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Influence of surface modification of alumina on bond strength in Al_2O_3/Al/Al_2O_3 joints

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The subject of the work was to study the effect of Ti thin film on alumina ceramic on mechanical strength and fracture character of Al_2O_3/Al/Al_2O_3 joints. The joints were formed by liquid state bonding of alumina substrates covered with titanium thin film of 800 nm thickness using Al interlayer of 30 μm thickness at temperature of 973 K in a vacuum of 0.2 mPa for 5 min. The bend strength was measured by four–point bending test at room temperature. Scanning and transmission electron microscopy were applied for detailed characterization of interface structure and failure character of fractured joint surfaces.

Result analysis has shown that application of the Ti thin film on alumina leads to decrease of bond strength properties of Al_2O_3/Al/Al_2O_3 joints along with the change either of structure and chemistry of interface or of failure character.

1. Introduction

Metal-alumina joints have found various practical applications in electronic devices and high technology industry for fabrication of metal matrix composites and bonding of ceramic components to metals [1]. When bonding together alumina and metals, the main problem is to ensure reliable joints. In liquid phase bonding process, the quality of bond is often limited by wetting phenomena of ceramic by liquid metal and formation of interfacial phases due to chemical reactions between both components. Additions of alloying elements to metal interlayer or metallization of ceramic surface may affect the interaction in the Al_2O_3/Al/Al_2O_3 assemblies by changes in interfacial energy and interface structure, which influence directly both the structure formation and the kinetics of the chemical reactions occurring at the interface. Moreover, the design of strong metal–ceramic joints requires coexisting material combinations with a minimum thermal mismatch and a high failure strength of each component in order to produce joints of high reliability [2–5].

The purpose of this work was to study the effect of Ti thin film on alumina ceramic on mechanical strength and fracture character in Al_2O_3/Al/Al_2O_3 joints.

2. Experimental procedure

The tested objects were Al_2O_3/Al/Al_2O_3 and Al_2O_3/Ti/Al/Ti/Al_2O_3 joints. The Al_2O_3/Al/Al_2O_3 joints were produced by liquid state bonding of sintered alumina blocks of 14 × 14 × 20 mm dimensions and the Al foil (99.99% purity) of 30 μm thickness. The Al foil was mechanically and ultrasonically cleaned in acetone for 10 min, rinsed in alcohol and then dried with warm air directly before bonding. The alumina blocks of 3.8 g/cm^3 density (almost 0% porosity) were produced by high temperature sintering at 1923 K from the powder, containing less than 0.1% impurities (0.009% CaO, 0.053% SiO_2, 0.0029% MgO, 0.023% Fe_2O_3, 0.0036% Na_2O). The surfaces to be joined were polished with diamond suspension up to 1 μm, ultrasonically cleaned in acetone, and finally the 800 nm thin titanium layer was deposited on the surface using PVD method. The Al foil was positioned between two alumina blocks and this assembly was placed in a graphite crucible situated in a vacuum chamber. The bonding was performed at 973 K in a vacuum of 0.2 mPa for 5 min. Subsequently, the assembly was cooled slowly to room temperature at a rate of 10 K min^{-1} to avoid crackings. Similar procedure was used for joining Ti-coated and uncoated alumina blocks. To reduce the interaction during cooling in the pair of joined alumina blocks in which the ceramic surface was activated by titanium, the assembly was cooled at faster (30 K min^{-1}) rate. The standard samples for bend test of 3 × 3 × 36 mm dimensions were cut from the joints and then examined surfaces were polished up to 1 μm. The 4-point bend tests were performed at room temperature using INSTRON 1115 machine with automatic recording of the applied load versus the corresponding displacement up to failure under constant load displacement rate of 1mm/min. Five specimens were tested from each pair of joined blocks.

The interfacial structure and chemistry of interfaces were characterized by means of optical microscopy, SEM and TEM analyses on fractured joint specimens cross sectioned in a plane inclined about 25 degrees to the joint surfaces. The fracture surface of the bend samples was estimated by SEM.

3. Results and discussion

Fig. 1 shows the bend test results of the Al_2O_3/Al/Al_2O_3 and Al_2O_3/Ti/Al/Ti/Al_2O_3 joints compared to the bulk Al_2O_3 specimen. For joints, the maximum bend stress values, corresponding to the fracture strength, decrease when the alumina surface is chemically modified by deposition of Ti thin layer, showing approx. 196 ± 10 and 297 ± 10 MPa for Ti-coated and uncoated alumina, respectively, compared to 308 ± 15 MPa for bulk Al_2O_3 specimen. These results demonstrate weakening of Al_2O_3/Al/Al_2O_3 joints due to surface modification of alumina by Ti film. In our previous investigations [6, 7] we found that application of 800 nm thin Ti layer on alumina substrate significantly improves both wetting and bond strength properties of Al/Al_2O_3 and Cu/Al_2O_3 couples examined by improved push off shear test of solidified sessile drop samples produced in wettability tests. On the other hand, another study [8] has demonstrated that alloying Al with 6 wt% Ti decreases both wetting and bond strength of Al-Ti/Al_2O_3 couples. It might be suggested that under conditions used in the study, the dissolution of Ti film in thin Al interlayer leads to disadvantageous alloying with Ti and consequent decrease in mechanical properties of the joints. In the case of Al/Ti/Al_2O_3 sessile drop samples, the amount of Ti dissolved in large Al drop (relative to thin Al layer) has no effect on interaction in the system. Furthermore, the difference in thermal expansion coefficient and Young's modulus of bonded materials may contribute to an increase of residual stresses that develop during cooling after bonding [3, 9]. On the contrary, for the Al_2O_3/Al/Al_2O_3 joints produced with uncoated ceramics allow one to obtain almost an ideal joint which has the same properties as bulk alumina.

It is interesting to note that the bend stress versus deflection curves have a parabolic character with a tendency towards the exhibiting linear behavior before failure. For Al_2O_3/Al/Al_2O_3 joints, the deflection value, corresponding to the maximum fracture stress, is approx. 113 μm contrary to approx. 68 μm for Al_2O_3/Ti/Al/Ti/Al_2O_3 joints. Moreover, fracture behavior of Al_2O_3/Ti/Al/Ti/Al_2O_3 joints is typical for ductile materials, while the joints made with uncoated alumina demonstrate sudden failure after reaching the maximum value of bend stress. Representative macrographs of the failure in the Al_2O_3/Al/Al_2O_3 and Al_2O_3/Ti/Al/Ti/Al_2O_3 specimens are shown in Fig. 1. It should be emphasized that the Al_2O_3/Ti/Al/Ti/Al_2O_3 specimens fractured completely inside Al layer, what is also evidenced by the large non-linear region in the stress-deflection curve after reaching the maximum value of bend stress as compared to these specimens, which failed by failure in the ceramic. Thus, plasticity in the metal layer of Al_2O_3/Ti/Al/Ti/Al_2O_3 joints is the valid parameter determining the integrity and lifetime of ceramic/metal/ceramic layered structure as discussed in [10, 11].

The structural and chemical characterization of Al/Ti/Al_2O_3 interface by means of optical microscopy (Fig. 2) and electron microscopy equipped with spectrometric system for microanalysis of chemical composition (Fig. 3) suggest that the negative effect of Ti layer on bond strength properties in the Al_2O_3/Al/Al_2O_3 joints results from unfavorable change of the structure and chemistry of the interface (mainly due to the

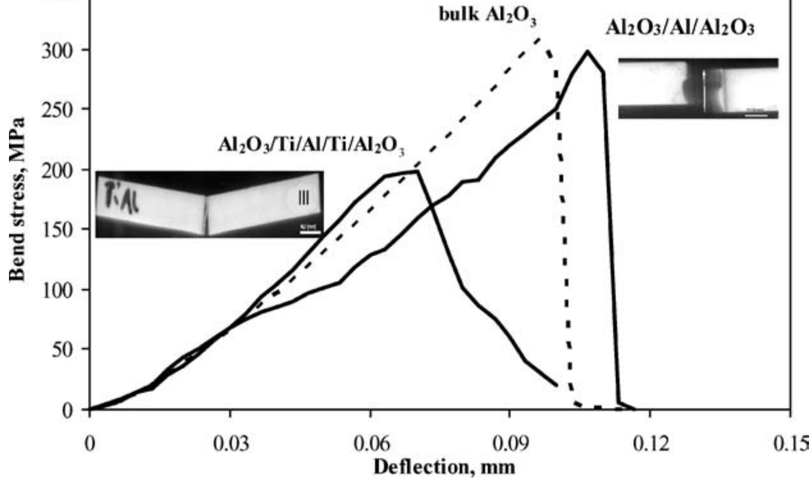

Figure 1 Bend test curves recorded for Al_2O_3/Al/Al_2O_3 and Al_2O_3/Ti/Al/Ti/Al_2O_3 joints compared to bulk Al_2O_3 specimen.

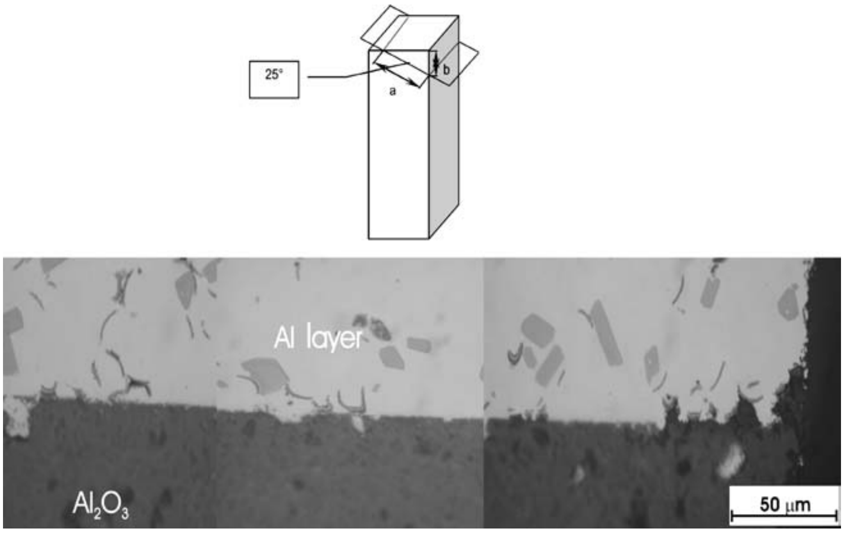

Figure 2 Microstructure of Al/Ti/Al_2O_3 interface after bonding, will corresponding scheme of cross section.

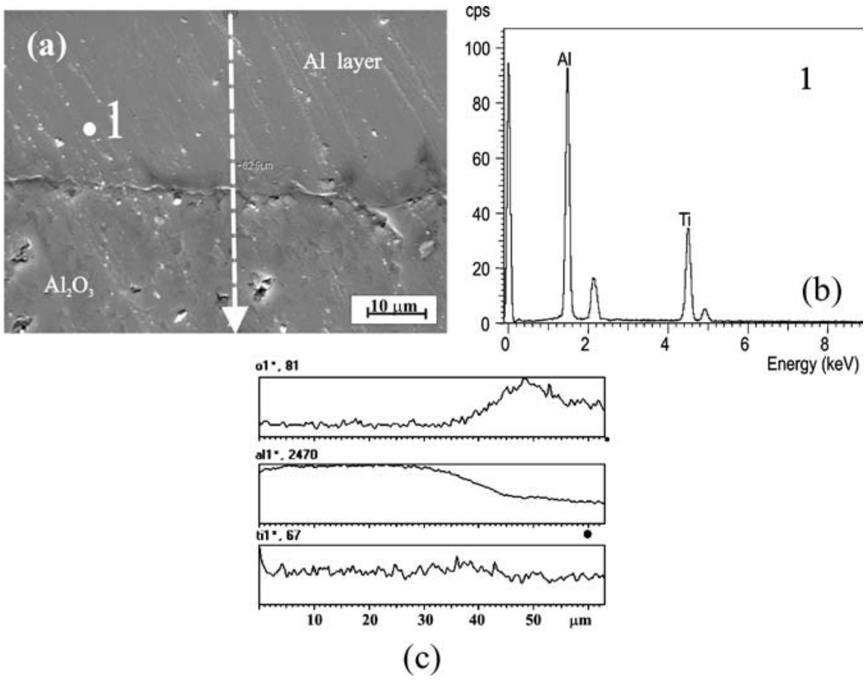

Figure 3 SEM results for characterization of the Al/Ti/Al_2O_3 interface: (a) general image, (b) EDS spectrum taken from point 1, (c) distribution of oxygen, Al and Ti along white line as marked in (a).

dissolution of Ti in molten Al leading to overalloying Al with Ti). The last statement was confirmed experimentally by TEM studies of interface in Al/Ti/Al_2O_3 system, because the presence of Al_3Ti precipitates noted in the region of the interface demonstrates the formation of Ti–rich alloy in the layer of the drop contacting Ti–coated substrate (Fig. 4).

The fracture surface of the Al_2O_3/Al/Al_2O_3 and Al_2O_3/Ti/Al/Ti/Al_2O_3 specimens after bend tests were observed by SEM (Figs 5 and 6). Fractographic analysis showed that the presence of Ti film affects the failure character of the joints. For Al_2O_3/Al/Al_2O_3 joints, the failure occurred by brittle cracking in the alumina near the interface (Figs 1 and 5). On the contrary, for Al_2O_3/Ti/Al/Ti/Al_2O_3 joints, the failure occurs in metal (Figs 1 and 6) either: (1) by ductile rupture of metal (void growth and coalescence- region A), (2) or by brittle rupture in metal (region B). This fracture type corresponds to the lower strength of the bond. Comparison of these results with our previous report on Al/Ti-coated Al_2O_3 couple fabricated in wetting experiments shows two important differences: (1) microstructural examination of the interface in the sessile drop samples did not indicate the presence of thick layer composed principally of the Al_3Ti precipitates in the region of the interface as in joined alumina blocks samples, (2) the positive effect of substrate surface modification by Ti coating on the wetting and bond strength properties of sessile drop samples was related with dissolution of Ti in molten Al, while good wetting in the couple improved its bonding. Thus, it may be suggested that dimensional factor plays an important role in interaction between metallized ceramics and liquid aluminium interlayer.

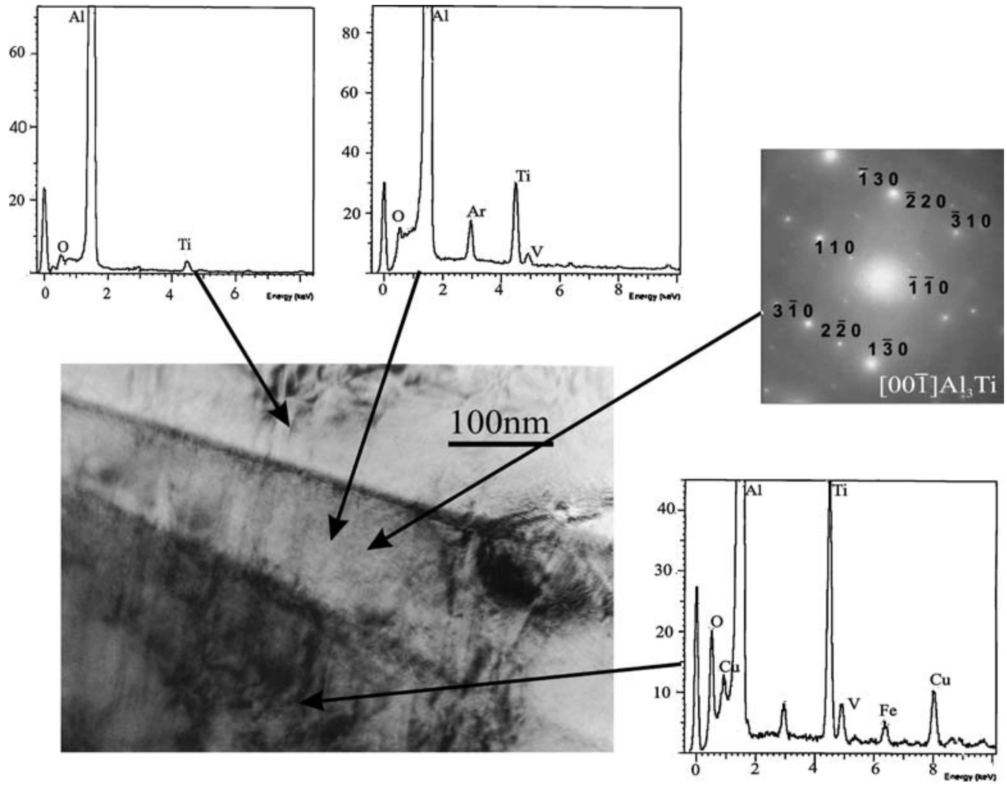

Figure 4 TEM analysis of $Al/Ti/Al_2O_3$ interface with corresponding EDS spectra taken from different places of the reaction products region; representative selected area diffraction pattern indicates the formation of Al_3Ti phase.

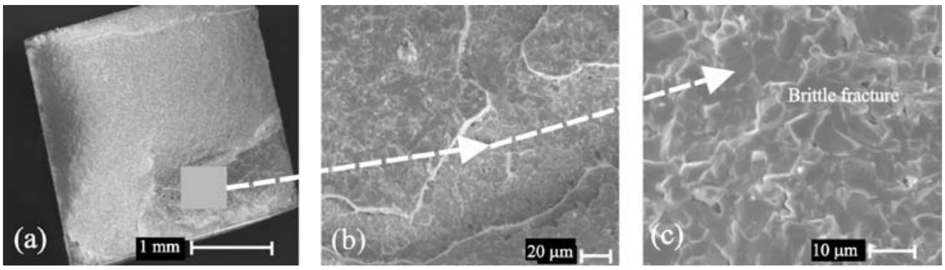

Figure 5 SEM micrographs of fracture surface of $Al_2O_3/Al/Al_2O_3$ joint after bend test: (a) general view, (b, c) under magnification as marked.

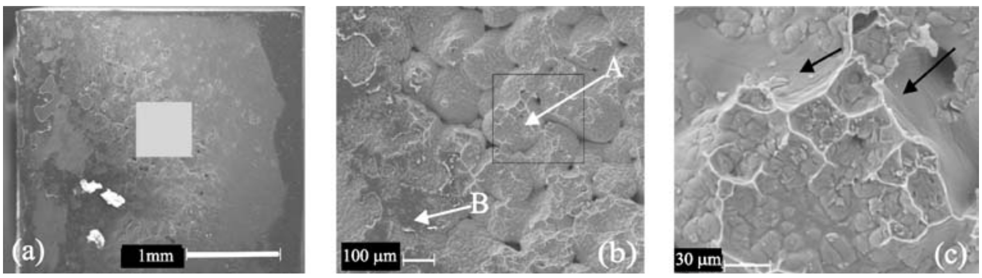

Figure 6 SEM micrographs of fracture surface of $Al_2O_3/Ti/Al/Ti/Al_2O_3$ joint after bend test: (a) general view, (b) area marked in (a) under magnification, (c) area marked in (b) under magnification. A—ductile rupture, B—brittle rupture, black arrows indicate void growth and coalescence.

4. Conclusions

Joining Ti-coated alumina by liquid state bonding with Al interlayer allows obtaining reliable joints in which the joining material is well adhered to the Al_2O_3 surface and the maximum bend stress value, corresponding to the fracture strength is approx. 200 MPa.

However, the joints produced with Ti-coated alumina blocks are weaker than those with uncoated ones. Compared to model sessile drop samples of Al/Al_2O_3 and $Al/Ti/Al_2O_3$ systems, the joints produced with alumina blocks demonstrate higher sensitivity to dimensional factor, showing different effects of ceramic substrate metallization on mechanical properties of interfaces.

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