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Microstructural characterization and phase constituents near the interfaces of TiC-Al_2O_3/Q235 diffusion bonds

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Abstract

Diffusion bonding of TiC-Al_2O_3 composite ceramics to Q235 low carbon steel using Ti/Cu/Ti as multi-interlayer has been carried out by liquid diffusion welding technology. The microstructure and phase constituents for TiC-Al_2O_3/Q235 diffusion bonded joint were studied via scanning electron microscope and X-ray diffraction. The results indicate that various new phases are formed near the interface of TiC-Al_2O_3/Q235 joint and these are TiO, Ti_3Al, Cu_2Ti_4O, and Cu_3Ti_3O. The joints were produced with average shear strength of more than 100 MPa. The maximum joint strength was obtained reaching a value of 143 MPa. The fracture reveals brittle cleavage features. The fracture initiated in TiC-Al_2O_3 near the interface and propagated further towards to the interface.

Key words: TiC-Al_2O_3/Q235 joint, diffusion bonding, microstructure, phase constituents

1. Introduction

TiC-Al_2O_3 composite ceramics have been widely used in various engineering fields because of their good comprehensive properties, such as high hardness, good chemical stability and excellent wear resistance [1, 2]. TiC-Al_2O_3 composite ceramics are promising materials as ceramic cutting tools, usually performing better in high speed machining and in the machining of high hardness workpiece materials as compared to high speed steel and carbide tools [3]. The industrial merit of this material would be further enhanced if complex shaped components can be made by diffusion bonding.

This study deals with the joining of TiC-Al_2O_3 composite ceramics to Q235 low carbon steel by using Ti/Cu/Ti as multi-interlayer. The microstructure, shear strength, fracture morphology and phase constituents for the TiC-Al_2O_3/Q235 diffusion bonded joint were analysed by scanning electron microscope (SEM), electron probe microanalysis (EPMA) and X-ray diffraction (XRD). This is important for enlarging the application of TiC-Al_2O_3 composite ceramics.

2. Experimental

The materials used in the present work were TiC-Al_2O_3 composite ceramics and Q235 low carbon steel. TiC-Al_2O_3 composite ceramics were made by hot pressure sintering (HPS) to a final circle plate having dimension ø 52 × 3.5 mm^2. The size of Q235 circle plate specimen is ø 52 × 1.5 mm^2. The chemical composition of Q235 steel is (wt.%): C 0.14, Si 0.10, Mn 0.5, S 0.035, P 0.035, and Fe balance. TiC-Al_2O_3 composite ceramics consist of Al_2O_3 matrix and TiC particles. The chemical composition of TiC-Al_2O_3 is 65.6 Al_2O_3 and 34.4 TiC (wt.%). The microstructure of TiC-Al_2O_3 composite ceramics is shown in Fig. 1. The dark TiC particles are dispersed in the white alumina matrix. Multi-interlayer of Ti/Cu/Ti at 60 μm thickness was employed to promote the joining between TiC-Al_2O_3 composite ceramics and Q235 low carbon steel and obtain tight metallurgic joint.

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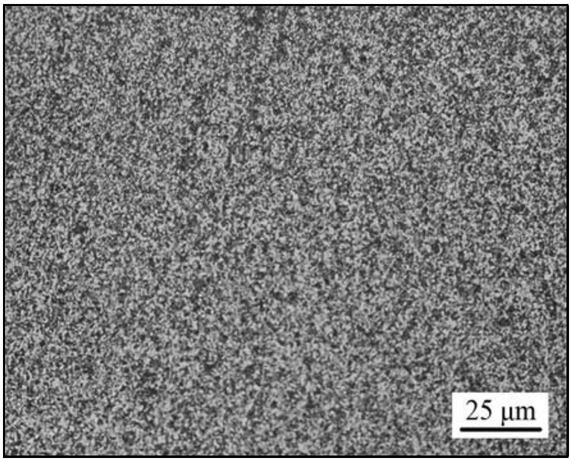

Fig. 1. Microstructure of TiC-Al_2O_3 composite ceramics.

The vacuum diffusion bonding equipment (Workhorse-II manufactured by Vacuum Industry Company) was used in the test. The heating power was 45 kW. Before diffusion bonding, the surfaces to be joined were ground with abrasive paper, and cleaned by immersing in acetone. After that, the test plates were overlapped and placed into a vacuum chamber. Technological parameters used in the test were: the heating temperature $T = 1100$–$1180\,^{\circ}C$, the pressure $P = 10$–15 MPa, the holding time $t = 30$–60 min and the vacuum degree of 1.33×10^{-4}–10^{-5} Pa. Under the conditions, the plastic flow formed in Q235 steel made the TiC-Al_2O_3/Q235 interface contact more tightly. Ti/Cu/Ti multi-interlayer melted instantly and reacted with TiC-Al_2O_3 at the interface, which promoted diffusive combination of TiC-Al_2O_3 and Q235. The cooling process was conducted in a vacuum chamber that was cooled by circuit water. When the chamber temperature was cooled to 100 °C, the TiC-Al_2O_3/Q235 diffusion bonded joints were taken out from the chamber. The resulting TiC-Al_2O_3/Q235 joints were cut into $10 \times 10 \times 8\,mm^3$ specimens for subsequent handling.

The TiC-Al_2O_3/Q235 specimens were ground and polished, mounted as metallographic samples, and then etched in 5 % HNO_3 + alcohol solution. The microstructure morphology of the TiC-Al_2O_3/Q235 interface was analysed by means of a JXA-840 scanning electron microscope (SEM). The interfacial shear strength was measured using a WEW-600E test machine at a shearing speed of $0.2\,mm\,min^{-1}$. A self-designed fixture was used in shearing, shown in Fig. 2. The phase constituents of fractured face were determined by XRD using D/MAXRC equipment. The fracture morphology was observed and the compositions on the fractured face were analysed by JXA-8800R electron probe microanalysis (EPMA).

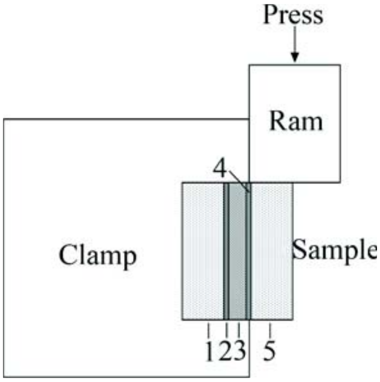

Fig. 2. Schematic graph in shearing: 1 – TiC-Al_2O_3, 2 – multi-interlayer, 3 – Q235, 4 – multi-interlayer, 5 – TiC-Al_2O_3.

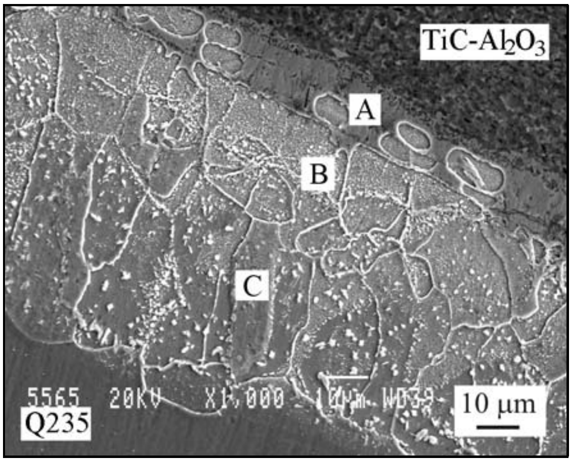

Fig. 3. Microstructure of the TiC-Al_2O_3/Q235 interface.

3. Results and discussion

3.1. Microstructural characterization near the TiC-Al_2O_3/Q235 interface

The microstructure near the interface of the TiC-Al_2O_3/Q235 diffusion bonded joint was observed using the SEM (see Fig. 3). The test result indicated that TiC-Al_2O_3 composite ceramics tightly combined with Q235 low carbon steel, and the interface both on the TiC-Al_2O_3 and Q235 side seems to be continuous, straight and free of the micro-voids, cracks and unbonded areas. The growth of diffusion and reaction layers takes place extensively into the low carbon steel but little into the TiC-Al_2O_3 composite ceramics because atomic diffusion is far easier in metals than in ceramics [4].

The interfacial transition region of the TiC-Al_2O_3/Q235 diffusion bonded joint is basically com-

Table 1. Shear tested results of the TiC-Al_2O_3/Q235 joint

Specimen number	Shear area (mm^2)	Maximum press (kN)	Shear strength (MPa)	Average shear strength (MPa)
1	100	9.7	97	108
2	100	8.0	80	
3	100	11.1	111	
4	100	14.3	143	

posed of three different regions: a transition region A on the TiC-Al_2O_3 side, a middle transition region B and a transition region C on the Q235 side (see Fig. 3). The transition region A on the TiC-Al_2O_3 side was narrower, elliptic light grey microstructures with the white outlines were discretely distributed on the dark grey microstructures, and the obvious boundaries formed both on the composite ceramics and the middle region B. The transition region C on the Q235 side was wider, which of grain shape was similar to that of low carbon steel, representing the united growing and interlocking feature at the interface between the region C and low carbon steel, and the white particles gathered on the grain boundary and dispersedly distributed inside the grains. The microstructure morphology of the region B is slightly different from that of the region C; the white particles are fewer and smaller. There is no obvious boundary between the region B and region C. All kinds of morphologic microstructures formed attributes to diffusion and reaction among the elements of the Ti/Cu/Ti multi-interlayer, TiC-Al_2O_3 composite ceramics and low carbon steel.

3.2. Shear strength and fracture

The interfacial shear strength was measured with a WEW-600E test machine. The stress was computed from the maximum load during testing (typical load at fracture) using standard static loading analyses for monolithic materials. The results are shown in Table 1. Joints were produced with average shear strength of more than 100 MPa. The maximum joint strength was obtained reaching a value of 143 MPa.

After strength testing, the fracture faces of the joint were analysed by EPMA in order to determine the fracture mode and localize the joint fracture propagation. The fracture morphology of TiC-Al_2O_3/Q235 joint is shown in Fig. 4a. The fracture reveals brittle cleavage features. There are typical cleavage steps on the fracture face, Fig. 4a. The composition of typical region (labelled by circle) on the fracture face was analysed by EPMA (the probe voltage: 4.9 keV), and analysed results are shown in Fig. 4b. It demonstrated strong peaks of Al, O, Ti, and low peaks of Mo and Cu. This indicated that the fracture of TiC-Al_2O_3/Q235 joint occurred mainly in the TiC-Al_2O_3 composite ceramics.

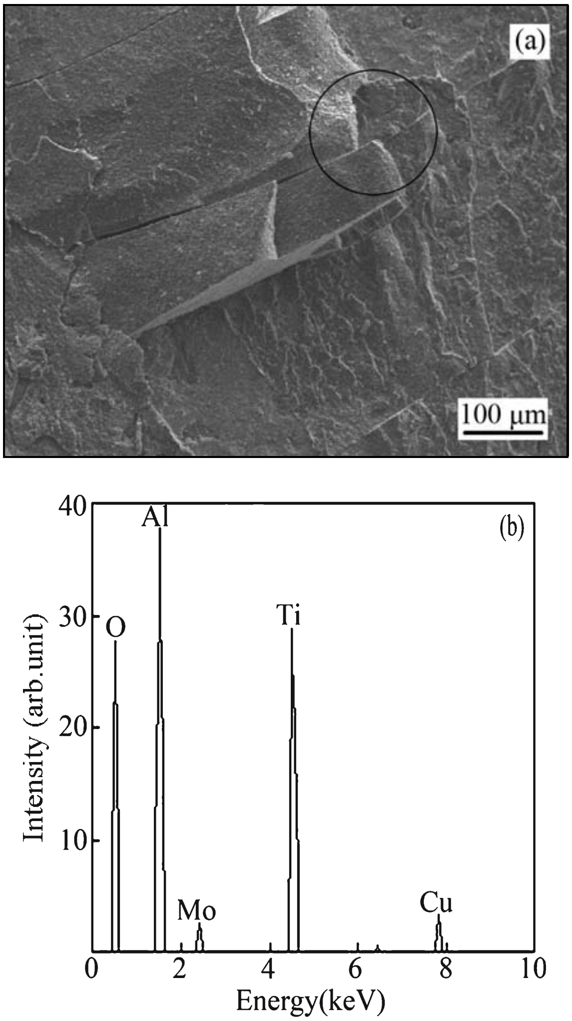

Fig. 4. Fracture morphology and EPMA analysis of TiC-Al_2O_3/Q235 joint: (a) fracture morphology (EPMA), (b) EPMA analysis of (a).

3.3. Phase constituents near the interface of TiC-Al_2O_3/Q235 diffusion bonds

The X-ray diffraction analysis of the TiC-Al_2O_3/Q235 fractured faces was carried out for identification of different phases developed at the interface during the joining process and fracture location. The diffractometer measurements were made on two sides, one near the TiC-Al_2O_3 and the other near the Q235.

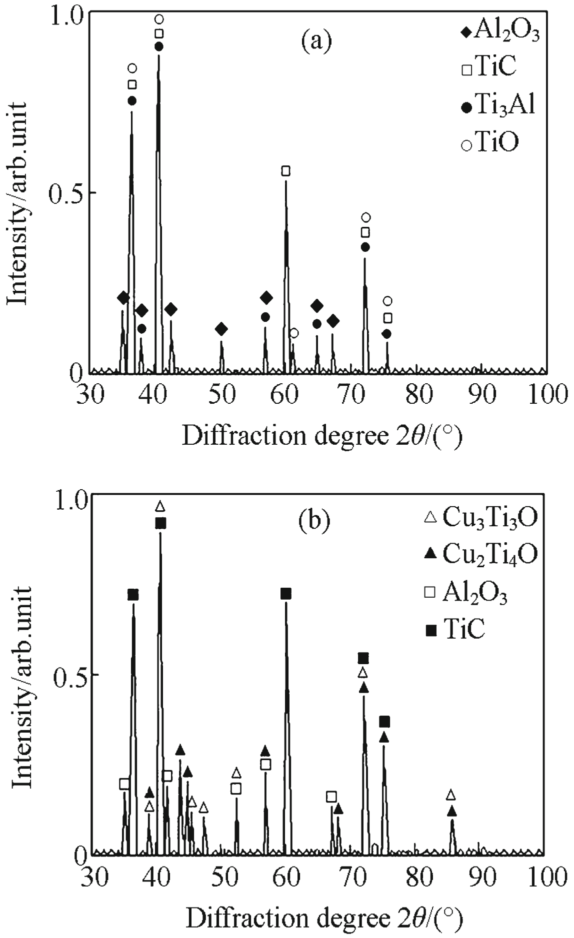

Fig. 5. X-ray diffraction results near the (a) TiC-Al_2O_3 and (b) Q235.

The XRD analysis was carried out using copper target under working voltage 60 kV and working current 40 mA. The X-ray diffraction analysis results are shown in Fig. 5. It can be seen from Fig. 5 that the phase constituents near the interface of TiC-Al_2O_3/Q235 joint were very complicated. The main peaks obtained are coming from the raw material and reaction products. Mainly, TiC-Al_2O_3 ceramics peaks are present. Various reaction products are also present, such as TiO, Ti_3Al, Cu_2Ti_4O, and Cu_3Ti_3O.

It can be concluded that Ti segregated from the molten Cu-Ti liquid phase to the TiC-Al_2O_3 surface because of its oxygen affinity [5] and reacted with O and Al from TiC-Al_2O_3 to form TiO and Ti_3Al. Once a continuous TiO and Ti_3Al thin layers were formed, it was difficult for titanium to diffuse further through the interface, therefore, the titanium concentrated in the vicinity of the TiO and Ti_3Al layer [6]. Simultaneously, O from TiC-Al_2O_3 is released and dissolved in the Cu-Ti liquid phase leading to the formation of Cu_2Ti_4O and Cu_3Ti_3O. The results indicated no Cu-Ti intermetallic compounds were detected and no Ti or Cu existed in metallic form, which suggested that Ti and Cu in Ti/Cu/Ti multilayer had reacted sufficiently. Fe-Ti brittle intermetallic compounds were also not detected on the Q235 side.

Al in TiC-Al_2O_3 diffuses towards the interface; the type of inter-metallic compounds formed between Al and Ti depends on local concentration of Al elements [7]. Al diffused from TiC-Al_2O_3 to the interface was limited, on the contrary, Ti largely congregated on the surface of TiC-Al_2O_3, leading to form Ti_3Al, not TiAl or the other inter-metallic compounds between Ti and Al.

Among all the titanium oxides, it is found that TiO is the most stable and wettable compound [5]. The Ti-O bonds that formed at the interface acted to “bridge” across the interface and join metal and ceramics [8]. Because the metallic nature of the TiO reaction layer results in a more metal-like transition between the TiC-Al_2O_3 and the Q235. Two kinds of complex oxides, Cu_2Ti_4O and Cu_3Ti_3O, were found on the fractured faces, Fig. 5b, in which, Cu_2Ti_4O and Cu_3Ti_3O are the M_6X-type compounds, which belong to the Fd3m space group [9], are isostructural with the η-phases [10] and have the Ti_2Ni-type structure stabilized by O [11]. These oxides are cubic in nature [10] and have the Fd3m space group, therefore the major peaks of both Cu_2Ti_4O and Cu_3Ti_3O more or less coincide (Fig. 5b). With regard to metallic behaviour of Cu_2Ti_4O and Cu_3Ti_3O phases, several investigators gave following explanations: because they are characterized by continuous metal lattices containing interstitial oxygen [10], Cu_2Ti_4O and Cu_3Ti_3O phases are likely to be metallic in nature, as suggested by Karlsson [9]; in Cu_2Ti_4O and Cu_3Ti_3O phases, oxygen concentration is not very high and we may expect mostly metallic behaviour [5]; Cu_2Ti_4O has a strong metallic gloss as reported by Kelkar and Carim [11], suggesting metallic bonding.

Therefore, the formation of reaction layer containing Cu_2Ti_4O and Cu_3Ti_3O phases in conjunction with a TiO layer can provide a more gradual transition and a more wettable surface that promotes bonding between the TiC-Al_2O_3 and Q235 than TiO would do alone [5, 10]. These M_6X phases, Cu_2Ti_4O and Cu_3Ti_3O, clearly play an important role in TiC-Al_2O_3/Q235 bonding. These M_6X phases, Cu_2Ti_4O and Cu_3Ti_3O, are known as the dilute ceramic compounds that are metal-rich compared to conventional ceramics, have properties intermediate between those of typical ceramics and typical metals[11] and are common transitional phases in ceramic-metal systems.

TiC and Al_2O_3 were found both on the TiC-Al_2O_3 and Q235 side, Fig. 5a, which suggests fracture initiated in TiC-Al_2O_3. TiO, Ti_3Al, Cu_2Ti_4O and Cu_3Ti_3O phases, which contain Al or O, were found on the fractured faces, which indicated fracture propagated towards the interface of TiC-Al_2O_3.

4. Conclusions

1. The interface of TiC-Al_2O_3/Q235 is continuous and straight without micro-voids, cracks and unbonded areas. The growth of diffusion and reaction layers takes place extensively into the Q235 low carbon steel but little into the TiC-Al_2O_3 composite ceramics.

2. Joints were produced with average shear strength of more than 100 MPa. The maximum joint strength was obtained reaching a value of 143 MPa. The fracture reveals brittle cleavage features. Fracture occurred in TiC-Al_2O_3 near the interface of TiC-Al_2O_3/Q235.

3. Two kinds of M_6X-type compounds of Cu_2Ti_4O and Cu_3Ti_3O were found on the TiC-Al_2O_3/Q235 interface. The reaction layer containing Cu_2Ti_4O and Cu_3Ti_3O phases in conjunction with a TiO layer can provide a more gradual transition and a more wettable surface that promotes bonding between the TiC-Al_2O_3 and Q235.

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