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Joining Silicon Carbide Plates by Titanium Disilicide-Based Compound

E. Jacques¹, Y. Le Petitcorps¹, L. Maillé¹, C. Lorrette^{1,2} and L. Chaffron²

¹ University of Bordeaux, Laboratoire des Composites Thermostructuraux, UMR 5801, 3 Allée de la Boétie, 33600 Pessac-France

² CEA-Nuclear Energy Division, Nuclear Material Department, Section for Applied Metallurgy Research, Technology for Extreme Materials Laboratory

KEY WORDS: Brazing silicon carbide, Ceramic metal interface, Ceramic Matrix Composites, Intermetallic and disilicide compounds.

ABSTRACT

The present work is part of the Fourth Generation reactors program where is described the methodology and the results for joining SiC substrates by metallic silicides with SiC powder reinforcements. The more severe temperature in service are in the range of 1000°C but short time incursions at 1600 or 2000°C have to be anticipated. One of the key issues is the joining of the SiC_f/SiC_m composites to seal the combustible cladding. We describe the results for joining SiC substrates at liquid state using TiSi₂. Joint integrity and joint strength can be improved by adding small SiC particles to the silicides powders. The assemblies are performed in an inductive furnace. Cross sections of the assembly, wettability tests, thermo-mechanical properties and 4-bending point mechanical tests are presented.

1. INTRODUCTION

Joining ceramic/ceramic composites (CMC's) is of interest for numerous applications in the aeronautic or nuclear fields. This joining may concern the assembly of two CMCs parts or the assembly of one CMC part against a metallic alloy. This study deals with the assembly of two silicon carbide/silicon carbide composite plates designed for a fuel cell component within the framework of the Fourth Generation Reactor Program [1]. In order to achieve this goal, many requirements have to be followed: (i) the joint must be tight enough to avoid the gas leaks in service and, (ii) has to withstand temperatures such as 1000°C for very long durations (service conditions) and 1500°C for short durations (accidental conditions) without damaging the CMC properties, (iii) the process has to be compatible with the nuclear constraints and must not damage the composite properties which are very sensible to fibre degradations. The composite material for this application is made of a woven silicon carbide fibre perform (Hi-Nicalon

Type STM or Tyranno SA3TM fibre) infiltrated by a silicon and carbon based precursor (CVI process). Both the matrix and the fibre are nearly pure silicon carbide which gives to the composite a good thermal stability up to 1600°C. The CMCs assembly research is not a new but this requirement in temperature stability (1500°C) makes the answer not trivial [2-5]. Conventional silicon based compositions cannot be used since the melting point of Si is too low. Thus a material selection study has been done in order to select compounds able to withstand these requirements. The selection of the right composition for the joint has been done from the physical properties, the thermodynamic data of the different candidates in order to avoid a too important mechanical and chemical mismatch with the silicon carbide. Various assembly processes have been evaluated (solid state bonding, liquid state bonding) and several ways of short heating have been tested (microwave, High Frequency heating). The quality of the assembly has been evaluated through metallographic observations on polished sections or with tomography images on the whole assembly.

2. MATERIALS AND METHODS

2.1. Material selections

The silicon carbide material is a covalent crystal characterised by a low coefficient of thermal expansion CTE, a high modulus, a high melting point and a intermediate toughness which makes the ceramic sensible to flaws (Table 1). Unfortunately the silicon carbide reacts at medium temperature (above 800°C) with most of the refractory metals. Thus these metals cannot be chosen which purpose. However the analysis of the ternary phase diagram involving Si, C and Metallic atoms indicates that some compositions can be in a thermodynamical equilibrium with SiC (Figure 1). The carbide (here TiC_x) and the disilicide (here TiSi₂) are thermodynamically stable with SiC in the solid state whereas the other silicides react with SiC. In addition to a poor thermal expansion mismatch with SiC, TiC has not been chosen due its too high melting point which could be a problem if the joint has to be melted for the assembly. The remaining choice is the metallic disilicide. This disilicide is related to a ionic-covalent crystals which exhibits an intermediate Young's modulus and a higher CTE than SiC. Due to this CTE mismatch, the joint will be in tension and the silicon carbide in compression on cooling. One way to reduce the CTE of the joint is to incorporate SiC particles within the silicide. By controlling the volume fraction of SiC added, the CTE and the Young's modulus can be tailored. Due to the relatively high thermal stability of TiSi₂, this disilicide has been selected. The TiSi₂ compound will be reinforced by SiC particles (less than 1µm in size) and with different volume fraction (from 0 to 30%).

2.2. Material heating

Since the disilicide do not react with the silicon carbide substrate in the solid state, the bonding between the joint and the SiC substrate should not be strong enough. In order to get this bonding, the melting of the joint should be done very rapidly in order to promote and to limit the chemical reaction with the silicon carbide substrate. A microwave furnace (2450MHz) can be used for melting ceramics which have an effective electrical conductivity between 1 and 10 Sm^{-1} such as semi-conductors. It is supposed to be a more economical process than the other conventional processes like the high frequency induction furnace. The principle was to selectively heat the joint and not the silicon carbide substrate. Thus the joint could be melted without heating the composite itself. Unfortunately the silicon carbide absorbs at the same wave than the titanium disilicide. Finally, a more conventional high frequency furnace has been chosen (60% of power – Five Celes HF MP3, 12kW). With that equipment, the joint between two silicon carbide plates is melted in less than 1 minute and cooled down for few seconds later after the melting. The assembly is maintained by a graphite susceptor inside a chamber under a purified argon atmosphere. The wettability is of great importance in the brazing technology and has been studied for the different compositions by recording the value of the contact angle during the spreading of the drop.

2.3. Material characterisations

In order to characterize the physical properties of the joint (CTE and Young's modulus) model materials of $\text{TiSi}_2 + x\%$ of SiC_p were prepared by spark plasma sintering. After sintering, the materials were nearly fully dense (95% of the theoretical density). The CTE has been measured by a TMA device (dilatometer Netzsch DIL402C) and the Young's modulus measured from the room temperature to 1000°C was determined by a Grindosonic apparatus (LEMMENS Inc.).

The quality of joining has been studied on polished cross sections by SEM and WDS analysis (Hitachi S4500). A particular attention has been paid to the occurrence of cracks in the joint, the thickness of the reaction zone between the melted joint and the SiC substrate, as well as its chemical composition. The macroscopic characterisation of the joint has been done by X ray tomography (Phoenix X Ray nanotom S).

3. RESULTS AND DISCUSSIONS

3.1. Decreasing the residual stresses in TiSi_2 by the addition of submicronic SiC particles (SiC_p)

Although the values are below the data expected from a rule of mixture (ROM), the SiC_p have a positive effect on the Young's modulus from room temperature to 1000°C (Figure 2). This difference from the ROM can be explained by the submicronic size of the particles, the aspect ratio of them which is closed to one and the presence of 5% of porosities. A 10% decrease in the CTE is also measured on TiSi_2 reinforced with 20% in Vo. of SiC_p .

The consequences on the network of cracks in the joint can also be seen on figure 3. In order to avoid a chemical reaction between the TiSi_2 and the SiC from both the particles and the substrate, the material was also prepared by spark plasma sintering at 1200°C . The presence of the SiC particles induces a decrease in the residual stresses within the joint which exhibits also a clear interface with the silicon carbide substrate.

3.2. Wettability of $\text{TiSi}_2 + \text{SiC}_p$

For pure TiSi_2 , the contact angle decreases from 80° to 12° , 4 seconds after the melting (Figure 4) TiSi_2 has a very low contact angle which is very useful for its infiltration in small cavities of the materials to join. The reason of this kinetic of wettability can be explained by the reactivity of the liquid Ti-Si alloy with the SiC substrate [8]. When SiC particles are added, the contact angle is increased to 30° for 30% Vo. of SiC_p after 4 seconds in the liquid state. The particles induce an increase in the viscosity as well as they react with the titanium disilicide matrix. However, the contact angle measured with 30% of SiC is very closed to the contact angle measured with pure silicon. The contact angle and the kinetic of spreading can be taylored by the amount of SiC_p . It is important to control the time during the melting step in order to limit the reaction zone thickness between the joint and the SiC substrate.

3.3. Reactivity of liquid (Ti,Si) + SiC_p with the SiC substrate

In the liquid state, the liquid containing Ti and Si atoms should react with SiC. From the SiC to the TiSi_2 , the interface is composed of Ti_3SiC_2 and TiC (Figure 5). If the joint contains SiC_p they also should react with the liquid and the liquid should be less aggressive against the SiC substrate. Consequently, the reaction zone thickness at the Joint/SiC interface decreases with an increase in SiC_p amount (Tableau 2).

3.4. Characterization of a real assembly between two plates of SiC

Two plates of SiC have been joined by melting the TiSi_2 mixture and followed by a rapid cooling. It can be seen that the brazing is nearly uniformly present even though some depletion of matter can be seen. No pressure was applied on the surface of the sample, a possible explanation of this lack of matter could be the presence of gas confined during the melting. However these first results are encouraging.

4. CONCLUSIONS

The aim of this work was to find an adequate brazing composition which is stable with SiC between 1000°C to 1500°C for long durations. Titanium disilicide can reach such a goal but due to the mismatch in the physical properties between SiC and TiSi_2 , the joint is drastically cracked. The addition of submicronic particles of SiC inside the joint reduces the tensile residual stresses. The SiC_p have also an

other beneficial effect on the chemical stability of the joint when the disilicide is melted during the brazing step. The interest of this type of brazing has now to be confirmed by the mechanical testing of the assembly.

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Figure 5: Reaction Zone thickness and morphologies at the SiC/TiSi_2 interface

Figure 6: X-Ray tomography images of the brazing between two plates of SiC

TABLES

Table 1: Physical Properties of the potential candidates for joining SiC [6]

Table 2: Influence of the amount of SiC_p on the Reaction Zone thickness at the TiSi_2/SiC interface

TABLE 1

	Si	SiC	TiSi ₂	TiC
Melting (°C)	1414	2700	1540	3140
CTE (10 ⁻⁶ /K)	5.7	4.5	10.4	7.6
Young's Modulus (GPa)	130	400	260	450

TABLE 2

	Pure TiSi ₂	TiSi ₂ + 10%SiC Vo.	TiSi ₂ + 20%SiC Vo.	TiSi ₂ + 30%SiC Vo.
Reaction zone thickness (μm)	20	6-7	4-5	4-5

FIGURE 1

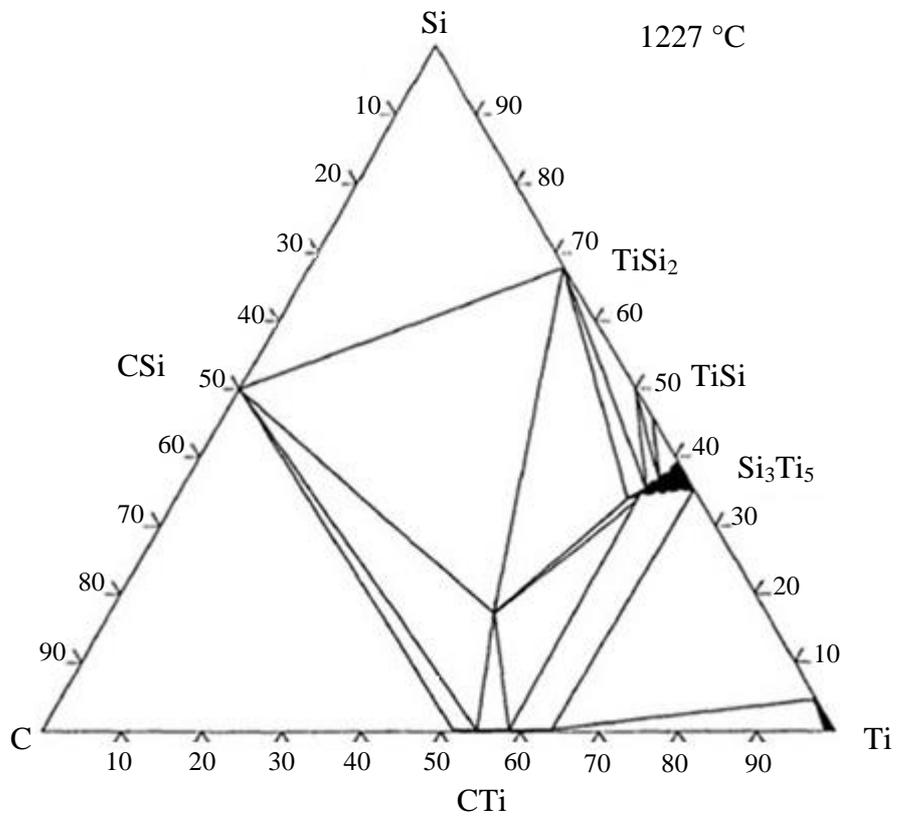


FIGURE 2

Young's modulus (GPa)

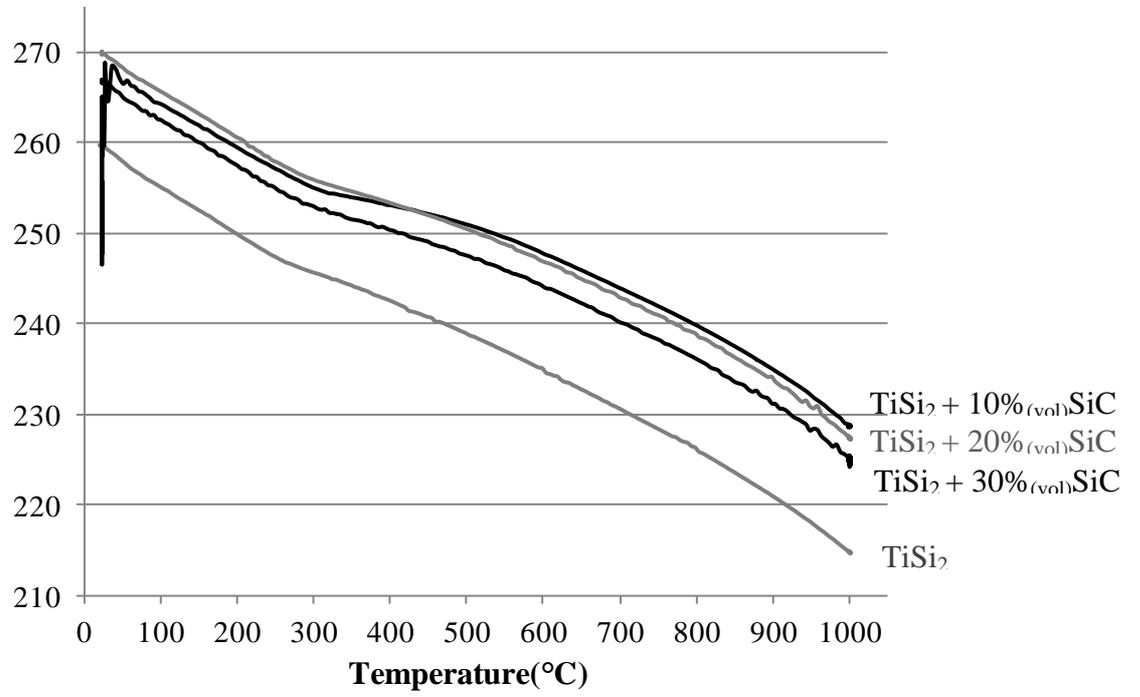


FIGURE 3

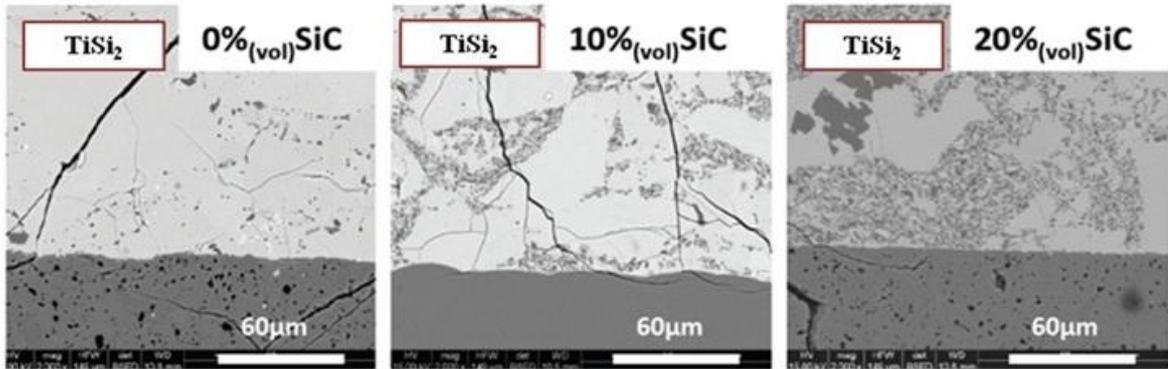


FIGURE 4

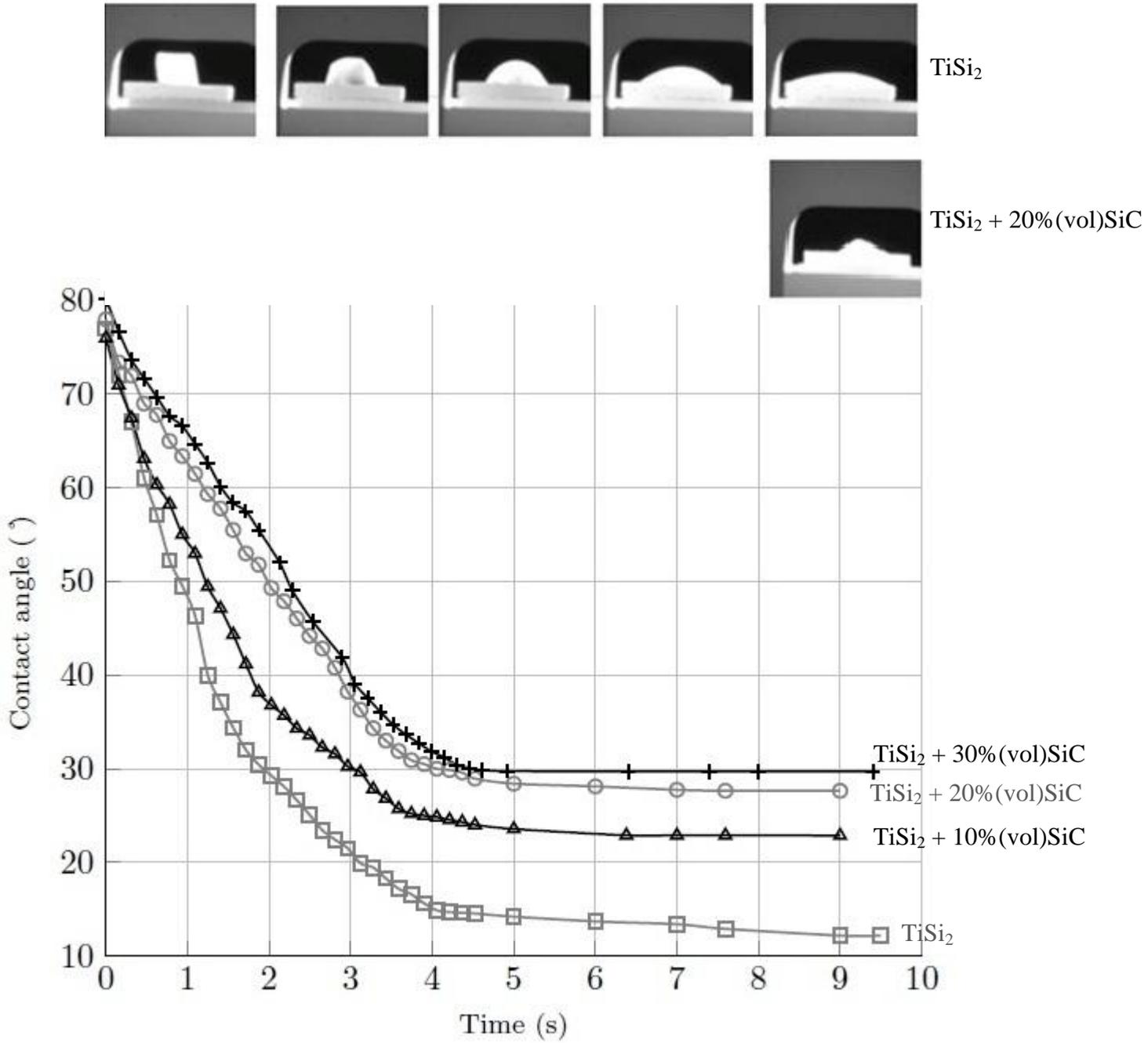


FIGURE 5

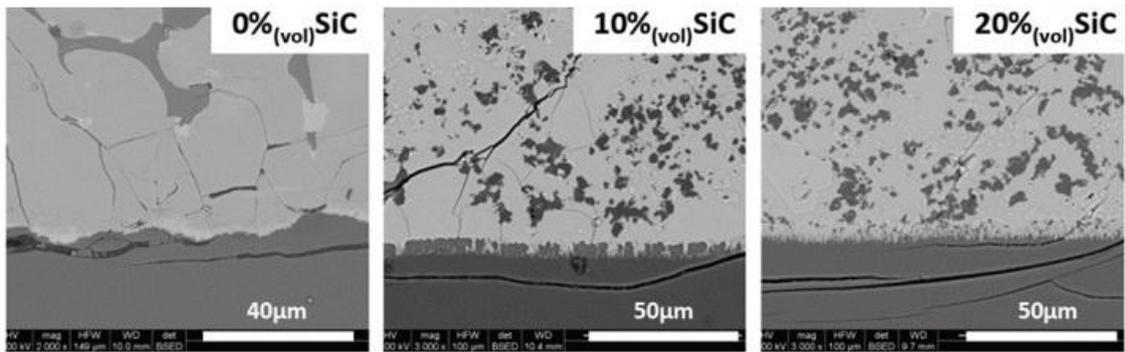


FIGURE 6

