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# Thermo-compression bonding of alumina ceramics to metal

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Alumina ceramics and Kovar with aluminum interlayer are pressed together under vacuum at temperatures around 600°C for joining. This process produces mechanically strong ceramic to metal bonds in one step in an economic manner. In order to arrive at the optimum conditions for solid-state bonding, effects of bonding temperature, pressure and time on the bond strength have been studied. Bonding kinetics is also elucidated. Irradiation of 99% $Al_2O_3$ ceramics by 4–5 MV X-rays has been found to increase the bond-strength sharply from 33 to 60 MPa with a dose of 15 k Rads for bonding temperatures around 540°C. The apparent activation energy for the bonding process ($Q_B$) depends strongly on the type of alumina ceramics. Irradiation of alumina ceramics (99%), prior to joining with Kovar, accelerates the solid-state bonding by reducing ($Q_B$) from 209 to 76 kJ/mole. © *2004 Kluwer Academic Publishers*

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## 1. Introduction

Solid-state bonding of alumina ceramics to metal by thermo-compression technique using aluminium interlayer is a convenient and fast process for producing mechanically strong joints. Thermo-compression is attractive because it requires only a single stage operation at relatively low temperatures (500–600°C) in comparison to other joining processes [1, 2]. Solid-state bonding is a diffusion-controlled phenomenon. It is well known that diffusion is accelerated in presence of defects [3]. High-energy irradiation is one method to introduce point defects in solids. Now-a-days irradiation is used to accelerate solid-state reactions [4]. Therefore, in this investigation the effect of radiation on solid-state bonding of alumina ceramics to Kovar has also been studied.

## 2. Experimental procedure

### 2.1. Bonding set-up

Fig. 1 gives a schematic view of the set-up for thermo-compression bonding. The bonding set-up was designed for pressing in hydrogen, inert or vacuum atmosphere. The envelope was made out of transparent quartz tube. The open ends were closed by stainless steel cups with Viton 'O'-ring seals. The end cup seals were so designed that those were free to slide without impairing the vacuum or gas atmosphere inside. The bottom cup had a groove to position the lower punch, a sheathed thermocouple for measurement of temperature and a gas purging port with stopper. The top cup had flexible vacuum connection and groove to align the upper punch. A ceramic ball was put between the top punch and the bonding pair to ensure alignment of the bonding faces. After loading the sample between the punches inside the quartz tube, terminated by the cups, the assembly was placed on a hydraulic ram and pressure was applied to the end cups on attainment of a vacuum of $1 \times 10^{-6}$ torr or better. Induction heating of the bonding pair was performed through the stainless steel susceptor inside the quartz tube. For evaluation purpose, samples were prepared by bonding grooved Kovar tubes to disk shaped alumina ceramics under vacuum.

### 2.2. Bonding process

Ceramics with three different percentages of alumina were used for bonding to Kovar using 20 $\mu$m thick aluminium interlayer by solid-state route. All the ceramic samples were lapped with 600 mesh silicon carbide powder to give uniform surface finish (Ra = 0.77 $\mu$m). After lapping, the ceramics were cleaned in trichloro ethylene (TCE) and dilute nitric acid before being fired in air at 800°C for one hour to burn out any trace of organic matter on the surface. After machining, the bonding faces of Kovar tubes were polished with 600 mesh silicon carbide powder to get

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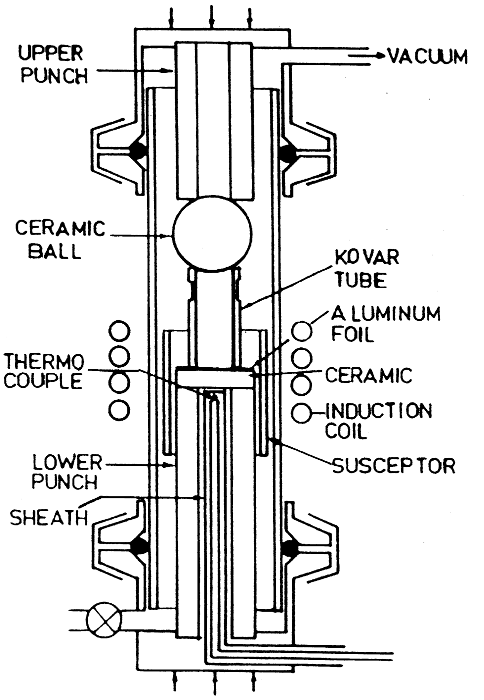


*Figure 1* Thermo-compression bonding set-up.

flat, smooth surface and then cleaned in TCE or acetone using ultrasonic agitation; aluminium foil was also cleaned in the same way. Cleaned pieces were assembled inside the vacuum envelope as shown in Fig. 1. The ceramic ball ensured proper alignment of the mating surfaces. Bonding variables, namely temperature, pressure, time, ceramic composition etc. were optimized by preparing test samples under different values of a particular variable, while keeping other variables unchanged and finding the effect of that on the tensile bond strength.

## 2.3. Radiation assisted bonding

Five batches of 99% alumina ceramics, prepared as mentioned earlier, were irradiated with respectively 0, 2 k, 5 k, 8 k and 15 k Rads of 4–5 MV Bremsstrahlung X-rays from a Linac. Using these ceramics, test bonds were made in the same way as mentioned in the previous section and characterized for tensile bond strengths.

## 2.4. Tensile bond strength

The technique of measurement is similar to that employed by Floid [5] with some modification. The bonded sample is clamped on the groove provided on the metal partner and the ceramic is de-bonded by application of force from inside. The force is transmitted to the ceramic via a rubber pad. The device is shown in Fig. 2.

The force on the ceramic is measured from the hydraulic oil pressure multiplied by ram area. Bond strength BS is given by the following relation:

$$\mathrm{BS} = p\left(\frac{A}{S}\right) - K \qquad (1)$$

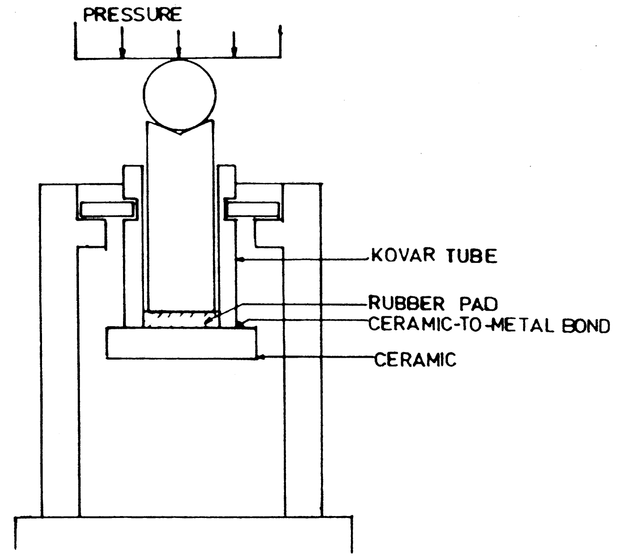


*Figure 2* Arrangement for measurement of bond-strength.

where, $P$ = oil pressure at which the bond fails. $A$ = area of the ram of the hydraulic press. $S$ = actual area of bonding, $K$ = a constant dependant upon the weight of the ram and friction between ram and the barrel.

## 2.5. Microstructural studies

In order to study the nature of the interface, bonded samples were fractured transverse to the bond and debonded along the interface by application of force. Broken surfaces were coated with thin layer of carbon and scanning electron micrographs (SEM) were taken.

## 3. Results and discussion

## 3.1. Effect of bonding temperature

For three types of alumina ceramics (90, 96 and 99% $Al_2O_3$), solid state bonding has been conducted at a pressure of 75 MPa for 10 min in the temperature range of 450 to 650°C and bond strengths are measured. The influence of bonding temperature on bond strength is shown in Fig. 3.

For each type of ceramics, bond strength goes through a maximum and the optimum bonding temperatures are 550, 512, and 575°C for 90, 96, and 99% alumina ceramics respectively. In contrast to this observation. Nicholas and Cripsin [6] reported that the bond strength of alumina-stainless steel couple increases monotonically with the fabrication temperature for a bonding time of 30 min. In our experiment, the origin of maxima in the bond strength versus bonding temperature curves is probably due to a balance between diffusion bonding and thermal stress. High temperature promotes diffusion bonding but it also gives rise to residual stresses in the bond, which will be more when cooled fast from higher bonding temperature: this would account for the fall in bond strength for samples made at higher temperature. In solid-state bonding, ultimate bond-strength is determined by the bulk strength of the ceramic involved. The flexural strength of alumina ceramics decrease from 310 to 280 MPa with

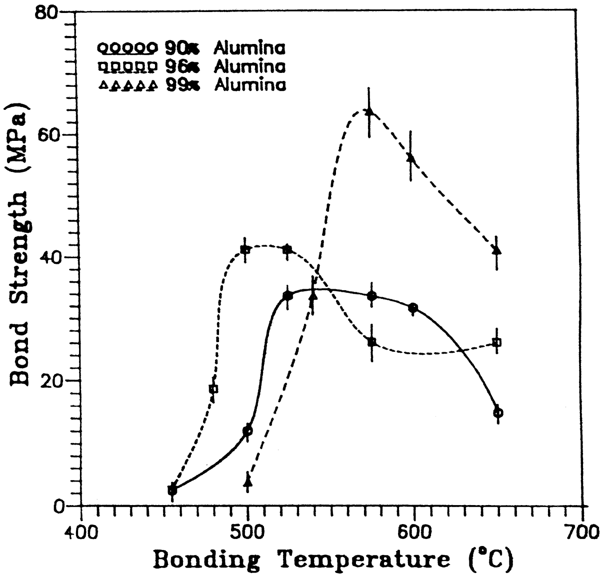


*Figure 3* Variation of bond-strength with temperature (Pressure 75 MPa. Time 10 min).

decrease in percentage of alumina from 99 to 90 in the body, as per the manufacturer's catalogue. Therefore, the maximum bond strength is observed to decrease from 99% alumina ceramic to 90% alumina ceramic. Though the bonds some times fail within the ceramic, the breaking strength obtained was much less than the flexural strengths. This may be due to immature failure caused by localised stress concentration. It is seen that, at a temperature lower than 500°C, 96% alumina produces stronger bonds than other two. A plausible explanation is that, in polycrystalline alumina, grains as well as grain-boundary glass are responsible for formation of bond with the metal. The composition of glassy phases and their percentage are different in three different ceramics. The temperature dependence of bond strength may be different for alumina-metal and glass-metal system. The difference in thermal expansion coefficient of different phases at the interface also plays a role in determining the bond strength. According to Nicholas and Crispin [6] the influence of fabrication temperature on the bond strength (BS) of joints failing at $Al/Al_2O_3$ interface can be represented by the expression

$$BS = BS_0 e^{Q_B/RT} \quad (2)$$

where $Q_B$ is the apparent activation energy. Following this approach, $Q_B s$ are determined from the slopes of the straight-line plots of $\ln B$ vs. $1/T$ upto the optimum bonding temperatures. The values of activation energies are given in Table I. It is observed that $Q_B$ is highly dependent upon the type of alumina ceramics. The 96% alumina ceramic shows the highest activation energy 295 kJ/mole as compared to 90 and 99% alumina ceramics.

Oishi and Kingery [7] calculated the activation energy for diffusion of $O^{-2}$ in alumina in the impurity-controlled or structure sensitive range for one set of samples. The energy varied depending upon the type of alumina. Though the activation energy for intrinsic oxygen diffusion in alumina is 635.4 kJ/mole a typical value of 241.2 kJ/mole was reported for non-intrinsic region [7]. The activation energy for intrinsic diffusion of Al in pure $Al_2O_3$ in the temperature range 1600 to 1850°C, estimated from diffusion coefficient versus inverse temperature plot, reported by Kingery *et al.* [8] is approximately 475 kJ/mole. As the apparent activation energies obtained for solid-state bonding are falling in the range of the activation energies for non-intrinsic diffusion in alumina, it can be inferred that diffusion in alumina controls the solid-state bonding. As the maximum bond strength of 63.7 MPa was exhibited by 99% alumina ceramics, for further studies on the effect of other variables, 99% alumina has been chosen.

TABLE I Activation energies for solid-state bonding

| Type of alumina | Apparent activation energy, ($Q_B$), (kJ/mole) | Bonding conditions |
|---|---|---|
| 90% | 176 | Present work: bonding pressure: 75 MPa; bonding time: 10 min; metal partner: Kovar; and interlayer: 0.02 mm thick aluminium. |
| 96% | 295 | |
| 99% | 209 | |
| 97% | 83.3 | Nicholas and Crispin [6]: bonding pressure 50 MPa: bonding time: 30 min metal partner: stainless steel: interlayer: 0.5 mm thick aluminium. |

## 3.2. Effect of bonding pressure

For studying the effect of bonding pressure, samples were prepared with 99% alumina ceramic at three different temperatures: 575, 600 and 650°C, employing pressures from 7.5 to 75 MPa. The dependence of bond-strength on bonding pressure for 99% alumina is shown in Fig. 4. In this case the bonding time was kept 10 min.

It is observed that bond strength increases monotonically with pressure. But there is a limit to the maximum

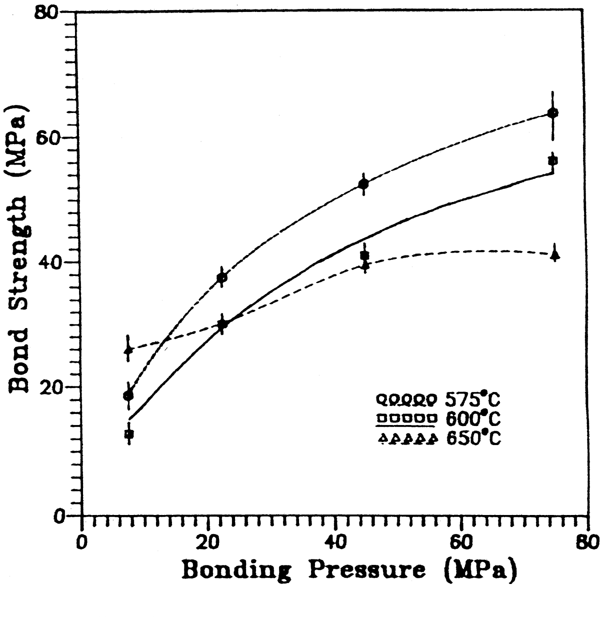


*Figure 4* Effect of bonding pressure and temperature on bond-strength.

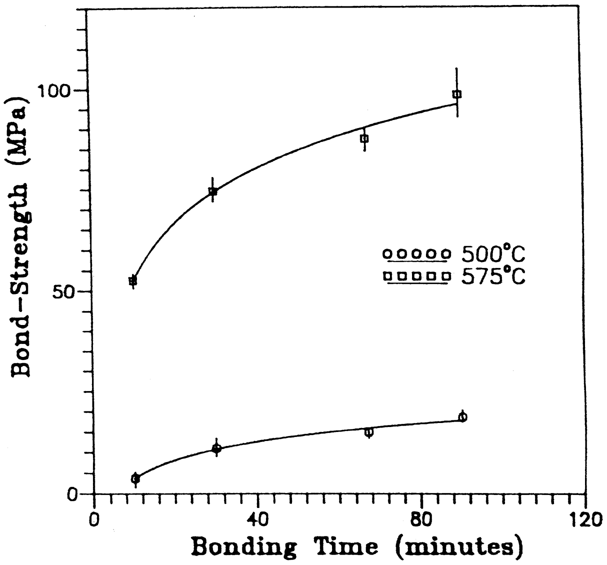


*Figure 5* Effect of bonding time and temperature on bond-strength (Ceramic: 99% alumina, bonding pressure: 45 MPa).

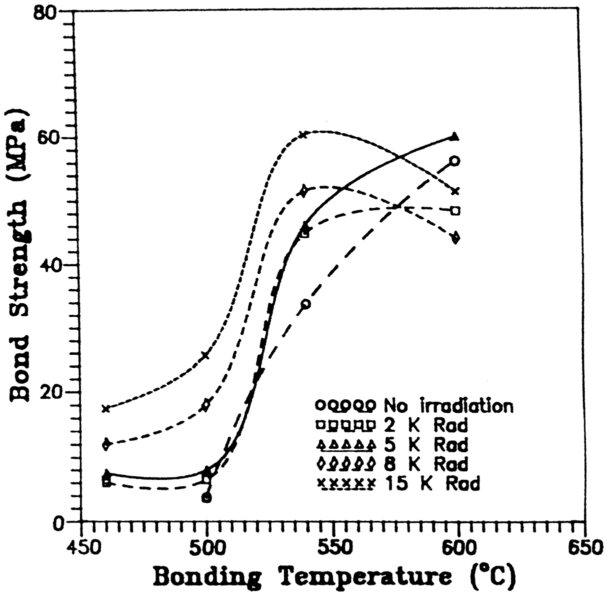


*Figure 6* Effect of irradiation on 99% alumina and bonding temperature on bond-strength (Radiation energy: 5 MV. Bonding pressure: 75 MPa. Bonding time: 10 min).

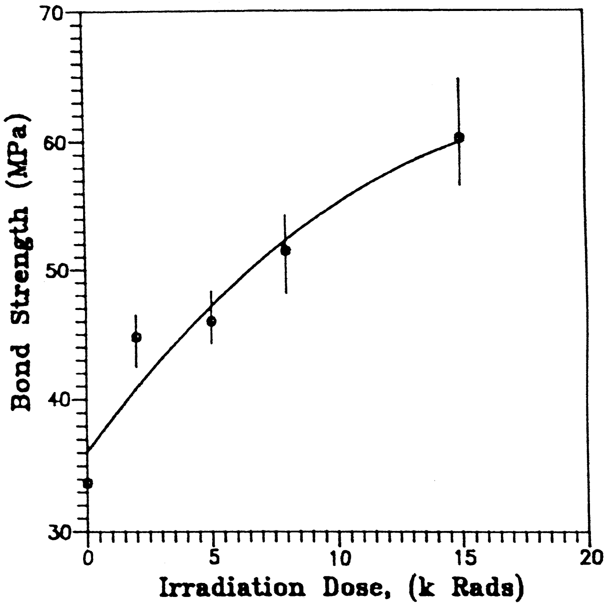


*Figure 7* Effect of dose of irradiation of 99% alumina ceramics on bond-strength (Radiation energy: 5 MV. Bonding pressure: 75 MPa. Bonding temp. 540°C. Bonding time: 10 min).

pressure, because a high pressure, particularly at high temperatures, causes deformation of the metal partner Kovar.

## 3.3. Effect of bonding time

The effect of bonding time has been studied on Kovar & 99% Alumina ceramic joints fabricated respectively at 500 and 575°C under 45 MPa pressure. Fig. 5 shows that the bond strength increases continuously with bonding time up to 90 min. The effect of bonding time at 600°C was not studied as 575 was the optimum bonding temperature for 99% alumina.

## 3.4. Radiation assisted solid-state bonding

Variation of bond-strength with bonding temperature after subjecting 99% alumina ceramics to increasing levels of irradiation is shown in Fig. 6.

It is seen that irradiation causes substantial improvement in bond strength in comparison to unirradiated samples. The effect of irradiation is most prominent at bonding temperatures about 540°C. At bonding temperatures higher than 540°C the effect of irradiation saturates. This may be attributed to annealing out of point defects caused by irradiation. The effect of irradiation on bond-strength is clearly brought out in Fig. 7. A monotonic rise in bond-strength up to a dose of 15 k Rads is observed which is around 179% of that of the unirradiated samples.

The activation energies for bonding ($Q_B$) are determined from the lnBS vs. inverse temperature plots. The influence of irradiation on the apparent activation energy is presented in the Fig. 8. It can be seen that ($Q_B$) reduces drastically upto a dose of 2 k Rad and there after slowly.

$\alpha$-alumina may be described as having crystal structure based upon the hexagonal closed-packing of oxygen ions, with two-thirds of the octahedral interstices being occupied by the aluminium ions. Pells and Phillips [9] have shown that the displacement energy for aluminium ion is considerably lower than that for the oxygen ion in $\alpha$-alumina. Based on high-voltage electron microscopy Das [10] has reported that at room temperature, the threshold electron energy for the formation of displacement induced defect clusters in $\alpha$-alumina is 240 to 300 keV depending upon the orientation. It has been reported that the electrons having energies between 200 and 400 keV can displace only aluminium ions. Oxygen ions will be displaced only for energies greater than 400 keV. It has been estimated that 1 MeV electrons produce five times more aluminium displacement than oxygen [11]. When high-energy photons interacts with any matter it causes photoelectric effect, pair-production, and

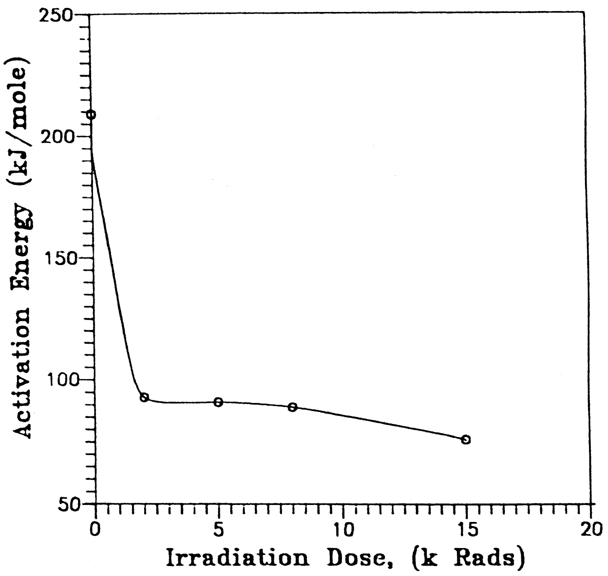


*Figure 8* Influence of irradiation on the apparent activation energy for solid-state bonding of 99% alumina to Kovar.

Compton-scattering. The last two predominates in 5 MeV photon energy. By Compton scattering the recoil electrons may have energies from 0 to a maximum of $2E(\lambda + 2)$(MeV) [12]. Where $E$ is the photon energy in MeV and $\lambda$ is the Compton wavelength, defined as:

$$\lambda = \frac{m_e \cdot c^2}{E} = \frac{0.511}{E}(\text{MeV})$$

$m_e$ = mass of electron and $c$ = velocity of light.

Therefore, 5 MV Bremssturlaung photons coming from Linac can easily produce electrons upto 4.7 MeV, which is more than sufficient to produce lattice defects by displacement of ions, particularly $Al^{3-}$ in $\alpha$-alumina.

In order to assess the damage of the lattice by irradiation, alumina substrate has been subjected to XRD analysis in three conditions as shown in Table II. This table clearly shows that irradiation modifies the alumina lattice due to substantial formation of defects and they do not get fully annealed out even at 900°C in 30 min. It may also be mentioned that the colour of the alumina substrate changes from off white to yellow after irradiation and to bluish white after annealing the irradiated specimen.

TABLE II Effect of irradiation on lattice parameters of 99% alumina

| Lattice parameters | Before irradiation | After irradiation with 5 M Rad of 5 MV X-rays | After irradiation and annealing |
|---|---|---|---|
| $a_o$(A) | 4.7392 | 4.6853 | 4.6842 |
| $c_o$(A) | 12.9571 | 13.0175 | 13.0547 |
| $c_o/a_o$ | 2.7340 | 2.7784 | 2.7870 |
| V($A^3$) | 252.03 | 247.47 | 248.06 |

Reported value of lattice parameters of $\alpha$-alumina (Sys: Trigonal, File: 10–173) are: $a_o$ = 4.758 Å, $c_o$ = 12.991 Å.

From the above discussion it may be assumed that the irradiation reduces the activation energy for diffusion (particularly of $Al^{3+}$ in $Al_2O_3$) by creating lattice defects. This phenomenon is reflected in the reduction of the apparent activation energy ($Q_B$) for solid-state bonding. In other words, the solid-state bonding process is strongly influenced by diffusion occurring at the $Al/Al_2O_3$ interface.

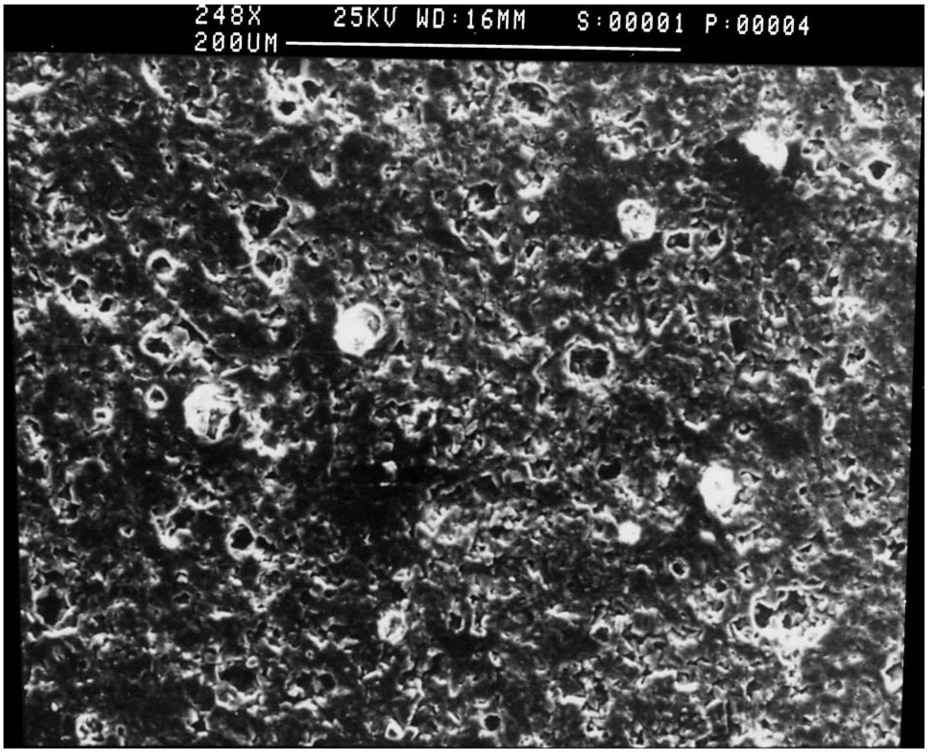
### 3.5. Microstructure of the interface

During the tensile testing of solid-state bonded alumina/aluminium/Kovar samples, failures were observed either in aluminium/alumina side or inside the ceramic, but never in aluminium/Kovar side. Therefore, for microstructure study, the alumina/aluminium interface has been chosen. Microstructural studies were conducted with unirradiated samples only, as irradiation did not produce any noticeable microstructural change.

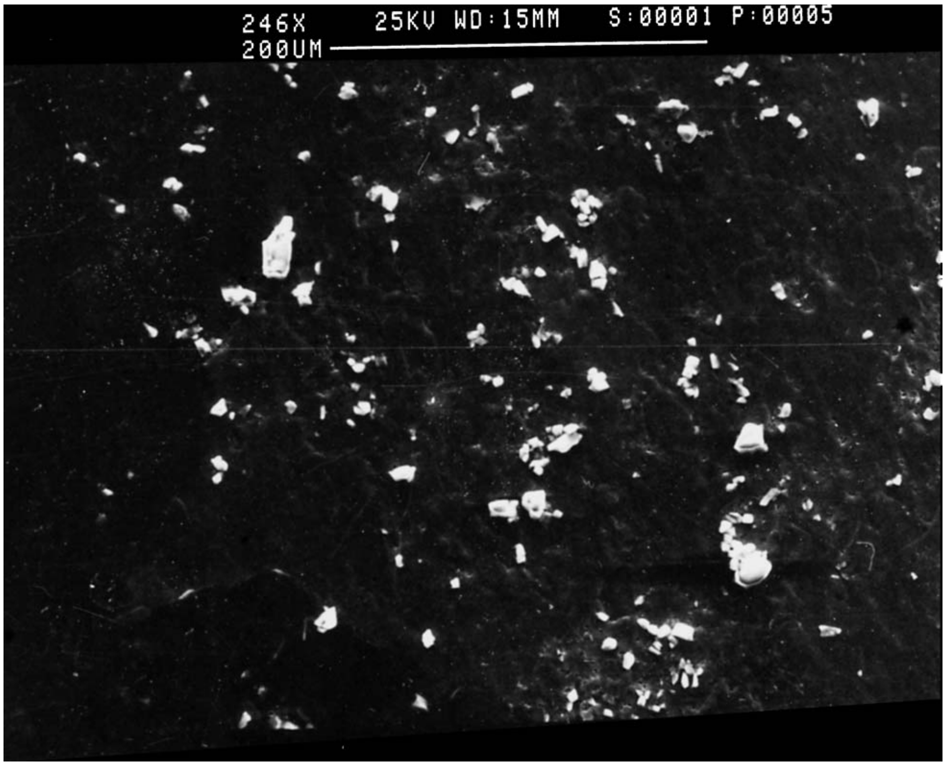
For taking SEM of the interface, 90 and 99% alumina ceramics were joined together using 0.02 mm thick aluminium interlayer at a constant pressure of 34 MPa for 45 min, but using two different temperatures: 500°C (type-I) and 550°C (type-II) respectively. The representative tensile strength sample, prepared under type-I conditions with 90% alumina ceramic exhibited bond-strength of 12 MPa only. Whereas, 75 MPa was obtained for 99% alumina ceramics bonded under type-II conditions.

After debonding the type-I sample, the aluminium foil remained with the 99% alumina. Fig. 9a shows the SEM of the 90% alumina side of debonded sample-1. Fig. 9b is the SEM of the other side (i.e., aluminium side) of the couple. Due to ceramic grain pull-outs void like features have formed (Fig. 9a). The aluminium side of the fractured surface clearly shows the pulled out alumina grains, which have been strongly bonded (Fig. 9b). The high magnification fractographs of this side show the pulled out alumina grains more clearly (Fig. 9c) and development of micro cracks (Fig. 9d). It also shows that at 500°C only partial bonding has taken place. Fig. 10a and b show the transverse section of type-II sample at different magnifications. After debonding of this sample, very little aluminium was visible at the interface: failure occurred on both the ceramics. Fig. 11a and b show the debonded 90% alumina side with pulled out grains from 99% alumina substrate. Grains of 99% alumina substrate are easily recognised because of their larger size. In these fractographs round pores (closed porosity), in the sintered alumina can be seen. Intergranular cracking of the ceramic is also noticeable. Thin (around 2 $\mu$m) aluminium layer is visible at the interface between 90 and 99% alumina substrates although starting aluminium foil thickness was 20 $\mu$m. Fig. 12a and b show the debonded 99% alumina side with pulled out grains from 90% alumina substrate. The flow of aluminium along the alumina grain boundaries is visible.

In order to identify the products of interfacial reaction between aluminium interlayer and Kovar by XRD technique, Kovar powder was mixed with aluminium flakes produced from the aluminium foil, compacted and vacuum hot-pressed under 75 MPa pressure at 600°C for 1/2 an hour to simulate the bonding condition. The XRD pattern obtained from the pellet using Cu K$\alpha$ radiation is shown in Fig. 13. From this diffractogram, one can identify the reaction products as $NiAl_3$ (shown as **N**) and $Ni_2Al_{18}O_{29}$ (shown as **O**) apart from the initial phases of Kovar (shown as **K**) and aluminium (shown as **A**). It can be inferred that $NiAl_3$

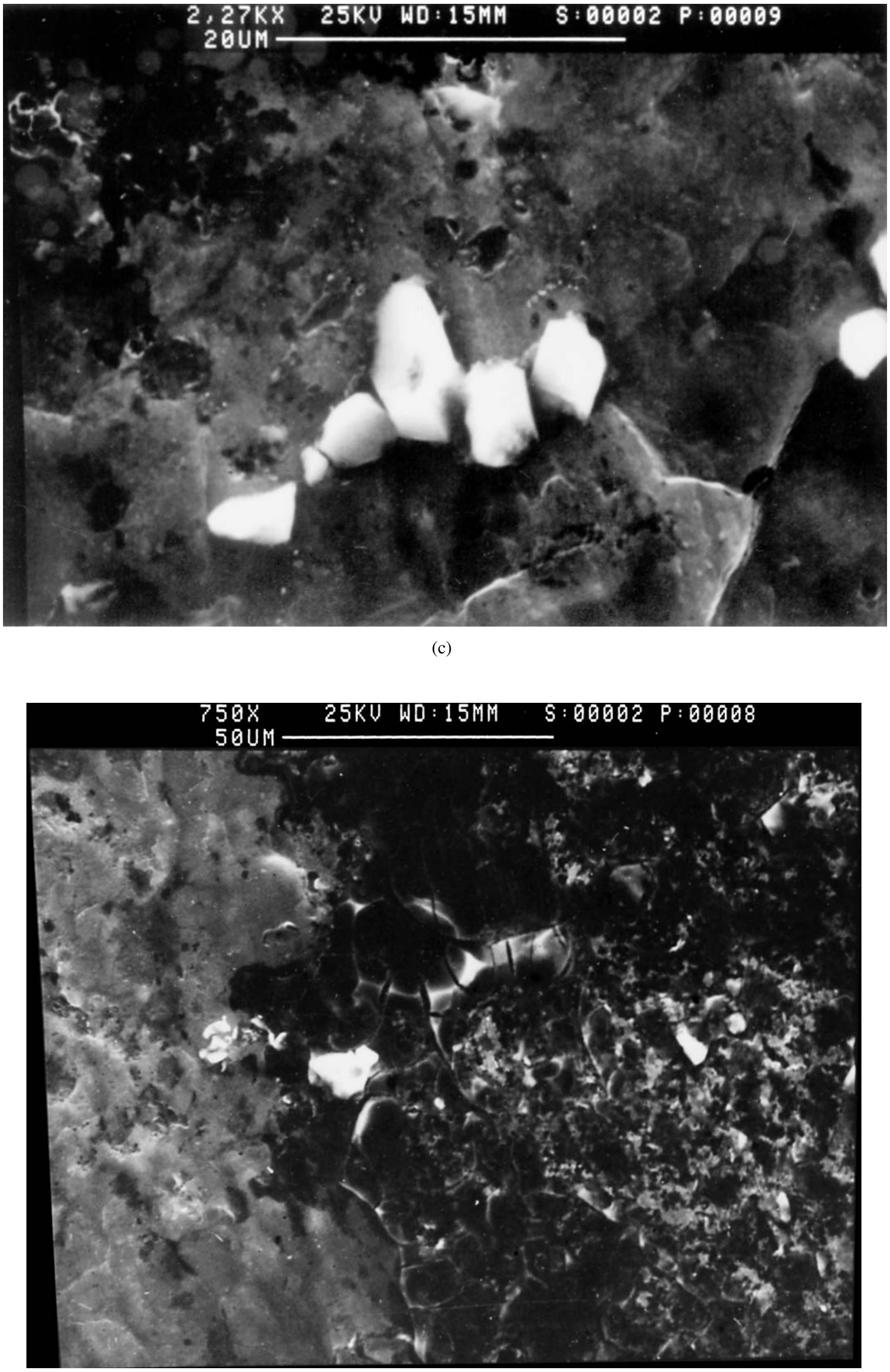


(a)



(b)

*Figure 9* SEM of debonded surfaces from the samples bonded in solid-state at 500°C under 34 MPa pressure for 45 min. (a) 90% alumina ceramic side: (b), (c) and (d) are aluminum side at different magnifications. (*Continued*)



(c)

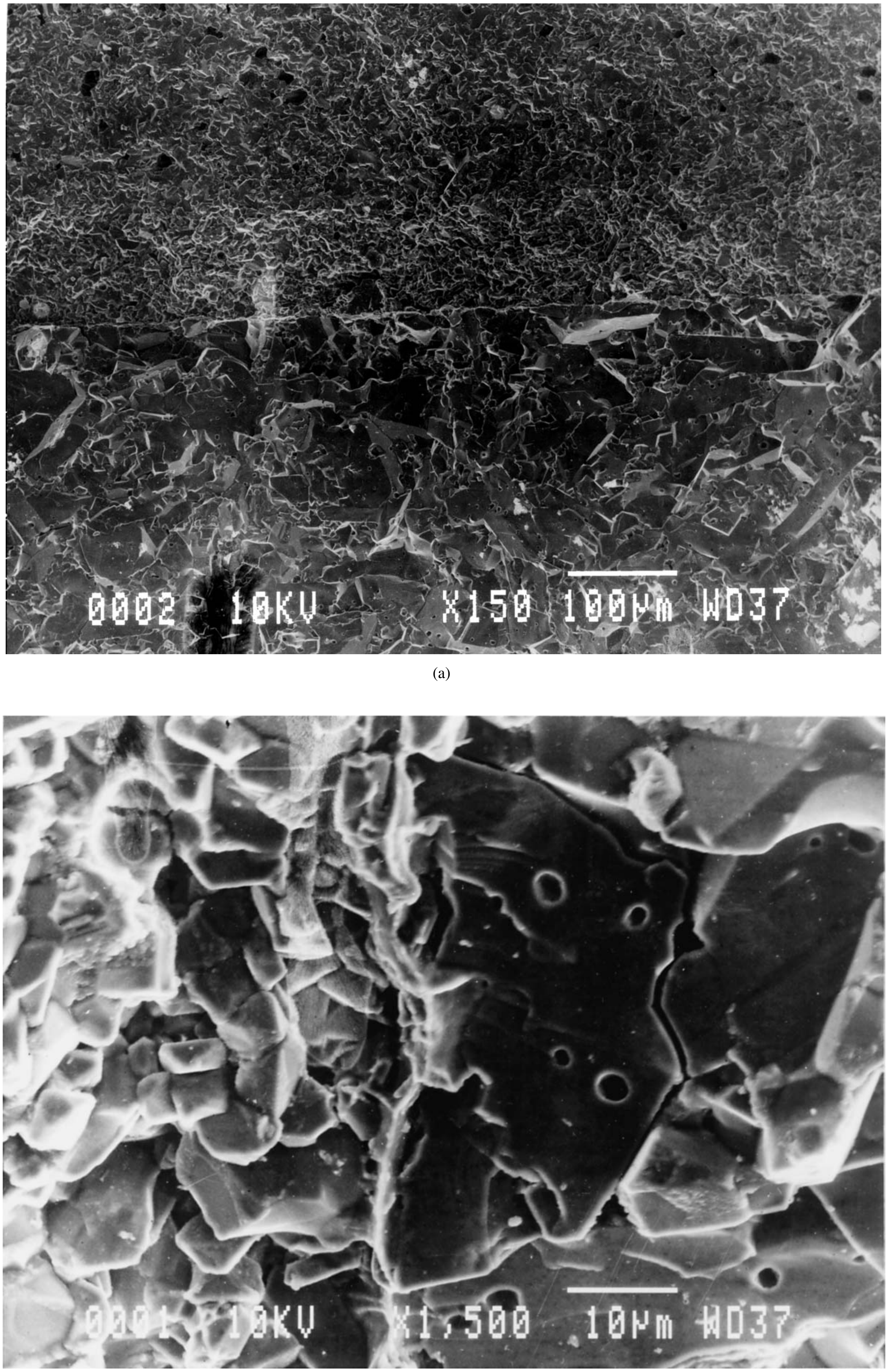
(d)

*Figure 9* (*Continued*).

and $Ni_2Al_{18}O_{29}$ are also forming during the solid-state bonding of Kovar/Al/$Al_2O_3$ seal at Kovar/Al interface.

## 4. Bonding mechanism by solid-state process

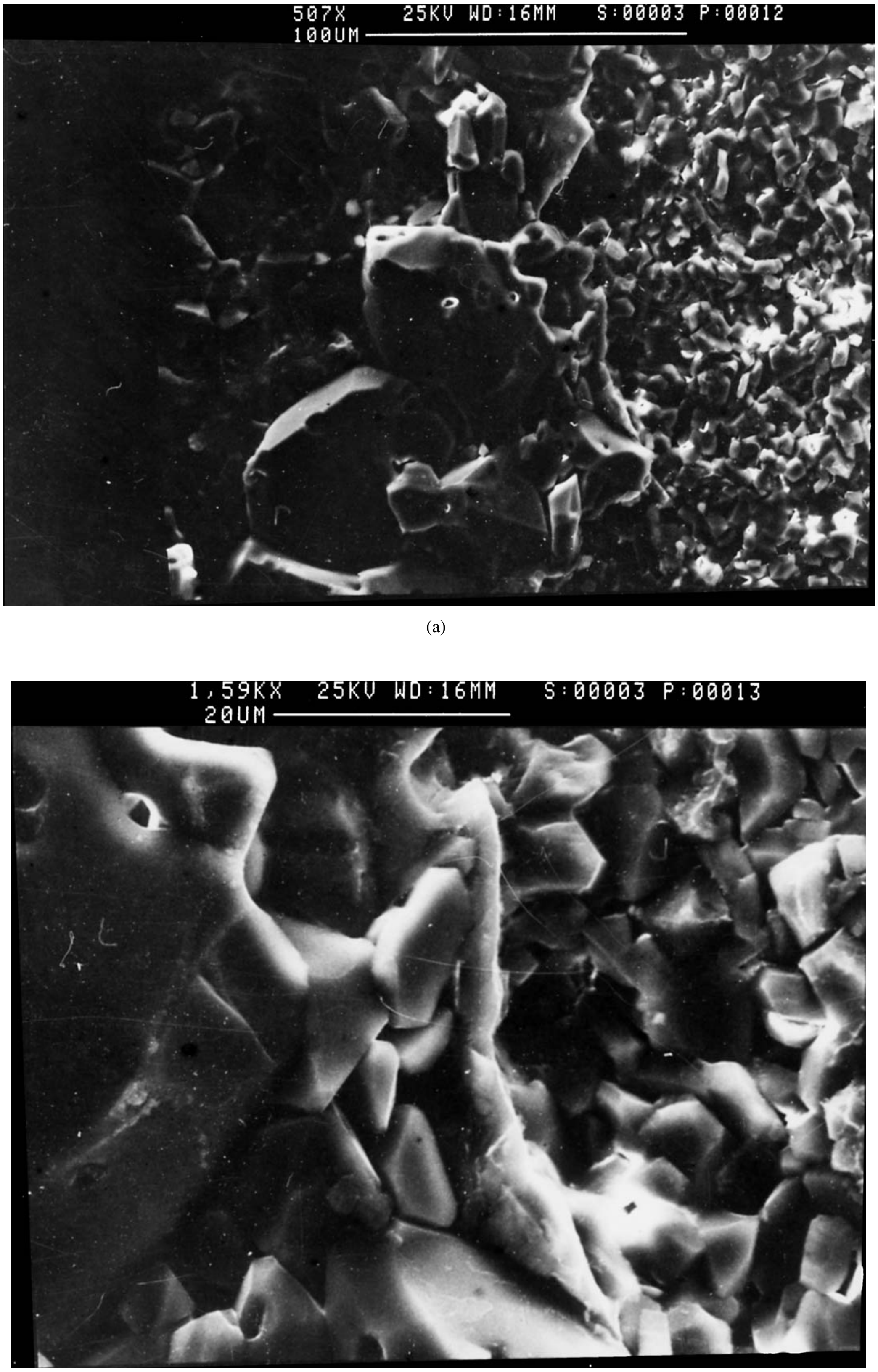
The bonding mechanism by thermo-compression technique is mostly due to the diffusion and plastic flow of metal in the ceramic asperities and consequent mechanical interlocking. In the present investigation, alumina ceramics have been bonded to Kovar using aluminium foil interlayer. The first step of bonding is the aluminium in contact with alumina ceramic and Kovar from opposite sides, at their asperities, come to an intimate interfacial conformity under applied



(a)

(b)

*Figure 10* (a) and (b) SEM of the transverse section of the joint formed between 90 and 99% alumina ceramics with aluminium interlayer in solid-state at 550°C under 34 MPa for 45 min at different magnifications.

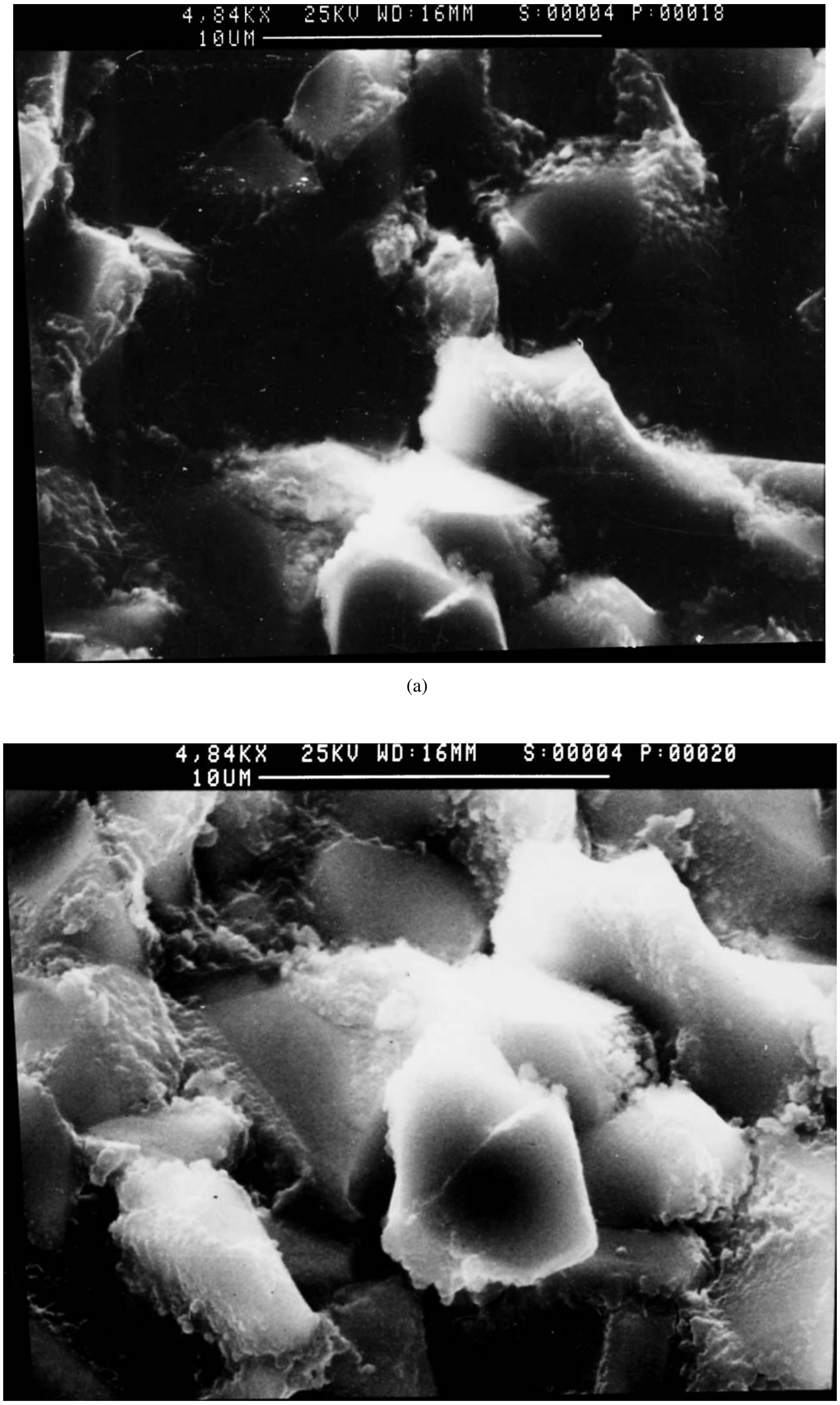


(a)

(b)

*Figure 11* (a) and (b) SEM at different magnifications of the debonded 90% alumina side with pulled out grains from 99% alumina substrate for the pair, transverse section of which is shown in Fig. 10.

pressure. Alumina, Kovar and aluminium have very different properties, the metal and alloy being much softer than the ceramic. Also at bonding temperature (≈600°C) the materials are respectively at 0.38, 0.41, 0.94 & of their melting points. Hence, it is obvious that during hot pressing, most of the deformation will occur in the aluminium. In the present investigation, fractographs of diffusion bonded joints have shown that aluminium flows into alumina grain boundaries. During the second step, interfacial diffusion occurs



(a)

(b)

*Figure 12* (a) and (b) SEM of different areas of the debonded 99% alumina side with pulled out grains from 90% alumina substrate for the pair, transverse section of which is shown in Fig. 10.

between Al foil and $Al_2O_3$ ceramics and reactions occur at the Al/Kovar interface resulting in the formation of $Ni_2Al_{18}O_{29}$ and $NiAl_3$. The apparent activation energy for the bonding process ($Q_B$) depends strongly on the type of alumina ceramics. The irradiation by photons of MeV energies has been found to affect the lattice parameters of $\alpha$-alumina due to creation of point defects. The irradiation of alumina ceramics (99%) prior to joining with Kovar accelerates the solid-state bonding by reducing ($Q_B$) from 209 to 76 kJ/mole. As the activation energy for self-diffusion of ions in alumina is comparable to the apparent activation energy of solid-state bonding and as the irradiation induced defect formation reduces it appreciably, it can be said that diffusion in alumina controls the solid-state bonding process.

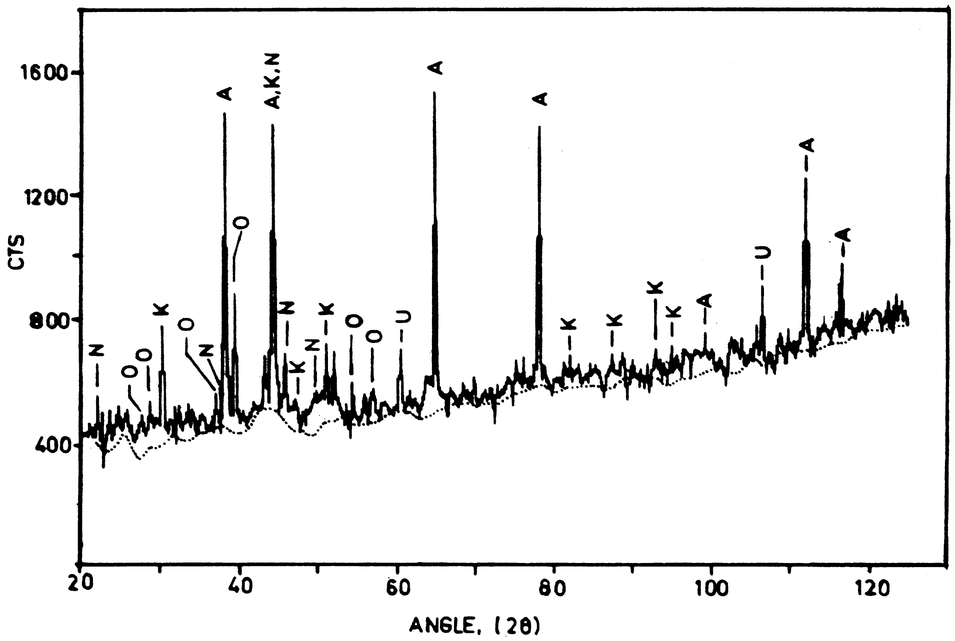


*Figure 13* XRD pattern (Cu $K_\alpha$) of thermo-compressed aluminium-Kovar mixture ($A$ = Aluminum; $K$ = Kovar; $N$ = $NiAl_3$; $O$ = $Ni_2Al_{13}O_{29}$, $U$ = Unknown).

## 5. Conclusions

Solid-state bonding between alumina and Kovar using aluminium interlayer has been successfully made by using thermo-compression technique. The main conclusions are as follows:

- To achieve high bond-strength by solid-state bonding under 75 MPa pressure for 10 min, the following temperatures should be used: (a) for 90% alumina −550°C (BS = 33.7 MPa): (b) for 96% alumina −515°C (BS = 41.2 MPa): and (c) for 99% alumina 575°C (BS = 63.7 MPa).
- The bond-strength has been found to increase with bonding time (10–90 min) and bonding pressure (7.5–75 MPa).
- During the bonding, interfacial diffusion occurs between Al foil and $Al_2O_3$ ceramics and reactions occur at the Al/Kovar interface resulting in the formation of $Ni_2Al_{18}O_{29}$ and $NiAl_3$.
- The irradiation of 99% $Al_2O_3$ ceramics by 5 MV X-rays has been found to increase the bond-strength sharply for bonding temperatures ≈540°C. The bond strength can be increased from 33 to 60 MPa by radiation with a dose of 15 k Rads.
- The apparent activation energy for the bonding process ($Q_B$) depends strongly on the type of alumina ceramics. The irradiation by 5 MV Bremsstrahlung X-rays has been found to affect the lattice parameters of $\alpha$-alumina due to creation of point defects. Thus irradiated alumina ceramics (99%), prior to joining with Kovar, accelerates the solid-state bonding by reducing ($Q_B$) from 209 to 76 kJ/mole.
- As the activation energy for self-diffusion of ions in alumina is comparable to the apparent activation energy of solid-state bonding and as the irradiation induced defect formation reduces it appreciably, it can be said that diffusion in alumina controls the solid-state bonding process.
- From SEM of the interface and the debonded surfaces it has been found that the flow of metal into ceramic grain boundaries contributes to the bond strength in addition to diffusion bonding.

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