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Structure and strength of AlN/V bonding interfaces

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AlN ceramics are bonded using vanadium metal foils at high temperatures in vacuum. Different bonding temperatures were used in the range 1373–1773 K with bonding times of 0.3–21.6 ks. The AlN/V interfaces of the bonded joints were investigated using SEM, electron probe microanalysis and X-ray diffraction. A bonding temperature of 1573 K was found to be suitable to activate both parts to initiate a phase reaction at the interface, because a thin V(Al) solid solution layer formed adjacent to the ceramic at 1573 K just after 0.9 ks, and a small flake-shaped V_2N reaction product formed inside the vanadium central layer. The formation of V(Al) and V_2N controls the interfacial joining of the AlN/V system at 1573 K up to 5.4 ks bonding time. The pure vanadium layer quickly changed to vanadium-containing V_2N. The diffusion path could be predicted for the AlN/V joints up to 0.9 ks at 1573 K following the sequence AlN/V(Al)/V_2N/V, while after 0.9 ks, the interface structure changed to AlN/V(Al)/$V_2N + V$ by the growth of V_2N into the vanadium. The AlN/V joints showed no ternary compounds at the interface. 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. At 7.2 ks, nitrogen, resulting from AlN decomposition, escaped and the remaining aluminium reacted with V(Al) to form V_5Al_8 intermetallic, which is attributable to the decrease in bond strength.

1. Introduction

Metal/ceramic joints are required in electronic field for insulators, capacitors or microchip substrates [1], and are in high operation for structural components where high toughness and good resistance to wear and corrosion at high-temperature operations [2] are required. In order to obtain reliable mechanical properties of ceramic and metal joints, the reaction phase and phase relations at the ceramic/metal interface have to be elucidated. Several results have been reported for ceramic/metal systems, such as SiC/Ti [3]. AlN has a high thermal conductivity [4], as well as high-temperature strength. Results of AlN reaction with titanium metal have also been reported [5], where the diffusion path followed the sequence of AlN/TiN/Ti_3AlN/Ti_3Al/Ti. The formation of another ternary compound, Ti_2AlN, after the consumption of titanium, was also reported. Vanadium is also an active element for reacting with ceramics, reacting with SiC to form V_5Si_3C, V_5Si_3, V_3Si, and V_2C [6]. Vanadium metal is preferred to join the AlN ceramics because it has a thermal expansion coefficient nearly equal to that of AlN. The present work attempted to reveal the reaction mechanism at the interface between AlN and V, in an effort to discuss the diffusion path at the AlN/V system, and to elucidate the bond strength variation with interface structure.

2. Experimental procedure

AlN ceramics containing Y_2O_3-based sintering additives (from Toukoyama Soda Corp., Japan) were joined using 25 μm vanadium foil. Prior to experiments, the surfaces of both AlN and vanadium were slightly polished to remove any stain or oxide film on the surface, especially the vanadium surface, which was found to prevent the interfacial reaction between AlN and vanadium. The AlN/V joint was placed in a vacuum furnace equipped with a graphite heating element in a sandwich form, where metal foil was placed between two ceramic cylinders each of 5 mm diameter and 4 mm length. The bonding temperatures varied from 1373–1773 K and the bonding time varied from 0.3–21.6 ks. The reaction products formed at the interfaces between AlN and vanadium metal were investigated by observing the microstructures using scanning electron microscopy (SEM), analysing elemental distribution using electron probe microanalysis (EPMA), and identifying the product phases using X-ray diffraction (XRD). Furthermore, bond strength measurements of the AlN/V joints were performed using the fracture shear test with 1.67×10^{-2} mm s^{-1} crosshead speed; this method was reported in detail elsewhere [7].

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3. Results and discussion

A solid state reaction at the AlN/V interface cannot be achieved on the as-received vanadium foil surface. The preliminary experiments performed to join AlN ceramics using vanadium metal foils indicated that a bonding temperature up to 1473 K is insufficient to activate the AlN ceramics and vanadium metal to initiate an interfacial reaction. When a bonding temperature of 1573 K is used for a short bonding time of 0.9 ks, the AlN/V interface of this joint showed the formation of a thin reaction layer adjacent to the ceramic part, and the appearance of another flake-shaped reaction product inside the vanadium central layer, as can be seen from the scanning electron micrograph of Fig. 1a. The EPMA analysis for this joint (Fig. 2a) indicating the first reaction layer to be a solid solution of 23 at % Al dissolved in vanadium, V(Al), 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. 3a. Calculation indicated the a- and c-values of the detected V_2N to be 4.875 and 4.554 nm, and 4.797 nm for V(Al), noting that the label V_3Al in the XRD data represents V(Al). Vanadium is known to dissolve a large amount of aluminium at high temperatures [8], when V(Al) exists at high temperatures during bonding and changed to V_3Al upon cooling to room temperature. At the AlN/V interface, V(Al) and V_2N are formed by the reaction of vanadium with AlN

$$V + Al \rightarrow V(Al) \qquad (1)$$

$$V + N \rightarrow V_2N \qquad (2)$$

Because the diffusion of nitrogen is faster than that of aluminium, V_2N and V(Al) grow at the vanadium side and AlN side, respectively.

When extending bonding time up to 1.8 ks at 1573 K, the interface reaction became clearer, where the V(Al) layer grew from the previous stage and the V_2N layer starts to occupy a larger space inside the vanadium, as can be seen from the scanning electron micrograph and EPMA of Figs 1b and 2b. In this step, nitrogen and aluminium continue to diffuse from AlN, and consequently V(Al) and V_2N grow.

In the next step, when the bonding time is doubled to 3.6 ks at 1573 K, growth of the V_2N reaction product was observed; its shape changed to a granular form as can be seen from the scanning electron micrograph of Fig. 1c. Also, the V(Al) solid solution layer grew slightly compared to the earlier step of the reaction, as can be observed from Fig. 1c. The XRD (Fig. 3b) performed on the fracture surface of this joint showed V(Al) ($a = 4.797$ nm) and V_2N ($a = 4.875$ nm, $c = 4.554$ nm) as well as vanadium ($a = 3.031$ nm).

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

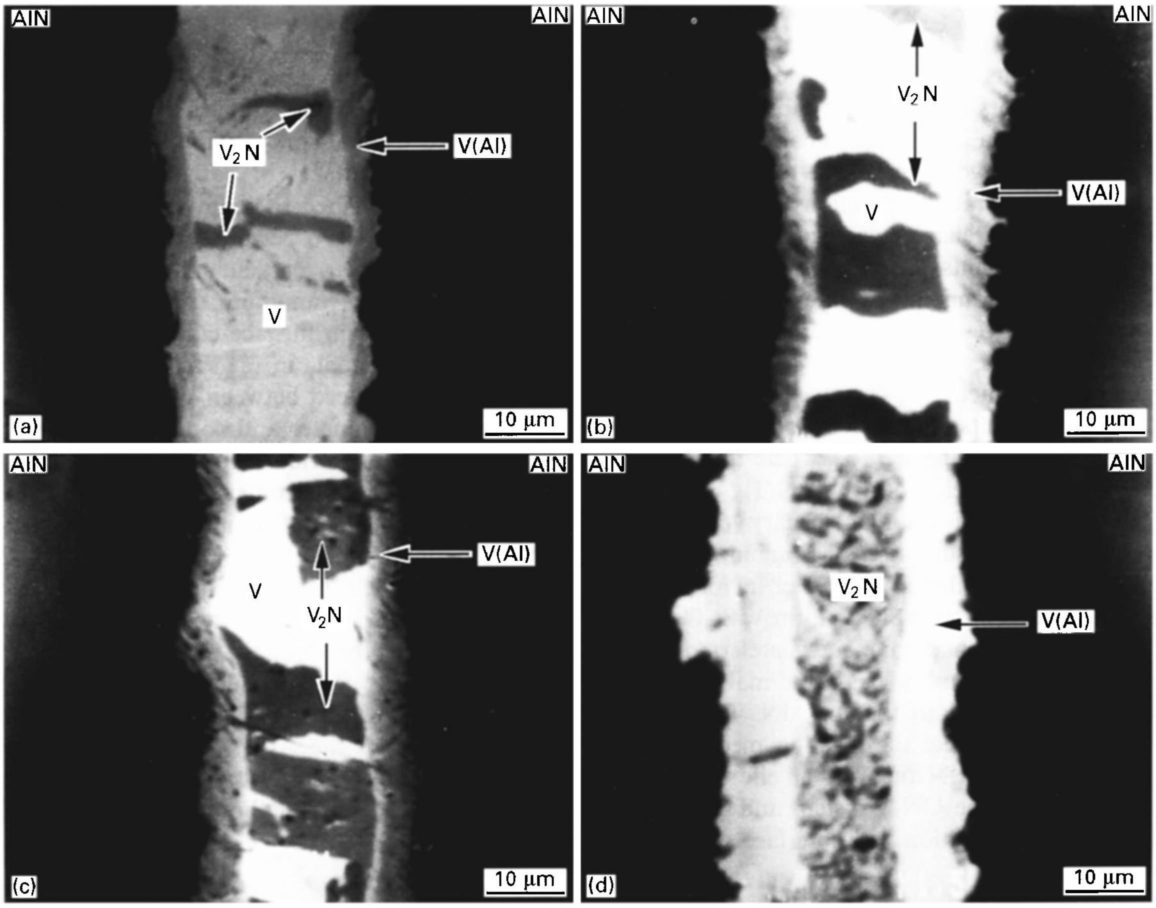

Figure 1 Scanning electron micrograph for joints bonded at 1573 K for (a) 0.9 ks, (b) 1.8 ks, (c) 3.6 ks, and (d) 5.4 ks.

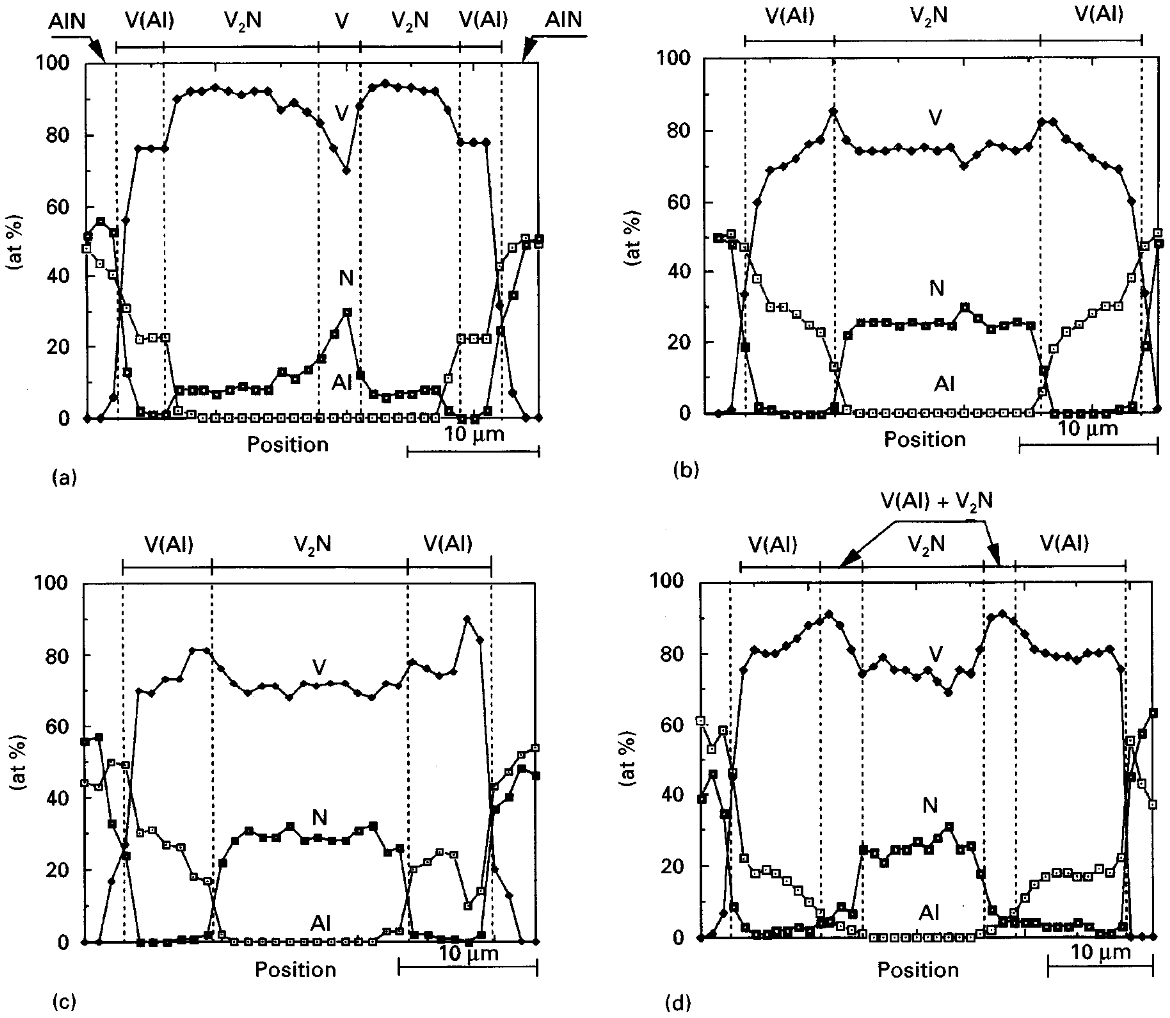

Figure 2 EPMA analysis for joints bonded at 1573 K for (a) 0.9 ks, (b) 1.8 ks, (c) 3.6 ks, and (d) 5.4 ks.

separated grains to fill almost the entire joint centre at the expense of vanadium (Figs 1d and 2d).

For the joint bonded at 1573 K, 0.9 ks, V(Al) solid solution grew as the layer structure, and V_2N formed inhomogeneously as grains inside the vanadium. While, the pure vanadium as a unique phase, quickly changed to vanadium containing V_2N. In other words, at 0.9 ks, the layer structure consists of AlN/V(Al)/V_2N + V (Fig. 1a), but in the initial stage before a bonding time of 0.9 ks, where the pure vanadium counterpart remains, the interface structure is predicted to be AlN/V(Al)/V_2N/V. Thus, the diffusion path connecting AlN with vanadium must follow the sequence of AlN/V(Al)/V_2N/V. These reaction phases (V(Al) and V_2N) are predicted from the ternary Al–V–N phase diagram [9] shown in Fig. 4. It is known that the reaction phases on the diffusion path described on the corresponding ternary phase diagram, will be formed at the ceramic/metal interface [10, 11]. V_2N nitrides started as flakes at 0.9 ks, grew significantly after 1.8 and 3.6 ks, and occupied a larger area in the joint centre after 5.4 ks as can be seen from Fig. 1. Before and at 5.4 ks, growth of V_2N is faster than that of V(Al). A gradual consumption of vanadium occurred on increasing the bonding time at 1573 K, until very few spots of it remained inside the V_2N; this is also shown in Fig. 1.

In AlN/V joints bonded at 1573 K, the diffusion path of AlN/V(Al)/V_2N/V predicted in Fig. 4 quickly disappeared, and the interface structure changed to AlN/V(Al)/V + V_2N, because V_2N quickly grew into the vanadium after a bonding time of 0.9 ks.

When extending the joining time to 7.2 ks, the thickness of the layer next to the AlN became thinner, as shown in Fig. 5, and the central part of the joint contains V_2N. The layer phase adjacent to AlN partly changed to V_5Al_8 intermetallic. XRD in Fig. 6 shows a cubic V_5Al_8 intermetallic having an a-value of 9.198 nm, which agrees closely with the literature values [12]. The formation of V_5Al_8 intermetallic is not predicted on the tie-line connecting AlN and vanadium in Fig. 12. This suggests that the unexpected reaction, such as the decomposition of AlN, took place during bonding. In other words, at this stage, AlN decomposed and nitrogen escaped to the atmosphere in the chamber and the remaining aluminium at the AlN interface preferentially reacted with V(Al) to form the V_5Al_8 intermetallic. The decomposition of AlN was also observed at the higher temperature of 1673 K for the longer bonding time of 7.2 ks or more. It was observed that AlN remained stable up to 39.6 ks at 1473 k, but started to decompose partially just after 3.6 ks at 1573 K, and became totally unstable after 3.6 ks at 1673 and 1773 K. For instance, XRD

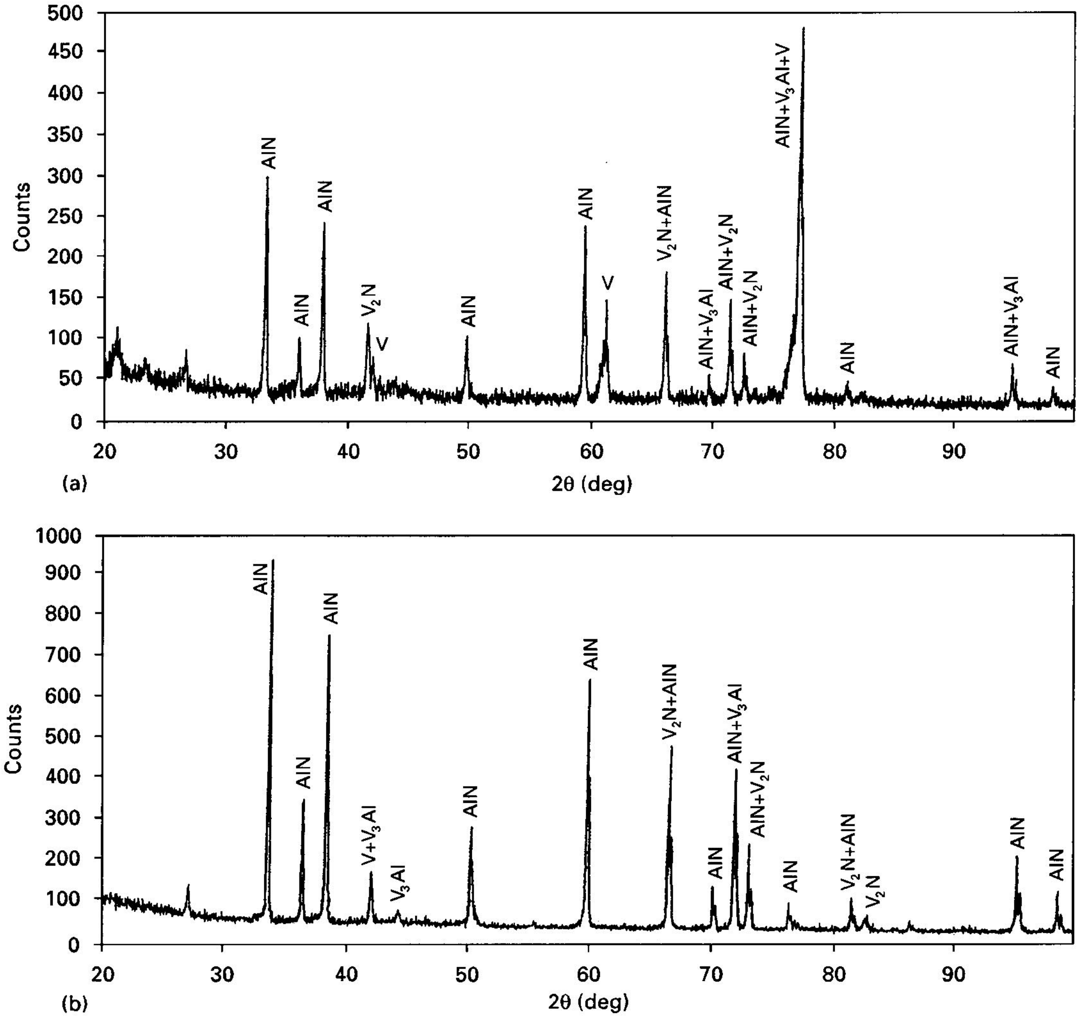

Figure 3 XRD pattern for joints bonded at 1573 K for (a) 0.9 ks, and (b) 3.6 ks.

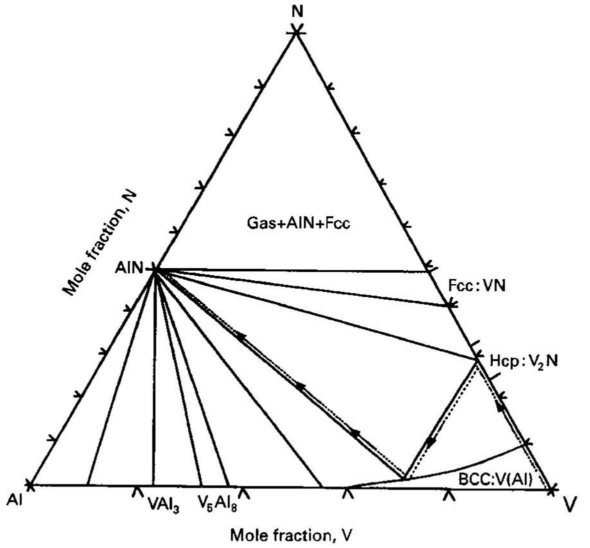

Figure 4 The diffusion path described on the Al–V–N ternary phase diagram by extrapolating from 1573 K [9].

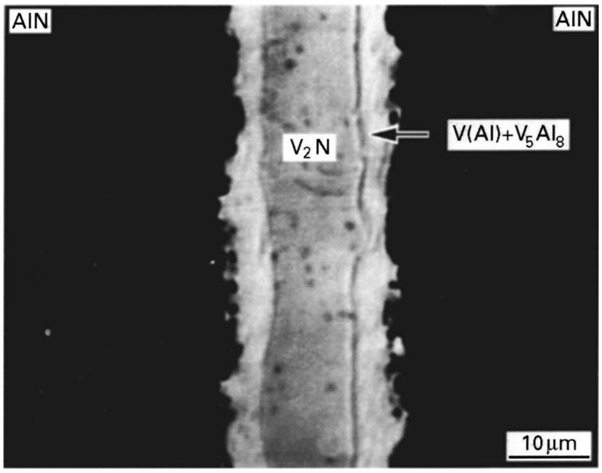

Figure 5 Scanning electron micrograph of a joint bonded at 1573 K for 7.2 ks.

(Fig. 7) performed on the fracture surface of a joint bonded at 1673 K for 21.6 ks shows the formation of V_5Al_8 and the existence of VN.

The appearance of the most stable nitride VN (cubic, $a = 4.0998$ nm) can be supported by the recent calculations and measurements performed by Yong *et al.* [9] for the Al–V–N phase diagram.

Upon increasing the bonding time at 1573 K, the strength of the joints increased, as shown in Fig. 8, until a maximum is obtained 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

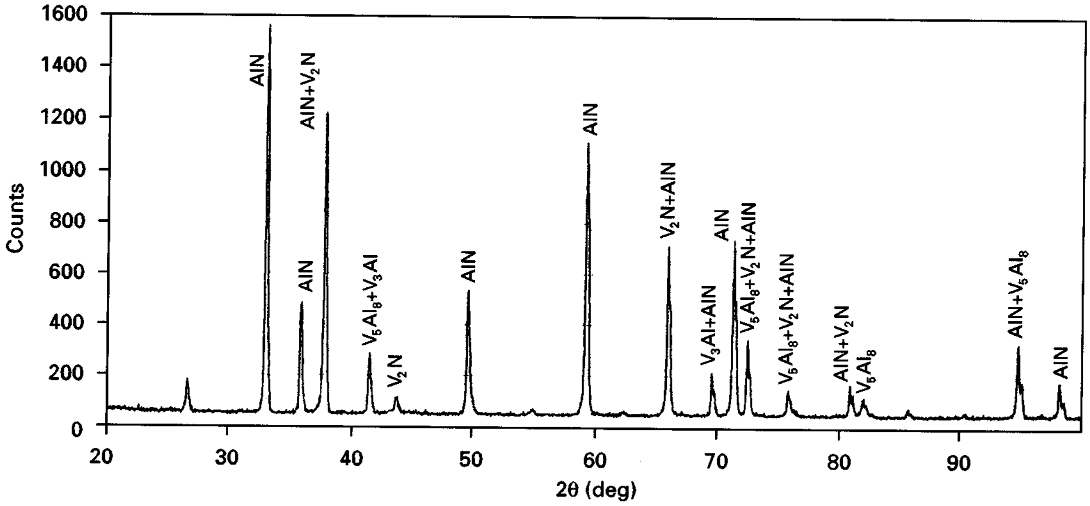

Figure 6 XRD pattern for the fracture surface of a joint bonded at 1573 K for 7.2 ks.

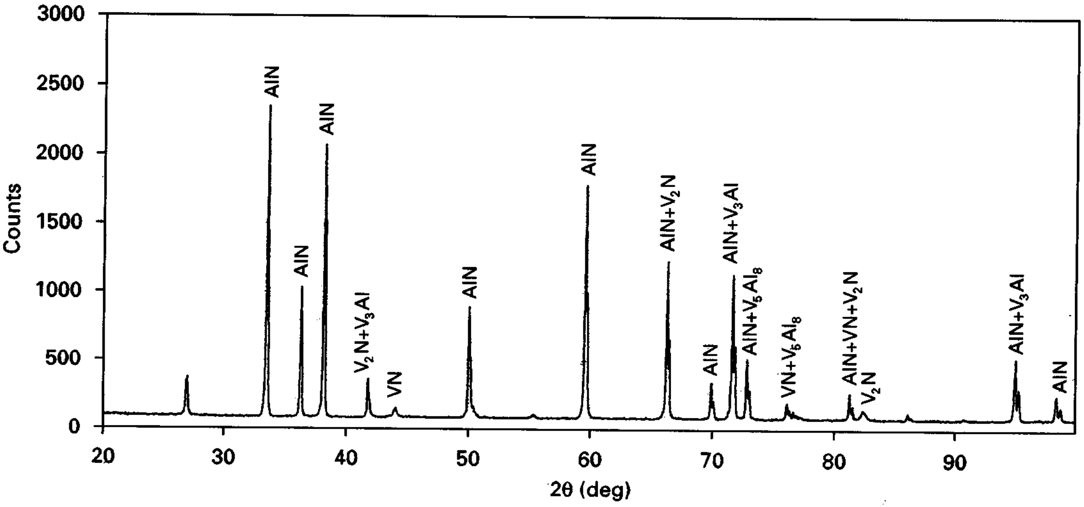

Figure 7 XRD pattern for the fracture surface of a joint bonded at 1673 K for 21.6 ks.

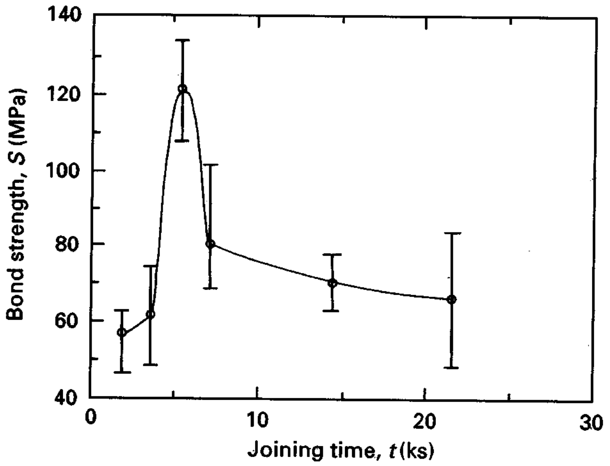

Figure 8 Results of bond strength for AlN/V joints bonded at 1573 K.

AlN/V(Al) interface became stronger than before, due to the saw effect, as shown in Fig. 1d; hence, the fracture occurred at the weaker V(Al)/V_2N interface as shown in Fig. 9b. For the joint bonded at 7.2 ks, the appearance of the brittle V_5Al_8 intermetallic caused a decrease in the bond strength. The bond strengths continue to decrease as joining time increases, which accelerates the consumption of vanadium and the growth of V_5Al_8. Because the thermal expansion coefficient of V_5Al_8 exceeds, by five times, that of the ceramic part, the contraction of the V_5Al_8 layer during cooling from the joining temperature will be larger than that of the AlN parts, which develop residual stresses at the AlN/V_5Al_8 interface. Consequently, the joint bond strength will decrease.

Three different joints bonded after 3.6, 5.4, and 7.2 ks were chosen for fracture surface observation (Fig. 9) in order to trace the effect of interface structure on the variation of bond strength. For the joint bonded for more than 3.6 ks (Fig. 9a), fracture originated and propagated along the AlN/V(Al) interface. For the joint bonded for more than 5.4 ks, which exerts the maximum bond strength, the crack propagated through the V(Al)/V_2N interface where fracture originated and traversed through V_2N to AlN, as shown in Fig. 9b. For the third joint bonded for more than 7.2 ks, fracture originated and

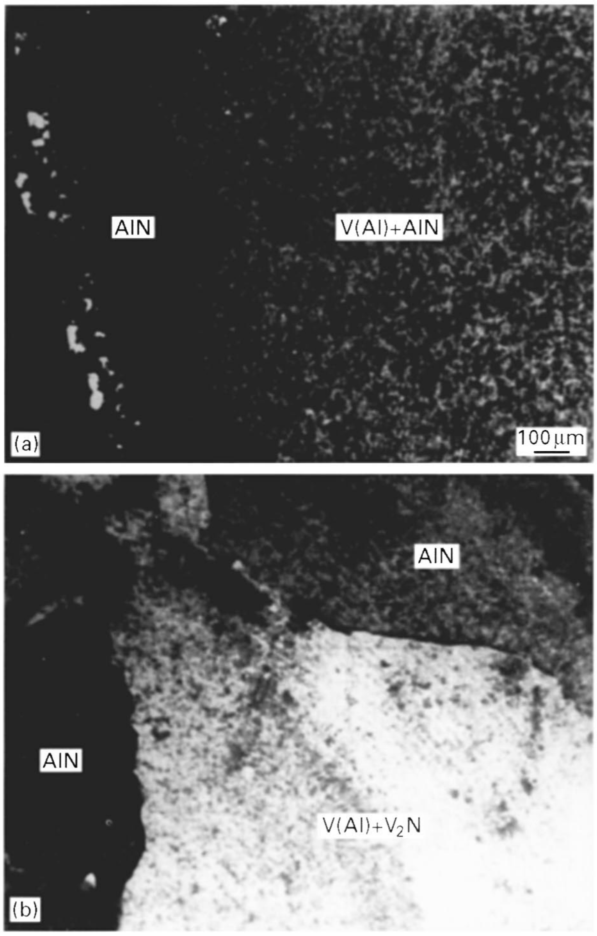

Figure 9 SEM image for the fracture surface of joints bonded at 1573 K for (a) 3.6 ks, and (b) 5.4 ks, showing the results of spot analyses.

propagated through the mixed phases in the joining layer, and traversed to AlN. Although, initially, the AlN/V(Al) interface was weak; the increase in irregularity of the interface improves the AlN/V(Al) interface strength. This increased the bond strength of the joint. By increasing the bonding time, AlN decomposed and nitrogen mainly escaped to the atmosphere, and the remaining aluminium reacted at the interface with V(Al) to form V_5Al_8 at 7.2 ks. The formation of a brittle V_5Al_8 intermetallic decreased the bond strength.

4. Conclusions

1. The phase reaction of the AlN/V system took place at a comparatively high temperature level, beginning from 1573 K. The formation of V(Al) solid solution adjacent to AlN, and V_2N nitride adjacent to vanadium, controls the interface joining of the AlN/V system. No ternary compounds were observed.

2. The diffusion path could be predicted for the AlN/V joints before 0.9 ks at 1573 K, following the sequence AlN/V(Al)/V_2N/V, which is described on the Al–V–N ternary phase diagram.

3. The bond strength for AlN/V joints bonded at 1573 K increased with bonding time, where a structure of AlN/V(Al)/V_2N + V existed at the interface, until a maximum is obtained at 5.4 ks. A further increase in bonding time causes decomposition of AlN, and the soft V(Al) changed gradually to V_5Al_8. The formation of V_5Al_8 consequently decreases the bond strength of the joint.

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