Research Article

Metal – Hard Ceramic Joining: Direct Bonding between 316-Austenitic Stainless Steel & α-SiAlON

Mita Biswas⁎, Gopa Chakraborty⁎⁎, Barun Haldar⁎ and Siddhartha Bandypadhyay⁎∗

⁎CSIR- Central Glass & Ceramic Research Institute, 196, Raja S.C. Mullick Road, Kolkata 700 032, India
⁎⁎Indira Gandhi Centre for Atomic Research, Kalpakkam, 603102, India.

Accepted 01 April 2014, Available online 10 April 2014, Vol.4, No.2 (April 2014)

Abstract

Diffusion bonding between a hard advanced ceramic material, oxygen rich compositional variety of yttrium stabilized silicon-aluminum-oxynitride (α-SiAlON), and 316 grade austenitic stainless steel has been attempted under gas pressure sintering & hot pressing. Diffusion interfaces were characterized by XRD, optical micrograph, SEM, EDS & microhardness measurements. Interdiffusion of elements in both ceramic and steel side and formation of intermediate compounds at the reaction interface indicate that bonding could be achieved between the ceramic and steel only under hot pressing condition at or above 1300°C. Role of diffusing elements vis-à-vis the interface formation is discussed.

Keywords: Diffusion Bonding; Reaction interface; α-SiAlON; 316-austenitic stainless steel

Introduction

Advanced ceramics are widely used to machine materials like highly alloyed grey cast irons, due to their excellent resistance to mechanical, chemical and thermal attacks [Bitterlich B. et al, 2008]. Nitride based ceramics with adequate hot hardness and thermal shock resistance are considered to be the most suitable ones for machining of cast iron in automobile industry and nickel based superalloys in aerospace industry [Silva R. F. et al, 1991 & Pashby R. I. et al, 1993]. SiAlON composites with improved combination of hardness and fracture toughness is a good alternative to sintered Si3N4 that offers increased performances in some sectors, for example, higher metal removal rate and longer tool life as compared to conventional cemented carbide cutting tools [Ekstrom T. et al, 1992 ].

For various applications, several ceramic wear parts and engine components often require interfacing with metallic machine parts and hence bonding with ductile metallic structure such as steels. However, large differences in coefficient of thermal expansion between ceramic and steel, along with relative chemical inertness of ceramic imposes restriction in metal to ceramic joining. Elaborate studies [Petves S. D., 1996, Polanco R. et al, 2004, Abed A. et al, 1995 & Vleugels J. et al, 1996] are available on different methods for joining of steels with the beta-variety of Si3N4 (β) or a solid solution SiAlON based on β-Si3N4 structure (β'), those are categorized amongst the most important candidate materials for cutting and tribological applications. There is another technologically important phase, alpha-SiAlON, a solid solution based on α-Si3N4 structure (α’) [Chen I. W. et al, 1997], the addition of which contributes towards improvement in hardness, thereby contributing towards much better performances of the composites than monolithic β or β’ [Ekstrom T. et al, 1992]. Although the interaction of 316-austenitic stainless with β/β’ variety is reported [Hussain P. et al, 2010] it might be interesting to know about its interaction with α’ ceramics. In this work, study on diffusion bonding between one oxygen rich variety of α-SiAlON and 316-austenitic stainless steel has been pursued under different firing conditions vis-à-vis the effects of diffusion of different atomic species across the bond interface.

Experimental

The starting powders- Si3N4 (E10, UBE Industries, Japan), AlN (Grade A, Starck, Germany), Al2O3 (Ceralox Corporation, USA) and Y2O3 (99%, Indian Rare Earth Ltd., India) were attrition milled for 4 h with silicon nitride milling balls and isopropyl alcohol as milling media. The composition of the investigated sample (equivalent substitution- Y: 2.95%, Al: 16.02%, O: 4.79%) was obtained by using adequate amounts of the corresponding oxides and nitriles. The powder was subsequently dried, sieved and green pressed under isostatic pressure of 200 MPa into 10 mm X 8 mm X 4 mm rectangular plate and was fired at 1800°C, 2 h and 1.5 bar nitrogen gas overpressure in graphite crucible. The surface to be joined was prepared by polishing to 1 μm diamond finish followed by characterization with X-ray diffraction (XRD), scanning electron microscopy (SEM) and energy disperse spectroscopy (EDS).
Diffusion bonding experiments were conducted with two sets of the interaction couple, first one for firing under gas pressure and the next one under hot pressing. For the first set, the polished section of the ceramic was taken in a cylindrical rubber mould of diameter 16 mm where the rest space is filled with the grade 316 steel powder. The entire assembly was isostatically pressed under 300 MPa. The resultant ceramic-steel green couple was taken for firing at a temperature range of 1000°C to 1300°C for 5 hours in a graphite resistance heating furnace under ambient to 1.8 MPa argon/nitrogen atmospheres. In the next set of assembly, the polished ceramic sample was encapsulated in stainless steel powder inside a 40 mm diameter graphite die coated with boron nitride as high temperature lubricant to avoid reaction between steel and carbon material of the die and punch. The sample was hot pressed under similar conditions of temperature and dwell time under 1.8 MPa punch pressure and by maintaining 1 bar nitrogen pressure inside the furnace. Ram pressure was withdrawn immediately when cooling started. In both cases the average heating and cooling rate were 5°C/min.

For microstructural studies under optical and electron microscope, the diffusion-bonded specimen was cross sectioned, polished by standard metallographic techniques and etched with oxalic acid. Distribution profile of elements viz. Fe, Cr, Ni, Si, etc. across the ceramic–metal interface was generated by EDS analysis. XRD using CuKα radiation was further employed for identification of the phases at the interface. Vickers microhardness indentations were taken across the bonding interface with 100 g and 1000 g load on the steel and ceramic surfaces respectively.

Results & Discussion

The α-SiAlON prepared under study is nearly fully dense (>98.5 %theoretical) with weight loss <1% during sintering indicating that the nominal composition is retained in the fired product. SEM micrograph of the α’ phase (Fig.1a) represents equiaxed nature of grains of size around 1 micron with a very few elongated grains. Phase characterization of α’ is confirmed through both XRD and EDS patterns during SEM (Fig.1b).

Firing under Argon atmosphere—Considering the fact that nitrogen saturation of steel during firing might cause reduced interaction with nitrogen bearing ceramics, the heat treatments of the first set of green assembly was undertaken at ambient argon gas atmosphere. No sign of development of bonding at the ceramic-metal join could be observed at 1000°C and above. This non-attachment happened irrespective of the firing temperature. A heat treatment at high temperature like 1300°C even produced the same result as it was at 1000°C. Some blistering occurred at the ceramic surface near metal side (fig. 2a); while in addition, a large amount of wool formation surrounding the sample was observed (fig. 2b). The blistering of the oxynitride phase may be ascribed to the decomposition of it under argon gas atmosphere, especially in presence of a metal. SiAlON phases undergo decomposition producing vapour species like SiO & N₂ with a corresponding shift in composition towards Al-richer phases. The stability of α-SiAlON phase in terms of SiO partial pressure, for example, has been estimated to be between 0.39 to 0.6 atm. assuming p_{O2}+p_{SiO} to be 1 atm. [Mitomo M. et al, 1979]. The effect was evident when sintering of the phase could be tailored from a fluffy mass to a densified body by changing the gaseous atmosphere [Bandyopadhyay S. et al, 1987].
Fig. 2(b): wool formation above the diffusion couple heat treated under argon atmosphere.

Therefore, depending on the low partial pressure or complete absence of nitrogen (as the present case to be) in firing atmosphere, the release of SiO seems logical. The presence of Si & O in the present case could be detected in the wool as evidenced from EDS under FESEM (fig. 2c). It is evident that some gaseous reactions at heat treatment temperature have predominated, leading to the wool formation.

Fig. 2(c): ED spectrum generated out of some portions of the wool

Firing under Nitrogen atmosphere-To avoid the decomposition of SiAlON, nitrogen was selected as the firing atmosphere during next set of runs. The change in firing atmosphere and variation of heat treatment temperature up to 1300°C could not however bring as well, any change in the nature of surface attachment. In subsequent runs, a higher pressure of nitrogen (1.8 MPa) was employed in otherwise similar conditions of heat treatment to the previous cases. Occasional bonding in discrete places could be observed only at 1300°C while majority of the ceramic-metal contour remained unattached. Both blistering and wool formation disappeared. Residual stress caused by the differential thermal expansion of the metal and ceramic seems responsible for the surface detachment. It is evident that gas pressure, even at the highest temperature applied in this study, is unable to produce as large extent of bonding as to keep the surfaces in contact. The ineffectiveness of the applied isostatic pressure from atmosphere may possibly be due to the porosities present, especially in the metal side.

Hot Pressing- In the second set of runs under hot pressing condition, where a carbon punch applies axial pressure over the samples confined in the carbon die, successful continuous surface attachment was observed only at 1300°C. Hot pressing below this temperature could produce only partly joined surfaces at around 1200°C while no interface formation took place at all below 1200°C. Optical and SEM micrographs of the bond interface obtained between 316 stainless steel and α-SiAlON in one typical samples produced at 1300°C are shown in Figs. 3a & b. Microscopy reveals a continuous and coherent interface between the ceramic and steel and the reaction layer is also clearly evident (Fig.3b). As metallographic studies indicate formation of reaction layer between the two phases, diffusion of different elements such as Fe, Cr and Ni in ceramic and Si in steel has been identified through EDS element line-scan at various locations across the interface.
Nitrogen solubility is influenced by the layers by enhancing extent of diffusion interface that ultimately the role of Fe has participated in the presence of alloying elements. Ni, Si, and C decrease the success rate into the iron alloy become rate controlling in two dissociation rate of the ceramic and the nitrogen solution reaction controlled [Peteves S. D. et al, 1992]. The improvement in strength value of the composite corresponded to the 3 to 14 wt% of Mo addition. The Mo content of 316-austenitic steel used in the present study coincides with the lower limit of this value. Hence, apart from the role of common members like Ni & Cr of other austenitic varieties, the presence of Mo is expected to induce additional features to make the 316-variety of austenitic steel to be an attractive bonding pair with SiAlION.

**Fig.4:** EDS line scan of α-SiAlION-316 stainless steel interface.

The interaction between SiAlION and iron alloy is reaction controlled [Vleugels J. et al, 1996]. The dissociation rate of the ceramic and the nitrogen solution rate into the iron alloy become rate controlling in two successive steps. Nitrogen solubility is influenced by the presence of alloying elements. Ni, Si, and C decrease the nitrogen solubility in steel, and on the other hand, Cr and Mo have the opposite effect. While Ni & Cr being the common member of other austenitic varieties, the presence of Mo is expected to induce larger extent of diffusion bonding. The different contributing roles of Mo in nitride-metal interaction systems may be referred to in this connection. The fact that Mo provides tendency towards thickening of interlayers by enhancing extent of diffusion was investigated earlier [Martinelli A. E. et al, 1999] through kinetic and microstructural observations during direct bonding of Mo with Si₃N₄. Successful bonding could be achieved in the temperature range of 1200°C to 1800°C through the formation of silicide phases in the interface. The extent of interface formation was proved to be dependent on the kinetics of the silicides- formation reactions. While the optimized Mo-promoted interlayer thickness was estimated [He Y. M, et al, 2010] to be within the range of 2 to 7 micron, the same value including the metal-protruded region in the present study corresponded to 5 to 8 micron (fig 3b).

In case of brazing (with the use of brazing alloy Ag–Cu–Ti), Mo in particulate form and when used in controlled quantity, also contributes to the successful joining of Si₃N₄ with 42CrMo steel. The role of Mo in such case is to control diffusion of Ti element towards the nitride, thereby controlling the thickness of the nitride-brazing interface that ultimately resulted in high joint strength [He Y. M. et al, 2010]. Similar improvement in strength property with the use of Mo in composition is reflected when FeMo70 alloy is used as a composite phase with β–SiAlION [Yang J. Z. et al, 2009]. Elemental Mo and FeMo react with FeSi to form Fe-Mo-Si alloy. The good mechanical properties are contributed by the interlocking structures of elongated Sialon grains with Mo–Fe–Si particles [Yang J. Z. et al, 2011]. The improvement in strength value of the composite corresponded to the 3 to 14 wt% of Mo addition. The Mo content of 316-austenitic steel used in the present study coincides with the lower limit of this value. Hence, apart from the role of common members like Ni & Cr of other austenitic varieties, the presence of Mo is expected to induce additional features to make the 316-variety of austenitic steel to be an attractive bonding pair with SiAlION.
silicides such as Fe₃Si, Cr-N (Cr₆N₃Si, Cr₆N₄Si₂) and Cr-nitride (CrN) in the reaction layer (Fig.5). The observation is similar to that occurs in case of β₃-Si₃N₄ against other grades of steel interaction [Vleugels J. et al, 1996]. However, intermediate compounds in the present case corresponding to silicides (MoSi₂) and nitrides (MoN, Mo₂N) of Mo could additionally be identified because of the migration of Mo towards ceramic side.

Characteristics of reaction layer: The influence of the appearance of intermetallic compounds at the reaction layer is further reflected through a high hardness value (Fig.6) at the reaction layer, much higher than austenitic stainless steel (~250 VHN) and marginally lower than the hardness of the ceramic phase (~2000 VHN). The initial improvement in the hardness curve in the metal side may be ascribed to the formation of nitrogen-pearlite caused by the diffusion of nitrogen (out of decomposition of SiAlON) into the stainless steel [Hussain P. et al, 2010].

Fig.6: Microhardness survey across α-SiAlON-316 stainless steel interface.

There is a sharp hardness gradient observed thereafter, between the ceramic and metal phases corresponding to the interaction layer: the hardness of the intermediate layer being contributed by the products of interdiffusion of ions across the interface. Apart from other common elements like Cr & Ni, 316-variety additionally contains different concentrations of Mo that form hard nitride phase. Hence the hardness value of the intermediate layer is expected to be higher in case of 316 in comparison to the other varieties of austenitic steel. Both contributions of Mo, firstly through higher hardness of the intermediate compounds and secondly through the promotion of greater extent of interaction of SiAlON into steel (via nitrogen diffusion, as discussed above), guides inferring that the 316-variety of austenitic steel might be a well suited pair for diffusion bonding with SiAlON.

Conclusion

From both the interdiffusion of alloy elements as evidenced through EDS analysis and microscopy studies, it may be inferred that direct diffusion bonding is possible to be achieved between oxygen richer variety of α-SiAlON and 316-austenitic stainless steel only in the temperature region of 1300°C and above. Combination of both temperature above a critical value and the application of die-pressure during hot pressing could lead to the continuous metal-ceramic surface attachment while an equivalent gas pressure (to that of hot pressing) was seen to remain ineffective for the purpose. Role of diffusion species is discussed in the formation of an intermediate layer between the steel and the ceramic. The presence of Mo, through its dual contribution towards larger extent of interface formation along with improvement in interface hardness, influences 316-grade of austenitic steel to become a suitable pair for diffusion bonding with SiAlON.

Acknowledgements

The authors thank Director, CGCRI for his support. Thanks are also due to M/S. Double Dee Technology, Mumbai for financial support.

References

Abed, A., Hussain, P. B., Jalham, I. S. and Hendry, A. (2001), Joining of silicon nitride to stainless steel (~250 VHN) and marginally lower than the hardness of the ceramic phase (~2000 VHN). The initial improvement in the hardness curve in the metal side may be ascribed to the formation of nitrogen-pearlite caused by the diffusion of nitrogen (out of decomposition of SiAlON) into the stainless steel [Hussain P. et al, 2010].

Fig.6: Microhardness survey across α-SiAlON-316 stainless steel interface.

There is a sharp hardness gradient observed thereafter, between the ceramic and metal phases corresponding to the interaction layer: the hardness of the intermediate layer being contributed by the products of interdiffusion of ions across the interface. Apart from other common elements like Cr & Ni, 316-variety additionally contains different concentrations of Mo that form hard nitride phase. Hence the hardness value of the intermediate layer is expected to be higher in case of 316 in comparison to the other varieties of austenitic steel. Both contributions of Mo, firstly through higher hardness of the intermediate compounds and secondly through the promotion of greater extent of interaction of SiAlON into steel (via nitrogen diffusion, as discussed above), guides inferring that the 316-variety of austenitic steel might be a well suited pair for diffusion bonding with SiAlON.

Conclusion

From both the interdiffusion of alloy elements as evidenced through EDS analysis and microscopy studies, it may be inferred that direct diffusion bonding is possible to be achieved between oxygen richer variety of α-SiAlON and 316-austenitic stainless steel only in the temperature region of 1300°C and above. Combination of both temperature above a critical value and the application of die-pressure during hot pressing could lead to the continuous metal-ceramic surface attachment while an equivalent gas pressure (to that of hot pressing) was seen to remain ineffective for the purpose. Role of diffusion species is discussed in the formation of an intermediate layer between the steel and the ceramic. The presence of Mo, through its dual contribution towards larger extent of interface formation along with improvement in interface hardness, influences 316-grade of austenitic steel to become a suitable pair for diffusion bonding with SiAlON.

Acknowledgements

The authors thank Director, CGCRI for his support. Thanks are also due to M/S. Double Dee Technology, Mumbai for financial support.

References