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Brazing of silicon nitride ceramic composite to steel using SiC-particle-reinforced active brazing alloy

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Abstract

Silicon carbide particles have been introduced as reinforcements in a commercially available active metal braze filler alloy (Incusil ABA, Wesgo Metals) used for the joining of ceramic-to-metal. The effect of particle reinforcement of the braze filler on the flexural strength of ceramic to metal joints has been investigated at room temperature and at elevated temperatures. An average four point flexural strength of nearly 400 MPa is achieved at room temperature when using Incusil ABA + 30 vol.% SiC (sandwich foil system) compared to 330 MPa with Incusil ABA alone. At a test temperature of 250 °C relaxation of residual stresses in the joints results in an average flexural strength of approximately 520 MPa when using Incusil ABA + 10 vol.% SiC. These values compare with an average room temperature flexural strength of nearly 800 MPa for the ceramic composite. The reaction products of the braze alloy at the joint interface were identified by SEM.

Keywords: A. Joining; C. Strength; D. Si_3N_4; D. SiC

1. Introduction

Advanced ceramics are widely used for extreme engineering applications were their particular properties including high strength, Young's modulus, hardness, corrosion resistance and wear properties can be utilised. Silicon nitride (Si_3N_4)-based ceramics are interesting as they have a good combination of mechanical properties and are suitable for use in harsh environments and at elevated temperatures [1]. In particular, applications of Si_3N_4 include cutting tools, ball bearings, reciprocating engine components and containers for hot metal handling (aluminium). However, Si_3N_4 components with complex shapes are difficult to produce as hot pressing is normally required to produce fully dense parts with the best properties and this technique allows for the production of simple shapes only. More complicated components can by produced by either joining simple ceramic pieces to other ceramic pieces (e.g. by glass sealing, diffusion bonding or brazing), or more commonly, to more complex metal pieces. The two main problems when joining ceramics to metals are firstly the poor wettability of ceramics by most metals and alloys, and secondly the differences in physical properties between ceramics and metals.

The first problem of wettability is overcome with the use of an activated braze alloy, where an active element, e.g. Ti, alters the surface chemistry of the ceramic by the formation of intermediate reaction layer and lowers the wetting angle of the molten braze on the ceramic [2]. The second problem, specifically the mismatch in the coefficient of thermal expansion (CTE) between the ceramic, the braze filler alloy and the metal, as well as differences in Young's modulus, can result in high critical residual stresses in the ceramic. The residual stresses are influenced by other factors including component size effects, braze area and braze gap thickness. The stresses in the ceramic are at maximum in the region near the joint interface, and this can lead to component fracture in this region, sometimes under very low, or even zero loads.

The active braze alloy filler used as the joining material is normally inserted as a foil between the joint partners, or is applied as a paste. In laboratory and industrial scale active alloy development, powder mixing of the alloy elements is often used to develop new compositions. Hence, care must be taken to

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prevent segregation and settling of the component elements when there are large differences in densities of the different elements.

The reinforcement of ceramics and metal alloys by ceramic particles has shown significant improvements in a number of materials properties over non-reinforced materials. These improvements include increased strength, fracture toughness, and creep and wear performance for both ceramic and metal matrix composites [3,4]. The use of ceramic particle reinforced Ti-activated alloy for brazing is thus a logical development. The use of SiC particles should theoretically lead to a lowering of the CTE of the braze alloy, in addition no brittle reaction products are observed when the commercial braze alloy Incusil ABA is used. A reduction in the CTE mismatch should result in lower residual stresses in the ceramic-to-metal joint and subsequently an increase in the flexural strength.

In the current work a Si_3N_4–TiN ceramic composite was joined to itself and to 14NiCr14 steel using the Ti-activated braze alloy Incusil ABA, with, and without silicon carbide (SiC) particle reinforcement. The Si_3N_4–TiN composite was used as it has a higher CTE than Si_3N_4 alone and therefore the CTE mismatch between the partners is reduced. Particle-reinforced braze foils were produced from starting powders via mixing and forming techniques. The microstructure of the brazed joints and their four-point bending strengths were investigated. Fractography of the flexural strength specimens was used to investigate the cause of failure with a view to improving the brazing procedure and obtaining a high joint strength. The goal of this work is to develop a brazing system which would be suitable for joining a cutting tool grade Si_3N_4–TiN ceramic composite to tool steel. Hence the brazed joint should have good strength at both room temperature and at typical elevated cutting tool operating temperatures of 200–400 °C.

2. Experimental

The ceramic braze partner consisted of a hot pressed Si_3N_4 ceramic composite reinforced with 30 wt.% of TiN particles. The metal braze partner was a 14NiCr14 steel (DIN 1.5752, ECN35, AISI 3310). The use of a Si_3N_4–TiN composite is preferred over Si_3N_4 since it has been shown that the addition of TiN particle reinforcements results in increases in certain physical and materials properties including fracture toughness, strength and more importantly the CTE over Si_3N_4 [5,6]. The CTE of the Si_3N_4–TiN ceramic composite is $3.8 \times 10^{-6}\ K^{-1}$ and the CTE of the steel is $14.1 \times 10^{-6}\ K^{-1}$. The ceramic and steel partners were surface ground into rectangular specimens 3 mm × 4 mm × 25 mm in dimension with a surface roughness $R_{max} < 2\ \mu m$ [7]. Joining was performed on the 3 mm × 4 mm face. Some basic materials properties of the joining partners are listed in Table 1.

Incusil ABA with chemical content Ag 59.0 wt.%, Cu 27.25 wt.%, In 12.5 wt.% and Ti 1.25 wt.% was purchased in the form of a foil of 50 μm thickness and also in a powder form with a particle size <325 mesh (Wesgo Metals, California, USA). Incusil ABA has a quoted CTE of $18.2 \times 10^{-6}\ K^{-1}$ which is higher than the CTE of the steel partner. SiC powder (Johnson Matthey, Zurich, Switzerland) with a CTE of $5.2 \times 10^{-6}\ K^{-1}$ and an average particle diameter of 10–20 μm was used as the particle reinforcement. The composite-reinforced braze foils were prepared from the Incusil ABA and SiC powders. The Incusil ABA powder was mixed with the SiC, cellulose nitrate and octylacetate and the mix was poured onto a rubber sheet. After drying, small foils were cut, uni-axially die pressed, and sintered at 640 °C in vacuum for 4 h. Incusil ABA foils with 10 and 30 vol.% of SiC powder reinforcement were prepared. Typically the Incusil ABA + SiC foils were approximately 300 μm thick.

Table 1
Comparison of selected materials properties of the joining partners

Material	*E* (GPa)	*α* [RT − 600 °C] ($10^{-6}\ K^{-1}$)	Strength (MPa)
14NiCr14	210	14.1	600
Si_3N_4 + 30 wt.% TiN	330	3.8	785 (± 51)
Incusil ABA	–	18.2	–
SiC	410	5.2	–

The braze filler alloys described above were used to join ceramic-to-ceramic and also ceramic-to-steel, with eight flexural strength specimens being manufactured for each brazing experiment. All brazing was performed in a Torvac vacuum furnace under a vacuum of 10^{-5} mbar, or better. The brazing equipment used to hold and align the brazing foil and partners has been described previously [8]. All braze partners were cleaned ultrasonically in acetone and then degassed in the vacuum furnace at temperatures of 950 and 1100 °C for the steel and ceramic, respectively. Brazing with the Incusil ABA foil was performed at 740 °C with a dwell time of 10 min for the ceramic-to-ceramic joints and for the ceramic-to-steel joints. Brazing with the Incusil ABA + SiC foils was performed at 750 °C with a dwell time of 10 min. The different brazing temperatures used for the different Incusil ABA based brazes are the optimized temperatures reached after extensive experimentation. A schematic illustration showing the four different braze joint systems investigated is presented in Fig. 1.

Brazed joints were characterised for four point bending strength using a 20/40 mm loading configuration and testing was performed as described in standard EN 843-1 [7]. Polished microstructures of the joints were examined with a Leo SEM equipped for EDX analysis. Fractography was performed with an optical stereo microscope and where necessary, further examination with a SEM was performed. Fractography was performed as recommended in the ASTM standard C1322-02a [9]. Elevated temperature flexural strength tests were performed over the temperature range of 200–400 °C based on EN 820-1 [10].

3. Results and discussion

3.1. Incusil ABA brazing

The Incusil ABA was used as the reference filler alloy primarily because of its relatively low brazing temperature of

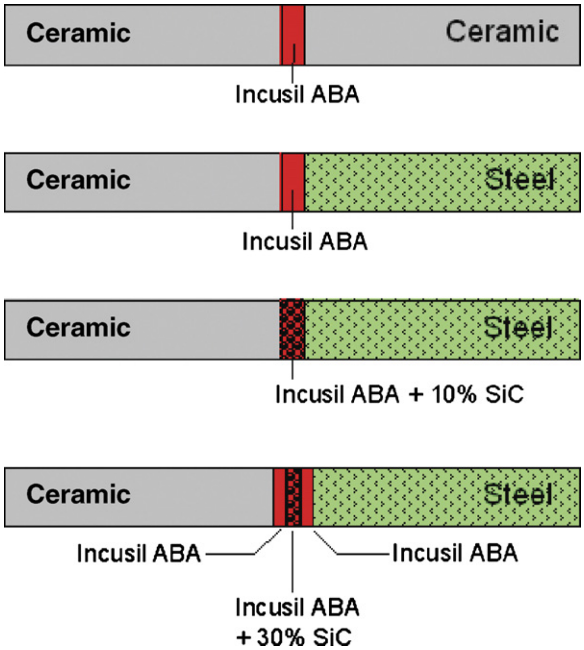

Fig. 1. Schematic illustration of the four different braze joints investigated.

740 °C compared to other commercial Ti-active alloys (e.g. Wegso Ticusil ~900 °C and Degussa CB6 ~1010 °C). The low brazing temperature hence prevents excessive reaction of the braze alloy with the steel partner, particularly embrittlement and the formation of Fe_2Ti type phases which have been observed when brazing at high temperatures [8].

Joining ceramic-to-ceramic with Incusil ABA resulted in an average flexural strength at RT of 461 MPa with a typical brazing gap thickness of 70–80 μm when two foils were used. When joining ceramic-to-ceramic the large CTE mismatch that exists when joining ceramic to metal is not as significant, since the braze gap is normally quite small and it is usual for ceramic-to-ceramic joints to have a higher strength than equivalent ceramic-to-metal joints. The results of the room temperature flexural strengths for all four types of joints investigated are presented in Fig. 2. Fractographic studies of the ceramic to ceramic specimens joined with Incusil ABA showed that failure was largely due to ceramic-type defects at, or near, the tensile loading surface. Typically these were small pores, machining marks or agglomerates of TiN (from the ceramic processing) on, or very close, to the tensile load surface of the ceramic.

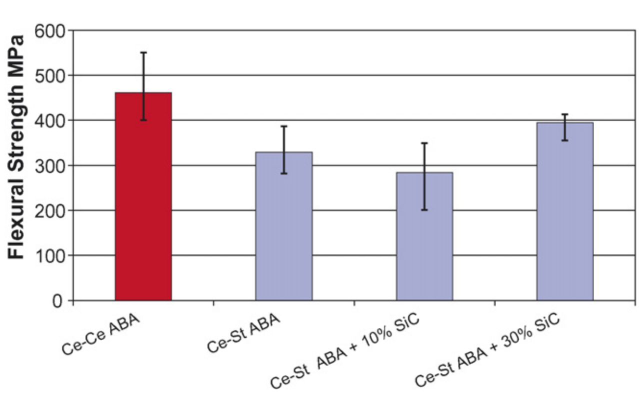

Fig. 2. Summary of the flexural strength results of the different brazed joints, with different braze filler setups (Ce–Ce, ceramic to ceramic joining; Ce–St, ceramic to steel joining; ABA, Incusil ABA).

When ceramic was brazed to steel using Incusil ABA the average flexural strength measured was 329 MPa (also when two foils were used). This decrease in flexural strength compared to the ceramic-to-ceramic joints is due to the higher CTE mismatch between the ceramic and steel partners. Failure of the ceramic-to-steel samples joined with Incusil ABA always occurred in the ceramic partner near the braze interface. When high thermal residual stresses are present in the ceramic partner in a ceramic to metal joint, a bowed crack path is observed after failure in the ceramic close to the braze interface. This typically results in very low strengths [11].

Fractography of the ceramic-Incusil ABA-steel joint sample with the lowest strength (282 MPa) showed little, or no, evidence of significant residual thermal stresses in the sample. In the ceramic-Incusil ABA-steel joint, the residual stresses in the ceramic are relatively low and this is shown by the relatively straight nature of the crack path on the ceramic partner after failure, as shown in Fig. 3a. The two fracture surfaces of the ceramic are shown in Fig. 3b and the approximate location of the fracture origin is highlighted. The fracture origin in this case was a pore on the tensile surface which was infiltrated by the braze filler alloy during joining (Fig. 3c). It appears that a reaction of braze alloy in the pore(s) leads to a further degradation of the local ceramic microstructure inside the pore which then acts as a stress concentration and promotes the start of crack formation and then propagation.

Metallography of the braze material in the joint shows Ag–In and Cu as the two main phases (Fig. 4a). In addition, a Cu phase rich in In and Ti is also observed in the braze filler alloy after joining. A fourth Cu–Ti phase is also observed (Fig. 4a). Another Ti-rich phase is observed in the form of either sub-micron particles, or as a thin discontinuous layer between the Cu and Ag phases (Fig. 4b). Any Ti-based reaction layers that may have formed on the ceramic, or steel partners were too thin to be observed by SEM.

3.2. Incusil ABA + SiC brazing

Ceramic-to-steel brazing with Incusil ABA + 10 vol.% SiC resulted in an average flexural strength of 284 MPa. The relatively low value of 10 vol.% SiC particle reinforcement is not sufficient in reducing the ‘‘composite CTE’’ of the braze filler alloy and hence not effective in reducing the CTE mismatch and residual thermal stresses between the ceramic and steel partners during the joining process. Finite element analysis of the joining system was performed and the findings are presented elsewhere [12]. Further, the finite element analysis indicated that the introduction of the highly stiff SiC particles into the Incusil ABA actually results in an increase in residual stresses. This is because the amount of plastic strain that can develop in the brazing alloy, in the joint, to release thermal residual stresses is effectively lowered, due to a stiffening effect of the SiC particles. This lowering of the possible plastic strain has a greater effect than any lowering of

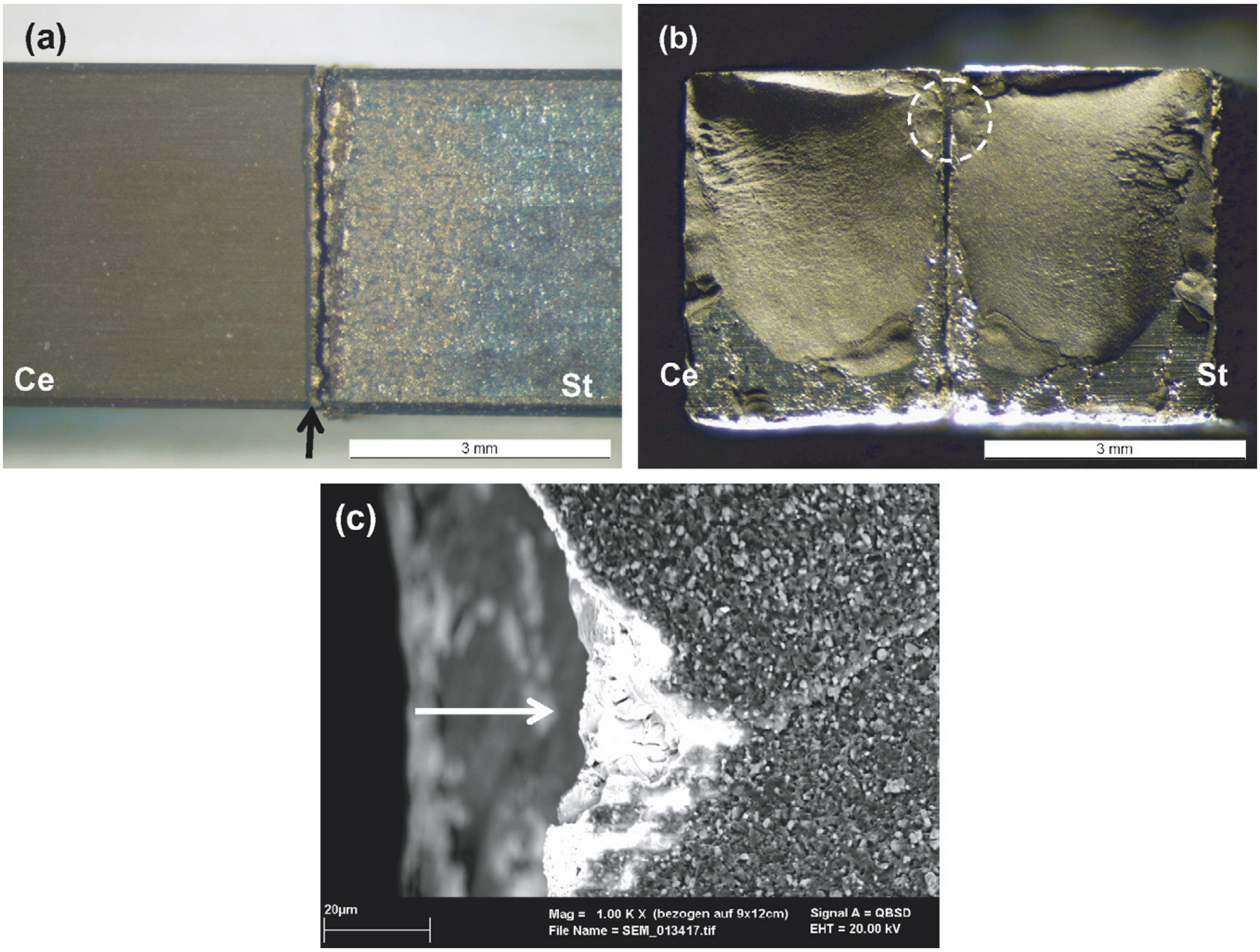

Fig. 3. Fractography of ceramic-to-steel joint: (a) side view of the straight crack path in the ceramic of the ceramic-to-steel joint with Incusil ABA, $\sigma_f = 282$ MPa; (b) view of the two main fracture surfaces from (a) showing that fracture occurs through the ceramic partner, the fracture origin is highlighted and (c) close up view showing the fracture origin is a pore which was infiltrated by the braze filler alloy.

the CTE of the braze alloy by the SiC particles. Fractography showed some evidence of thermally induced residual stresses in the form of slightly bowed facture patterns in the ceramic partners in the specimens with the lowest strengths (e.g. Fig. 5). Failure was concluded to be due to the combination of residual thermal stresses and existing ceramic defects at, or near, the tensile load surface.

In order to overcome the problem of the lower allowable plastic strain and to utilise the CTE lowering effect of the SiC particles, a sandwich foil solution was proposed. Here a foil of Incusil ABA + 30 vol.% SiC particles was sandwiched between two 50 μm foils of Incusil ABA and inserted in to the brazing gap. This results in a ceramic-to-steel brazing with an average flexural strength of 395 MPa. This average strength is approximately 110 MPa (~28%) higher than Incusil ABA + 10 vol.% SiC and 65 MPa (~17%) higher than the Incusil ABA alone. In addition the deviation in the strength is now less than half of that recorded, when the Incusil ABA + 10 vol.% SiC, or the Incusil ABA are used (see Fig. 2).

Two different types of failure were observed with the specimens brazed with the Incusil ABA + 30 vol.% SiC (sandwich). Fracture occurred either mainly through the braze interface, as shown in Fig. 6a (this has not been observed previously), or through a combination of the ceramic and braze alloy (Fig. 6b). Both failure modes show a straight fracture path with no evidence of significant residual thermal stresses.

Metallography of the brazed joint shows a wider braze gap than previously, with the SiC particles spread fairly randomly throughout the braze alloy (Fig. 7a). The SiC particles are generally not observed in contact with the braze partners (steel or ceramic) and are more concentrated in the centre of the braze joint. High magnification SEM observation showed that a Ti-rich layer of approximately 200 nm thickness forms on the surface of the SiC particles (Fig. 7b). This reaction layer product is hence thicker than any formed on the surface of the Si_3N_4–TiN ceramic partner, where it was not possible to observe any reaction products by SEM.

While care has to be taken when comparing strengths with other workers due to differences in braze area, specimen and test geometry and braze gap thickness, comparisons can be made with previously published data, and the results from this study compare favourably. Kim et al. reported a maximum strength of 310 MPa when brazing Si_3N_4 to 316 stainless steel with Cusil ABA (also with a braze area of 3 mm × 4 mm) but

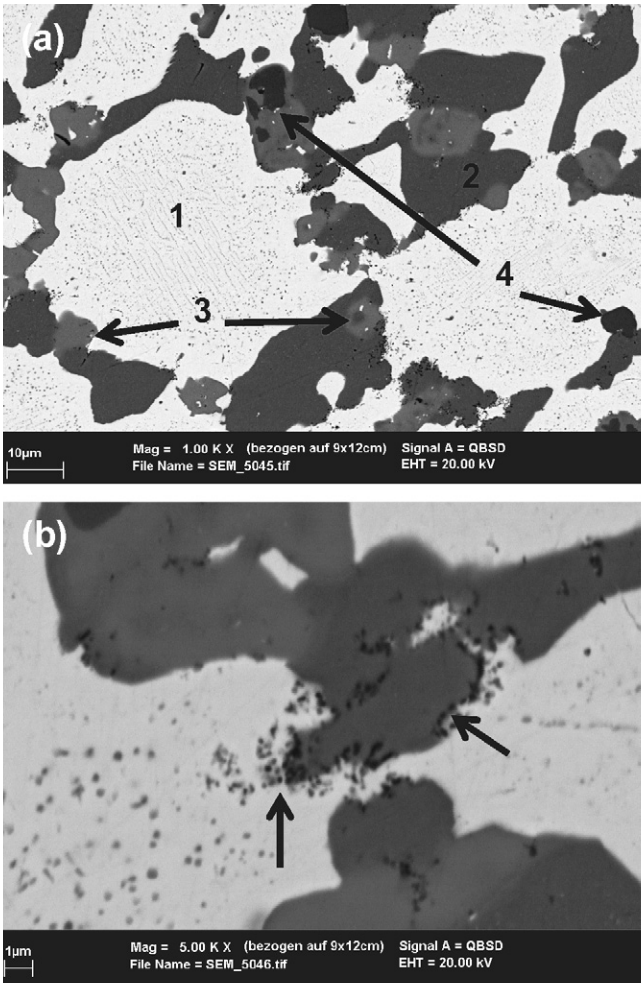

Fig. 4. SEM micrographs of an Incusil ABA braze alloy: (a) showing an overview with one Ag–In phase, two Cu phase, three Cu–In–Ti phase and four Cu–Ti phase and (b) higher magnification view showing the Ti-rich phase as nano particles and thin layer (arrowed).

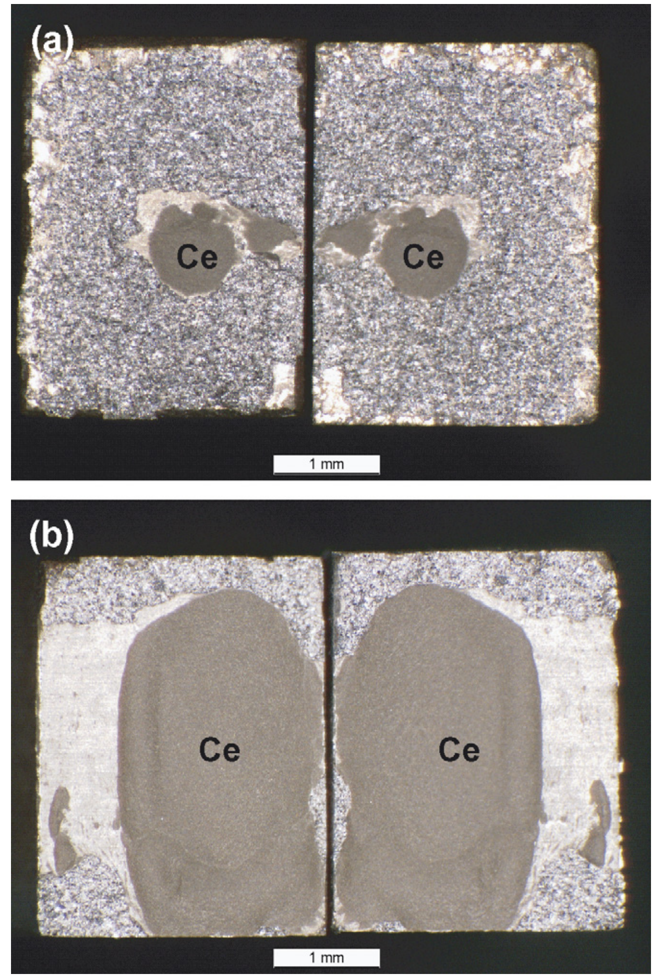

Fig. 6. Fracture surfaces of ceramic-to-steel joints with the Incusil ABA + 30 vol.% SiC sandwich system: (a) fracture can occur mainly through the braze filler alloy and (b) fracture can also occur through both the ceramic and the braze.

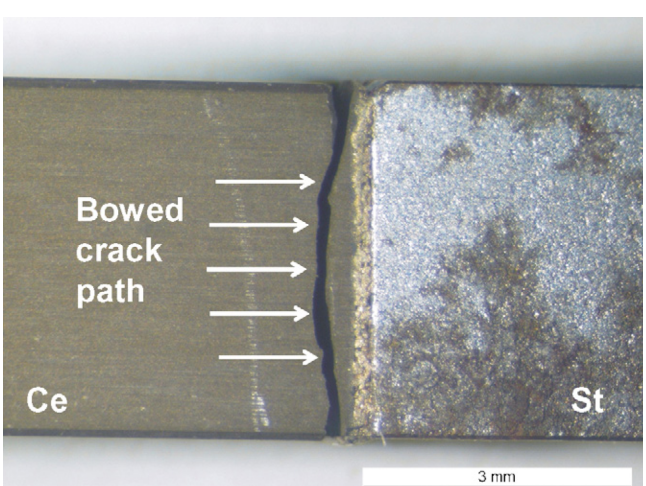

Fig. 5. Fractography of a ceramic-to-steel joint with Incusil ABA + 10 vol.% SiC showing a bowed crack path indicating significant residual stresses in the ceramic partner after joining.

gave no average strength or deviation values [13]. The highest reported flexural strengths observed with Incusil ABA for brazing of stainless steel to a β-sialon are approximately 500 MPa, however the specimen test geometry used (braze area of 5 mm × 5 mm) was different from the current set up [14]. These compare to the maximum values of 387 MPa for Incusil ABA and 413 MPa with the Incusil ABA + 30 vol.% SiC (sandwich system) in the current work.

3.3. High temperature properties

One of the aims of these experiments was to develop a brazing filler which could be used to join a ceramic cutting tool tip to a steel holder. The typical operating temperature for this type of cutting tool is in the region of 200–400 °C even when cooling lubricants are used. Hence flexural strength tests of ceramic and steel partners joined with Incusil ABA, Incusil ABA + 10 vol.% SiC and Incusil ABA + 30 vol.% SiC (sandwich system) were performed at elevated temperatures. The measured high temperature flexural strengths are presented in Fig. 8. In general an increase in the average flexural strength for all systems up to a

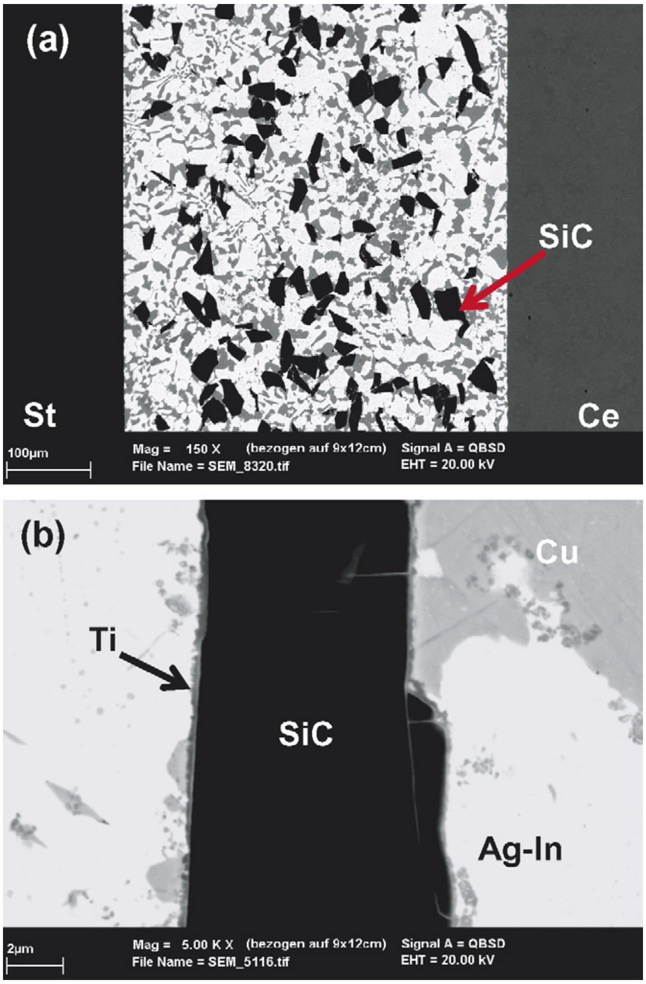

Fig. 7. (a) Micrograph of a ceramic-to-steel joint brazed with Incusil ABA + 30 vol.% SiC (sandwich system) showing random distribution of SiC particles (black) in the centre of the braze gap ~300 μm thick and (b) high magnification of a SiC particle showing a 200 nm thick Ti rich reaction layer on the SiC particle.

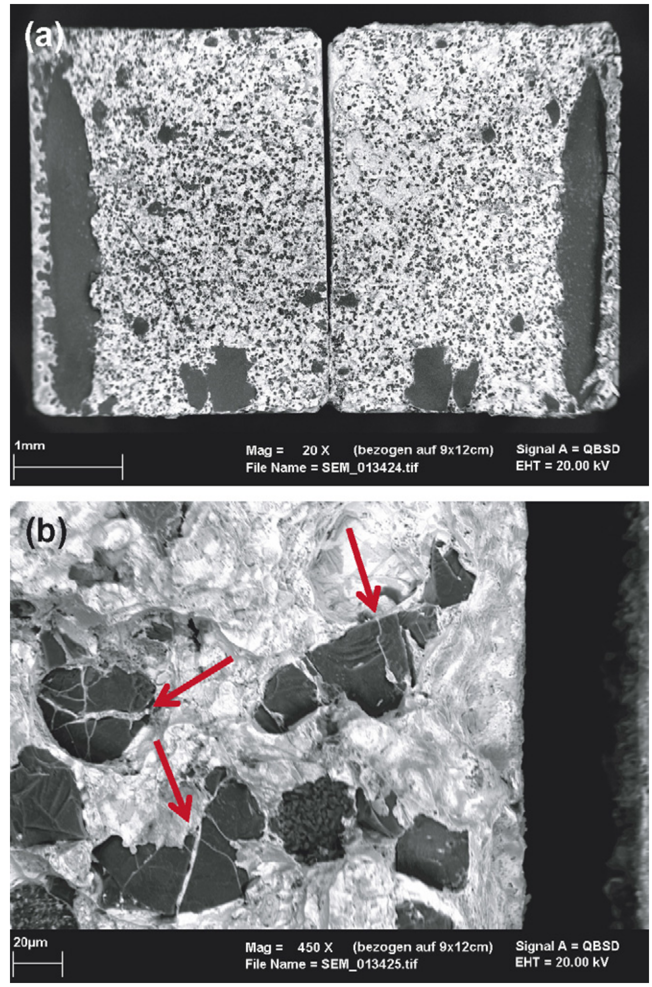

Fig. 9. Ceramic to steel joint brazed with Incusil ABA + 10 vol.% SiC tested at 250 °C: (a) fracture surfaces showing failure through the composite braze and (b) individual SiC particles have been fractured during brazing and the resulting cracks filled with braze alloy (arrowed).

test temperature of 250 °C is observed. The increase in average strength with test temperature compared to the room temperature tests is expected as heating the specimens results in a relaxation of thermal residual stresses in the ceramic. In addition the increase in test temperature leads to a softening of the braze filler alloy resulting in a more ductile failure mechanism. The high temperature results show that overall the Incusil ABA + 10 vol.% SiC has the best strength results. The highest average strength of 523 MPa was achieved at 250 °C, at higher temperatures the average strength gradually decreases again. At 300 °C the specimens joined with the Incusil ABA + 30 vol.% SiC (sandwich) have both the highest average strength (505 MPa) and the highest measured strength. These results indicate that the application of temperature leads to a relaxation of the residual stresses and an increase in strength up to 250 °C and to some extent 300 °C. Above 300 °C the braze alloys soften considerably, leading to the observed decrease in strength (Fig. 8).

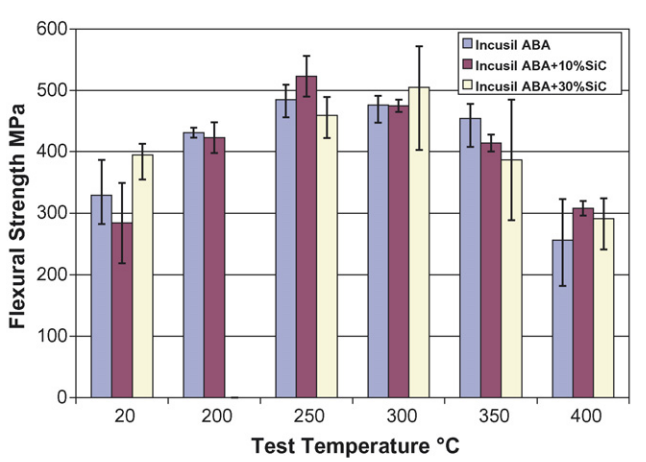

Fig. 8. Summary of the high temperature flexural strength results of ceramic-to-steel joints.

Fractography also showed the effect of the increase in test temperature on relaxtion of the residual thermal stresses, with failure now occurring through the braze filler material. An example of the fracture surfaces of a ceramic-to-steel joined with Incusil ABA + 10 vol.% SiC tested at 250 °C is shown in Fig. 9a. This shows clearly that fracture occurred through the composite braze filler alloy. Higher magnification shows that the reinforcing SiC particles appear to be cracked (Fig. 9b). It seems that some of the SiC particles cracked during the brazing process and were infiltrated subsequently by the molten braze alloy.

4. Conclusions

It has been shown that the use of ceramic SiC particles to reinforce a commercially available Ti-activated brazing filler can affect the flexural strength of ceramic-to-steel braze joints. The reaction between Incusil ABA and the SiC particles is limited to the formation of a thin Ti rich layer. No detrimental reaction occurs between the steel and the SiC, or Incusil ABA. For ceramic-to-steel joining:

- The use of Incusil ABA results in an average flexural strength of 329 MPa at room temperature.
- The use of Incusil ABA + 10 vol.% SiC reduces the amount of plastic strain possible in the joint and results in a lower average flexural strength of 284 MPa, hence for room temperature strength SiC particle reinforcement by itself is detrimental.
- Direct SiC particle reinforcement leads to a increase in joint strength at elevated temperatures, with the highest average flexural strength of 523 MPa achieved with Incusil ABA + 10 vol.% SiC at 250 °C.
- The use of Incusil ABA + 30 vol.% SiC in a sandwiched foil system results in a higher average flexural strength of 395 MPa at room temperature and lower residual thermal stresses due to an increase in allowable plastic strain and results in a change in fracture behaviour.
- Thus, the use of SiC particles in a sandwich system appears to lead to a lowering of the residual stresses between the steel and ceramic partners.
- Processing could be improved further as the optimum SiC content and SiC particle size and sandwich layer thicknesses remain to be determined.

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