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Effect of Nb + Ti coating on the wetting behavior, interfacial microstructure, and mechanical properties of Al/Al_2O_3 joints

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Abstract The subject of the work was to study the effect of Nb + Ti thin film deposited by PVD method on alumina substrates on the wetting behavior, bond strength properties, and structure of interface in the Al/Al_2O_3 joints. Applying the sessile drop method, the wetting behavior of molten Al (99,999%) on coated alumina substrates was studied in the temperature range between 953 and 1373 K under a vacuum of 0.2 mPa for 30 min of contact. The sessile drop samples were used to examine the interface structure, shear strength, and interfacial fracture toughness under the concentrated load. The introduction of the thin Nb + Ti film layer of 900 nm thickness: (1) greatly improves the wettability of alumina by molten Al at above 1223 K and the shear strength of Al/Al_2O_3 joints produced at 1223 K, (2) has positive effect on structure transformation in the interface and leads to fabrication of reliable metal–ceramic joints. Microstructural investigations of the interface indicated that the precipitates of Nb and Ti-rich intermetallic phases were formed at the Al/Al_2O_3 interface, which influenced strengthening of these joints. Hence a conclusion can be drawn that the interface structure influences the durability increase in Al/Al_2O_3 joints.

Introduction

Problems regarding the wetting behavior, structure, and mechanical properties of metal–ceramic joint interface are of great practical importance for the technology by which composite materials and metal–ceramic joints are fabricated [1, 2]. So far, several techniques have been mastered to produce joints between alumina ceramics (Al_2O_3) and metals. In practical applications, the most popular are vacuum techniques of solid state bonding, i.e., diffusion welding [3, 4], liquid state bonding with active solders [5, 6], and "SQ brazing"—utilizing squeeze casting [7]. When different materials are joined together, the technology used most often is liquid state bonding, where the main requirement is good wettability of the ceramic surface by liquid metal and the residual stress relaxation response of metal–ceramic joint.

Numerous works have been carried out to investigate the possibilities of improving the wettability [8, 9] and strength of Al/Al_2O_3 joints through addition of various alloying elements to aluminum (e.g., Cu, Ag, Au, Mg), or by chemical modification of the substrate surface through application of metallic coatings (e.g., Ti, Cu, Au, Ni) onto the Al_2O_3 ceramics. The effect of Al_2O_3 surface metallization with titanium and niobium on an improvement of the alumina wettability by liquid aluminum was discussed in [10, 11]. The low-temperature wettability of Al/Al_2O_3 system obtained by this technique results from high solubility of the coating material in liquid aluminum, producing strong chemical interaction due to the formation of transient diffusion layers, improving the adhesive power of metal–ceramic interface and physico-chemical compatibility of different bond components, which are solid solutions of titanium and niobium in aluminum, and local Al_3Ti and Al_3Nb phases. From the data available in technical

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literature it follows that the use of titanium as an alloying addition to aluminum does not improve the wettability and reduces the strength of an Al/Al_2O_3 joint [12]. The drop in bond strength is due to the presence of numerous brittle precipitates of the Al_3Ti phase and voids that occur in the joint interface area.

Therefore, the aim of the study was to investigate what effect the thin layer of Nb + Ti deposited on the surface of ceramic material may have on the wettability, strength, and structure of an Al/Al_2O_3 joint interface.

Materials and methods

The following materials were used in the investigations: aluminum (99.9999 wt%) and 98% pure polycrystalline alumina acting as a substrate, fabricated at the Faculty of Materials Science and Ceramics, University of Science and Technology in Cracow, by sintering at the temperature of 1923 K, powders of the following starting composition: 99.9085% α-Al_2O_3, 0.009% CaO, 0.053% SiO_2, 0.0029% MgO, 0.023% Fe_2O_3, and 0.0036% Na_2O. The ceramic substrate contained 2% of Mg(SiO_4) and $MgAl_2O_4$ spinels. The alumina substrate was characterized by the following physical and mechanical properties: porosity almost 0%, density 3.8 g/cm^3, Young's modulus 300 GPa, Poisson ratio 0.24.

To produce a bond between the ceramic substrate and metal, a wettability experiment was carried out by the sessile drop method. The wettability studies were carried out in the temperature range of 953–1323 K for 30 min of contact under dynamic vacuum of 0.2 mPa, the oxygen partial pressure $P(O_2)$, measured by a Prisma QMS residual gas analyzer, was found to be in the order of 10^{-8} mbar for a dry atmosphere. Every tested object was introduced to the furnace and heated at a rate of 10 K/min from room temperature to the required experimental temperature. After wettability examinations, the Al/Al_2O_3 couple was cooled at a rate of 10 K/min to reduce the effect of thermal stresses.

Metal specimens in the form of $\phi 5 \times 4$ mm rods weighing 0.20 g were used in the tests. The samples of Al_2O_3 substrates of $\phi 17 \times 5$ mm dimensions were polished with diamond paste of 1 μm grain size to an average roughness $R_a = 0.3$ μm. As a next step, the films of Nb + Ti were deposited by rf sputtering of a mosaic Nb + Ti target in argon gas atmosphere onto different substrates, such as Corning glasses and alumina, depending on the requirements imposed by further applications, i.e., the determination of the film thickness and composition. The film thickness was measured mechanically by means of a Talystep profilometer. The final thickness of the film was found to be in the order of 900 nm. The thin film composition was determined from X-ray diffraction (XRD) patterns. The weight ratio of Nb/(Nb + Ti) was found to be equal to 0.625 (62.5 wt%). The specimens of both metal and ceramics without the deposited films were washed for 5 min in an ultrasonic cleaner directly before being placed in a vacuum apparatus.

On joint specimens examined by the sessile drop method, the bond strength was next tested by a shear method (modified push-off technique). The solidified sessile drop specimens are carefully bisected perpendicular to the drop–substrate interface at the mid-plane of the contact circle by means of Accutom 50 automatic precision cut-off machine and the shear test is done on one half of the specimen. Additionally, the other half of the same bisected droplet may be used to carry out detailed structure and chemistry characterization of interfaces formed under conditions identical to the corresponding wettability test [13]. The structure was examined on specimens after the wettability test (metal drop–ceramic substrate couple) using light microscopy, scanning electron microscopy, and transmission electron microscopy. The examinations of structure were restricted to the areas adjacent to metal–ceramic interface. The bond structure morphologies present in the interface were determined by scanning microscopy on a STEREOSCAN 420 scanning microscope with analysis of the metal–ceramic interface chemical composition done on an EDS LINK ISIS 300 X-ray microanalyzer. The application of X-ray microanalysis enabled the linear, local and surface distribution analyses to be carried out in the examined microregions of metal–ceramic interface. Detailed structural examinations of the joints were also carried out on a JEOL 2010 ARP transmission electron microscope equipped with energy dispersive X-ray spectrometer (EDS). The TEM examinations of the structure of metal–ceramic joints were made on thin foils prepared in a Gatan PIPS691V3.1 ion thinner by standard methods, i.e., by cutting out 3 mm diameter discs, thinning by dimpler, and ion polishing [14].

The hardness at the interface of the joints was measured using the Hanemann Micro-Vickers hardness tester installed on a Neophot 2 microscope under a vertical loading from 5 to 10 G. Hardness was measured along the metal drop–ceramic substrate interface at a distance of 5 μm from the border of the joint. For every sample 5–7 measurements were taken (the measurements of the hardness of the new precipitated phases were not taken into consideration).

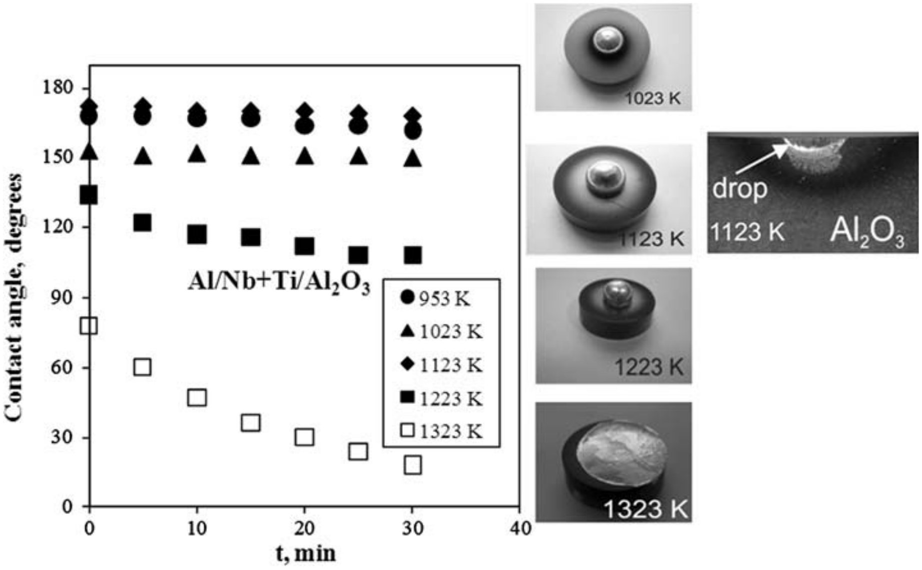
The bond strengths were determined using an INSTRON 1115 machine with automatic recording of the applied load versus the corresponding displacement up to failure under constant displacement rate of 1 mm min^{-1}. The shear strength was calculated dividing the applied load by the initial substrate interfacial area measured under 10× magnification using an AMSLER magnifying system.

Fracture toughness of the Al/Nb + Ti/Al_2O_3 system produced at $T = 1223$ K was also investigated under the effect of concentrated force (of 6 kN value) exerted by a ball-shaped indenter of the hardness tester [15].

Results and discussion

Figure 1 shows the wetting kinetics of Al/Nb + Ti/Al_2O_3 system at different temperatures, i.e., at 953, 1023, 1123, 1223, and 1323 K during 30 min of contact. Within the temperature range of 953–1223 K, the contact angle is larger than 90°. On the other hand, at $T = 1323$ K, after 30 min of contact, the contact angle is 18°, which proves good wettability and intense chemical reaction proceeding at that temperature. It is worth noting that, applied onto ceramic material, the Nb + Ti coating has improved the wettability at high temperature (1323 K) and, compared to the base system (Al/Al_2O_3) at the same temperature, reduced contact time necessary to obtain this wettability [13]. The main reason accounting for the lack of low-temperature wettability in the examined system is the presence of a continuous and compact oxide coating, formed on the surface of an aluminum drop and acting as a natural barrier in achievement of true contact between the aluminum drop and substrate, thus contributing to an increase of the contact angle values. In an Al/Al_2O_3 system, the temperature increasing in vacuum (especially above 1143 K) destroys the coating continuity or even removes it totally, mainly as an outcome of several processes occurring simultaneously: (1) oxide coating solving in liquid aluminum, (2) mechanical destruction of the coating continuity due to differences in the coefficients of thermal expansion of Al and Al_2O_3, and (3) the formation of gaseous products of Al_2O reaction [16]. In a basic system of Al/Al_2O_3, within the investigated range of wettability temperatures, i.e., 953–1050 K, the contact angle decreases gradually with increasing temperature and time of contact [13]. On the other hand, in a system including the coating, i.e., Al/Nb + Ti/Al_2O_3, the wetting behavior, especially in the temperature range of 1023–1123 K, is of a much more complex nature. After 30 min of contact, the contact angle at $T = 1023$ K is smaller than it is at $T = 1123$ K and has the value of 150° and 168°, respectively. A similar temperature–contact angle relationship at relatively low temperatures was observed in an Al/Nb/Al_2O_3 system [17]. The examinations of wettability at $T = 1123$ K in an Al/Nb + Ti/Al_2O_3 system revealed on the Al_2O_3 substrate surface the presence of typical "reaction rings"; their number was most probably corresponding to the number of Al drop contractions (Fig. 1). The authors of Ref. [18] suggested that in an Al/Al_2O_3 system, the phenomenon of the drop spreading and shrinking during the wettability test was caused by aluminum dissolving the Al_2O_3 substrate. The dissolution of the Al_2O_3 substrate results in the formation of dimples at a ternary bond point. In spite of the fact that under these experimental conditions, carried out in a system of Al/Nb + Ti/Al_2O_3, the negative effect of oxide coating on the surface of an Al drop has been considerably mitigated, the main layers of an oxygen-rich metal are forming at the interface. The precipitates of the fine crystals of Al_2O_3 are well visible on a cross-section at large magnifications (Fig. 2b), while the Nb + Ti film formed on Al_2O_3 substrate is of an almost continuous character (Fig. 2a).

Fig. 1 The wetting kinetics of Al/Nb + Ti/Al_2O_3 system



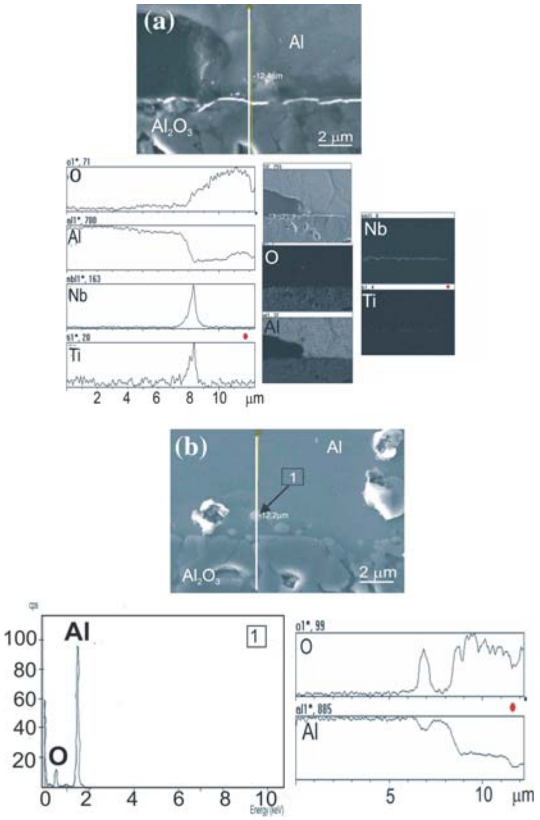


Fig. 2 SEM analysis of the Al/Nb + Ti/Al_2O_3 system interface after wettability test at $T = 1123$ K, **a** the peripheral part of the interface with distribution of the oxygen, aluminum, niobium, and titanium along *white line*, and oxygen, aluminum, niobium, and titanium X-ray maps; **b** the central part of the interface with corresponding EDS spectra taken from marked area point *1*—Al_2O_3 crystals in the drop near the interface, and distribution of the oxygen and aluminum along *white line*

The results of the structural examinations carried out in the region of Al/Nb + Ti/Al_2O_3 bond by the method of optical microscopy, combined with SEM and TEM, are shown in Figs. 3, 4, and 5. Observations under a magnification of 500× revealed in the area of bond interface the presence of new and discontinuous phases, marked in the microstructural images with arrows (Figs. 3a, 4a). On the other hand, structural examinations of metal–ceramic interface carried out at large magnifications (Figs. 3b, 4b, c) have proved the formation of new phases containing Nb, Ti, and oxygen. Local analysis of the chemical composition of the above-mentioned precipitates has indicated that these are the precipitates of $Al_3(Nb,Ti)$ (e.g., items 2 and 1 in Figs. 3b, 4b, respectively). This statement is consistent with the results of analysis of the Al–Nb–Ti phase equilibrium diagrams [19] based on the reaction at 1273 and 1373 K and of the precipitates of Al_2O_3 (items 3 and 2 in Figs. 3b, 4b, respectively). Here it is worth noting that, because of difficulties in correct estimation of the chemical composition of an Al drop after its interaction with the Nb + Ti film coating, it has been assumed that the thin film layer (about 900 nm thick) results in the actual Nb + Ti content lower than 1 wt%. The structural analysis of the Al/Nb + Ti/Al_2O_3 bond interfaces produced at the temperatures of 1223 and 1323 K indicates that the wettability effect at $T = 1323$ K has been accompanied by very intense chemical interaction between the Nb + Ti film-coated substrate and liquid aluminum. The mechanism of this interaction is probably due to dissolution of the Nb + Ti coating in liquid aluminum. The result is diffusion and, as a consequence of this diffusion, a reaction between aluminum and the metallic coating, resulting in the formation of new phases in the form of large, block crystals. On the other hand, in bonds obtained at lower temperature ($T = 1223$ K), the precipitates of Al_2O_3 and fine precipitates of phase rich in Nb and Ti are predominant at the metal–ceramic interface.

High-accuracy structural examinations carried out by high-resolution electron microscopy, where, applying the EDS technique, local chemical composition was examined in the most interesting structural elements of metal–ceramic interface, revealed partial dissolution of Nb + Ti coating in liquid aluminum and formation by reactive diffusion of new reaction products in the form of Nb- and Ti-rich phase precipitates. Phases rich in both Nb and Ti were present in the form of highly dispersed precipitates of the size between 10 and 100 nm (Fig. 5); occasionally precipitates of the size from 100 to 300 nm appeared as well. It has to be remembered that, within an area of the interface, the investigations are carried out locally, and so they do not reflect the complex phase chemical composition of an Al/Nb + Ti/Al_2O_3 joint interface. Probably, in the area of an interface, the compounds of Al_3Nb and Al_3Ti, are present, the fact which does not exclude the existence of complex $Al_3(Nb,Ti)$ phases. The possibility of an occurrence of complex phases is due to the effect of interdiffusion, high temperature, and fluctuations in the chemical composition during wettability test. A characteristic feature of the bond microstructure is the presence of separate, isolated particles of Al_2O_3 oxide (Fig. 5b). At this point of the investigations, a hypothesis can be put forward that Al_2O_3 oxide nucleates within the interface area in the presence of oxygen originating from the dissolved oxide coating deposited on Al drop and from the surface of the ceramic material. Oxygen originating from the oxide coating present on Al drop and/or from the surface of the ceramic material is entrapped in the area of the interface, and its presence may result in the achievement of local thermodynamic equilibrium, which promotes the formation of Al_2O_3 oxide. Due to these changes, oxygen is sucked off, and its absence in the interface area arrests the reaction, resulting in the formation and growth of oxides. This

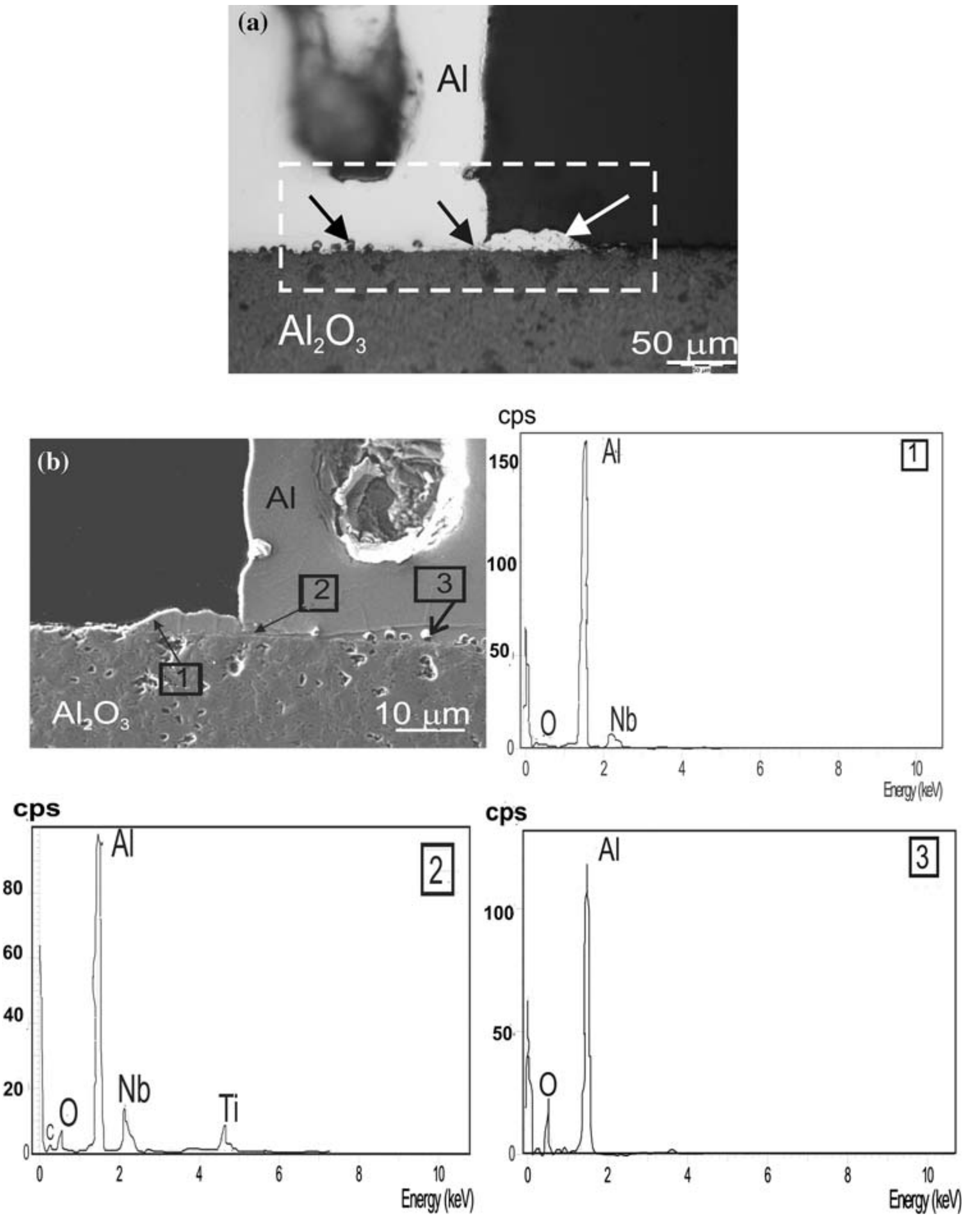


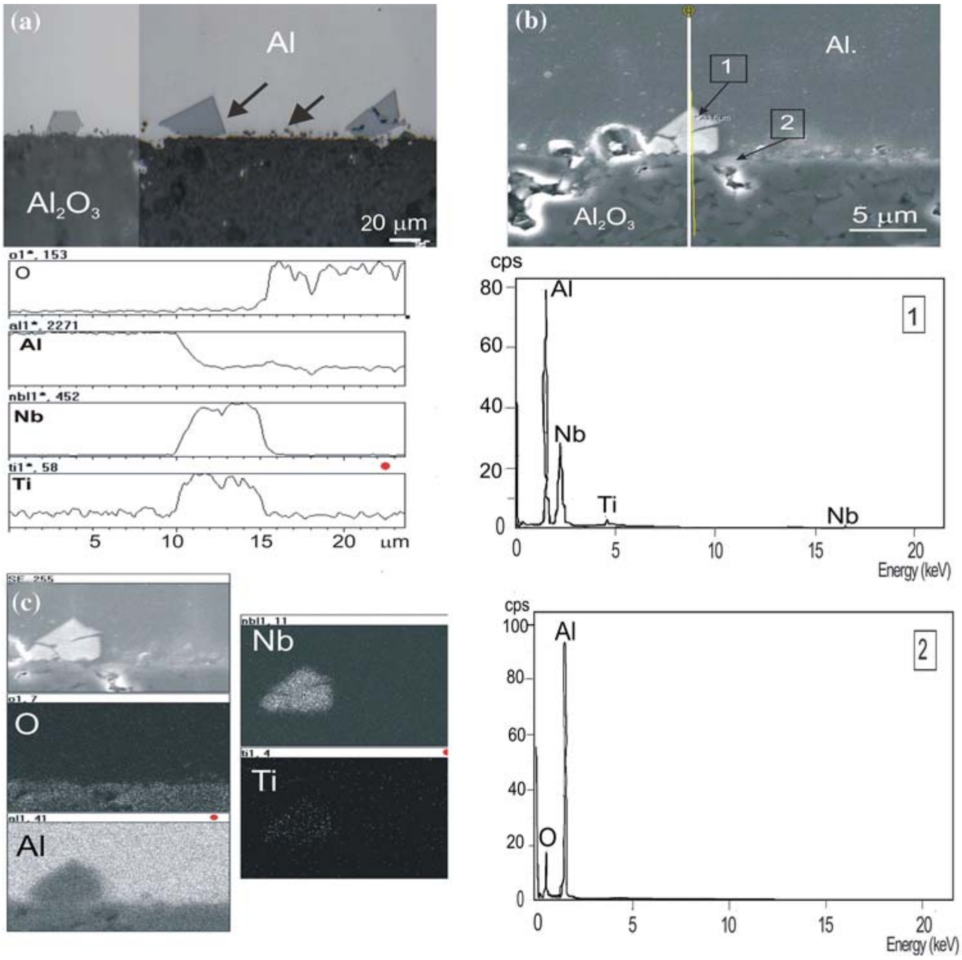
Fig. 3 **a** Microstructure of the Al/Nb + Ti/Al_2O_3 system interface after wettability test at $T = 1223$ K, **b** scanning micrograph of interface with EDS analysis at points *1*, *2*, and *3*

is why the composition and morphology of phases formed at the Al/Nb + Ti/Al_2O_3 bond interface depend on temperature, pressure, atmosphere, contact time, and partial oxygen pressure $P(O_2)$ [8].

Microhardness measurements taken by Vickers method in the interface area of Al/Nb + Ti/Al_2O_3 bond obtained after the wettability tests at different temperatures have proved that, in the range of $T = 1023$–1323 K, hardness in this area reaches higher values with increasing temperature of the wettability test. And so, at a distance of 5 µm from the metal–ceramic interface, the mean hardness values amounted to 238, 263, and 338 µHV0.005 at temperatures of 1023, 1123, and 1323 K, respectively, and to 640 µHV0.01 at a temperature of 1223 K. It should be mentioned here that the values of microhardness for a bond with Nb + Ti coating are higher than the values obtained for a bond produced at the same temperature but without modification of the Al_2O_3 substrate surface. And so, in Al/Al_2O_3 and Al/Nb + Ti/Al_2O_3 bonds produced at $T = 1223$ K, the mean values of hardness in the boundary contact zone amount to 60 and 640 µHV0.01, respectively. The application of Nb + Ti coating onto an Al_2O_3 substrate results in obvious hardening of the metal–ceramic interface at $T = 1223$ K.

The results of a shear test, carried out by modified *push-off* technique on an Al/Nb + Ti/Al_2O_3 system, are compared in Fig. 6 as a shear stress–displacement relationship at different wettability test temperatures. Bonds between the metal and ceramic substrate obtained at a temperature above $T = 1023$ K are of a permanent character.

Fig. 4 **a** Microstructure of the Al/Nb + Ti/Al_2O_3 system interface after wettability test at $T = 1323$ K; **b** scanning micrograph of interface with EDS analysis at points *1* and *2*; **c** oxygen, aluminum, niobium, and titanium X-ray maps



Maximum values of the shear stresses after wettability tests carried out at the temperatures of 1123, 1223, and 1323 K amount to 23, 58, and 22 MPa, respectively. Compared with basic system [13], the values of the maximum shear stresses are higher for the Al/Nb + Ti/Al_2O_3 bonds only at the wetting test temperature equal to 1223 K. From the run of the shear test curve plotted for an Al/Nb + Ti/Al_2O_3 bond produced at $T = 1223$ K it follows that the hardening effect has been proceeding more rapidly in the shear zone (short range flow on the curve). The reason is most probably the presence of fine precipitates of Nb- and Ti-rich phases, having more prominent effect on hardening of this zone, as well as the presence of Al_2O_3 crystals that adhere more strongly to the interface, thus improving the general adhesive behavior of a metal–ceramic system. The microstructural examinations enabled the formulation of a conclusion that the higher is the test temperature, the larger are the dimensions of the Al_3(Nb,Ti) precipitates. Particularly intense effect was observed at the peak test temperature of 1323 K. At this temperature, large and thick layers of the precipitates of Al_3(Nb,Ti) were observed to form in the area of an interface. This proves that at higher temperatures the diffusion processes proceed much more rapidly in the interface area. Changes observed in the interface area structure at $T = 1323$ K affect in a significant way the drop of shear strength in an Al/Nb + Ti/Al_2O_3 joint. At 1223 K, the visible precipitates of an Al_3(Nb,Ti) phase occupy much smaller areas of the interface than the structure of bond interface produced at a higher temperature of 1323 K. Higher temperature of the wettability test favors the formation of large precipitates of an Al_3(Nb,Ti) phase, distributed along the interface. The nucleation and growth of an Al_3(Nb,Ti) phase is the additional factor that weakens the metal–ceramic bond and results in stress concentration within an area of the Al/Nb + Ti/Al_2O_3 joint interface. Numerous cracks observed on the surface of Al_3(Nb,Ti) precipitates are also caused by the internal stresses due to volume shrinkage. This type of structure observed in an Al_3(Nb,Ti) phase is one of the reasons that account for the drop of shear strength in Al/Nb + Ti/Al_2O_3 bonds formed at the highest temperature. Lu et al. [20] in their investigations also observed the presence of numerous voids at the Ti–Al/Al_2O_3, interface, accompanying the formation of an $AlTi_3$ phase.

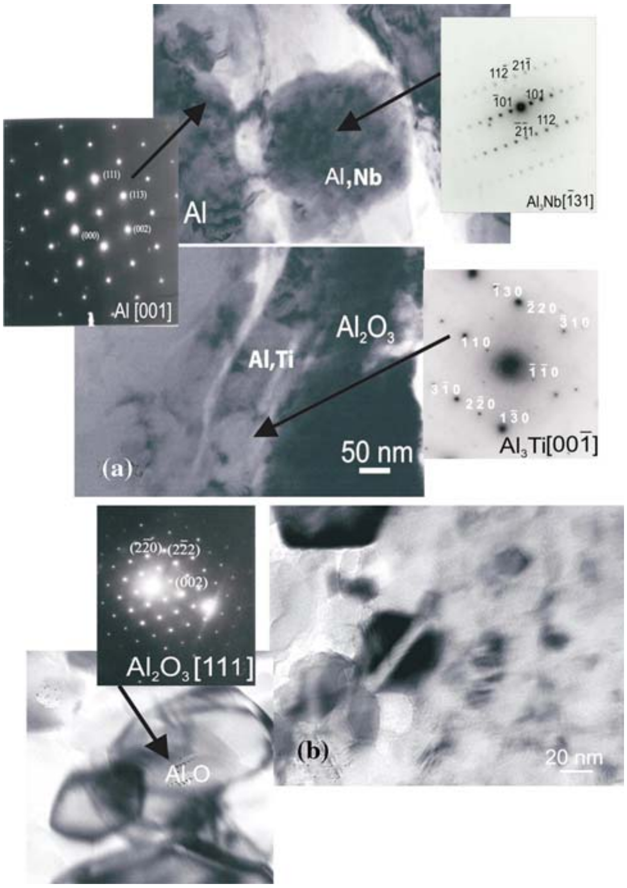


Fig. 5 TEM analysis of the Al/Nb + Ti/Al_2O_3 system interface after wettability test at: **a** $T = 1223$ K, **b** $T = 1323$ K

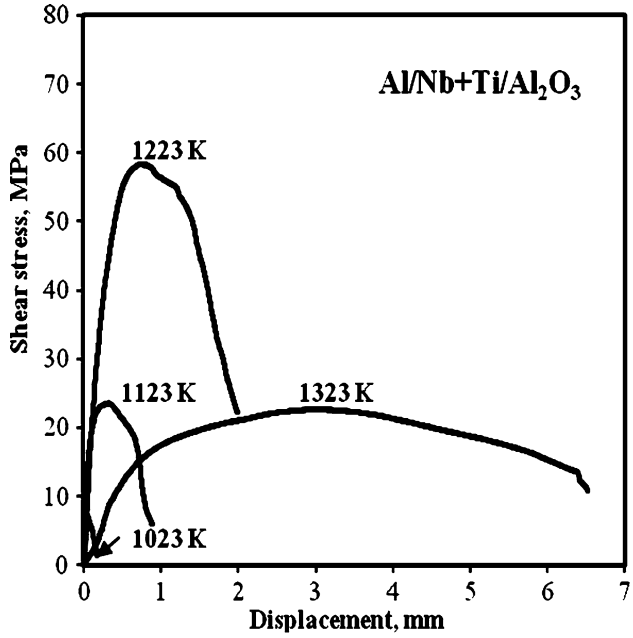


Fig. 6 Shear stress as a function of displacement for the Al/Nb + Ti/Al_2O_3 system after wettability test at different temperatures

Therefore, the large and hard precipitates of Nb- and Ti-rich phases embedded in soft aluminum matrix, obtained in a bond produced at $T = 1323$ K, are not so effective in hardening of the interface as the fine and distributed in a more uniform manner precipitates of phases rich in Nb, Ti, and oxygen are. Summing up it can be observed that it is not always necessary to produce a bond between materials characterized by very high wettability; sometimes it is enough to have a bond in the zone of interface, characterized by high strength and optimum wettability.

Fracture toughness tests, carried out on Al/Al_2O_3 and Al/Nb + Ti/Al_2O_3 systems after the wettability test at the same temperature of 1223 K, applying the effect of concentrated force of the hardness tester indenter, revealed satisfactory values of this parameter in bonds of both types. In the bond with Nb + Ti coating, small cracks were observed to appear near the indentation, but without any tendency to penetrate inside the examined material (Fig. 7b). No cracks or damages were observed to be present along the bond length. This proves good metal-substrate adhesive behavior.

Tests and comparison of the characteristic features of the layered Al/Nb + Ti/Al_2O_3 and Al/Al_2O_3 systems indicate that there are no significant differences in the properties of both systems, except for the crystals of new phases precipitating at relatively high temperatures of the metal–ceramic interface and a near-interface increase of microhardness. The presence of complex phases rich in Nb and Ti (niobium and titanium precipitates of new phases*)* proves the occurrence of reactive diffusion during the process of material-bonding, consequently resulting in the precipitation hardening of contact zone. The precipitates of new phases play an important part in conferring simultaneously both hardness and ductility to the interfacial zone. Good plastic properties of the contact zone accompanied by strong hardening are due to the character of the precipitates/matrix interface, to the deformability of precipitates, and to the slowed down propagation of cracks in the area of high residual stresses close to the interface.

Conclusions

The conclusions from the experiments can be summarized in the following items:

1. Modification of the Al_2O_3 substrate surface through application of a thin film of Nb + Ti (900 nm thick):
   - cannot improve the wettability of alumina by liquid aluminum in the temperature range of 953–1223 K, but it can improve the wettability at $T = 1323$ K. It was found that the thin film actively reacted with aluminum over 1223 K and

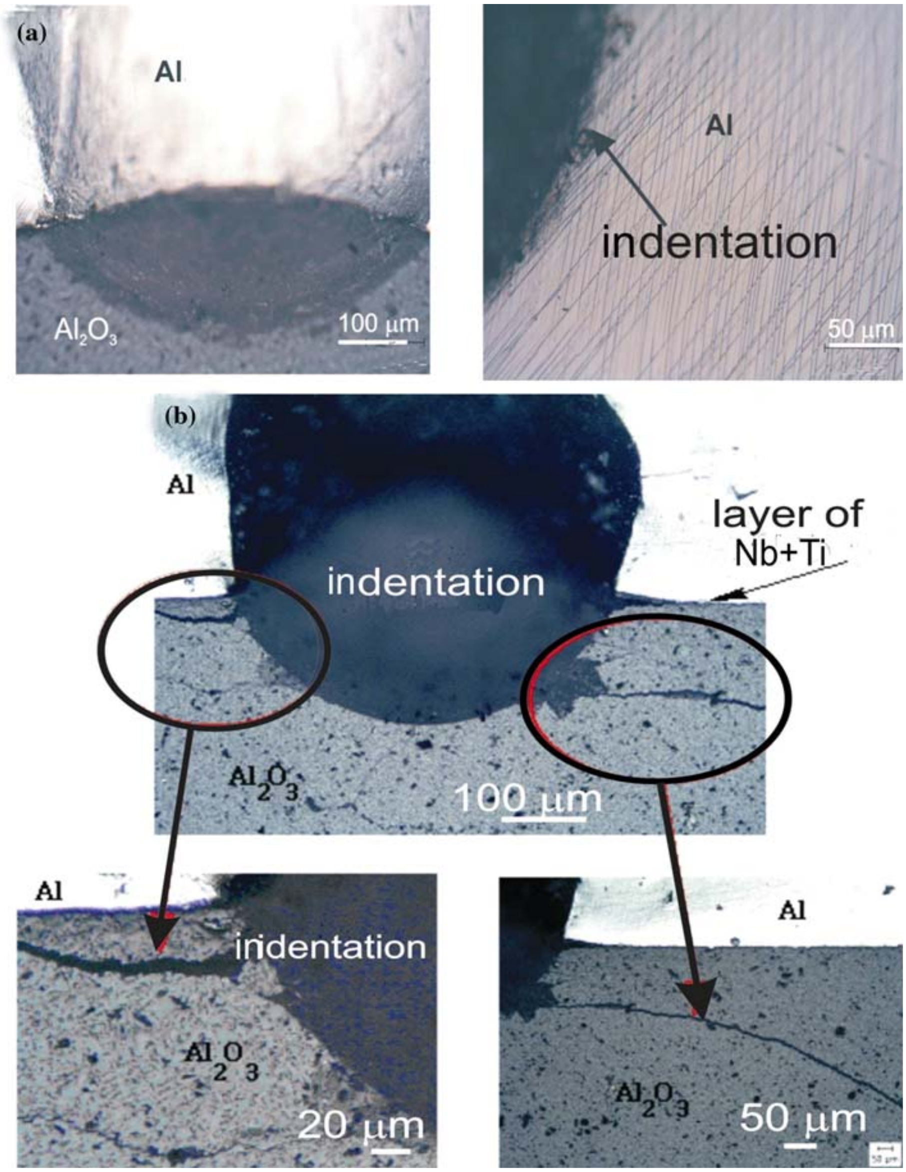


Fig. 7 Fracture toughness behavior: indentations obtained under the effect of concentrated force of the hardness tester indenter in bonds produced at $T = 1223$ K: **a** Al/Al_2O_3, **b** $Al/Nb + Ti/Al_2O_3$

Nb- and Ti-rich phases as well as the precipitates of aluminum oxide were formed at the interface,

- increases hardness of the Al/Al_2O_3 contact zone within the range of the investigated wettability test temperatures. The bonds with coating are characterized by good adherence to the substrate, resulting from their diffusive character, confers high mechanical strength to the bond at a temperature of 1223 K, resulting from the structure of the joint area, containing fine precipitates of $Al_3(Nb,Ti)$ phases.

2. The structure of the joint interface area changes with temperature. The examinations of bond interface structure carried out by optical microscopy, scanning microscopy, and transmission microscopy on an $Al/Nb + Ti/Al_2O_3$ system have revealed the dissolution of Nb + Ti coating in liquid aluminum, accompanied by the formation of new phases rich in Nb and Ti and the precipitation of $Al_2O_{3,}$ crystals, which play an important part in simultaneous hardening and plastic softening of the interface, a consequence of which is the bond of superior quality.

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