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Interfacial Phenomena and Bond Strength in Joining AlN Ceramics Using Metals Foils†

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Abstract

The solid state reaction between AlN ceramic and metals foils such as vanadium and titanium was studied between 1323 K and 1773 K for times ranging from 0.3 ks to 72 ks. Reaction couples consisting of AlN/Nb and AlN/Ni were also studied. Bonding was carried out in vacuum, in the shape of discs, maintained in contact under a pressure of 7.2 mPa. Titanium reacts with AlN at 1323 K by forming a thin nitride layer composed of TiN, beside which another reaction layer composed of Ti_3Al existed. The complete diffusion path was observed at 1473 K after 7.2 ks having a sequence of AlN / TiN / Ti_3AlN / Ti_3Al / Ti. The nitride layer grew slowly with bonding time, on the other hand, the Ti_3Al layer grew following Fick's law with an activation energy of 146 kJ/mol. A maximum bond strength of 128 MPa was obtained in a joint bonded at 1473 K after 28.8 ks. The phase reaction in the AlN/V couples started at a temperature of 1573 K. Formation of V(Al) and V_2N controls the interface joining of the AlN/V joints. A complete diffusion path could be predicted before 0.9 ks at 1573 K, following a sequence of AlN / V(Al) / V_2N / V. A maximum bond strength could be obtained for a joint bonded at 1573 K after 5.4 ks having a structure of AlN/V(Al)/V_2N+V. No ternary compounds could be observed at the AlN/V joining interfaces. Joining AlN using Nb foils requires high temperature(1673 K or above). Nb_2Al, Nb_2N, and Nb_3Al_2N were observed at the interface between AlN and Nb. A complete diffusion path was established at 1673 K for 7.2 ks, having a sequence of AlN / Nb_3Al / Nb_3Al_2N /Nb_2N/Nb. Upon increasing the bonding ttemperature to 1773 K, cracks and voids which reduce the bond strength are usually found. A flat interface was observed in the AlN/Ni joints bonded at 1673 K after 21.6 ks, indicating no formation of compounds except for a trace of Al-Ni solid solution. The bond strength in AlN/Ni couples was poor.

KEY WORDS: (Ceramic/metal joining) (Interface structure) (Reaction mechanism) (Diffusion path) (Bond strength) (Aluminum nitride)

1. Introduction

Ceramics are under active consideration for structural use in hot machinery such as gas turbines. In these applications, ceramics and metals will be in contact with each other at high temperatures. Different methods of joining ceramics to metals have been developed: high temperature diffusion bonding, brazing with liquid phase or reactive solid components, friction welding, etc. During diffusion bonding the pieces are brought into contact at high temperature and under pressure. Chemical reaction and elemental diffusion at the ceramic/metal interface yields extended interdiffusion zones or even new product phases. These chemical reactions affect the interface properties. The reaction between SiC and metals like Ti and Nb have been studied in literature[1, 2] and that of Si_3N_4 with Ti has been studied also[3]. AlN ceramic as a promising candidate having a high thermal conductivity[)] is to be bonded using various metals. The metals used to join AlN were Ti, V, Nb, and Ni which, except for Ni can react with nitrogen to form nitrides. They can also react with AlN by forming a broad range of reaction products like nitrides and intermetallics[5]. The aim of the present work is to investigate compounds formed by phase reaction at the interfaces between AlN and the various metals. It is also aimed at studying the reaction mechanisms and diffusion paths at these AlN/metals couples. In addition, factors affecting bond strength of the joints in their connection to the interface structure will be discussed.

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2. Experimental work

Various metals foils (Ti-50 μm or 20 μm, V-25 μm, Ni-20 μm, Nb-25 μm) have been used to join AlN ceramics (contains Y_2O_3-based sintering additives from Toukoyama Corp., Japan). The AlN/metal joints were placed in a vacuum furnace equipped with a graphite heating element in a sandwich form, where metal foil is placed between two ceramic cylinders 5 mm in diameter and 6 mm in length. The bonding temperatures varied according to the metal type but generally were in the range from 1323 K up to 1773 K and bonding times varied from 0.3 ks to 72 ks. The reaction products formed at the interfaces between AlN and the various metals were investigated by observing their microstructures using scanning electron microscopy(SEM), analyzing elemental distribution using electron probe microanalyses(EPMA), and identifying the product phases using X-ray diffraction. Furthermore, bond strength measurements of the AlN/metals joints were performed using fracture shear test with a 1.67×10^{-2} mm/sec cross head speed.

3. Results and discussion

3-1 Joining of AlN with Ti

Figure 1 shows a typical microstructure observed in the interface region of the AlN/Ti joints bonded at 1473 K for 7.2 ks. There are some reaction products among the reaction phases with layered structure of 5 μm in total thickness adjacent to the Ti foil.

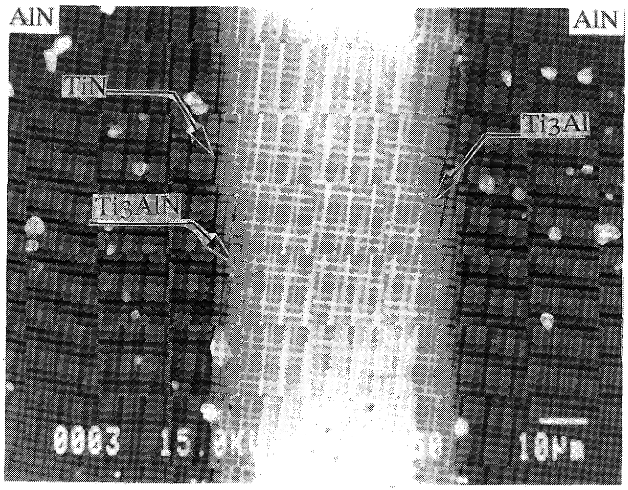

Fig. 1 SEM microphotograph for an AlN/Ti joint bonded at 1473 K for 7.2 ks.

Figures 2 and 3 show the results of analysis by EPMA and X-ray diffraction method, respectively. From these data the reaction phase having layered structure was identified as Ti_3Al.

There is a slight change of composition in between the Ti_3Al and Ti phases, whilst a minimum value of Al content exists in the transition region between Ti_3Al and AlN. This depleted zone of Al is formed by the formation of TiN of 1 μm in thickness, which was detected by X-ray diffraction analysis shown in Fig. 3. A further reaction product layer with a thickness of 1 μm is thought to exist between the thin TiN layer and Ti_3Al. The composition of this layer corresponds to the composition of the ternary phase τ_1-Ti_3AlN which was detected also by XRD shown in fig. 3.

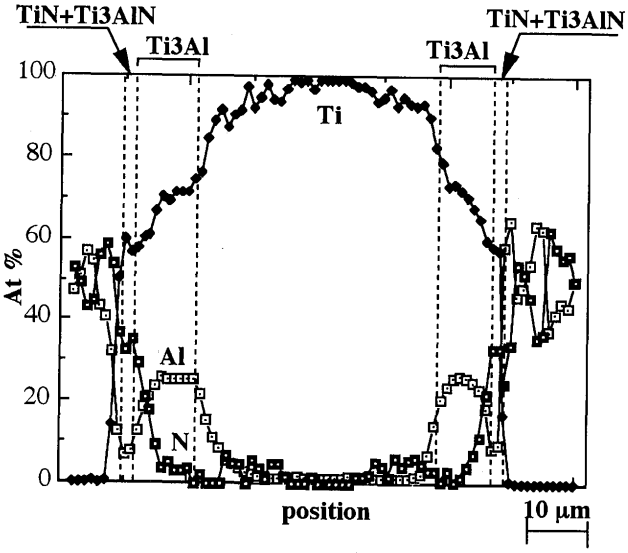

Fig. 2 EPMA analysis for an AlN/Ti joint bonded at 1473 K for 7.2 ks.

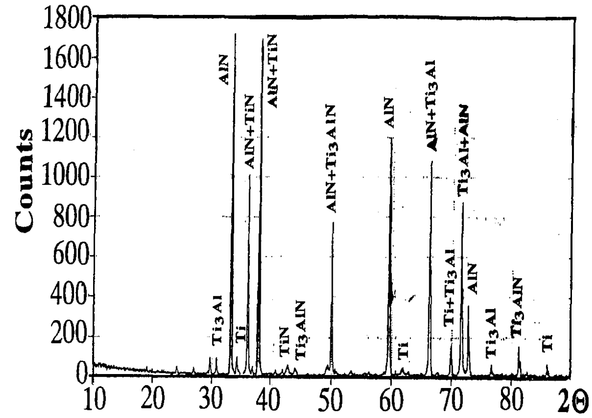

Fig. 3 XRD at the fracture surface of an AlN/Ti joint bonded at 1473 K for 7.2 ks.

As for the thickness of each reaction phase, the nitride layer (TiN and Ti_3AlN) phase remained almost the same thickness regardless of the bonding time, but Ti_3Al phase showed a remarkable increase in thickness with bonding time. From these observations, the interfacial structures of the AlN/Ti joint bonded at 1473 K for 7.2 ks is concluded to have a sequence of AlN/TiN/τ_1-Ti_3AlN/Ti_3Al/Ti, and the sequence is well explained by the diffusion path indicated by the dashed lines on the phase diagram (fig. 4) of the present ternary system [6].

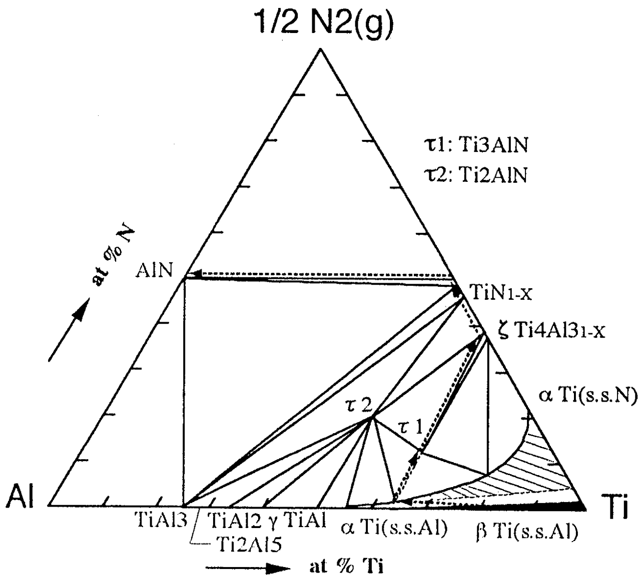

Fig. 4 Diffusion path observed for the AlN/Ti joints shown on the Al-Ti-N ternary phase diagram (isotherm at 1473 K)

Figure 5 shows the dependence of the fracture strength of AlN/Ti/AlN joints on bonding time at a constant temperature of 1473 K. There is a maximum value of 128 MPa for the joint bonded at 28.8 ks, and remarkable decrease of the strength occurred for the further increase in bonding time.

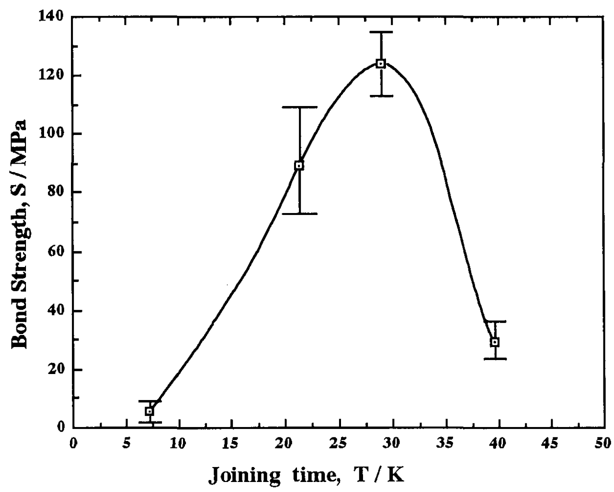

Fig. 5 Fracture strength dependence on bonding time at 1473 K.

Figure 6 shows the result of X-ray diffraction analysis on the fractured surface of the joint bonded at 1473 K for 39.6 ks. In this specimen, Ti has been consumed completely and τ_2-Ti_2AlN is newly observed in addition to Ti_3Al, TiN and τ_1-Ti_3AlN phases. These results show that a bonding longer than 28.8 ks produces the brittle τ_2-Ti_2AlN phase by consuming the ductile Ti foil which relaxes concentrated stress in the central portion of the joint. As a consequence, this phase change would result in the decrease of the bonding strength of the joint.

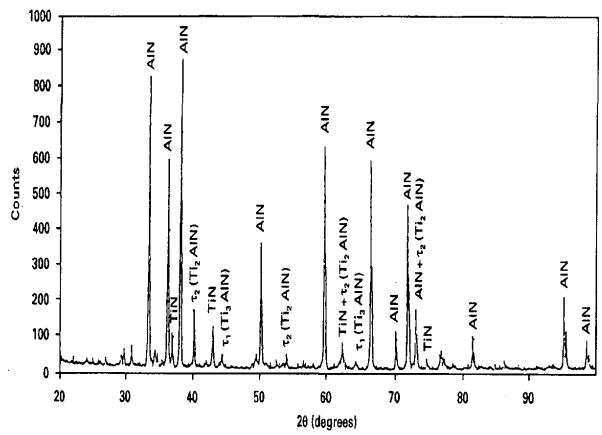

Fig. 6 XRD data at the fracture surface of a joint bonded at 1473 k for 39.6 ks.

3-2 Joining of AlN with V

The preliminary experiments to join AlN ceramics using vanadium metal foils indicated that a bonding temperature up to 1473 K is not enough to activate the AlN ceramics and vanadium metal to start an interfacial reaction. A bonding temperature of 1573 K was used and the reaction could then proceed in a short bonding time of 0.9 ks indicating a fast reaction rate. The AlN/V interface of this joint showed a formation of a thin (3 μm in average) reaction layer adjacent to the ceramic part, and the appearance of another reaction product of a flake shape inside the vanadium central layer, as can be seen from the SEM microphotograph of fig. 7. The EPMA analysis for this joint (fig. 8) indicates the first reaction layer as a solid solution V(Al) dissolving Al content of 23 at. % in vanadium, while the other reaction product inside the vanadium central part was vanadium nitride V_2N. Both V(Al) and V_2N were also identified by XRD as shown in fig. 9.

The V(Al) solid solution layer continued to grow with further increase in bonding time until 9 μm after 5.4 ks and at the same time V_2N also grew from small separated grains to fill almost the entire joint center at the expense of vanadium, as shown by the SEM microphotograph of fig. 10.

For the joint bonded at 1573 K, 0.9 ks, V(Al) grew as the layer structure and V_2N formed inhomogeneously as grains inside V. While the pure V, as a unique phase, quickly changed to V containing V_2N. In other words, at 0.9 ks the layer structure consists of AlN/V(Al)/V_2N+V (fig. 7), but at the initial stage before 0.9 ks where the pure vanadium counter part remains, the interface structure is predicted as AlN / V(Al) / V2N / V. Thus, the diffusion path connecting AlN with V must follow a sequence of AlN / V(Al) / V_2N / V as shown in fig. 11. This reaction path is predicted as the dotted lined drawn on the ternary Al-V-N phase diagram (fig. 11) [7].

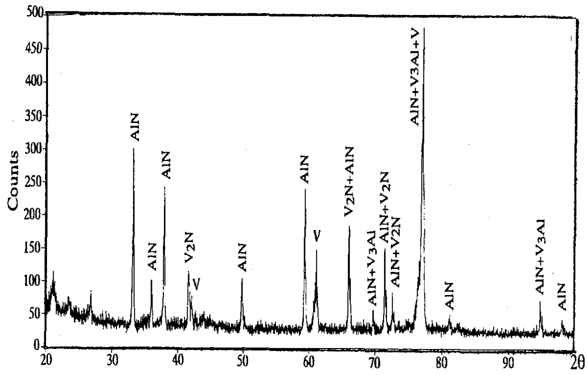

Fig. 9 XRD at the fracture surface of an AlN/V joint bonded at 1573 K for 0.9 ks.

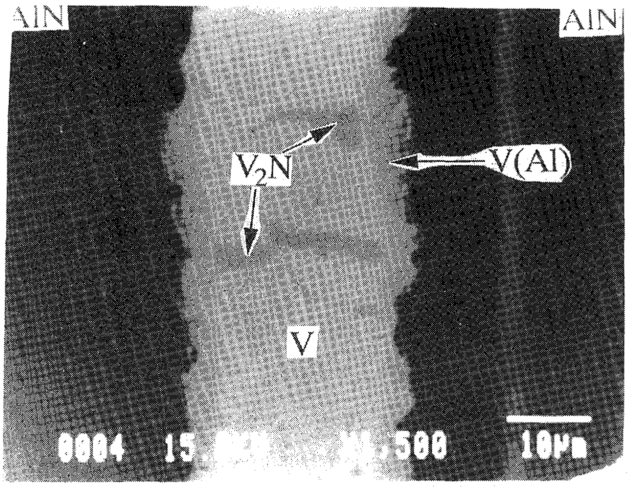

Fig. 7 SEM microphotograph for an AlN/V joint bonded at 1573 K for 0.9 ks.

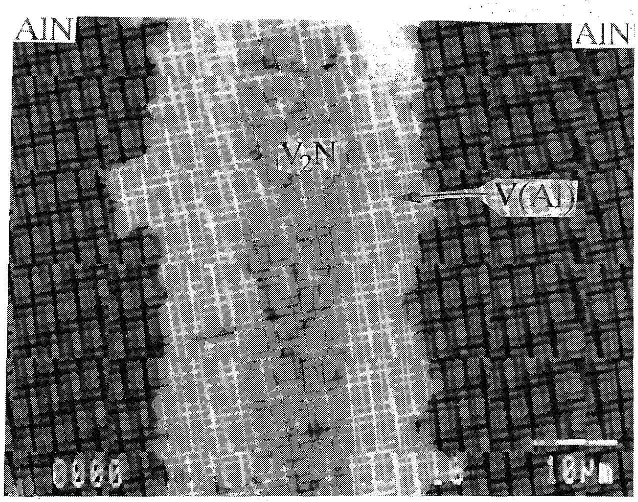

Fig. 10 SEM microphotograph for an AlN/V joint bonded at 1573 K for 5.4 ks.

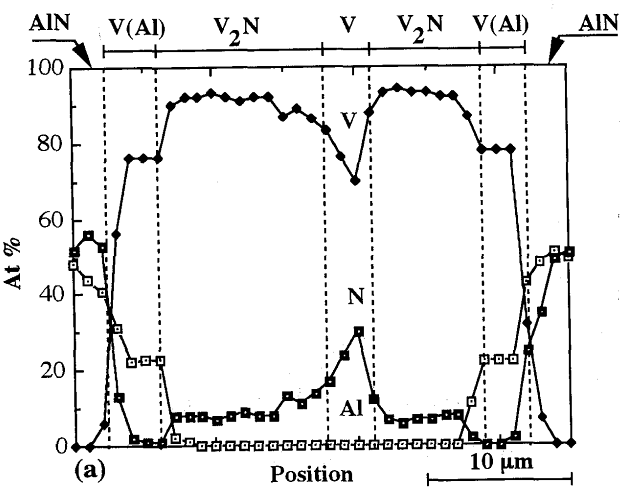

Fig. 8 EPMA analysis for an AlN/V joint bonded at 1573 K for 0.9 ks.

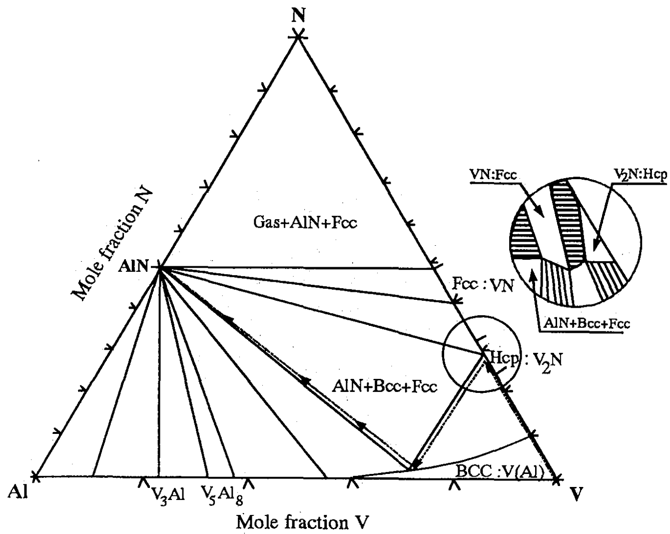

Fig. 11 Diffusion path observed for the AlN/V joints shown on the Al-V-N ternary phase diagram.

When extending the joining time to 7.2 ks at 1673 K, the thickness of the layer next to the AlN became thinner, as shown in fig. 12, and the central part of the joint contained V_2N. The layer phase adjacent to AlN partly changed to V_5Al_8 intermetallic having an a-value of 9.198 nm which fits closely with the literature values[8]. The formation of V_5Al_8 is not predicted on the tie line connecting AlN and V and this suggested that an unexpected reaction, such as the decomposition of AlN, took place during bonding. In other words, at this stage, AlN decomposed and N escaped to the atmosphere of the bonding chamber and the remaining Al at the AlN interface preferentially reacted with V(Al) to form the V_5Al_8 intermetallic.

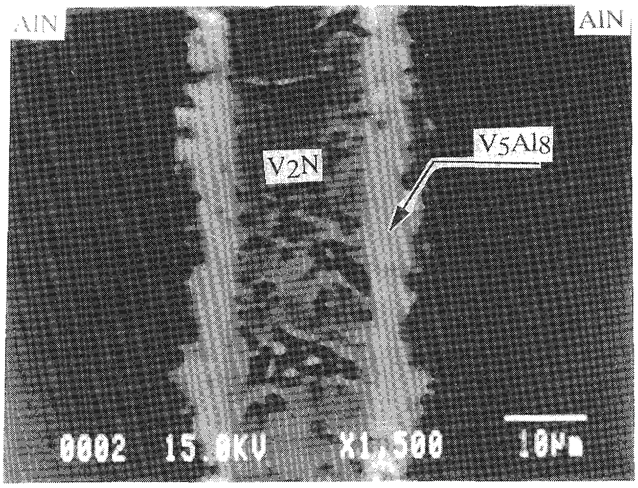

Fig. 12 SEM microphotograph for an AlN/V joint bonded at 1673 k for 7.2 ks.

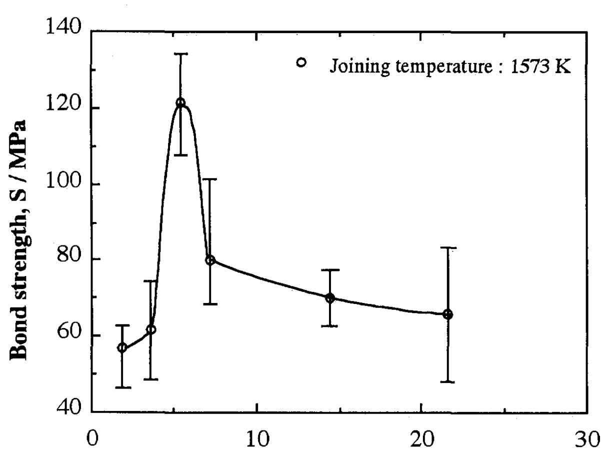

Fig. 13 Fracture strength dependence on bonding time for AlN/V joints bonded at 1573 K.

By extending the bonding time at 1573 K, the strength of the joints increased as shown in fig. 13, until a maximum was reached after 5.4 ks. Although the phases at the interface did not change up to 5.4 ks, the strength of the joints increased, this might be due to an increase in the interface irregularity.

At 5.4 ks the AlN/V interface became stronger than before due to saw effects shown in fig. 10, hence, the fracture occurred at the weaker V(Al)/V_2N interface. At 7.2 ks, the appearance of the brittle V_5Al_8 intermetallic caused a decrease in the joint bond strength. As the bonding time increases, consumption of the vanadium and growth of V_5Al_8 increases, and consequently, the bond strength continue to decrease. Since the thermal expansion coefficient of V_5Al_8 exceeds by five times that of the ceramic part, the contraction of V_5Al_8 layer during cooling from the joining temperature will be larger than that of the AlN parts, so that residual stresses developed at the AlN/V_5Al_8 interface, lead to a decrease in the joints bond strength.

3.3 Joining of AlN with Nb

The interface reaction in the case of AlN/Ti and AlN/V joints could proceed at bonding temperatures of 1473 K and 1573 K respectively. When AlN is bonded using Nb foils, a temperature up to 1573 K results in a some evidence of reaction in the form of nitride spots inside the Nb central layer, this could be observed at 14.4 ks.

Nb start to react with AlN at the interface by forming a thin reaction layer of 2 μm in average thickness adjacent to the AlN. Beside this layer, another reaction product of a spherical shape was formed at the Nb central layer. This can be seen from the SEM microphotograph of fig. 14, for a joint bonded at 1673 K for 3.6 ks. The EPMA data for this joint given in fig. 15 showed that the first reaction layer was Nb_2Al and the other reaction product was Nb_2N.

When extending the bonding time up to 7.2 ks, another reaction layer within 1 μm started to appear between Nb_2Al and AlN, the composition of this reaction layer corresponds to the composition of the ternary compound Nb_3Al_2N.

By extending the bonding time up to 14.4 ks, the thickness of the Nb_2Al layer decreased and that of the ternary compound increased, and Nb_2N grew to fill the entire joint center, while the Nb counter part almost consumed, except for a few spots remaining inside the

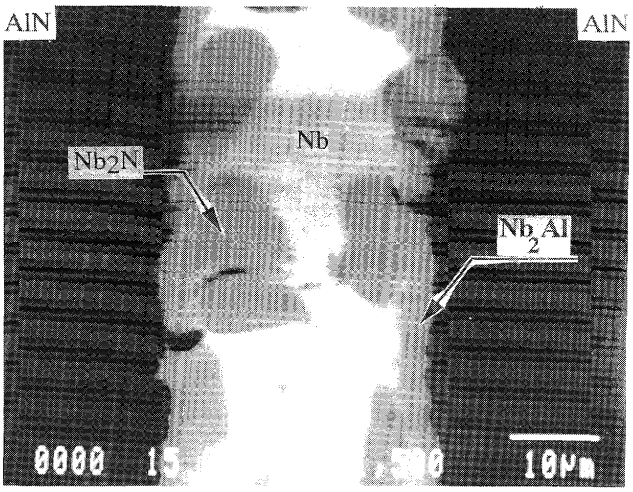

Fig. 14 SEM microphotograph for an AlN/Nb joint bonded at 1673 K for 3.6 ks.

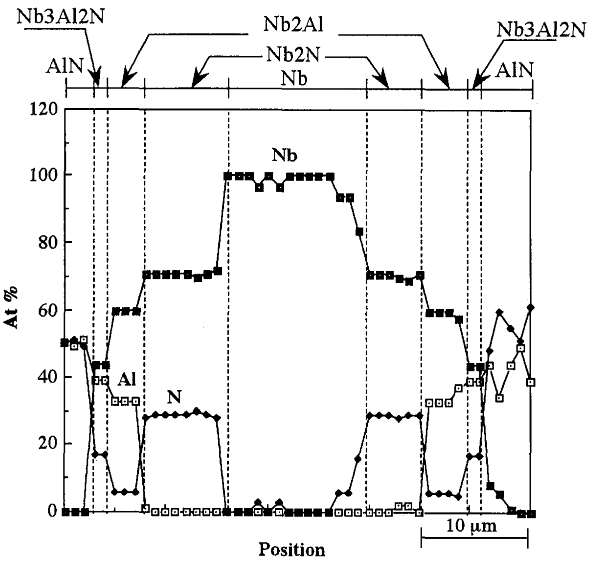

Fig. 15 EPMA analysis for an AlN/Nb joint bonded at 1673 K for 3.6 ks.

V_2N central layer. This can be observed from the SEM microphotograph of fig. 16 and the EPMA data of fig. 17.

From the previous observation, the interface structure of the AlN/Nb joints bonded up to 1673 K for 7.2 ks is concluded to have a sequence of AlN / Nb_3Al_2N / Nb_2Al / Nb_2N / Nb, which represents a complete diffusion path for the AlN/Nb joints. This path is indicated by the dashed lines on the Al-Nb-N ternary system[9] shown in fig. 18.

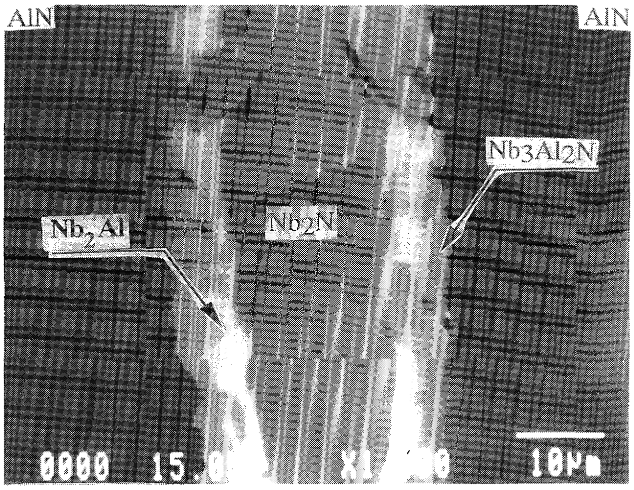

Fig. 16 SEM microphotograph for an AlN/Nb joint bonded at 1673 K for 14.4 ks.

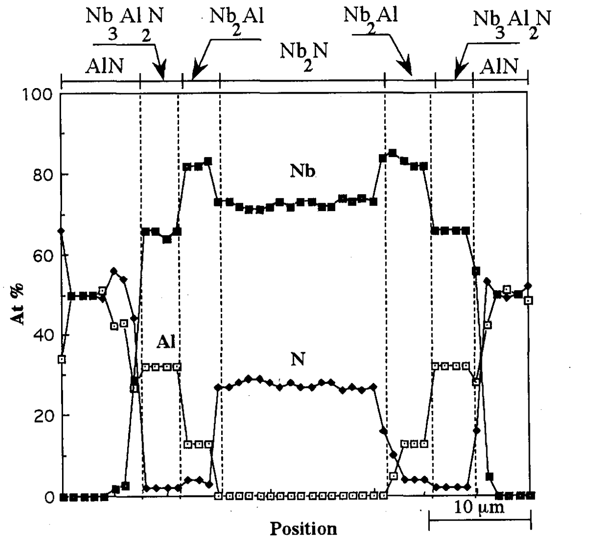

Fig. 17 EPMA analysis for an AlN/Nb joint bonded at 1673 K for 14.4 ks.

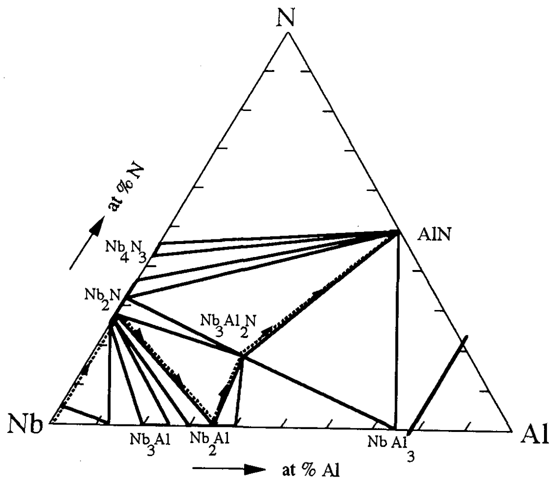

Fig. 18 Diffusion path observed for the AlN/Nb joints shown on the Al-Nb-N ternary phase diagram.

3.4 Joining of AlN with Ni

A flat fracture surface due to the formation of cracks was observed after the fracture test of AlN/Ni joint which was bonded at 1673 K for 21.6 ks. This result means that there is no formation of reaction products phase other than Ni-Al solid solution and severe decomposition of AlN.

In addition, no intermetallic phase was observed in the interface region of this joint, and this was confirmed by X-ray diffraction analysis on a polished surface of the specimen.

In the present study, joining of AlN with metals depends mainly on the formation behavior of nitrides in the joint region. As nickel is known to be non-nitride former, no reaction product is thought to exist.

4. Conclusions

Joining of AlN ceramics using various types of metals foil such as Ti, V, Nb and Ni has been studied by examining the interfacial structures and bonding strength, and the results are summarized as follows;

(1) A complete reaction path between AlN and Ti was established at 1473 K for 7.2 ks bonding time following a sequence of AlN / TiN / Ti_3AlN / Ti_3Al / Ti. A maximum bond strength of 128 MPa could be obtained in a joint bonded at 1473 K for 28.8 ks. Beyond this, the soft Ti was completely consumed and another brittle ternary compound Ti_2AlN was formed.

(2) The phase reaction in the AlN/V system took place at a temperature of 1573 K or higher. Formation of V(Al) solid solution adjacent to the AlN and V_2N nitride adjacent to the V, controls the interface joining. No ternary compounds were observed. The diffusion path could be predicted for the AlN/V joints up to 0.9 ks at 1573 K following a sequence of AlN / V(Al) / V_2N / V. Bond strength at 1573 increased with bonding time until a maximum was obtained at 5.4 ks.

(3) Nb reacts with AlN at higher temperatures, starting from 1673 K. A diffusion path is established between AlN and Nb at 1673 K for 7.2 ks, following a sequence of AlN / Nb_3Al_2N / Nb_2Al / Nb_2N / Nb.

(4) The interface between AlN and Ni showed no formation of reaction phases except for Ni-Al solid solution since nickel is a non-nitride former.

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