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Oxidation of aluminium nitride substrates

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The growth of oxide films on two types of aluminium nitride substrates of different origin has been studied as a function of temperature. At a given set of oxidation reaction parameters, the oxide layers grown on substrates with a relatively large grain size and high concentrations of Y–Al–O-based liquid sintering aid phases (type I substrates) were observed to be thicker and more diffuse than those obtained on substrates with an average particle size of approximately 3 μm and low liquid sintering aid concentrations (type II substrates). The effects of the oxygen partial pressure variation on the oxide film growth have been investigated for the oxidation of type II AlN substrates. The kinetics of the growth of oxide films on such substrates were analysed and determined to fit best to a linear rate law. This type of rate law indicates that the rate-limiting step in the growth of oxide films on high-quality type II aluminium nitride substrates is an interface reaction-controlled process.

1. Introduction

Aluminium nitride has a number of properties that make it an attractive material for use as heat sinks and computer chip substrates. In particular, the thermal conductivity of aluminium nitride is 320 $W m^{-1} K^{-1}$ [1, 2], which is much higher than that of alumina (20–40 $W m^{-1} K^{-1}$ [3, 4]). Additionally , the thermal expansion of aluminium nitride is $\sim 4.5 \times 10^{-6} C^{-1}$ from 20–400 °C [5] and is close to that of silicon [6]. The thermal conductivity is adversely affected by oxygen impurities inherent in the preparation of aluminium nitride [2, 7]. Although the presence of oxide films on aluminium nitride substrates is generally considered detrimental to the use of such substrates for heat removal, it has been shown to be beneficial for the adhesion of copper films [4].

The role of oxygen in aluminium nitride has received considerable attention in the literature [4, 8–13]. The kinetics of the oxidation of aluminium nitride powder, recently studied by Suryanarayana [13], was shown to depend on particle size. The data obtained from this study were fit to a mixed kinetic model based on transport and an unspecified interface process as important rate-limiting steps.

In this paper, results from a new investigation of the oxide layer growth on two aluminium nitride substrates with distinctly different characteristics are reported. The effects of the different sample characteristics on the oxide film growth and the growth kinetics (where relevant) will be discussed.

2. Experimental procedure

In this study of the oxidation of aluminium nitride substrates, two different types of substrate were considered. The first type of substrate (type I) was prepared by an AlN substrate manufacturer as an experimental substrate by hot isostatically pressing aluminium nitride powder with a liquid sintering aid, Y_2O_3. The second type of substrate (type II) was an experimental aluminium nitride substrate supplied by Carborundum. Owing to their proprietary nature, the exact manufacturing methods of these substrates are not known to the present authors.

Samples of the substrates were prepared for oxidation experiments by first cutting pieces of type I substrate with a diamond wheel to approximately 1 cm × 1 cm × 1 cm. Because the type II substrates were received with dimensions of approximately 1 cm × 1 cm × 3 mm, cutting of these samples was not required. When lapped surfaces had to be prepared, samples were affixed to glass slides which were then mounted to a lapping machine (GMN, Nürnberg, Germany), equipped with a grinding wheel with 65 μm diamonds, on a planar vacuum chuck. The samples surfaces were then ground plane-parallel to within 0.001 mm. To decrease the surface roughness, a second lapping was performed with a second, identical lapping machine, but equipped with a grinding wheel with 15 μm diamonds. Oxidation studies were performed on lapped samples of types I and II and on as-received samples of type II.

Samples were oxidized in a muffle furnace in air at temperatures between 1000 and 1300 °C for up to 48 h and also in a controlled atmosphere furnace in "argon" at 1200–1400 °C for up to 50 h. The notation "argon" refers to a purified argon gas containing about 200 p.p.m. oxygen as an impurity, corresponding to an oxygen partial pressure of about 2×10^{-4} bar at a total pressure of 1 bar. Samples to be oxidized in air were heated from room temperature to the desired temperature in 3 h. The time at which the furnace reached the desired temperature was taken as $t = 0$. At $t = 0$, one sample was removed from the

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furnace and quenched to room temperature. Thereafter, samples were removed from the furnace after regular time intervals and quenched to room temperature. Samples oxidized in "argon" were introduced into a tube furnace which was at the desired oxidation temperature and containing a flowing "argon" gas atmosphere. It took approximately 5 min for a sample to be moved into the hot zone. The time at which the sample reached the hot zone was taken as $t = 0$. After completion of a pre-selected oxidation annealing time, the sample was cooled to room temperature in the "argon" atmosphere by retracting the sample holder from the hot zone of the furnace.

After cooling, each oxidized substrate sample was mounted in a cold mounting material (Struers Triofix). Then, the sample was ground parallel to the growth direction of the oxide film in steps of 0.25 mm with a diamond wheel using a universal cutting and grinding machine (Conrad, Clausthal-Zellerfeld, Germany). This grinding was repeated until approximately 0.5 cm material was removed. By grinding away about half of the sample, the centre of the sample became available for analysis of the morphology and thickness of the grown oxide films. This was done, because, in the middle of the sample, the influence of edges and sides on the oxidation process is expected to be minimal. After grinding, the samples were polished using successively finer grit SiC paper (320, 500, 1200 and 4000 grit) on a polishing and grinding machine (Planapol-3 equipped with a Pedamax, Struers, Danmark). Between each polishing step, the samples were ultrasonically cleaned in water.

To prepare the polished samples for microprobe analysis they were coated with approximately 20 nm carbon. The thickness and quality of the grown oxide films were determined by obtaining oxygen concentration maps and backscattered electron images on an electron microprobe (Joel 733) equipped with a lead stearate crystal that is sensitive to light elements. The chemical makeup of the oxide film was examined by oxygen and nitrogen line profiles.

The phases present in oxide films grown on aluminium nitride substrates and within these substrates were identified by X-ray diffraction using a theta–theta diffractometer with a CuK_{α} X-ray source (Scintag PADX). X-ray patterns were obtained in the normal mode for as-received samples and in the 2θ mode with the omega angle fixed at 2° for samples with a thin oxide film. In the first mode of operation, the patterns are obtained by simultaneously scanning both the omega and theta angles. In the second case, the patterns are obtained by fixing the incident angle of the X-ray source. By using this thin-film configuration, the X-ray patterns are more sensitive to the oxide film. The surfaces of the oxidized and as-received substrates were used as they were, and aligned for the measurement of X-ray patterns in the usual way.

3. Results and discussion

The characteristics of the two types of substrate investigated with regard to their oxidation behaviour are different, as can be seen by visual inspection and in backscattered electron images of both materials, see Fig. 1. The micrographs reveal that the type I substrate material contains more yttria (liquid sintering aid) than the type II material. Also, the particle size is more uniform and smaller in the case of type II substrates. X-ray diffraction patterns confirm that the two types of substrates contain yttria which has been introduced as liquid sintering aid. Based on the analysis of X-ray patterns, see Fig. 2, it was found that the two types of substrate differ with regard to the secondary phases created by using yttria as sintering aid. In type I substrates, only $Y_4Al_2O_9$ was detected as secondary phase. In type II substrates, both $Y_4Al_2O_9$ and $Y_3Al_5O_{12}$ were observed as secondary phases. The compositions of the secondary phases present were confirmed by a semi-quantitative analysis using energy dispersive spectroscopy on an electron microprobe (Joel 733). The different observations with regard to secondary phases in types I and II substrates must be related to the different approaches used to produce these substrates and possibly also to different

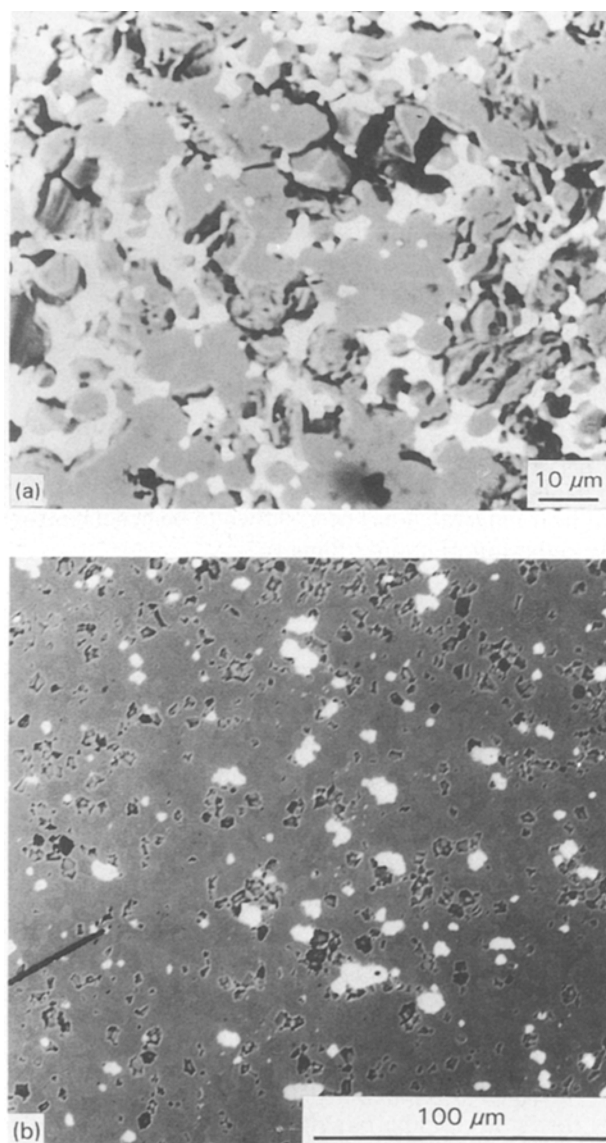
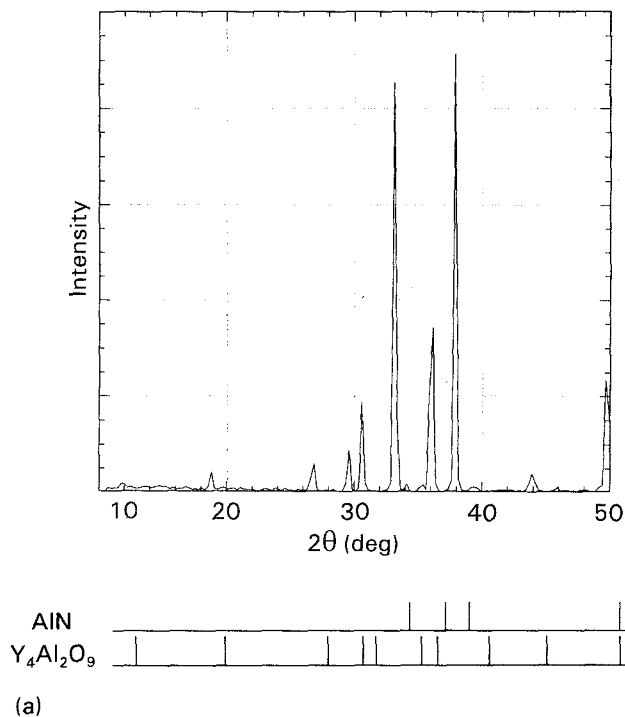


Figure 1 Backscattered electron images of samples of as-received substrates: (a) type I substrate, (b) type II substrate.



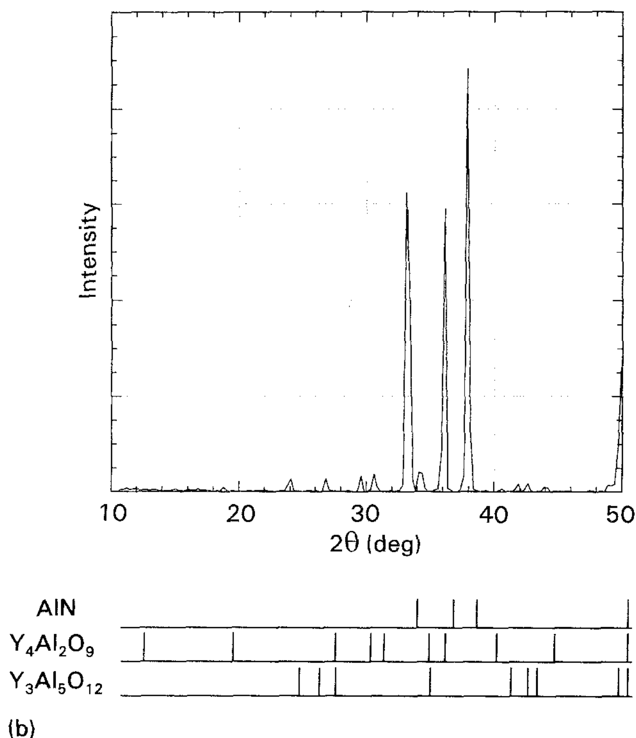


Figure 2 X-ray diffraction patterns of samples of as-received substrates: (a) type I substrate, (b) type II substrate.

sintering aid additions during processing. The overall yttria concentrations of the as-received substrate materials were determined to be approximately 4 vol% for type I substrates and approximately 2 vol% for type II substrates by energy dispersive X-ray spectroscopy on an electron microprobe (Joel 733).

3.1. Oxide film formation

After oxidation in air, the quality and the thickness of the oxide films grown on the substrates were determined through oxygen concentration maps and backscattered electron images by using an electron microprobe (Joel 733). Initial observations of the oxide films very clearly showed that the quality of the oxide layers depends on the characteristics of the substrate used. For example, the oxidation of a type I substrate resulted in more diffuse and thicker oxide layers than of a type II substrate annealed under identical conditions, see Figs 3 and 4. The differences in the substrate materials that were believed to affect the film formation were the quantity of the liquid sintering aid phases present, the particle size and possibly also carbon impurities. (Type I substrates were lightly greyish in colour.) In type I substrates, a defined oxide layer was not observed until the oxidation temperature was higher than 1200 °C, see Figs 3 and 4. For type II substrates, defined oxide films were observed as low as 1000 °C, see Fig. 5.

Additional electron microprobe analysis of oxide films grown by oxidation has been performed on type II substrates only, because uniform oxide films were observed over a broader temperature and time range

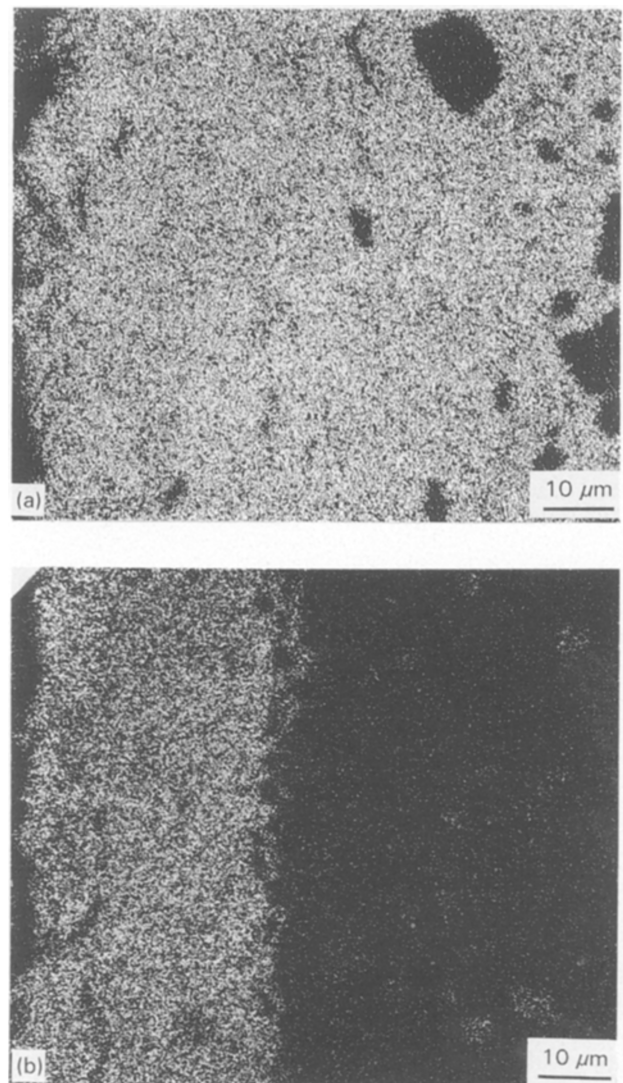


Figure 3 Oxygen maps for samples of substrates that were oxidized at 1300 °C in air at atmospheric pressure for 1 h: (a) type I substrate, (b) type II substrate.

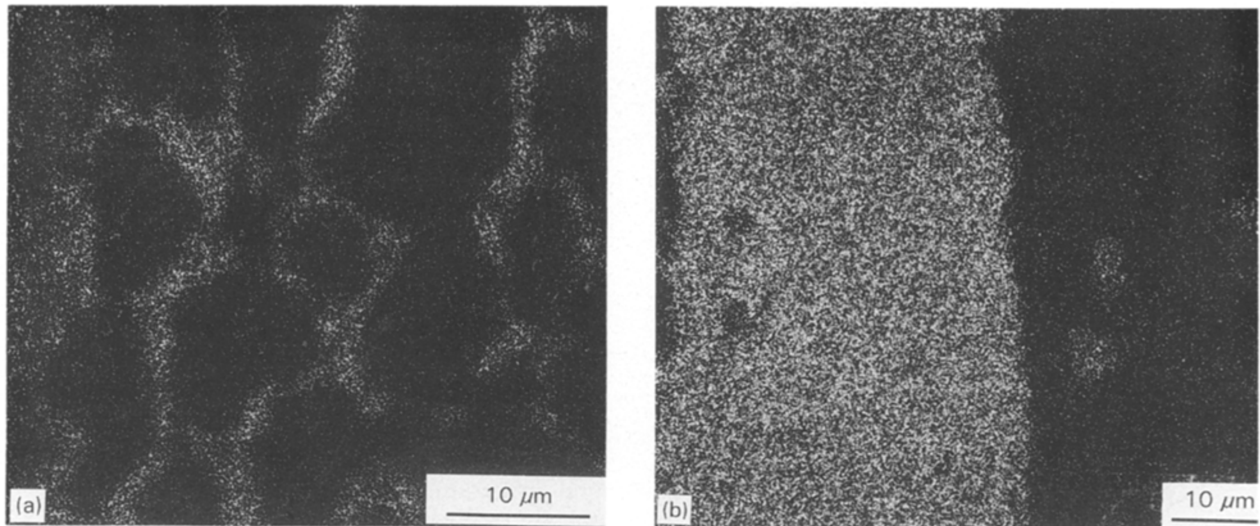


Figure 4 Oxygen concentration maps of substrate samples that were oxidized at 1200 °C in air atmospheric pressure for 12 h: (a) type I substrate, (b) type II substrate.

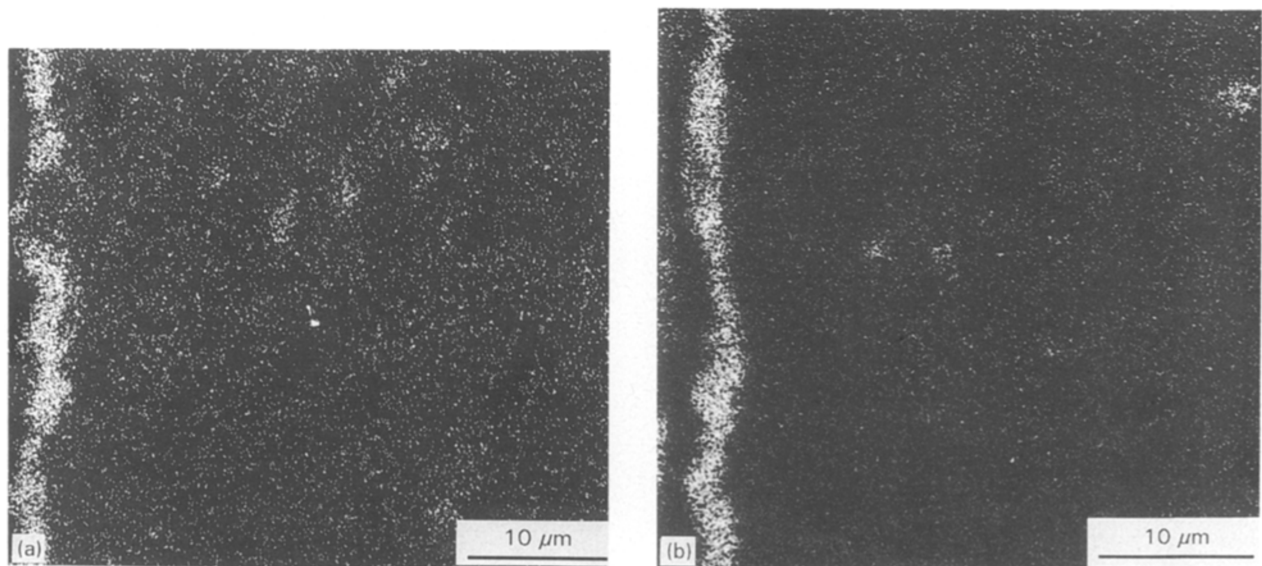


Figure 5 Oxygen concentration maps of type II substrate samples oxidized in air at atmospheric pressure (a) at 1000 °C for 24 h, and (b) at 1100 °C for 12 h.

than in the case of type I substrates. Oxide film thicknesses, Δx, were measured from the edge of the sample to the abrupt decrease in oxygen concentration which was clearly evident in the case of type II substrates but not in the case of type I substrates, see Figs 3–5. When uniform oxide films were obtained, the layers were examined with regard to oxygen and nitrogen concentration line profiles. The profiles were obtained by using an electron beam with a width of about 10 μm in a Joel 733 electron microprobe. Typical nitrogen and oxygen profiles are schematically shown in Fig. 6. It should be noted that the location of the original top surface (later Al_2O_3/gas) was easily identified by the abrupt increase in the oxygen intensity when moving the electron beam from the mounting material to the alumina film. The AlN/oxide film interface was identified by the abrupt decrease in oxygen intensity and the sharp increase in nitrogen intensity occurring when passing this interface from the alumina to the aluminium nitride side. Based

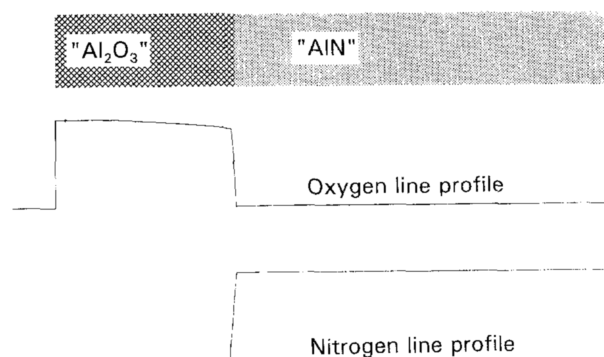


Figure 6 Schematic representation of typical nitrogen and oxygen line profiles for a uniform oxide film formed by direct oxidation on an aluminium nitride surface.

on the line profiles obtained, it is concluded that the oxide films formed contain predominately Al_2O_3; aluminium oxynitride phases were not present in a significant concentration. Within the resolution of the

electron microprobe, no aluminium oxynitride phases could be identified between AlN and Al_2O_3.

The compositions of the oxide films on types I and II substrates were determined by X-ray diffraction (XRD). Typical XRD patterns for oxide films grown on types I and II substrates are shown in Fig. 7. These XRD patterns contain two sets of peaks. The first set of peaks is of a relatively low intensity and can be attributed to the substrate material below the oxide film. The second set of more intense peaks has to be assigned to α-Al_2O_3 for both types of substrate. It should be noted that no peaks for any aluminium oxynitride phases could be identified. Based on the analysis of the XRD patterns and of the electron microprobe data, it is concluded that the oxide films formed by high-temperature oxidation of aluminium nitride substrates consist predominately of α-corundum. These results do not preclude the possibility of the presence of a very thin layer consisting of one or more aluminium oxynitride phases at the AlN/oxide film interface.

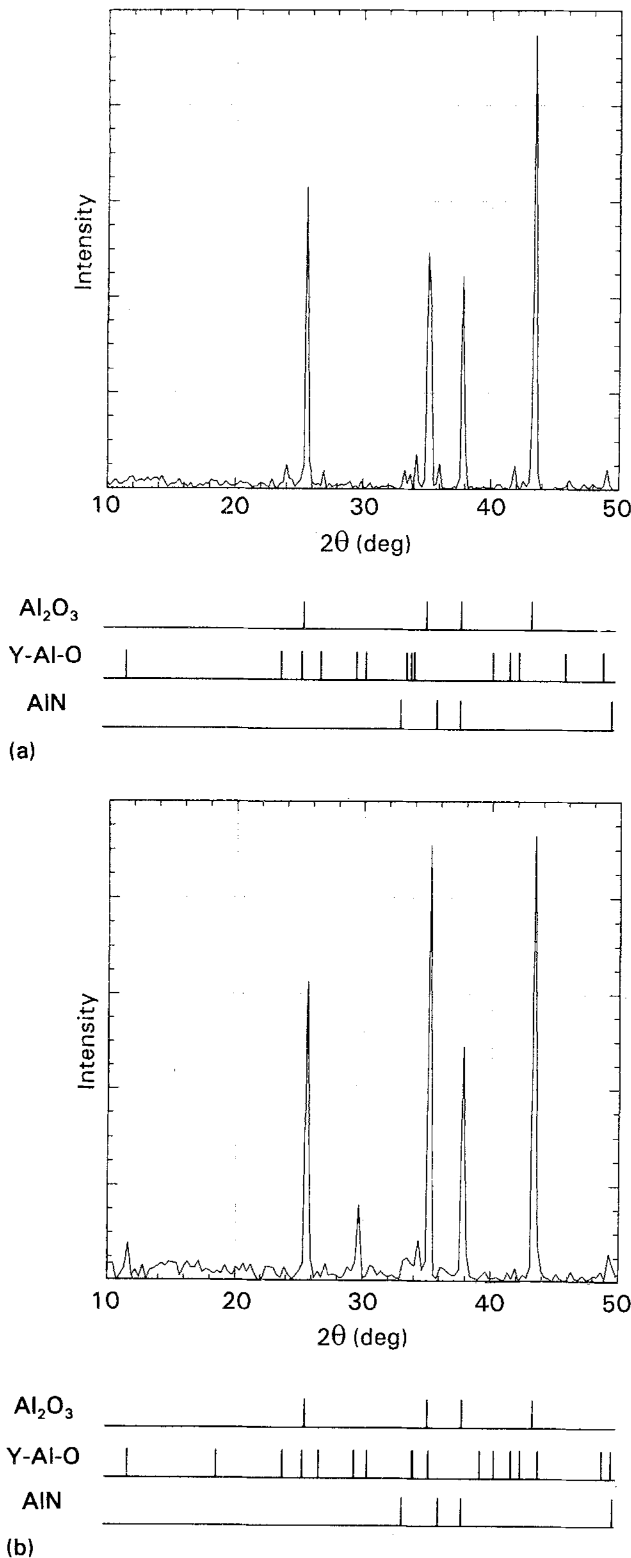


Figure 7 Thin-film X-ray diffraction patterns of aluminium nitride samples covered with an oxide film formed by direct oxidation of (a) type I substrate, (b) type II substrate. The patterns were obtained with a fixed omega angle of 2°.

3.2. Kinetics

In the oxidation of aluminium nitride to aluminium oxide without any oxynitride formation, three different processes are involved and may be rate limiting. These processes are: (i) a reaction at the AlN/Al_2O_3 interface; (ii) transport of nitrogen and oxygen ions across the "Al_2O_3" layer; and (iii) a reaction at the gas/solid interface. The overall driving force for the oxide formation process is given by the differences between the chemical potentials of both oxygen and nitrogen in the gas phase and in the aluminium nitride phase at the nitride/oxide interface. The situation outlined above can also be envisaged as three resistors in series with a battery representing the overall driving force, see Fig. 8. The current in this circuit is limited by the total resistance of the circuit given by the sum of individual resistances. If the resistors are similar in size, all of their resistances are important for the overall current. If one resistor has a much larger resistance compared with those of all others, this resistor limits the current. The current through the electrical circuit discussed before is analogous to the rate of oxide film growth. This rate is limited by a combination of subsequent reaction and transport processes. If the rates of these processes are similar, all processes are important for the overall oxidation reaction rate. If the rates of these processes are very different from each other, the slowest process becomes rate determining. For the rate-limiting process, there exist three possibilities: (1) $R_2 \gg R_1, R_3$, (2) $R_1 \gg R_2, R_3$, and (3) $R_3 \gg R_1, R_2$. Case 1 refers to a transport-controlled growth obeying a parabolic rate law,

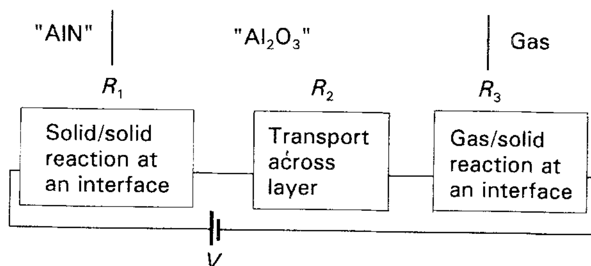


Figure 8 Schematic representation of the contributions of different types of processes involved in the high-temperature oxidation of aluminium nitride to the overall oxidation rate.

$\Delta x^2 \propto t$ [14, 15] and cases 2 and 3 correspond to two different interface process-controlled growth kinetics following a linear rate law, $\Delta x \propto t$ [16].

Usually, it is expected that at high temperatures the growth of a new phase by oxidation is diffusion controlled and therefore will follow parabolic rate kinetics, i.e. the square of the thickness of the oxide film linearly increases with time. A typical plot of the square of the thickness of oxide films grown in air on lapped, polished type II aluminium nitride substrates at 1250 °C is shown in Fig. 9 as a function of time. The data shown clearly demonstrate that the kinetics do not follow a parabolic rate law. In Fig. 10, data for different temperatures are plotted in the form of thickness versus time. From this plot it follows that the kinetics of the oxide film growth fit closely to a linear rate law. This result suggests that the kinetics of the overall oxidation reaction is almost completely controlled by one or more interface processes. No conclusion can be made whether one or more interface reaction steps are rate controlling. The rate-limiting process is probably not occurring at the gas/solid interface because the rate of oxidation is only slightly different for lapped and as-received substrates, see Fig. 11.

The rate of the oxidation of type II aluminium nitride substrates increases rapidly with increasing temperature. For temperatures above 1300 °C, an estimate of the rate of the oxidation in air at atmospheric pressure can be obtained from the Arrhenius-type plot shown in Fig. 12. This plot has been derived

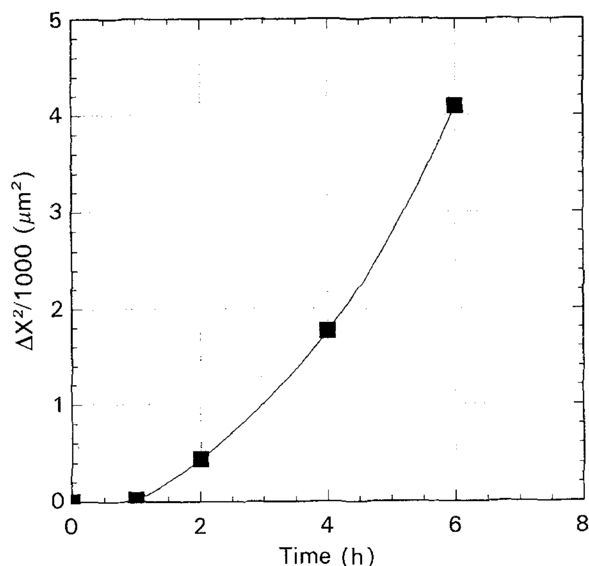


Figure 9 Square of observed oxide layer thicknesses as a function of time for the oxidation of aluminium nitride in air at atmospheric pressure at 1250 °C (type II substrates).

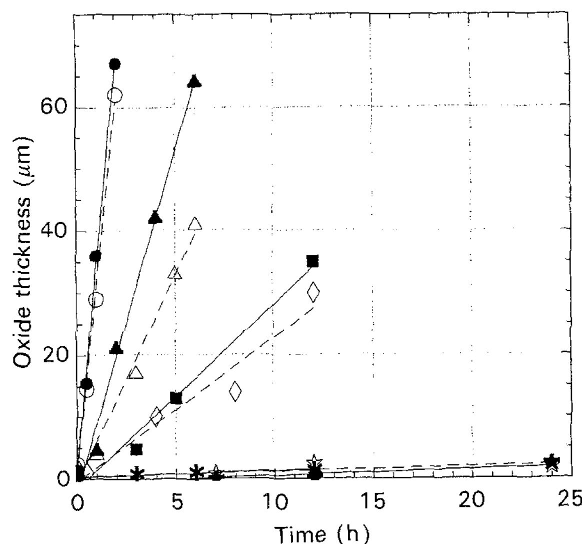


Figure 11 Comparison of observed oxide layer thicknesses as a function of time for the oxidation of (solid symbols) lapped and (open symbols) as-received samples of type II substrates for temperatures between 1000 and 1300° C. T: (○, ●) 1300 °C, (△, ▲) 1250 °C, (◇, ◆) 1200 °C, (☆) lapped, 1100 °C, (☆, ★) 1000 °C.

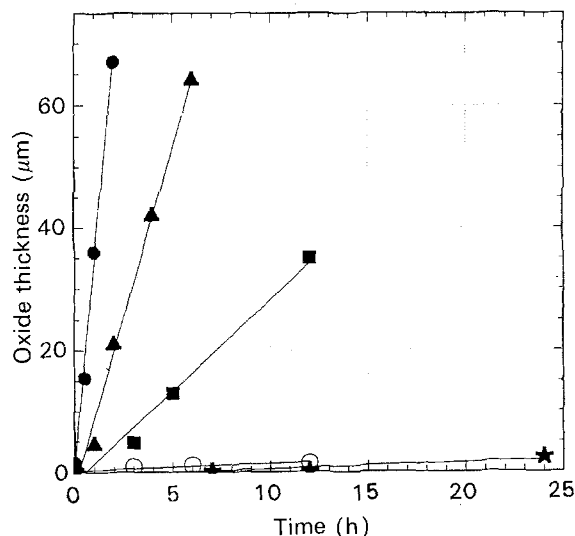


Figure 10 Observed oxide layer thicknesses as a function of time for the oxidation of lapped, type II substrate samples in air at atmospheric pressure at temperatures between 1000 and 1300 °C. T: (●) 1300 °C, (▲) 1250 °C, (■) 1200 °C, (○) 1100 °C, (★) 1000 °C.

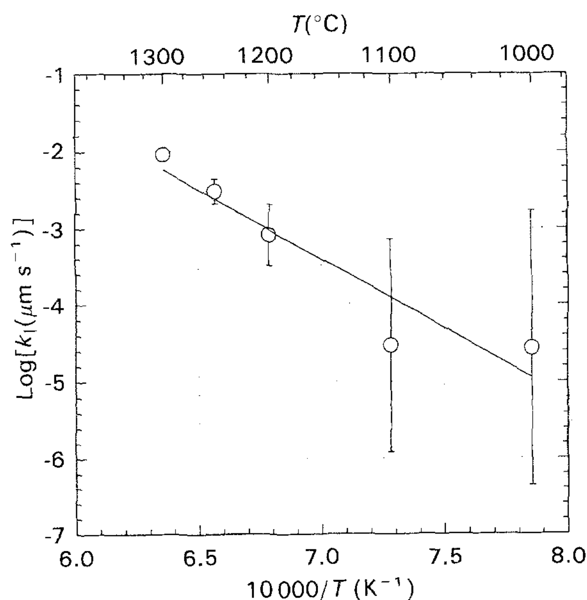


Figure 12 Temperature dependence of linear rate constants, k_l, for the oxidation of lapped aluminium nitride substrates (type II) derived by assuming completely linear reaction kinetics.

by assuming that the reaction kinetics for the growth of an oxide film on as-received type II AlN substrates, Fig. 10, were completely linear, i.e. $\Delta x = k_l t$; k_l is a linear rate constant.

The activation energy for the oxidation of lapped type II samples following a linear rate law, i.e. for k_l, has been determined to be $347 \pm 20\ \mathrm{kJ\,mol^{-1}}$. In the case of as-received type II substrates, a value of $311 \pm 20\ \mathrm{kJ\,mol^{-1}}$ was obtained. The difference between these two activation energies is just within the experimental error. Therefore, it cannot unequivocally be concluded that the surface characteristics have a significant influence on the oxidation kinetics, although it is likely.

It is interesting to compare these activation energy values for the oxidation of an experimental aluminium nitride substrate with those for commercially available substrates. In a brochure published by The Carborundum Company [17], data have been reported for the oxidation of commercially available aluminium nitride substrates of this company. The data refer to the oxidation in air at temperatures of 1000 and 1200 °C and can be fit very well by a linear rate law (but not by a parabolic rate law). By assuming an Arrhenius-type temperature dependence of k_l a value of $320\ \mathrm{kJ\,mol^{-1}}$ was calculated for the activation energy of k_l, i.e. not very different from the values derived from the experiments in this study. A reason for this observation may be that the characteristics of the experimental, type II substrate and Carborundum's commercial substrate are not very different.

In contrast to the observations in this study and the data reported in [17] from two studies on the oxidation of different aluminium nitride samples [9, 10], other authors have reported that they have observed kinetics which follow non-linear rate laws. Lavrenko and Alexeev [10] reported that the oxidized aluminium nitride samples had a porosity of 12%–16% before oxidation. The samples were prepared by isostatically pressing and sintering aluminium nitride powders with a particle size of 7.5 μm. The characteristics of these samples are significantly different from those of type II samples, for example, with regard to density and grain-size distribution. The kinetics of the oxidation of such aluminium nitride samples in pure oxygen at atmospheric pressure were reported to follow a parabolic rate law. This is clearly a different behaviour from that observed in this study and must be attributed to the existing differences between the samples oxidized. Billy *et al.* [9], reported the oxidation in air of aluminium nitride samples prepared by sintering aluminium nitride powder and using 1 wt% yttria as a sintering aid. The samples contained yttrium aluminates as secondary phases, similar to the substrates considered in this study. The oxidation kinetics observed at temperatures below 1550 °C cannot be described by a simple rate law, i.e. a linear or parabolic one. A detailed analysis of the data with regard to parabolic and linear rate constants and their temperature dependence was not reported by Billy *et al.* [9]. The authors claim, without proof, that an area exists where the data follow a parabolic rate law and they even report a value for an activation energy for the parabolic rate constant. Based on the details given by Billy *et al.* [9], it is questionable whether an appropriate analysis has been performed and whether the activation energy reported has a significant physical meaning.

In a study by Suryanarayana [13], the kinetics of the oxidation of aluminium nitride powders in air was investigated. The samples were fine- (0.5 μm) and coarse-grained (3.1 μm) powders. The kinetic data were fit to a mixed kinetic model, i.e. including contributions from transport and interface reaction processes. In view of the oxide layer thicknesses studied here, only the data for the coarse-grained samples are compared with those from this study. For the activation energy of the process leading to a linear rate law contribution, Suryanarayana reported an activation energy of $297\ \mathrm{kJ\,mol^{-1}}$, i.e. a value which is of the same order of magnitude as the values derived from the present study. However, it is unclear what this agreement really means in view of the different sample characteristics, for example, different (unknown) impurity levels.

An important goal of this study was to identify suitable conditions to grow 1–5 μm thick oxide films with well-defined thicknesses on aluminium nitride samples for future interdiffusion studies. From the data shown in Fig. 10 it follows that the rate of oxidation in air above 1300 °C is too large to control well the thickness of oxide films by adjustment of the oxidation time. This is because small deviations from the calculated oxidation time would result in significantly different thicknesses. Below 1200 °C, the rate of oxidation becomes very slow and it has been observed that uniform oxide films cannot be formed on all substrates. Based on these observations, it is concluded that the growth of thin (1–5 μm), uniform oxide films on aluminium nitride substrates by direct oxidation in air can best be accomplished at temperatures between 1200 and 1300 °C with reaction times between 0.1 and 10 h.

For comparison, type II substrates were also oxidized in flowing "argon" (argon with approximately 200 p.p.m. oxygen) at atmospheric pressure. Because of the lower oxygen activity, the temperatures selected to be studied were 1300 and 1400 °C. The as-grown oxide films were found to be of the same quality as those prepared in air, see Fig. 13. Results of XRD studies of the oxide films grown by oxidizing in "argon" are similar to those obtained for oxide films grown in air and reveal that the oxide layers grown by oxidation in "argon" also consist predominately of α-Al_2O_3. Data for this oxide film growth as a function of time are shown in Fig. 14. They can also be approximated by a linear rate law as in the case of oxide film growth by oxidation in air.

As expected, the rate of oxide film growth is significantly affected by the oxygen partial pressure. For example, when as-received type II substrates are oxidized at 1300 °C in air and in "argon", the linear rate constants, k_l, have values of 8.4×10^{-7} and $1.15 \times 10^{-8}\ \mathrm{cm\,s^{-1}}$, respectively. This variation in the reaction rates by a factor of almost 75 may allow a

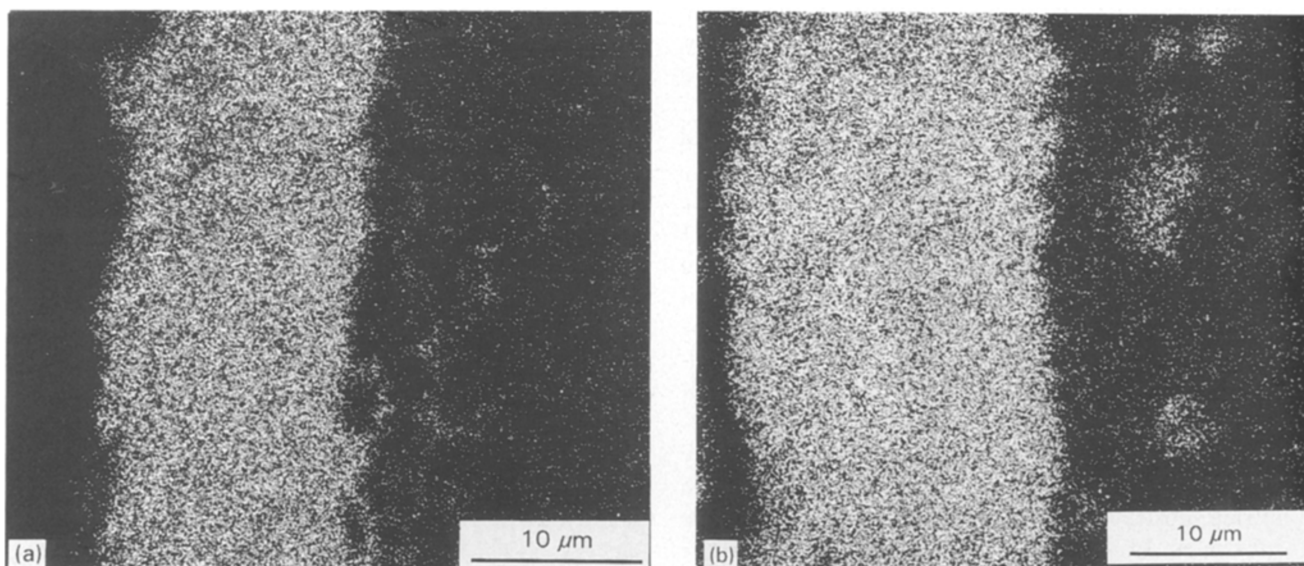


Figure 13 Oxygen concentration maps of samples of type II substrates oxidized in "argon" (containing about 200 p.p.m. oxygen) (a) at 1300 °C for 57 h and (b) at 1400 °C for 5 h.

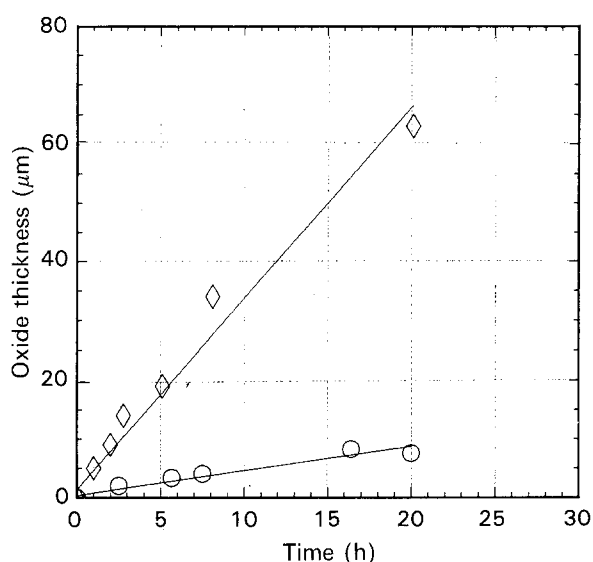


Figure 14 Observed oxide layer thicknesses as a function of time for the oxidation of samples of type II substrates in "argon" (containing about 200 p.p.m oxygen) at atmospheric pressure at (○) 1300 °C, $a_{O_2} = 10^{-3.65}$, and (◇) 1400 °C, $a_{O_2} = 10^{-3.29}$.

better control of the formation of thin, uniform oxide films on aluminium nitride substrates. In fact, we have observed that uniform oxide films can even be formed of type I substrates by selecting appropriate conditions with regard to temperature and oxygen partial pressure.

At 1200 °C in air, the rate of the oxide film growth on as-received type II substrates, k_l, is 6.5 $\times 10^{-8}$ cm s^{-1}. This value can be compared with one derived from the data reported elsewhere [17]. To make this comparison, the data from [17] had to be converted from mass changes per square centimetre to oxide film thicknesses. This has been done by assuming that the oxide films formed were fully dense. The resulting value for k_l was 4.6×10^{-8} cm s^{-1}, i.e. similar to the value found in this study. The difference between the two values for k_l may be attributed to microstructural and impurity concentration differences between the different substrates. However, it may also have been caused by differences between the actual temperatures present during the different measurements. A temperature difference of about 20 °C would be sufficient to cause the deviation observed.

4. Conclusions

The growth of oxide films on two types of aluminium nitride substrates of different quality and origin has been studied as a function of temperature. It was found that type I substrates produced an inferior quality oxide film as compared to type II substrates. The major differences between the two types of substrate were particle size and liquid sintering aid concentration. Analysis of the grown oxide films by X-ray diffraction and wavelength dispersive spectroscopy revealed that the composition of the film was primarily Al_2O_3. Aluminium oxynitride phases could not be identified within the resolution of the experimental techniques used.

The growth kinetics were analysed for type II substrates only, because the films on such substrates were well defined as opposed to diffuse ones usually observed on type I substrates. The thickness of the oxide films grown as a function of time could not be fit to a parabolic rate law, but instead were best approximated by a linear rate law. This type of rate law suggests that the rate-limiting step is an interface-controlled process, most likely occurring at the AlN/Al_2O_3 interface.

The effect of oxygen partial pressure on the growth of oxide films on aluminium nitride substrates was also studied. The kinetics of the growth of oxide films in "argon" (with 200 p.p.m. oxygen present) at atmospheric pressure can also be approximated by a linear

rate law. By lowering the oxygen partial pressure, the oxide film growth is slowed significantly.

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References
1. G. A. SLACK, R. A. TANZILL, R. O. POHL and J. W. VANDERSANDE, *J. Phys. Chem. Solids* **48** (1987) 641.
2. G. A. SLACK, *ibid.* **34** (1973) 321.
3. R. W. RICE, J. H. ENLOE, J. W. LAU, E. Y. LUH and L. E. DOLHERT, *Ceram. Bull.* **71** (1992) 751.
4. N. IWASE, K. ANZAI and K. SHINOZAKI, *Solid State Technol.* **29**(10) (1986) 135.
5. L. M. SHEPPARD, *Ceram. Bull.* **69** (1990) 1801.
6. F. MIYASHIRO, N. IWASE, A. TSUGE, F. UENO, M. NAKAHASHI and T. TAKAHASHI, *IEEE Trans. Components, Hybrids, Manuf. Technol.* **13** (1990) 313.
7. A. V. VIRKAR, T. B. JACKSON and R. A. CUTLER, *J. Am. Ceram. Soc.* **72** (1989) 2031.
8. J. H. HARRIS, R. A. YOUNGMAN and R. G. TELLER, *J. Mater. Res.* **5** (1990) 1763.
9. M. BILLY, J. JARRIGE, J. P. LECOMPTE, J. MEXMAIN and S. YEFSAH, *Rev. Chim. Miner.* **19** (1982) 673 (in French).
10. V. A. LAVRENKO and A. F. ALEXEEV, *Ceram. Int.* **9**(3) (1983) 80.
11. A. D. KATNANI and K. I. PAPATHOMAS, *J. Vac. Sci. Technol.* **A5** (1987) 1335.
12. D. SURYANARAYANA, L. J. MATIENZO, and D. F. SPENCER, *IEEE Trans. Components, Hybrids, Manuf. Technol.* **12** (1989) 566.
13. D. SURYANARAYANA, *J. Am. Ceram. Soc.* **73** (1990) 1108.
14. G. TAMMANN, *Z. Anorg. Allg. Chem.* **111** (1920) 78 (in German).
15. N. B. PILLING and R. E. BEDWORTH, *J. Inst. Metals* **29** (1923) 529.
16. H. SCHMALZRIED, "Solid State Reactions," 2nd Ed. (Verlag Chemie GmbH, Weinheim, 1981) pp. 171–8.
17. "Aluminium Nitride - The Choice for Thermal Management Applications", Brochure A-14065A (The Carborundum Company, Electronics Materials Group, Substrates Division. Sanborn, NY, USA, 1990).

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