Flash joining of CVD-SiC coated Cf/SiC composites with a Ti interlayer

Journal of the European Ceramic Society 37 (2017) 3841–3848

Contents lists available at www.sciencedirect.com

Journal of the European Ceramic Society

journal homepage: www.elsevier.com/locate/jeurceramsoc

Flash joining of CVD-SiC coated C_f/SiC composites with a Ti interlayer

Peter Tatarko[a,b,c,*], Salvatore Grasso[a,*], Theo G. Saunders[a], Valentina Casalegno[d], Monica Ferraris[d], Michael J. Reece[a]

[a] School of Engineering & Materials Science and Nanoforce Technology Ltd., Queen Mary University of London, Mile End Road, London E1 4NS, United Kingdom
[b] Institute of Inorganic Chemistry, Slovak Academy of Sciences, Dúbravská cesta 9, 845 36, Bratislava 45, Slovakia
[c] Institute of Materials Research, Slovak Academy of Sciences, Watsonova 47, 04001 Košice, Slovakia
[d] Politecnico di Torino, Department of Applied Science and Technology, Corso Duca degli Abruzzi 24, 10129 Torino, Italy

ARTICLE INFO

Article history:
Received 12 April 2017
Received in revised form 4 May 2017
Accepted 10 May 2017
Available online 26 May 2017

Keywords:
Flash joining
Diffusion bonding
Spark plasma sintering
C_f/SiC composites
Ti interlayer

ABSTRACT

Flash joining of CVD-SiC coated C_f/SiC samples with a Ti interlayer was achieved using a Spark Plasma Sintering machine. The influence of different heating powers and discharge times were investigated. The sample flash joined at a maximum heating power of 2.2 kW (peak electric current of 370 A) within 7 s showed the highest apparent shear strength of 31.4 MPa, which corresponds to the interlaminar shear strength of the composites. A maximum joining temperature of ~1237 °C was reached during the flash joining. An extremely rapid heating rate of 9600 °C/min combined with a very short processing time hindered any reaction between the CVD-SiC coating and the Ti interlayer. The formation of a metallic joint (Ti based) in the absence of any detectable reaction phase is proposed as a new joining mechanism. For a conventionally joined SPS sample, the formation of titanium silicide phases inhibited the formation of a bond.

1. Introduction

Ceramic matrix composites (CMCs) with a SiC matrix reinforced with carbon fibres (C_f/SiC) and silicon carbide fibres (SiC_f/SiC) are materials of great interest for aerospace and nuclear applications. These materials possess superior mechanical properties, resistance against high temperatures and low density. CMCs are usually coated with an outer SiC protective layer to improve their oxidation and ablation resistance as they are used in extreme environments, involving high temperatures, mechanical stresses and oxidative atmospheres [1,2]. However, their manufacture as large components with complicated shapes is extremely difficult and expensive. Therefore, the application of CMCs depends on the ability to join them in order to assemble large components in complex structures [2–4]. In many cases, CMCs are required to be joined with metals, especially to Ti alloys for aerospace applications [5].

A number of techniques for joining of CMCs to themselves or dissimilar materials (e.g. metal components) have been developed. This includes brazing [6], diffusion bonding [7], transient eutectic phase routes [8], glass-ceramic joining [9], adhesion [10], pre-ceramic polymer routes [11], MAX phase bonding [2], and mechanical fastening [12]. These techniques are based on pressureless or pressure assisted technologies, which normally require high temperatures and relatively long dwell times, limiting their industrial applications.

Electrical current assisted processes have been developed for more than a century and they have been used to consolidate particles or analogously bond together bulk materials. The first use of resistance welding might go to James Prescott Joule when he introduced the Joule heating effect (1840), and later in 1856 when he attempted the first arc welding experiments by passing currents through various metal wires surrounded by charcoal [13]. Elihu Thomson is considered to be the father of resistance welding with his work published in 1886 [14]. As stated: “All that was required was a transformer with a primary to be connected to the lighting circuit and a secondary of a few turns of massive copper cable”. He perfected the process into what is now known as resistance welding [15]. In resistance welding, the heat in the components to be joined is generated by the electric current flow. To facilitate the formation of a strong bond and good electrical contact, uniaxial forces are employed.

* Corresponding authors at: School of Engineering & Materials Science and Nanoforce Technology Ltd., Queen Mary University of London, Mile End Road, E1 4NS, United Kingdom.
E-mail addresses: peter.tatarko@savba.sk, tatarko.peter@gmail.com (P. Tatarko), salpolito@hotmail.it (S. Grasso).

http://dx.doi.org/10.1016/j.jeurceramsoc.2017.05.016

Early works on electric current assisted sintering (ECAS) have been reviewed in Refs [16,17]. The most recent evolution of ECAS is Flash Sintering that allows the densification of particulate materials in a very short time (<60 s) under the application of an electric field due to the localised Joule heating developed within the consolidating particles using a die-less configuration [18]. So far, most of the resistive joining/welding works using a very rapid processing (discharge time <10 s) have been limited to metals or cermet. This is because: i) some of the ceramics may require sufficient preheating to become conductive enough to allow suitable flow of electric current and activate the thermal runaway process seen in semiconductors; ii) ceramics may not tolerate the thermal shocks as in the case of resistive welding of metals. Therefore, most of the works to join ceramics in the presence of an electric current have applied low voltages (1–10 V) and high electric current through electrically conductive graphite dies with the specimen under a mechanical pressure using Spark Plasma Sintering (SPS) apparatus. The electric current heats graphite dies and the ceramic pieces to be joined are subsequently heated indirectly through the graphite dies with resulting processing times of the order of few tens of minutes. Contrary to the previously developed techniques to join ceramics, both a rapid heating and a short processing time allow an accelerated bonding process. SPS processing has recently been successfully used to join both monolithic SiC [19–22] and CMCs [2,7,23]. However, the resulting processing time was still of the order of few tens of minutes rather than a few seconds as in the case of flash processing.

The purpose of this work is, for the first time, to bring together the flash processing and the joining of ceramic materials, thus allowing flash joining of ceramics. The inherent advantage of flash processing is the rapid temperature transition that would limit degradation of materials susceptible to thermal degradation, such as C_f/SiC composites. A pair of C_f/SiC materials was successfully joined with a Ti foil using a SPS machine with a discharge time of less than 10 s.

2. Experimental procedure

2.1. Starting materials

Keraman® C_f/SiC composites containing T300 1 K (Torayca®) carbon fibres were manufactured at MT Aerospace (Germany) using the standard gradient Chemical Vapour Infiltration (CVI) process and supplied as disc shaped samples with a diameter of 20 mm and a thickness of 2 mm. After the gradient CVI process, all sides of the samples were coated with a protective CVD β-SiC layer. After coating, the nominal density of the samples was 2.00 ± 0.1 g/cm^3.

High purity Ti foil (99.99%) with a thickness of 130 μm (Goodfellow, UK) was used as a joining material. The foil was cut into the discs with a diameter of 20 mm, to match the diameter of the C_f/SiC samples.

2.2. Flash SPS joining process

The C_f/SiC materials as well as the titanium foil were ultrasonically cleaned in acetone before joining. The Ti foil was interposed between two C_f/SiC samples as a sandwich and the whole assembly wrapped by a graphite felt. As usual, graphite felt was used to thermally insulate the sample, but in this case to allow preheating of the sample as well. The sample wrapped in the graphite felt was then placed between two graphite punches with a diameter of 30 mm and a constant uniaxial force of 5 kN (16 MPa) was applied throughout the joining process. The flash joining process was performed using a SPS machine (HPD 25/1, FCT systems, Germany). Due to difficulties in measuring the actual sample temperature during the flash joining process, the process was controlled by limiting the power to a preset value and the joining process was terminated by interrupting the electric power after a certain time was reach. The samples were discharged under a peak power of 2–7 kW. Three samples were discharged using a relative power limit of 80% and the process was interrupted at different times for each sample (22 s, 19 s, 12 s). Two samples were discharged using a power limit of 60% and the process was interrupted after 13 s and 7 s, respectively. After the best joining conditions were identified, the joining process was repeated twice to investigate reproducibility of the process. The same joining parameters were then also applied for samples with a K thermocouple (Ø 1 mm) inserted into the joining interface (up to ~2 mm from the edge of the sample) in order to measure maximum joining temperature of the process. In addition, in this case a data logger (National Instruments USB-6221, UK) was used to obtain a more accurate measurement of the electric current. The schematic illustration of the joining set-up and the heat dissipation during the joining process with the individual parameters for each step is shown in Fig. 1.

Apart from the flash joining process, a joining assembly was also heated up to 1250 °C using a standard SPS die set (the samples were placed in a graphite die) with a heating rate of 100 °C/min, and then immediately cooled down without any dwell time at the maximum temperature. This experiment was performed to compare the chemical composition of the Ti interlayer after the flash joining and SPS joining processes with the similar maximum joining temperatures.

2.3. Characterisation of the joints

The polished cross-sections of the joined areas and fracture surfaces after the mechanical tests were characterized using a Scanning Electron Microscopy (SEM) FEI Inspect-F equipped with Energy Dispersive Spectroscopy (EDS) detector.

X-ray diffraction (XRD) patterns of the Ti foils adhered to the surface of C_f/SiC were obtained on the fracture surfaces after the mechanical tests using an X-ray diffractometer (Siemens D5000 using Cu Kα radiation).

Each of the joined discs was cut into two cuboids with a cross sectional joining area of 90 mm^2 (10 × 9 mm; middle part of the disc) and 30 mm^2 (10 × 3 mm), respectively. The mechanical strength of the joined samples was evaluated using a single lap offset shear test. The tests were performed at room temperature with a compression machine (SINTEC D/10) with a cross-head speed of 0.5 mm/min, according to method adapted from ASTM D1002-05 standard. Copper bars were used as an artificial step on the joined C_f/SiC rectangles to enable the single lap offset configuration, as schematically shown in our previous work [2]. The maximum force was then divided by the joining area (90 or 30 mm^2) to calculate the apparent shear strength of the joints.

3. Results and discussion

Table 1 summarises all of the flash joining parameters (SPS data) used for the individual samples. The samples Nos. 6 and 7 were joined using similar parameters as those for the sample No. 5. On the other hand, the sample No. 8 was joined using the same parameters, but in this case the temperature and electric current were monitored using a Ø 1 mm thermocouple and an external logger, respectively. It is believed that a heat dissipation during the flash joining process in the SPS machine was the same as occurred during flash SPS sintering of β SiC [24]. This suggests that, as schematically shown in Fig. 1, in all cases the sample was probably preheated through the surrounding graphite felt as the electric current passed the punches and the side felt at the beginning of process due to their

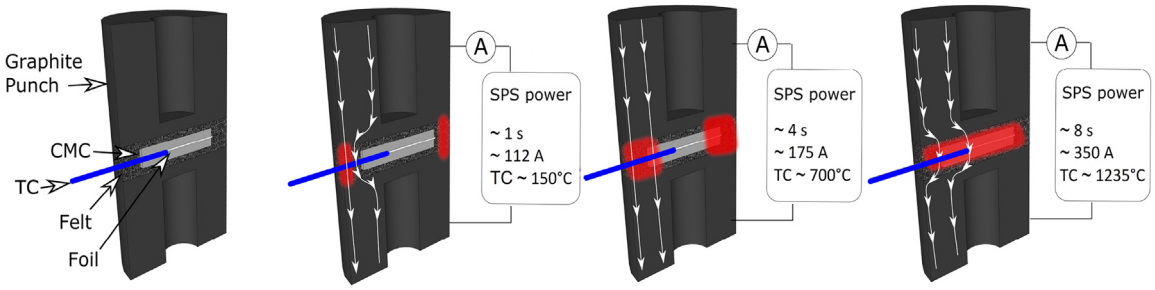

Fig. 1. Schematic illustration of the flash joining process using SPS with a Ø 1 mm thermocouple embedded in the joining interface. In each figure the qualitative temperature profile is sketched, and corresponding electric current, temperature (TC) and time are also provided.

Table 1
Summary of the SPS data recorded during the flash joining of C_f/SiC samples with a Ti interlayer. Summarizing remarks on the quality of the corresponding joints are also listed.

Sample No.	Relative Power limit [%]	Max. heating power [kW]	Max. current [A]	Time [s]	Final resistance [mOhm]	Remarks
1	80	7.3	1110	22	5.9	SiC heavily damaged
2	80	6.8	1030	19	6.4	SiC heavily damaged
3	80	5.7	850	12	7.9	SiC damaged
4	60	3.4	570	13	10.3	SiC damaged
5	60	2.2	370	7	16.2	Strong joint
6	60	2.0	310	6	21.2	Weak joint
7	60	2.0	320	7	19.1	Weak joint
8*	60	2.1	340	7	18.0	TC inserted in the interface

*An external Ø 1 mm thermocouple and a data logger were used for this sample to more accurately monitor both the temperature and the electric current, respectively (the results are shown in Fig. 2).

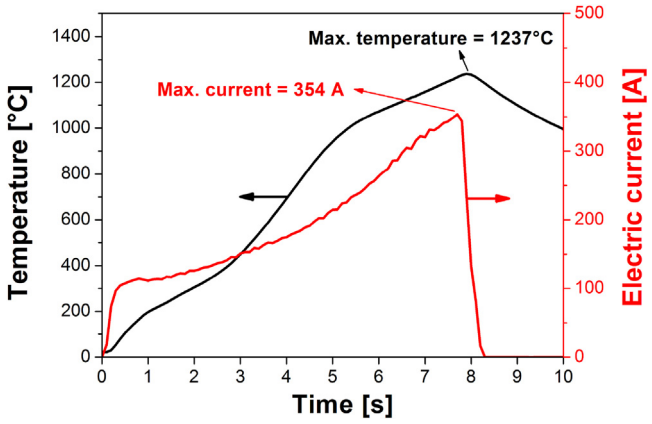

Fig. 2. The temperature and electric current measured during the SPS flash joining process of the C_f/SiC sample No. 8 using a Ø 1 mm K thermocouple and the external data logger, respectively (not SPS data). The heating rate was determined to be ~9600 °C/min (160 °C/s), while the cooling rate from the maximum temperature was ~6000 °C/min (100 °C/s).

lower resistance compared to the C_f/SiC joining couple. According to the data logger results for sample No. 8 (Fig. 2), this might have happened after approximately 0.5 s from the start of the discharge. At this point, the electric current was starting to stabilize at an almost constant value of ~112 A for a very short time (~0.5 s), see Fig. 2. The current then continued increasing, and a more significant increase of the current was observed after ~4 s. It is believed that at this point the current started passing through the sample, as the sample became conductive enough at this temperature (~700 °C; ~175 A). This is in accordance with the previous work of Grasso et al. on flash sintering of α and β-SiC who reported that β-SiC became conductive enough at 500–600 °C during the flash sintering [24]. The CMCs investigated in the present work contained a significant amount of β-SiC, which is the reason why similar behaviour could be expected. As the flash joining process continued, the resistance of the sample decreased rapidly, and at this point there was maximum heat dissipation in the sample as its resistance was significantly lower than the resistance of the felt. This allowed the sample to be heated up to a sufficient temperature to enable diffusion bonding of the C_f/SiC samples with the Ti interlayer. In the final stage, the resistance of the samples further decreased with the increasing processing time, while the maximum current increased (Table 1).

The external logger allowed us to monitor electric current more precisely than the SPS machine due to its faster response (0.1 s versus 1 s for the logger and the SPS, respectively). A maximum current of 354 A was recorded using a data logger after 7.7 s from the start of the discharge, which resulted in a maximum joining temperature of 1237 °C (Fig. 2). This current was slightly higher than the current recorded by the SPS machine for the same sample (340 A; see sample No. 8 in Table 1). This was caused by the fact that both the heating power and the sample resistance changed rapidly during the flash joining process; see Fig. 3 for both 80% and 60% of power limit. Therefore, even a slight difference in the processing time (fraction of second) had a significant effect on the final measured heating power and the resistance of the samples. Fig. 3 also shows that the sample resistance decreased more rapidly when the higher power limit was used. The drop in resistance is due to the semiconductor behavior of β-SiC [24]. As shown in Fig. 2, the average experimentally recorded heating rate was ~9600 °C/min (160 °C/s), while the cooling rate from the maximum temperature was around 6000 °C/min (100 °C/s). At present, the SPS machine employed is able to control the set program within a unit of seconds. The rapid variation of resistance/temperature made it difficult to precisely control the maximum output power, which was in the range of 2–2.2 kW. This caused a variability of the final resistance/temperature for the samples No. 5–8.

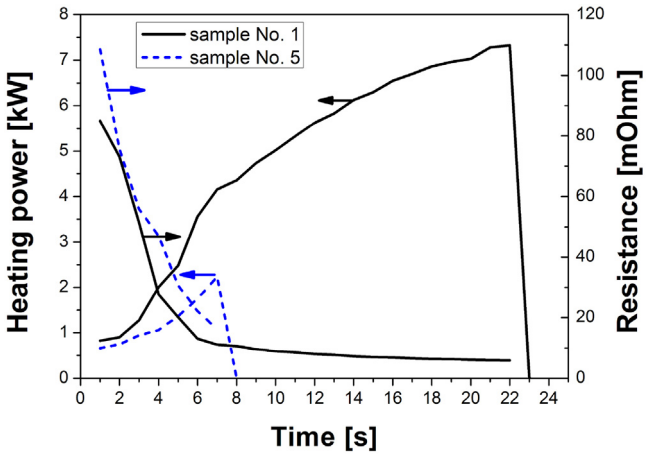

Fig. 3. The heating power and the resistance of the C_f/SiC samples recorded during the flash joining process in the SPS machine for different power limit used (80% versus 60% for the sample No. 1 and the sample No. 5, respectively). These results were recorded by the SPS machine.

3.1. Cross-section analysis of the flash joined C_f/SiC samples with a Ti interlayer

In all cases, the samples were joined after the joining process, and survived cutting and polishing. However, a significant thermal degradation of SiC resulted in the sublimation of SiC in the samples joined using a relative power limit of 80%. Moreover, melting accompanied by a significant infiltration of Ti into the pores of both halves of the C_f/SiC composites was also observed. Similar features were observed for all the samples joined using the same power limit of 80%, the only difference was in the extent of the damage (for details, see Supplementary material, S.1).

Therefore, for the further investigations the relative power limit was reduced to 60% and the joining process was interrupted after either 13 or 7 s. Fig. 4 shows back-scattered SEM images of the polished cross sections of these samples. Despite lowering the power limit, a heating power of 3.4 kW (electric current of 570 A) was still slightly higher than the optimum value when the joining process was interrupted after 13 s. This resulted in the joining temperature being sufficiently high to melt the Ti foil (T_m 1668 °C), which then infiltrated into the pores of the C_f/SiC (Fig. 4a). The infiltration led to a significant shrinkage of the joining interface and a porous interlayer (Fig. 4b). On the positive side, no decomposition of either the CVD-SiC or SiC in the matrix was found in the cross sections (no formation of nanopores in the SiC). The interface between the Ti interlayer and the CVD-SiC coating was not smooth, but there was some sign of nanoinfiltration of Ti into the CVD-SiC. This suggests that there were some reactions between the Ti and CVD-SiC, followed by a slight dissolution of CVD-SiC and infiltration of Ti over a small distance from the joining interface.

On the other hand, a lower heating power (with a maximum electric current of 370 A) was reached after a processing time of 7 s; and this resulted in a uniform, crack/pore-free joining interface (Fig, 4c and 4d). According to Fig. 2, these parameters led to a final joining temperature of ~1250 °C. In this case only, the final thickness of the Ti interlayer matched the initial thickness of the Ti foil (~130 μm). This confirms that the joining temperature was not high enough to melt the Ti (T_m 1668 °C) foil and no long-distance infiltration of Ti was therefore observed. Moreover, there was no sign of any reaction between the Ti foil and the CVD-SiC. The interface between these two was without any visible reaction zone or nanoinfiltration of Ti into the CVD-SiC (Fig. 4d). On the contrary, the joining temperature was high enough to allow plastic deformation of the Ti foil to conform to the rough surface of the CVD-SiC (Fig. 4c and d), and/or to fill the surface cracks that are always present in CVD-SiC coatings on C_f/SiC composite (due to the different thermal expansion coefficients of the coating and the matrix). The cracks were filled up by the Ti interlayer due to the so-called hot forging effect (see the inset in Fig. 4c). On the other hand, despite a difference between thermal expansion coefficients of Ti and CVD-SiC, neither parallel nor perpendicular cracks were observed in the Ti interlayer (Fig. 4c and d). The most plausible explanation of the absence of these cracks is that the metal was still ductile and the stresses were distributed within the material forming dislocations. Further work is required to confirm this hypothesis. The present work indicates that, when the right processing conditions are applied, a solid-state diffusion bonding could be obtained within 7 s during the flash joining of C_f/SiC composite with the Ti foil at a maximum temperature of ~1250 °C ± 100 °C.

Table 2
Apparent shear strength of the C_f/SiC samples joined with a Ti interlayer within ~7 s using SPS flash joining with a maximum current in the range of 310–370 A. Samples labeled as *a* were cut from the middle of the joined discs while the sample labeled *b* were cut from the edge of the joined discs.

Sample No.	Apparent shear strength [MPa]
5*a*	31.4
5*b*	18.1
6*a*	3.3
6*b*	N/A*
7*a*	8.4
7*b*	N/A*

*N/A – non-applicable; the sample delaminated during testing shortly after a load was applied.

3.2. Apparent shear strength of the flash joined C_f/SiC samples

The apparent shear strength was not measured for the samples with significant damage detected by the SEM analysis of the cross sections in the previous steps. Therefore, the shear strength was measured for the samples Nos. 5–7. Two cuboids were cut out of each disc after joining; one from the middle of the disc (labeled as *a*) while the other one from the area close to the edge of the disc (labeled as *b*). The other parts of the discs were used for both SEM and XRD analysis.

The results of the apparent shear strength measurements (Table 2) show two important facts: i) in all cases, the middle part of the flash joined samples was stronger than the edge part (*a* versus *b*); ii) strength was strongly affected even by a very slight change in the maximum current and/or the sample resistivity. The first point reveals that the joining temperature was not homogenous throughout the sample. This is in agreement with modelling results presented in Ref. [24], where the sintering temperature was not homogenous in a radial direction and a maximum temperature was localised in the centre of the samples because the heat was radiated radially. In addition, this also means that the maximum joining temperature in the middle of the joints might have been slightly higher than that detected by the thermocouple (1237 °C), as this was inserted right into the interface up to ~2 mm from the edge of the joining couple. A higher temperature in the middle of the joints resulted in greater plasticity of the Ti foil, which allowed the foil to be more conformed to the C_f/SiC surface, allowing the filling up of the surface cracks (Fig. 4c). This then led to a higher shear strength of the joints measured for the middle part of the flash joined discs. Regarding the second point, although all three samples were flash joined in approximately the same time (~7 s) even a slight difference in time (fraction of seconds) led to a rather significant difference in the final heating power and the sample resistance reached. The highest apparent shear strength (31.4 MPa) was measured for the middle part of the sample flash joined with

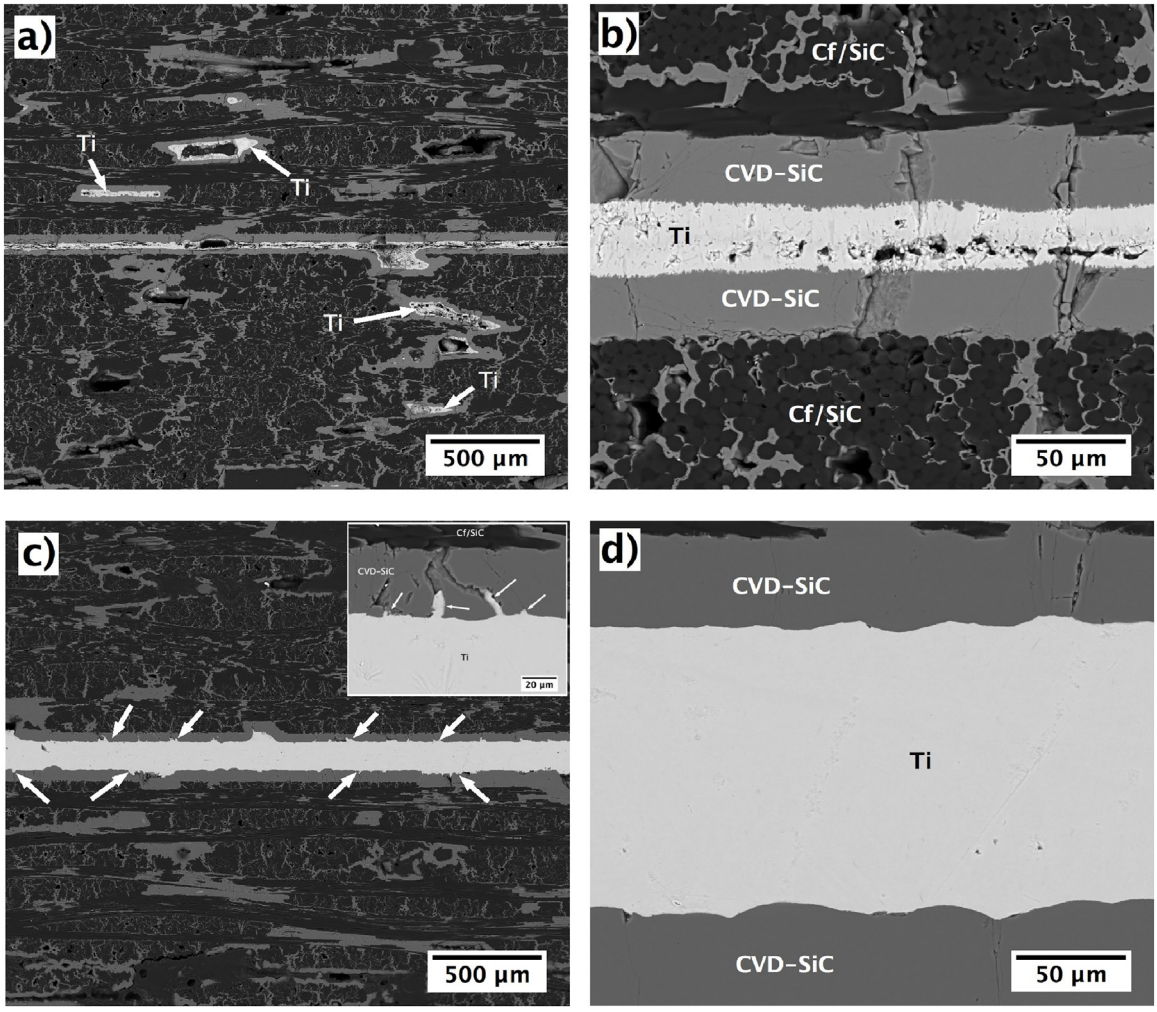

Fig. 4. Back-scattered SEM images of the polished cross sections of the flash joined C_f/SiC with a Ti interlayer using a power limit of 60% for: a), b) 13 s; c), d) 7 s. A porous interlayer with some reaction between the Ti foil and CVD-SiC and infiltration of Ti into the pores of C_f/SiC were found after 13 s. On the other hand, a uniform, crack or pores-free joining interface was defected after 7 s (with a maximum current of 370 A). A diffusion bonding with the filled up surface cracks of the CVD-SiC coating on the C_f/SiC was observed (shown by arrows in Fig. c) and in the inset).

the maximum electric current/power and the lowest resistivity among all of the three investigated samples (sample No. 5: 2.2 kW, 370 A, 16.2 mOhm). On the other hand, two other samples that were joined using the flash-SPS process with a lower heating power (2.0 kW) showed a significantly lower apparent shear strength. In both cases, the Ti foil completely delaminated from one side of the coated C_f/SiC, while it remained strongly attached to other side (bottom piece of the SPS joining assembly); this was most likely caused by the Peltier effect, which produces an asymmetric temperature distribution with respect to the sample mid-thickness plane described in Ref. [25]. The poor strength of these two joints was further confirmed by the fact that the pieces cut from the area closer to the edge of the joined disc delaminated shortly after a load was applied. The lowest strength was measured for the sample flash joined using the lowest electric current (310 A), resulting in the highest resistivity (21.2 mOhm) among all three samples. This all indicates that the final strength of the joints was significantly affected by the maximum current/heating power reached. The higher the heating power the higher the temperature and the lower the resistivity of the samples.

Unlike a complete delamination of the Ti foil from one part of the C_f/SiC observed for the samples Nos. 6 and 7, a mixed crack propagation was observed after the mechanical test of the sample No. 5, i.e. a crack propagated along the Ti/CVD-SiC interface and through the C_f/SiC composite (Fig. 5). Fig. 5a shows that the fracture surface is partially covered by the CVD-SiC coating with a Ti layer, while a significant part of the fracture surface shows typical interlaminar composite failure. In addition, Fig. 5b shows a higher magnification of the fracture surface area shown by the rectangle in Fig. 5a. The conformal behaviour of the Ti foil can be seen as the morphology of the Ti foil reproduced the rough surface of the CVD coating. Interestingly, after the failure the Ti foil remained strongly attached to the CVD-SiC coating in the valleys of the rough surface, while it was partially detached from the other parts. Fig. 5b also shows that the CVD coating was cracked (shown by arrows) during the testing and the presence of a Ti layer had no effect on the crack propagation. It can be concluded that the flash joining parameters, such as the heating power of 2.2 kW, and the electric current of 370 A, were the optimal parameters to obtain a sound joint. Using the SPS apparatus and the joining set up as shown in Fig. 1, such conditions could be reached in ~7 s. The corresponding joining temperature was ~1250 °C. When a lower heating power was used (samples Nos. 6 and 7) the maximum joining temperature was not sufficient to enable a strong diffusion bonding. On the other hand, when a higher heating power was used (samples Nos. 1–4), the corresponding maximum temperature was too high, resulting in melting of the Ti and a significant infiltration of Ti into

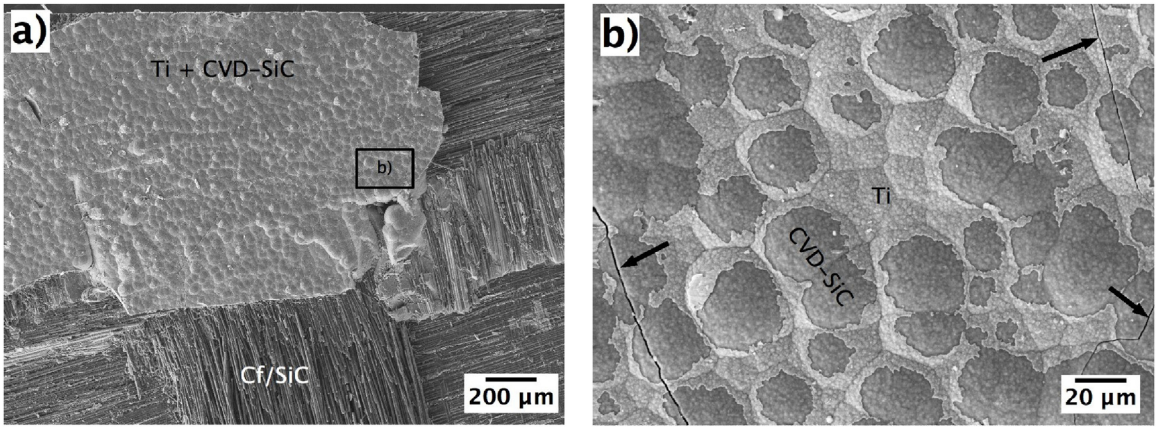

Fig. 5. SEM images of the fracture surface of sample No. 5, the flash joined C_f/SiC with a Ti interlayer using a heating power of 2.2 kW (an electric current of 370 A) for 7 s. a) A mixed crack propagation (along the interface and through the composite) was observed on the fracture surface. b) A higher magnification of the fracture surface area highlighted by the rectangle in a. After the mechanical test, the Ti foil was strongly adhered to the SiC-CVD coating in the valleys of the rough surface. The CVD coating was cracked during the testing as shown by arrows in b).

the pores of C_f/SiC composites, as well as a decomposition of the CVD-SiC coating and SiC matrix (see Supplementary material, S.1).

3.3. *Flash joining mechanism and the flash joining potentials*

It is well known that a Ti/SiC interface is thermodynamically unstable and reacts at elevated temperatures (~600 °C) to form a diffusion reaction zone that is dominated by the presence of titanium silicides (mainly Ti_5Si_3), titanium carbide (TiC) and, under certain conditions, a ternary phase (Ti_3SiC_2) [26,27]. It was reported that Ti reacts with SiC even at a temperature as low as 700 °C if they are in contact for a sufficiently long time (71 days) [26]. Recently, Yang et al. showed that Ti reacted with pressureless sintered SiC to form TiC even during SPS joining at a temperature of 600 °C for 5 min [27]. They reported that the reaction always starts with the formation of TiC, followed by the formation of Ti_5Si_3 (>900 °C) and finally Ti_3SiC_2 (>1000–1100 °C), depending on the joining temperature. Similarly, the formation of titanium silicide ($TiSi_2$), TiC, and Ti_3SiC_2 was found in the interlayer after joining of C_f/SiC samples with a Ti foil using the SPS process at 1700 °C for 3 min [7]. In addition, multicomponent phases (TiC, $Ti_5Si_3C_x$, and Ti_3SiC_2) were formed in the Ti interlayer after the reactions with the SiC matrix during hot pressing at the temperatures between 1400 and 1600 °C in the SiC-Ti-Re sandwich joints [28]. In all cases, the presence of the reaction phases was easily detected by the different contrast on the back-scattered SEM images and/or EDS and XRD analysis.

Contrary to these previous works, no obvious reaction layer was found on the cross section of the flash joined C_f/SiC with a Ti interlayer within 7 s (Fig. 4d). The XRD pattern of the Ti foil adhered to the surface of coated C_f/SiC after the mechanical test of the sample flash joined in 7 s (No. 5) is shown in Fig. 6. When compared to the fracture surface of the composite (second pattern in Fig. 6), the only difference was the presence of Ti (besides hexagonal/graphitic carbon and cubic β SiC from the matrix). No other secondary phases (no possible reaction products) were found in the Ti interlayer. In addition, the EDS analysis performed on the same sample (EDS elemental maps are shown in Supplementary material, Fig. S.2) confirmed there was no significant diffusion of the elements and/or any visible reaction layer at the joining interface. Nevertheless, the EDS point analysis revealed a small amount of Si (~0.30–0.40 wt.%) in the Ti interlayer very close (up to 15 µm) to the interfaces with the CVD-SiC coating of a pair of the C_f/SiC joined together. Similarly, a small amount of C (~5.00 wt.%) was detected uniformly across the Ti interlayer (this must be considered with caution as EDS is not a reliable technique to detect such a light element). This clearly confirms some diffusion of both Si and C into the Ti interlayer during the flash joining, but the concentration of these two was not high enough to enable the formation of a reaction zone or phases. This all indicates that the coated C_f/SiC samples were joined with the Ti foil in a very short time (~7 s) via solid state diffusion bonding and the Ti interlayer did not react with the CVD-SiC coating of the CMCs.

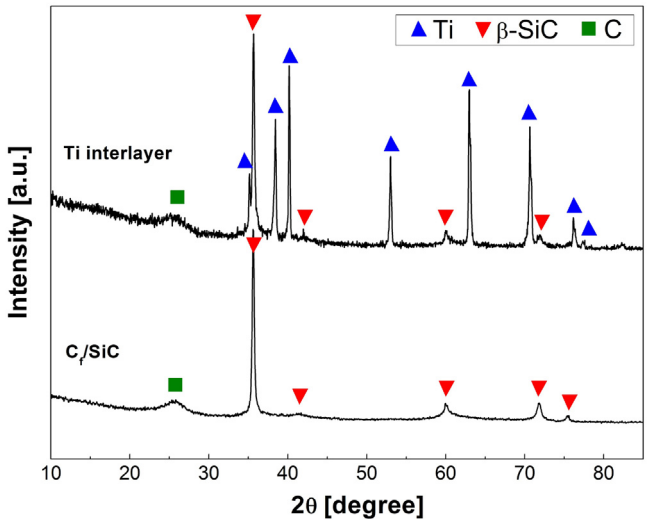

Fig. 6. XRD patterns of the Ti interlayer adhered to the fracture surface of the flash joined C_f/SiC using a heating power of 2.2 kW for 7 s (sample No. 5) and the fracture surface of the same composite without the Ti layer adhered to the surface. Except Ti, no other secondary phases were detected in the Ti interlayer (no evidence of the possible reaction products). JCPDS cards No.: Ti (hexagonal, 01-089-4893); β-SiC (cubic, 01-075-0254); C (hexagonal, 00-008-0415).

A different situation occurred when the same joining assembly (the Ti foil interposed between the two C_f/SiC samples) was heated to the same corresponding temperature of 1250 °C using standard SPS processing with a heating rate of 100 °C/min. Unlike the die-less flash SPS process, this time the sample was placed in a graphite die and an external load of 5 kN (16 MPa) was applied through the graphite punches. Similarly to the flash joining, the sample was cooled down immediately after reaching the final temperature without any dwell time. It must be pointed out that the samples were not joined after this SPS joining process, and the Ti interlayer delaminated from both parts of the coated C_f/SiC samples. The SEM analysis of the cross-section of the delaminated Ti

foil revealed a layer of titanium silicides (up to ~10 μm from the edge) present in the Ti interlayer at both interfaces adjacent to the CVD-SiC coating of the C_f/SiC composite. The SEM images of the cross section along with the results of EDS analysis are given in Supplementary material, Fig. S.3 and Fig. S.4. This result of the SPS joining experiment confirmed that when heated relatively slowly (~100 °C/min), the Ti interlayer reacts with the CVD-SiC coating at a temperature as low as 1250 °C despite the fact that there was no dwell time at the highest temperature.

On the other hand, when the same CVD-SiC coated C_f/SiC samples were joined with a Ti interlayer using flash joining, the diffusion of both Si and C into the Ti interlayer was suppressed due to both a rapid heating rate (~9600 °C/min) and a rapid cooling rate (6000 °C/min). This avoided an oversaturation of Si and C in the Ti interlayer to form either silicides or carbides or ternary phases. The fact that Ti remained Ti, with the absence of any possible reaction products that would have prolonged the necessary time for diffusion bonding, was the main reason why such a metallic kind of bonding was obtained in such a short time by flash joining. Our analysis was mostly done on the length scale comparable with the foil thickness and the temperature was probed using a Ø 1 mm thermocouple. The interfacial electrochemical phenomena occurring between TiO_2 covering the foil and SiO_2 covering the SiC are not understood in the present work and further investigation is needed. A simplified hypothesis is that the oxides phases might have been electrochemically reduced by the application of an electric field [18]. The electrochemical reduction might have been also enhanced by the preferential heating occurring at the interfaces in combination with the presence of carbon in SiC.

The demonstration that a sound joint can be obtained for C_f/SiC within 7 s using the SPS flash joining process (current passes through sample) is the main result of the present work. The SPS flash joining process as developed and outlined in this work has some difficulties in reproducibility and obtaining a homogenous temperature throughout the joining assembly. However, this is the first work reporting the possibility of joining the industrially very important materials, ceramic matrix composites, in just 7 s. In addition, such a joint may possess a relatively high apparent shear strength (31.1 MPa), which matches the interlaminar shear strength of the composites (in the range of 25–35 MPa, according to the supplier MT Aerospace, Germany). It must be pointed out that this apparent shear strength of the flash joint was higher when compared to the apparent shear strength of the C_f/SiC joints when joined with Ti (both 30 and 130 μm) using a standard SPS process at 1700 °C with a heating rate of 150–200 °C/min and a dwell time of 3 min [7]. Moreover, the apparent shear strength of this joint was in the same range as the average shear strength value of the coated C_f/SiC joined with a Ti_3SiC_2 interlayer at 1300 °C using a standard SPS process (31.1 ± 4.0 MPa), reported in our previous work [2]. In both works, the overall apparent shear strength of the joints was predetermined by the interlaminar shear strength of the C_f/SiC composites (in both cases the joints were stronger than the initial composites). This is the reason why almost the same apparent shear strength of the joints was measured in both works. In the present work, the C_f/SiC samples were joined with the Ti interlayer within 7 s compared with more than 45 min (whole SPS process) in [2,7], and at a temperature corresponding to ~1250 °C rather than 1700 °C for the same Ti foil [7] or 1300 °C for Ti_3SiC_2 foil [2]. This constitutes a significant reduction in a processing time and the maximum temperature required, which in turn leads to energy savings and potentially higher industrial interest in the process. More importantly, as the quality of the joints strongly depends on the heating power used, all the drawbacks of the current joining set-up may be easily diminished using a more appropriate apparatus than SPS offers. This includes the use of a machine with better power control and operating with AC current instead of DC, and more accurate temperature measurement at the joining interface.

The present work shows, for the first time, that it is possible to obtain sound joints within 7 s using the current joining set-up (using SPS) when the right processing conditions are applied. However, further work, including a detail analysis of the interface and the interlayer is needed to fully understand the flash joining mechanism. All drawbacks of this joining set-up (related to reproducibility and obtaining a homogenous temperature throughout the samples) might be diminished using a more appropriate apparatus than SPS. The potential of flash joining has been clearly demonstrated here, and it could be an inspiration for future work. The present flash joining approach is not only limited to C_f/SiC materials, but due to the rapid temperature transition, it is also suitable for other materials susceptible to thermal degradation, such as CVD-SiC, etc.

4. Conclusions

The Flash joining process was developed to join CVD-SiC coated C_f/SiC samples with a Ti interlayer using an SPS apparatus. The samples to be joined were wrapped in graphite felt, which acted as thermal insulation, but more importantly, it produced preheating of the joining samples in the initial stages of the process. Different heating powers were achieved using different power limits and discharge times. The following joining parameters were found to be the optimal ones: heating power of 2.2 kW, electric current of 370 A. The samples (each having a diameter of 20 mm and a height of 2 mm) were joined within 7 s from the start of the discharge. The corresponding temperature, ~1237 °C, was measured using an external thermocouple inserted into the joining interface. An extremely rapid heating rate (9600 °C/min) was achieved, and a very short processing time prevented any reaction between the CVD-SiC coating and the Ti interlayer from occurring. No reaction phases were found in the Ti interlayer after the flash joining, and the interlayer only contained some elemental C while a trace amount of Si (~0.30–0.40 wt.%) was also detected up to 15 μm from the interface with the CVD-SiC coating of C_f/SiC. Therefore, a sound joint was obtained by forming a metallic kind of joint (Ti-based) at the joining temperature, in which the absence of any reaction phases (titanium silicides, carbides or ternary phases) significantly shortened the time necessary for the solid-state diffusion bonding. The maximum apparent shear strength of the joints was 31.4 MPa, which matches the interlaminar shear strength of the C_f/SiC composites. This indicates that the joint was stronger than the initial composites.

Acknowledgments

The research leading to these results has received funding from the European Community's 7th Framework Programme FP7 2007–2013 under the grant agreement No.: 609188, within the European project ADMACOM (Advanced manufacturing routes for metal/composite components for aerospace). P.T. gratefully acknowledges the contribution of the Slovak Research and Development Agency under the grant No.: APVV-15-0469 and the Scientific Grant Agency of the Slovak Republic under the grant No.: VEGA 2/0189/15. S.G. and M.J.R. were supported by EPSRC (EP/K008749/1, XMat). T.S. was supported by EC FP7 2007–2013 (ADMACOM). MJR would like to acknowledge the support of Sunchon National University, South Korea, through the BK21+ programme; Seventh Framework Programme. The authors are very grateful to Dr. Karin E. Handrick (MT Aerospace, Germany) for the supply of CMC materials to be joined.

Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.jeurceramsoc.2017.05.016.

References

[1] C. Jiménez, K. Mergia, M. Lagos, P. Yialouris, I. Agote, V. Liedtke, S. Messoloras, Y. Panayiotatos, E. Padovano, C. Badini, C. Wilhelmi, J. Barcena, Joining of ceramic matrix composites to high temperature ceramics for thermal protection systems, J. Eur. Ceram. Soc. 36 (2016) 443–449.

[2] P. Tatarko, V. Casalegno, C. Hu, M. Salvo, M. Ferraris, M.J. Reece, Joining of CVD-SiC coated and uncoated fibre reinforced ceramic matrix composites with pre-sintered Ti_3SiC_2 MAX phase using Spark Plasma Sintering, J. Eur. Ceram. Soc. 36 (2016) 3957–3967.

[3] X. Shi, X. Jin, H. Lin, J. Jing, L. Li, C. Wang, Joining of SiC nanowires-toughened SiC coated C/C composites and nickel based superalloy (GH3044) using Ni71CrSi interlayer, J. Alloy Compd. 693 (2017) 837–842.

[4] F. Lan, K. Li, H. Li, L. Guo, Y. He, L. Zhang, High-temperature property of carbon/carbon composite joints bonded with ternary Ti-Si-C compound, J. Alloy Compd. 480 (2009) 747–749.

[5] X. Song, H. Li, X. Zeng, Brazing of C/C composites to Ti6Al4 V using multiwall carbon nanotubes reinforced TiCuZrNi brazing alloy, J. Alloy Compd. 664 (2016) 175–180.

[6] T.T. Ikesoji, Brazing of carbon–carbon (C/C) composites to metals, in: D.P. Sekulic (Ed.), Advances in Brazing: Science, Technology and Applications, Woodhead Publishing, Cambridge, UK, 2013, pp. 395–417.

[7] S. Rizzo, S. Grasso, M. Salvo, V. Casalegno, M.J. Reece, M. Ferraris, Joining of C/SiC composites by spark plasma sintering, J. Eur. Ceram. Soc. 34 (2014) 903–913.

[8] Y. Katoh, L.L. Snead, T. Chenga, C. Shih, W.D. Lewis, T. Koyanagi, T. Hinoki, C.H. Henager Jr., M. Ferraris, Radiation-tolerant joining technologies for silicon carbide ceramics and composites, J. Nucl. Mater. 448 (2014) 497–511.

[9] M. Ferraris, M. Salvo, V. Casalegno, A. Ciampichetti, F. Smeacetto, M. Zucchetti, Joining machined SiC/SiC composites for thermonuclear fusion reactors, J. Nucl. Mater. 375 (2008) 410–415.

[10] M. Salvo, S. Rizzo, V. Casalegno, K. Handrick, M. Ferraris, Shear and bending strength of SiC/SiC joined by a modified commercial adhesive, Int. J. Appl. Ceram. Technol. 9 (2012) 778–785.

[11] P. Colombo, B. Riccardi, A. Donato, G. Scarinci, Joining of SiC/SiCf ceramic matrix composites for fusion reactor blanket applications, J. Nucl. Mater. 278 (2000) 127–135.

[12] H. Böhrk, U. Beyermann, Secure tightening of a CMC fastener for the heat shield of re-entry vehicles, Compos. Struct. 92 (2010) 107–112.

[13] J.G. Crowther, British Scientists of the Nineteenth Century, Routledge Library Editions: History & Philosophy of Science, Routledge, 2013, ISBN: 1135028788 9781135028787.

[14] E. Thomson Apparatus for electricwelding (1886) US Patent US 347, 142.

[15] K.T. Compton, Elihu Thomson, 1853–1937 (March 20th, 1942) Science 95, No. 2464, 285–289.

[16] S. Grasso, Y. Sakka, G. Maizza, Electric current activated/assisted sintering (ECAS): a review of patents 1906–2008, Sci. Technol. Adv. Mater. 10 (5) (2009), 053001 (24pp).

[17] E.A. Olevsky, E.V. Aleksandrova, A.M. Ilyina, D.V. Dudina, A.N. Novoselov, K.Y. Pelve, E.G. Grigoryev, Outside mainstream electronic databases: review of studies conducted in the USSR and post-soviet countries on electric current-assisted consolidation of powder materials, Materials 6 (10) (2013) 4375–4440.

[18] M. Yu, S. Grasso, R. McKinnon, T. Saunders, M.J. Reece, Review of flash sintering: materials, mechanisms and modeling, Adv. Appl. Ceram. 116 (2017) 24–60.

[19] P. Tatarko, Z. Chlup, A. Mahajan, V. Casalegno, T.G. Saunders, I. Dlouhý, M.J. Reece, High temperature properties of the monolithic CVD β-SiC materials joined with a pre-sintered MAX phase Ti_3SiC_2 interlayer via solid-state diffusion bonding, J. Eur. Ceram. Soc. 37 (4) (2016) 1205–1216.

[20] X. Zhou, Y.H. Han, X. Shen, S. Du, J. Lee, Q. Huang, Fast joining SiC ceramics with Ti_3SiC_2 tape film by electric field-assisted sintering technology, J. Nucl. Mater. 466 (2015) 322–327.

[21] S. Grasso, P. Tatarko, S. Rizzo, H. Porwal, C. Hu, Y. Katoh, M. Salvo, M.J. Reece, M. Ferraris, Joining of β-SiC by spark plasma sintering, J. Eur. Ceram. Soc. 34 (7) (2014) 1681–1686.

[22] Y. Yu, H. Dong, B. Ma, Q. Ren, W. Ma, Effect of different filler materials on the microstructure and mechanical properties of SiC–SiC joints joined by spark plasma sintering, J. Alloy Compd. 708 (2017) 373–379, http://dx.doi.org/10.1016/j.allcom.2017.03.039.

[23] X. Zhou, H. Yang, F. Chen, Y.-H. Han, J. Lee, S. Du, Q. Huang, Joining of carbon fiber reinforced carbon composites with Ti_3SiC_2 tape film by electric field assisted sintering technique, Carbon 102 (2016) 106–115.

[24] S. Grasso, T. Saunders, H. Porwal, B. Milsom, A. Tudball, M. Reece, Flash spark plasma sintering (FSPS) of α and β SiC, J. Am. Ceram. Soc. 99 (5) (2016) 1534–1543.

[25] A. Becker, S. Angst, A. Schmitz, M. Engenhorst, J. Stoetzel, D. Gautam, H. Wiggers, D.E. Wolf, G. Schierning, R. Schmechel, The effect of Peltier heat during current activated densification, Appl. Phys. Lett. 101 (1) (2012), art. no. 013113.

[26] F. Goesmann, R. Schmid-Fetzer, Temperature-dependent interface reactions and electrical contact properties of titanium on 6H-SiC, Semicond. Sci. Technol. 10 (1995) 1652–1658.

[27] H. Yang, X. Zhou, W. Shi, J. Wang, P. Li, F. Chen, Q. Deng, J. Lee, Y.-H. Han, F. Huang, L. He, S. Du, Q. Huang, Thickness-dependent phase evolution and bonding strength of SiC ceramics joints with active Ti interlayer, J. Eur. Ceram. Soc. 37 (4) (2017) 1233–1241.

[28] J.H. Kim, D.S. Kim, S.T. Lim, D.K. Kim, Interfacial microstructure of diffusion-bonded SiC and Re with Ti interlayer, J. Alloy Compd. 701 (2017) 316–320.