400 °C) and not significant gaseous production and bubbling was observed.

Time and temperature for glass deposition were chosen accordingly with DTA results obtained on bulk and on powdered glass (38 μm < size < 106 μm and size < 38 μm). The glass transition temperature, T_g, is found to be at 925 °C, the crystallization temperature is about 1220 °C and the melting point is about 1370 °C, as shown in Fig. 2: these results are in good agreement with data reported in Ref. [19]. From these data, the joining process temperature of 1375 °C was chosen in order to have a low viscosity for better glass deposition on the composite.

The dwell time for the isothermal treatment at 1235 °C was 1 h in order to obtain a glass–ceramic material: the glass can crystallize as a glass–ceramic during the joining process. Differential thermal analysis (Fig. 2) and TMA of SAY after heat treatment at 1375 °C for 20 min and 1235 °C for 1 h did not reveal any amorphous phase since

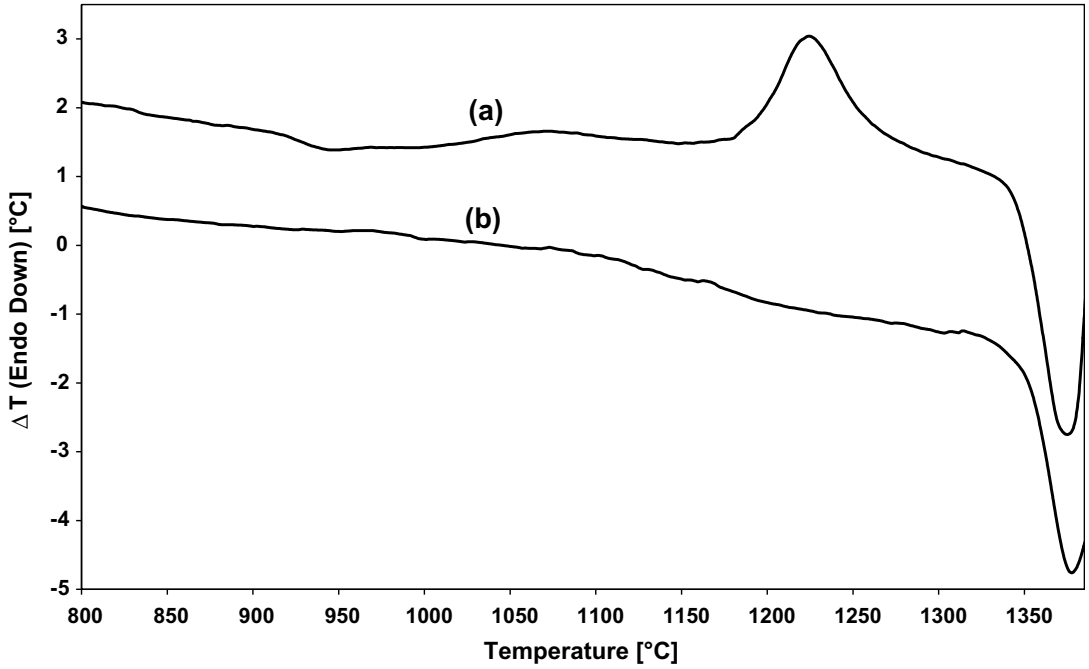

Fig. 2. Differential thermal analysis of SAY (a) glass (grain size < 38 μm) and (b) SAY glass–ceramic obtained by heat treatment at 1375 °C for 20 min and at 1235 °C for 1 h. DTA heating rate of 20 °C/min.

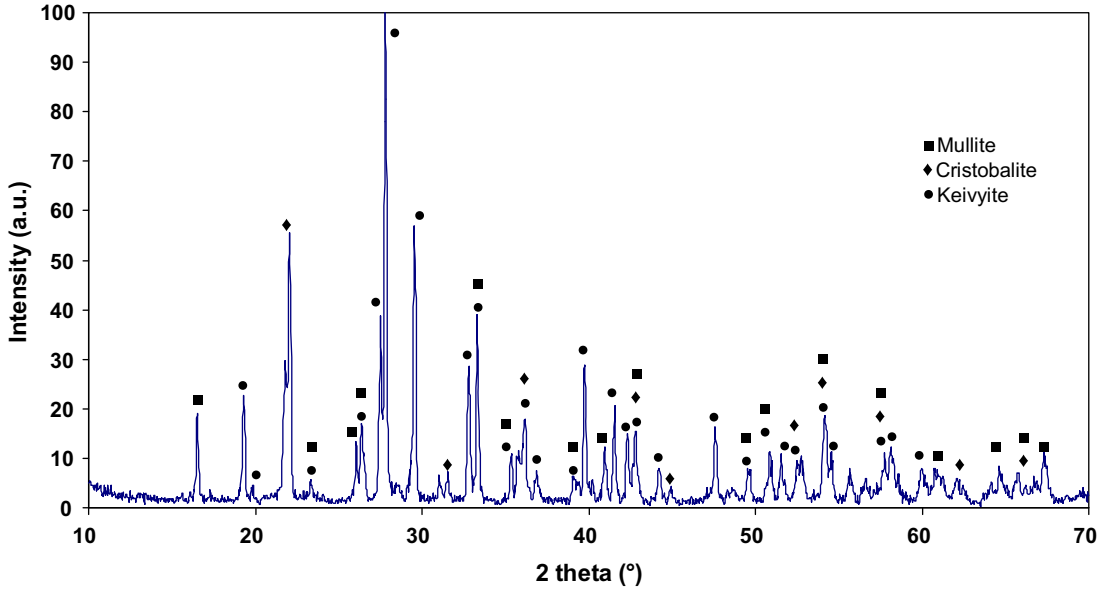

Fig. 3. XRD measurement conducted on the SAY glass–ceramic after heat treatment at at 1375 °C for 20 min and at 1235 °C for 1 h, heating rate of 1000 °C/h, in Ar flow.

no glass transition was detected (TMA max temperature was 1000, DTA max temperature was 1400 °C). A melting temperature of 1377 °C was found, according to the phase diagram (not reported here) of the system SiO_2–Al_2O_3–Y_2O_3 [24] where it can be noticed that the glass composition used here is in the eutectic transformation region of the diagram. The CTE of the glass–ceramic was 5.49×10^{-6} °C^{-1} (between 400 °C and 700 °C). The XRD of SAY after heat treatment shows three crystalline phases: cristobalite, mullite and yttrium disilicate (keivyite) no residual amorphous phase could be detected by this analysis (Fig. 3). The absence or the very low quantity (undetectable by thermal analysis and XRD) of glassy phase could be beneficial for the irradiation behavior of this material but excludes the possibility of a self-sealing behavior.

Fig. 4 shows a back-scattered SEM cross-section of a SAY joined SiC/SiC sample: grey and white zones can be observed. EDS analysis on white areas detected silicon, yttrium and oxygen, while silicon, aluminum and oxygen were found in the grey regions. This confirms the XRD analysis which reveals the presence of mullite and yttrium disilicate (keiviite).

The interfaces between CVD and SiC coated SiC/SiC and the glass–ceramic material are defect free and the joining material is homogeneous and crack-free. The average thickness of the joint is 70 μm. Some pores are detectable in the glass–ceramic layer due to gas entrapped between the joined sample; smaller voids are due to shrinkage of the glass–ceramic during sintering: their reduced size allows a dense joining material (pictures not reported here).

SiC/SiC joined samples were manufactured not only in the sandwich-like joined structure, but several geometries were tested, as reported in Fig. 1, to couple the reliability of a machined joint with sealant properties of the glass–ceramic joining material. Moreover, the joined surface is enhanced (Table 2), thus improving mechanical resistance of the joint. In Ref. [25], it is reported that a simple butt joint is not expected to fail in a tough manner, but lapped joints are required to obtain tough behavior. A sufficient number of shear surfaces must be introduced to prevent the 'unzipping' of the joint; moreover, the joint geometry must be symmetric about the centerline to avoid bending of the sample. Macrograph of joined SiC/SiC samples is shown in Fig. 1.

Mechanical characterization of the joined samples was carried out using four-point flexural test: the results are shown in Table 2 and Fig. 5.

Table 2
Four point bending test results on SiC/SiC joined by SAY glass–ceramic, three different joint configurations (thickness = 2.6 mm, width = 5.2 mm, length = 45 mm)

	Joint type (top view)	Joined area (mm^2)	Average flexural strength (MPa)
1		13.5	24 ± 2
2		39.5	122 ± 10
3		39.5	149[a]

[a] Two samples tested.

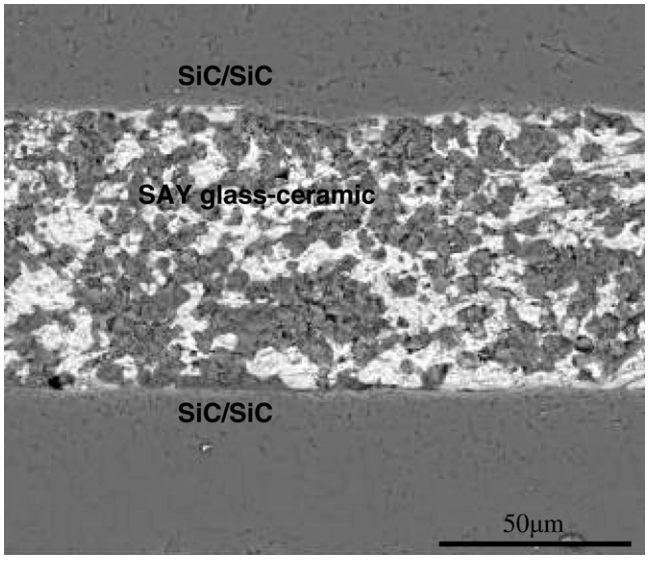

Fig. 4. Back-scattered SEM micrograph of the cross-section of a SiC/SiC joined by SAY glass–ceramic.

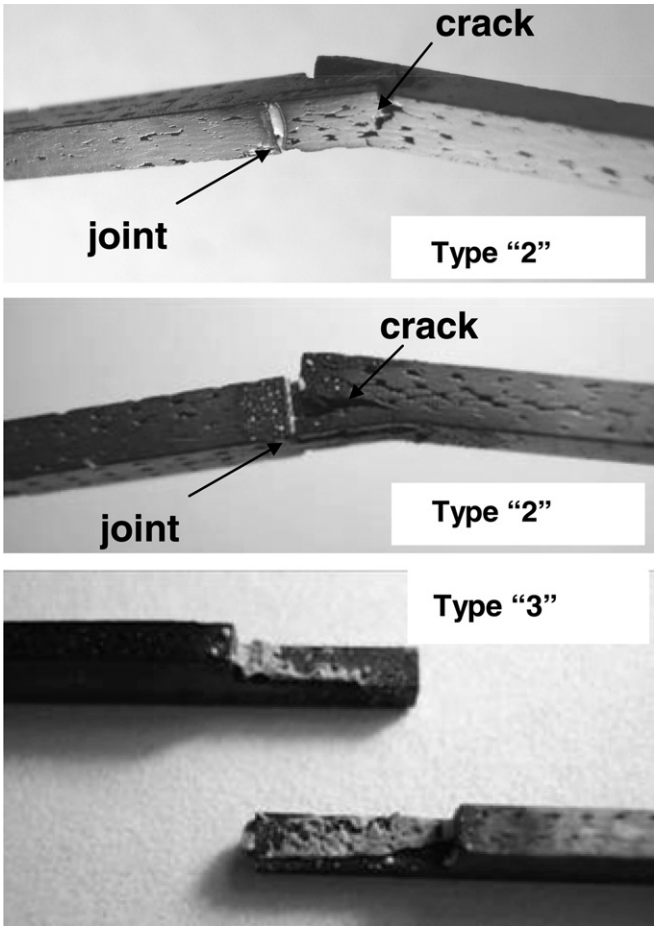

Fig. 5. Fractured samples (type 2 and 3) after four-point bending tests. See Fig. 1 for sample size.

The mechanical tests performed on joint type '1' (simple butt joint) show considerably lower flexural strength values (24 ± 2 MPa) in comparison with those obtained with joint type '2' and '3', 122 ± 10 MPa and 149 MPa, respectively. The increase of joint strength for the last two configurations can be explained by the increased joined area (39.52 mm^2 for type 2 and 3; 13.52 mm^2 for type 1). Macrographs of samples after mechanical test are reported in Fig. 5; with reference to type '2' joined samples cracks always occurred inside the composite and the joined area resulted not detached. Fracture surface analysis of samples '3' showed that on both sides it is evident the presence of the glass–ceramic material; failure occurs within the joining material.

4. Conclusions

This work proposes a low-activation silica–alumina–yttria glass to join SiC/SiC composites; the wettability and adhesion of the glass were very good on the SiC/SiC substrate. The joining process at 1375 °C leads to a glass–ceramic joining material, which is thermomechanically compatible with the composites to be joined. Three different kinds of joined samples have been manufactured to couple the reliability of a machined joint with sealant properties of the glass–ceramic joining material.

The bending strength results with the joint configuration 2 and 3 are very promising: higher shear strength than 120 MPa at room temperature were obtained. In type 2 joined samples, the fracture always occurred in the composite.

Future developments will be addressed towards the irradiation tests on joining material and joined samples.

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