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Joining of CVD-SiC coated and uncoated fibre reinforced ceramic matrix composites with pre-sintered Ti_3SiC_2 MAX phase using Spark Plasma Sintering

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ABSTRACT

CVD-SiC coated and uncoated ceramic matrix composites (C_f/SiC and SiC_f/SiC) were joined to their counterparts with a pre-sintered Ti_3SiC_2 foil using Spark Plasma Sintering. For the first time pre-sintered Ti_3SiC_2 foil was used as a joining filler. The joining parameters were carefully selected to avoid the decomposition of Ti_3SiC_2 and the reaction between the joining filler and the CVD-SiC coating, which would have deteriorated the oxidation protective function of the coating. Conformal behaviour of the Ti_3SiC_2 foil during the diffusion joining and the infiltration of the joining filler into the surface cracks in the CVD β-SiC coating allowed the filler to be more integrated with the matrix material. While diffusion bonding occurred during joining of the coated composites, a combination of both solid-state reaction and diffusion bonding was observed for the uncoated C_f/SiC composites. This produced the lower shear strength (19.1 MPa) when compared to the diffusion bonded CVD-SiC coated C_f/SiC (31.1 MPa).

1. Introduction

Advanced ceramic matrix composites (CMCs) based on a SiC matrix reinforced either with carbon fibres (C_f/SiC) or silicon carbide fibres (SiC_f/SiC) are materials of great interest for aerospace [1–3] and nuclear applications [4–6] due to their superior mechanical properties, resistance against high temperatures and their light weight. CMCs typically consist of straight or woven ceramic fibres embedded in a ceramic matrix with a weak bond between them resulting in the improved fracture toughness of the materials [7]. They are being used as thermo-structural materials in different fields, such as components of heat exchangers, gas turbines for power plants, heat shields for space vehicles (thermal protection system), inner wall of plasma chamber of nuclear fusion reactors, aircraft brakes, body flaps, leading edges, heat treatment furnaces, etc. [1,2]. Since the CMCs are used for applications operating at very high temperatures, they are usually coated with an outer CVD-SiC protective layer to improve their oxidation and ablation resistance [2]. In many cases, however, their application will depend on the ability to join them because the manufacture of these materials as large components with complicated shapes is extremely difficult and expensive. A critical issue of the wider use of CMCs is thus the development of inexpensive, reliable and user-friendly joining methods to assemble them as large components in complex structures [1].

In recent years, Spark Plasma Sintering (SPS, also referred to as field-assisted sintering technology (FAST)) has attracted considerable interest as a new advanced technique for joining of both monolithic SiC [8–10] and CMC materials [1,9,11]. In this case, both a rapid heating and a short processing time allow a highly controllable reaction of the interlayer with the materials to be joined. At the same time, the electric field can accelerate self-diffusion, and promote the migration of ions through the joining interface [10].

In order to develop new techniques to join advanced CMC materials that can perform in extreme environments, a critical issue is to develop new filler materials with high melting points and good oxidation resistance. A family of layered ternary materials, so-called

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"MAX" phases ($M_{n+1}AX_n$, where n is 1, 2 or 3, M is an early transition metal, A is an A-group element, and X is either C or N) have attracted increasing attention during the last decade. MAX phases exhibit a unique combination of metallic and ceramic properties, such as damage tolerance, machinability, high temperature oxidation resistance, good electrical and thermal conductivities, thermal shock resistance, and wear resistance [12]. Moreover, they exhibit large plastic deformation at temperatures above 1200 °C [13,14]. Since MAX phases also possess excellent irradiation/corrosion resistance, they are regarded as promising structural materials for fusion reactors and lead-cooled fast reactors [15]. Among all of the MAX phase materials, Ti_3SiC_2 is considered the most promising candidate structural material for nuclear and high-temperature applications [10]. More recently, Ti_3SiC_2 has attracted considerable attention as a joining filler for SiC [8–10] and CMC materials [9,11].

Dong et al. [9] used a pre-synthesized Ti_3SiC_2 powder (containing ~10 vol.% of TiC) to join both SiC and C_f/SiC materials using hot-pressing technology at different temperatures from 1300 °C to 1600 °C with a dwell time of 30 min. The flexural strength of the joints increased with increasing joining temperature and reached a maximum value of ~110 MPa for SiC joined at 1600 °C. However, at the same time, the amount of Ti_3SiC_2 decreased with increasing joining temperature, while the amount of TiC and $TiSi_2$ increased due to chemical reaction and decomposition of Ti_3SiC_2. On the other hand, no chemical reaction took place at a joining temperature of 1300 °C, but the lowest flexural strength was measured. Zhou et al. [10] joined pressureless sintered SiC with a Ti_3SiC_2 tape film (made using a commercial Ti_3SiC_2 powder) using SPS at temperatures from 1300 °C to 1600 °C with a dwell time of 5 min. Similar to the previous work, flexural strength increased with increasing joining temperature and reached a maximum value of ~99 MPa at a joining temperature of 1500 °C. Again, they suggested that interface reactions were beneficial for achieving a higher joining strength. In recent work, a Ti_3SiC_2 tape was also used to join C_f/C composites using SPS processing at temperatures between 1100 °C and 1400 °C [11]. A relatively high shear strength (~26 MPa) was measured for the components joined at 1200 °C and 1300 °C. This was attributed to the strong bonding between the interlayer and the matrix resulting from the reaction between them. The reaction was the result of a partial decomposition of Ti_3SiC_2, followed by a reaction with carbon from the C_f/C matrix to form SiC and cubic TiC at the interface. The shear strength significantly dropped when the joining temperature was 1400 °C due to almost complete decomposition of Ti_3SiC_2.

In all these works, the joining procedures relied on the reaction between the joining filler and the matrix to obtain a good strength of the joined components. However, decomposition of Ti_3SiC_2 occurring at the joining temperatures along with the chemical reactions with the matrices led to a decreasing amount of Ti_3SiC_2 in the joining area. In other words, although the Ti_3SiC_2 powders were used to join SiC and CMC materials, the best results were achieved when the amount of Ti_3SiC_2 used decreased due to both its partial decomposition and chemical reaction with SiC and C. It should also be pointed out that this reaction could cause damage to the SiC oxidation protective outer layer on the CMCs.

Although CMC materials are usually covered by a CVD-SiC oxidation protective coating for their final applications, to the best of the authors' knowledge, there has been no reported study on the joining of CVD-SiC coated CMCs with Ti_3SiC_2 MAX phase. Therefore, the aim of the present work was to develop a technique to join SiC_f/SiC and C_f/SiC composites, both coated with an oxidation protective layer of CVD β-SiC. In order to conduct a systematic study, two uncoated C_f/SiC composites with different types of carbon fibres were also joined using the same technique for the sake of comparison. Unlike the synthesized Ti_3SiC_2 powder [9] or Ti_3SiC_2 tapes [10,11], this is the first report of using pre-sintered Ti_3SiC_2 to join CMC materials via solid-state diffusion bonding. The Ti_3SiC_2 foil was pre-sintered from a synthesized Ti_3SiC_2 powder [16] (containing a small amount of impurities in the form of TiC and Ti_5Si_3) using SPS and then ground down to 80–100 μm thickness. After the SPS sintering, the amount of impurities (TiC and Ti_5Si_3) was negligible and the Ti_3SiC_2 foil containing ~4.8 wt.% Al_2O_3 was used as a joining filler. The addition of Al for the synthesis of Ti_3SiC_2 was reported to significantly decrease the quantity of the TiC impurity [16] as well as to improve the oxidation resistance of Ti_3SiC_2 [17]. Similarly, the presence of Al_2O_3 in the final synthesized Ti_3SiC_2 powder should improve the hardness, strength and fracture toughness of Ti_3SiC_2 material [16]. The advantages of using pre-sintered foil rather than powder and/or tape lie in the fact that due to the solid-state diffusion bonding (no melting of joining material) there is neither densification nor reaction required to obtain sound joints. This should rule out the possibility of undesirable shrinkage of the joining interlayer as well as its reaction with the CVD-SiC coated CMCs. Any reaction between the filler and the matrix could cause damage to the external CVD β-SiC layer, which would deteriorate the oxidation protective function of the coating. Therefore, the joining parameters were carefully chosen to avoid decomposition of Ti_3SiC_2 and the reactions between the joining filler and CVD coating. Using such an approach, sound joints with a high joining strength were obtained by diffusion bonding using SPS at a temperature as low as 1300 °C, with an external pressure of 50 MPa and a dwell time of 5 min.

2. Experimental procedure

2.1. Materials to be joined

Four different CMCs (all supplied by MT Aerospace, Germany) were joined with the Ti_3SiC_2 pre-sintered foil using the SPS. All samples were manufactured at MT Aerospace using the standard gradient Chemical Vapour Infiltration (CVI) process and supplied as rectangular shaped samples. As-received CMCs materials are summarized in Table 1. All of the CMCs materials contained characteristic natural flaws, such as macro pores between the individual fabric layers or surface cracks in the CVD β-SiC layer; see Supplementary material, S.1.

Besides Keraman® SiC_f/SiC and C_f/SiC samples coated with a protective CVD β-SiC layer, two uncoated C_f/SiC ceramic composites were joined and investigated with two main aims: (i) to investigate the influence of a SiC coating on the joining process as well as the mechanical performance of the joints; (ii) to investigate the influence of different carbon fibres on the joining process and mechanical performance of the joined components. Regarding the second point, the main difference between the carbon fibres was their different stiffness's. According to the supplier, the M40J 3K carbon fibres (tensile modulus of ~230 GPa) are much stiffer than the T300 1K ones (tensile modulus of ~135 GPa). The stiffer carbon fibres led to a greater separation of the individual fabric layers in the initial CMC materials, see Fig. S.1 in Supplementary material.

2.2. Material for joining filler

Details about the synthesis of the Ti_3SiC_2 powder used in this work and its characterisation are reported elsewhere [16]. Briefly, the powder was synthesized from the elemental powders in the molar ratio of 1.0 Ti / 1.2 Si / 0.3 Al / 2.0 TiC in flowing argon at 1200 °C for 2 h using a tungsten furnace. This powder was sintered using SPS at a temperature of 1300 °C with an external pressure of 50 MPa and a dwell time of 5 min in vacuum. The as-sintered Ti_3SiC_2 bulk samples were then ground and polished using a final diamond suspension of 3 μm to produce the thin foils with a final thickness of 80–100 μm. Afterwards, the pre-sintered foils were cut into a

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