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Feature Article

Joining of C/SiC composites by spark plasma sintering technique

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Abstract

CVD–SiC coated C/SiC composites (C/SiC) were joined by spark plasma sintering (SPS) by direct bonding with and without the aid of joining materials. A calcia-alumina based glass–ceramic (CA), a SiC + 5 wt% B_4C mixture and pure Ti foils were used as joining materials in the non-direct bonding processes. Morphological and compositional analyses were performed on each joined sample. The shear strength of joined C/SiC was measured by a single lap test and found comparable to that of C/SiC.

Keywords: Joining; C/SiC composites; Spark plasma sintering

1. Introduction

Ceramic matrix composites (CMC), e.g. SiC/SiC, C/SiC and C/C, are being considered as the primary candidates for components and subsystems in the field of satellite (near-sun) missions, defence, aerospace missions (e.g. body flaps, nose cones, wings, leading edges, turbine components) and for terrestrial/industrial applications under extreme environmental conditions (e.g. valves, shaft sleeves for pump sliding bearings, heat exchangers, nuclear plant components, etc.).[1–3] A critical issue for a wider use of CMC is the development of inexpensive, reliable and user-friendly joining methods to assemble large components into more complex structures.[4]

There are many possible techniques for joining CMC to themselves and to dissimilar materials: diffusion bonding;[5] transient eutectic phase methods such as nano-infiltration and transient eutectic-phase (NITE);[6] transient liquid-phase diffusion bonding;[7] pressure-less glass–ceramic joining;[8–10] solid state displacement reactions;[11] adhesive and preceramic polymer routes;[12,13] reaction forming;[14] brazing.[15] Brazing is the most commonly used joining and integration method for CMC and extensive research on brazing of CMC for their joining and integration has been done by Singh and his group at NASA.[16–18] High temperature brazing alloys are based on gold, nickel and copper and are often used for joining CMC to cobalt, titanium alloys and nickel-based superalloys.[19] TiZrNiCu, Ni and Ag–Cu–Ti have been used as filler to join C/SiC to Ti–6Al–4V.[20]

Whatever the joining process is, the joined interfaces need to be thermodynamically stable: reactions and diffusion of species from the joining material and the CMC can affect the quality of the joints and their performance in service.[21] Furthermore, the coefficient of thermal expansion (CTE) of CMC is lower than that of most metals and considerable residual stress is generated in the joint during the cooling process or when exposed to high temperature environments, thus leading to cracks or failure of the joints. In order to deal with the CTE mismatch between CMC and brazing alloys, several options have been proposed: different layers with a gradually changing CTE from CMC to the metal alloy;[22] compliant metallic layers (e.g. Cu); composite brazing alloys obtained by adding short fibres or particles to the brazing alloy.[22,23]

Selecting the optimal braze filler metal for brazing CMC to metals is not easy because the application requires a high brazing temperature (high thermal load in operation predicted), while the prevention of high joining stresses should require a low brazing temperature. Moreover, the wetting of CMC often

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requires the addition of active elements (i.e. Ti, Cr ...), but the formation of brittle intermetallic phases based on these elements must be avoided.

The CMC employed in this study consist of carbon fibre reinforced SiC matrix composites coated with a chemical vapour deposited (CVD) layer of SiC. The aim of this work is to investigate the effectiveness of using spark plasma sintering (SPS)[24] to join CMC. The SPS has recently been employed to achieve high quality joint[25].The intrinsic advantages of SPS as a joining process for CMC, especially if the materials are electrically conductive, are:

- localized heating generated by joule heating;
- low energy consumption due to both the rapid processing and the localized heating;
- limited deformation of the joined parts;
- both the rapid heating and short processing time allow highly controllable reaction of interlayer formed between the joined materials.

The effect of the electric field has been reported to enhance the diffusivity by electro migration phenomena,[26] thus promoting migration of ions trough the joining interface.

The joining by SPS represents a novelty in the field of composites. Several works on SPS as a manufacturing technique for SiC based composites have been published in the last few years.[27–31] In the field of materials joining, research has focused on metal-to-metal, ceramic-to-ceramic or ceramic-to-metal joints[32–34] using conventional techniques, but at present there is still a lack of scientific experience on joining of composites by SPS.

Direct bonding and three different types of joining materials (a metal, a glass–ceramic and a ceramic powder mixture) were tested by SPS in order to have an overview of this technique for joining C/SiC.

If the use of a joining material cannot be avoided, several options can be proposed to join C/SiC. The first choice would be using SiC as joining material in order to maintain the same thermo-mechanical properties of the composite matrix. However, both the direct bonding process and the use of SiC as joining material need high temperature, high pressure and long processing time.

In this work, SPS was used to obtain a quick and effective direct bonding between C/SiC and a sound SiC based joint by using a typical SiC sintering aid (B_4C).

The room temperature electrical and thermal conductivity of C/SiC composite is 0.5×10^4 S/m and 135 W/m K,[35] which is comparable to the constitutive graphite of the SPS mould (6×10^4 S/m and 70 W/m K). The lower electrical conductivity of C/SiC compared to graphite results in a preferential current across the SPS moulds, however according to modelling results up to 10% of the total current might still able to flow across the sample, in particular at the joined region where there is initially a contact resistance that will lead to a favourable increase in Joule heating.[36] In addition the high thermal conductivity of C/SiC compared to graphite allowed an even temperature homogenization during heating/cooling.

The choice of a $CaO–Al_2O_3$ glass–ceramic (CA) as joining material have been discussed in:[8,37,38] CA was designed to have a suitable wettability and CTE towards SiC based materials; CA can be almost completely crystallized after a pressure-less joining process, thus providing a potential high temperature resistant joining material, also suitable in a neutron environment.[8,37] SPS was used here as an alternative joining technique for CA based joints, which is able to provide a localized quick heating of the joined region.

Titanium was chosen as a joining material for C/SiC because of its refractory properties and well known reactivity with SiC. Ti–SiC system finds use in the joining of ceramics and metals because the interfacial reaction provides good bonding effects.[39] Halbig et al. developed a SiC joining technology, where titanium interlayers were used to form diffusion bonds between CVD–SiC substrates.[40]

Ti–SiC and Ti–C reactions can limit the joint thermodynamic stability and must be carefully controlled. The reaction products between Ti and SiC are very brittle with the exception of TiC and Ti_3SiC_2.[41] Furthermore, coherent phase boundaries have been found between SiC and Ti_3SiC_2, thus leading to very good adhesion of this ternary phase on the SiC.

The main purpose of this paper is to explore the influence of pressure, surface and intermediate joining materials used for bonding C/SiC by SPS. Since SPS involves rapid heating rates, a direct comparison with other bonding techniques is difficult. These results can be useful in order to define the SPS joining processing window, the intermediate joining materials and to guide future research.

In this study CVD–SiC coated C/SiC composites have been used because oxidation protected C/SiC is the final status of C/SiC for most of the applications and the coating deposition is directly performed by the manufacturer (MT Aerospace, Germany).

2. Experimental

CVD–SiC coated C/SiC composites used in this study were manufactured in disk shaped samples with diameter of 20 mm and height of 5 mm by MT Aerospace (Germany).[42] C/SiC were joined by SPS technique (SPS-HP D 25, FCT HP, Germany) using joining materials or by direct bonding. Specimens joined by direct bonding were hand polished up to 3 μm diamond slurry in order to reduce roughness and maximize the contact surface.

The as received samples were ultrasonically cleaned in acetone. Before inserting the samples in a graphite mould (hollow die, diameter of 2 cm) an interlayer was interposed between the materials to be joined. The temperature was probed by an optical pyrometer installed on the top side of the SPS machine. This configuration allowed a precise measurement of the sample temperature. According to experimental/modelling results, the temperature was measured by an optical pyrometer focused at 4 mm from the sample top surface, as detailed in Ref. [43].

All SPS joining process parameters are summarized in Table 1. All the joining surfaces were ultrasonically cleaned to remove dust particles, grease and any other contaminants.