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Author(s)	Ohmori, Akira; Arata, Yoshiaki; Piech, Stan et al.
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Al₂O₃ Diffusion Bonding Using TiH₂ Powder Interlayer†

Akira OHMORI*, Yoshiaki ARATA**, Stan PIECH*** and Wladyslaw WLOSINSKI****

Abstract

The possibility of intensification of Al₂O₃-Ti diffusion bonding process through application of Ti and TiH₂ powder interlayers was investigated. The bonds obtained by direct diffusion bonding and with powder interlayers were examined by SEM-EDX and XRD methods. The strength of joints was determined by 4-point bending test. The beneficial effect of application of powder interlayer can be connected with three factors: increase of a real contact area between metal and ceramic, change of diffusion coefficients and compensation of some amount of residual stresses.

KEY WORDS: (Diffusion Bonding) (Sinterwelding) (Al₂O₃) (Ti) (Ti Powder) (TiH₂ Powder)

1. Introduction

The ceramic to metal diffusion bonding can be considered in two categories. In the first case metal serves only as an interlayer enabling to join, for instance, two ceramic parts or ceramic to different than interlayer metal. The pressure exerted during bonding causes plastic deformation and creep of metal what ensures a high area of real contact and considerably increases the bond strength. Here one of the most popularly used metals is aluminium which is rather unsuitable as a bonding agent for components exposed to high temperatures or caustic environments. In the second case both ceramic and metal serve as constructional parts and then exerted pressure should be reduced just enough to minimize the deformation of a metal part. This usually results in decreasing of bond strength.

One of the methods which allows to reduce temperature and pressure during diffusion bonding of metals is so called "sinterwelding". In this method a powder interlayer is inserted between components to be joined. The essential part of the process is to obtain a good sintering and bonding of the powder mass between and with components. This method was reported to be successful in some cases [1-3].

The formation of a thermodynamically stable interface is the most important requirement during ceramic to metal bonding. The second factor which greatly influences the strength of the bond is the presence of thermally induced residual stresses [4, 5]. They result predominantly from differences in the values of thermal coefficients of expansion. During heating in high temperatures

relaxation of stresses occurs mainly due to metal creep. During cooling, however, when creep and plastic deformation are not possible stresses arise which grow as the cooling continues. The second type of thermal stresses is connected with unequal distribution of temperature in materials. The value of these stresses depends mainly on a cooling rate, which is connected not only with the type of bonded materials but also with their dimensions and shapes.

The aim of this investigation was to examine, basing on joining of Al₂O₃ ceramic with titanium, whether the application of a powder interlayer may be beneficial for the process of ceramic to metal diffusion bonding. Two main preliminary conditions were taken: the pressure exerted during bonding cannot cause macroscopic deformation of metal and the sintered interlayer is composed from the same metal as the part to be joined, in this case — from titanium.

There are two possibilities to obtain such an interlayer. The first — by straight insertion of Ti powder and the second — by application of TiH₂ powder which decomposes during process into titanium and hydrogen. The dehydriding process is accompanied by structural changes (Fig. 1 [6]). During this conversions the Ti atoms are in the state of increased mobility which should contribute to enhanced diffusion and thereby to creation of a sound bond.

2. Materials and Experimental Procedure

Materials used in this investigations were commercially pure Ti (alpha to beta transformation temperature at

† Received on May 8, 1989

* Associate Professor

** Emeritus Professor

*** Foreign Research Fellow (Warsaw Technical University)

**** Visiting Professor (Warsaw Technical University)

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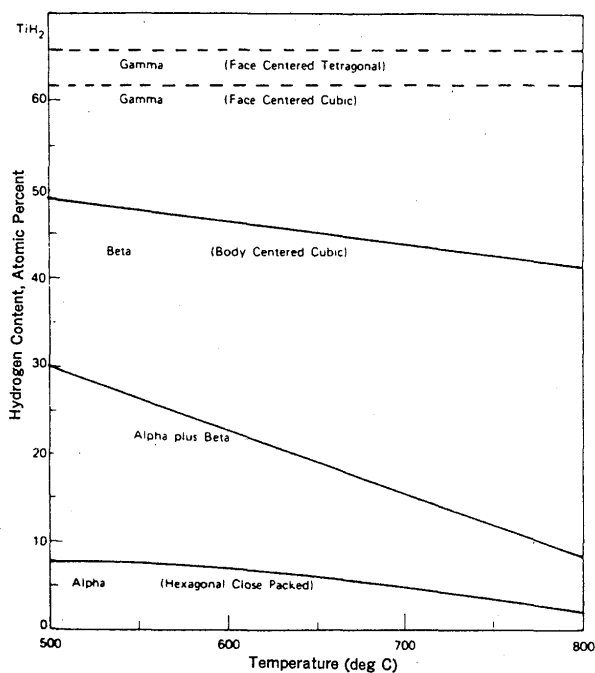


Fig. 1 Dependence of TiH_2 structure on temperature [6].

Table 1 Chemical composition of Ti

C	Fe	N	O	Ti
0.020	0.04	0.0055	0.08	Bal.

1155K) and 99.6% Al_2O_3 ceramic. The chemical composition of Ti is shown in Table 1. Ti and TiH_2 powders used in the experiments were of 325 mesh.

Mixtures of Al_2O_3 -50%wtTi and Al_2O_3 -50%wt TiH_2 heated separately in a vacuum of 2×10^{-3} Pa, temperature of 1123K and 1173K for one hour, were examined by X-ray diffraction method to obtain informations about possible reactions between Al_2O_3 and Ti in similar to bonding conditions.

The process of diffusion bonding was carried out using three types of specimens – A, B and C (Fig. 2).

Type A served for structural examinations. It enabled to produce simultaneously two types of joints: by direct bonding of Al_2O_3 to Ti and with the application of a powder interlayer. It gave the possibility to compare the structure of resultant bonds obtained in exactly the same conditions.

Specimens of type B (both joints obtained by direct diffusion bonding) and of type C (both joints with powder interlayers) were used for mechanical testing.

Surfaces of ceramic components were ground with emery paper and polished with $1\mu\text{m}$ diamond paste. Both surfaces of titanium discs were polished with up to 1500 grade emery paper and $0.05\mu\text{m}$ Al_2O_3 slurry. After polishing the specimens were degreased in acetone with an ultrasonic bath. The mixture of Ti (TiH_2) powder with binding agent was put on the surfaces of both ceramic and metal components to be joined through the interlayer.

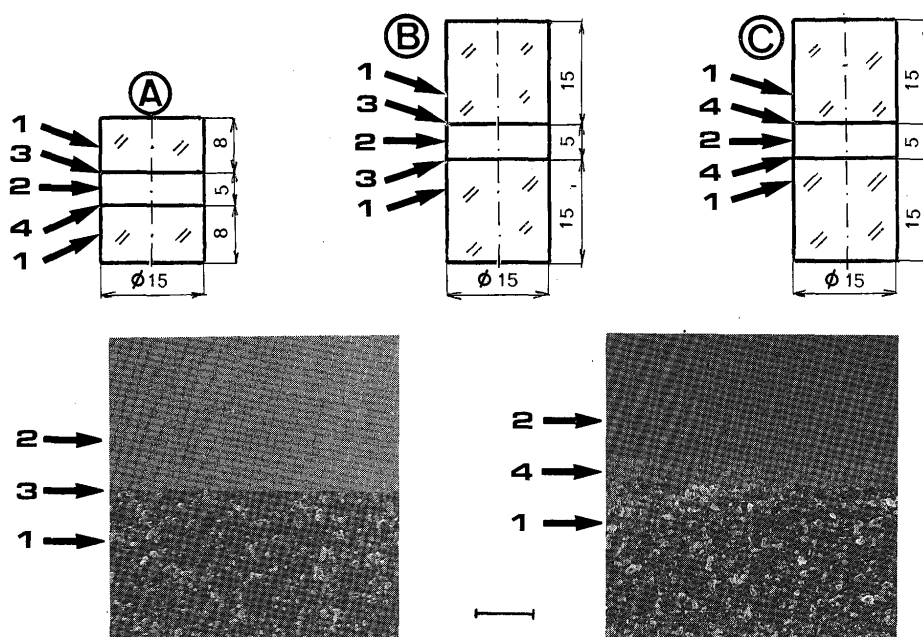


Fig. 2 Types of specimens used in experiments and structure of bonds

- 1 – Al_2O_3 ceramic
- 2 – Ti
- 3 – direct Al_2O_3 bond
- 4 – sintered Interlayer

After drying the whole assembly was set together. The bonding was carried out in an apparatus already presented in [7, 8].

The main parameters of the process (temperature, force, vacuum) were measured, respectively, by K type thermocouple, load cell and ionization type vacuum meter. All the measured values were recorded with three channel X-Y type recorder. After putting the assembly into the bonding apparatus the chamber was evacuated up to 7×10^{-3} Pa. Then the assembly was heated to the required temperature with a stabilized rate of 50 K/min. (Fig. 3). When type A or C specimens were heated the binding agent was evaporating at about 200°C (973 K). Then (for TiH₂) the decomposition of the powder was starting at about 400°C (673 K). When the bonding temperature was obtained and the vacuum was again in the range of 7×10^{-3} Pa the external pressure was exerted. The value of the applied force was chosen experimentally in order to not deform the titanium discs. For temperatures 1173 K (Ti of Body Centered Cubic structure) and 1123 K (Ti of Hexagonal Close Packed structure) the forces were 785 N and 1225 N respectively. The bonding time, equal to the time of force exertion, was changing in range from 600s

to 3600s. The cooling rate of specimens was stabilized to the value of 10 K/min.

Generally type A specimens were used for structural observations with SEM and microanalysis with EDX. To reveal structure of the sintered layer the specimens were etched for 10s with reagent of composition: 2% HCl, 2% NH₄OH, 1% HF, 95% H₂O. 4-point bending test was chosen to evaluate the strength of joints. The rectangular testpieces of height $h = 3$ mm, width $w = 10$ mm and length $l = 35$ mm were cut off from the bonded specimens of type B and C. The test was carried out in a device shown in Fig. 4 using Instron type machine. The load P was applied at 0.5 mm/min. rate. The bend strength was determined by equation

$$\delta = 1.5P \frac{b-a}{wh^2}$$

where: P — load at which specimen fractured,
 b — distance between the lower rolls,
 a — distance between the upper rolls,
 w, h — width and height of a specimen, respectively.

This kind of test assured that the whole joint, including ceramic parts, metal, interlayer and interfaces was loaded with an equal moment. Fractured surfaces were examined by SEM-EDX method and X-ray diffraction to reveal the character of bond.

To check if the sintered interlayer may compensate same amount of residual stresses, two specimens of type A and two specimens of type B were spontaneously cooled from the bonding temperature to room temperature by switching off the heating during bonding process. Line V3 in Fig. 3 presents the recorded cooling rate. Also one specimen, similar to type A but with dimensions of specimens B and C, was treated in this way. The dimensions were change to preserve the same cooling conditions as for specimens B and C. After longitudinal cutting the

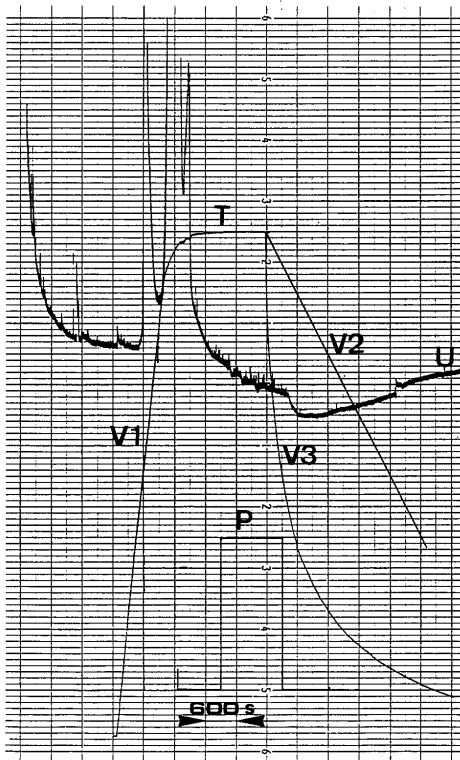


Fig. 3 The course of process parameters during diffusion bonding using TiH₂ powder interlayer
 U — Vacuum
 V1 — heating slope
 V2 — cooling slope (drawn in)
 V3 — spontaneous cooling slope
 T — bonding temperature
 P — force

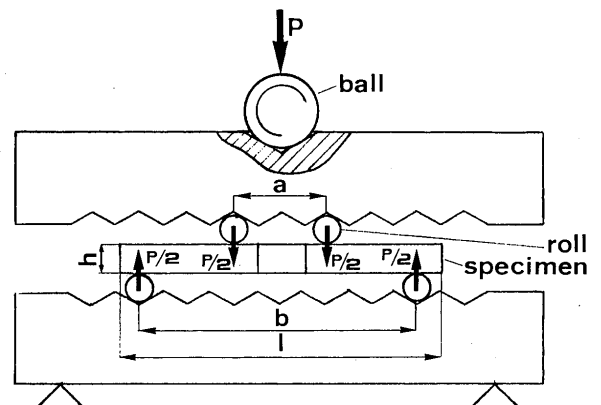


Fig. 4 Device for 4-point bending test

revealed surfaces were observed to check the presence of cracks.

3. Results and Discussion

X-ray diffraction patterns obtained from heated in vacuum Al_2O_3 -50wtTi and Al_2O_3 -50wtTiH₂ powder mixtures revealed the formation of AlTi_2 compound. Comparing the results for temperatures of 1123K and 1173K no difference was observed for one type of mixture. Although the intensity of the AlTi_2 signals is quite weak there was a slight difference to TiH₂ powder advantage at both temperatures. Figure 5 shows the XRD patterns obtained for powder mixtures heated in 1173K.

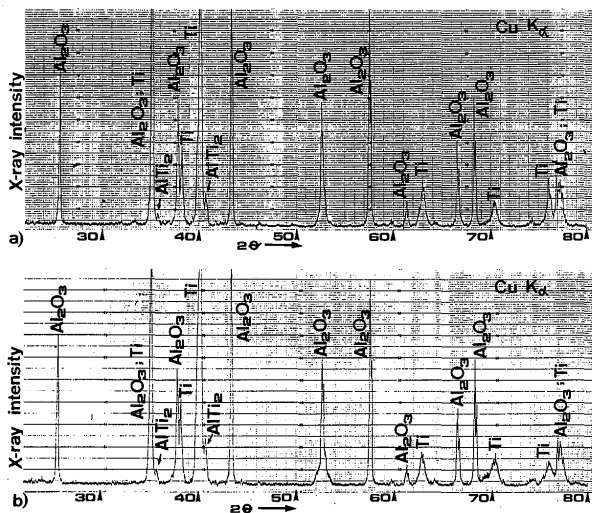


Fig. 5 XRD pattern of powders mixtures heated in vacuum (2×10^{-3} Pa) in 1173K for one hour;
a) Al_2O_3 - 50wtTi b) Al_2O_3 -50wt%TiH₂

Diffusion bonding using Ti powder interlayer was carried out only for specimens of type A. Figure 6 shows the microstructure of bond obtained at 1173K with bonding time of 2400s. A crumbled layer between sintered Ti powder and ceramic is clearly visible. Such kind of layer existed also at bonds obtained in other conditions and it was regarded as an unfavourable structural change. Figure 7 shows line analysis of this bond and corresponding to it direct diffusion bond. Also point analysis of crumbled layer is included.

Such type of layer was not observed when TiH₂ powder had been used. Figure 8 shows microphotographs from joint obtained in the same conditions, that means 1173K and 2400s. The line analysis of this bond and corresponding to it direct diffusion bond is presented in Fig. 9. The visible shift of lines into ceramic was registered also for other bonding conditions. It was associated with the fact that diffusion coefficient of powder X to plate Y is higher than from plate X to plate Y. And diffusion coefficient from plate Y to powder X is smaller than from plate Y to plate X. This results among others from higher surface energy of powdered material and activating effect of exerted pressure. In the early phase of the bonding process arise initial contact areas. In the beginning they are considerably small since powder grains are in touch with a solid, flat surface. The contact areas are significantly smaller than in case of two flat, polished surfaces. When the pressure is exerted very high forces start to work at the contact areas between powder grains and ceramic. The grains are quickly deformed and the contact area grows. The free space between powder grains and the surface of ceramic is intensively filling up due to plastic deformation and creep. This results in high area of

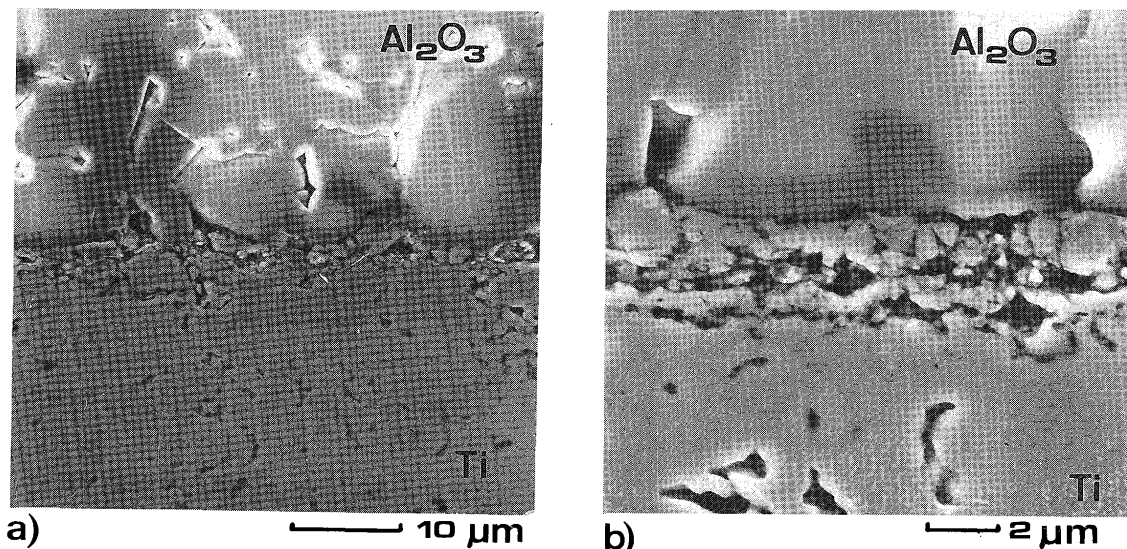


Fig. 6 Microstructures of bond between sintered from Ti powder layer and Al_2O_3 ceramic (1173K, 2400 s)

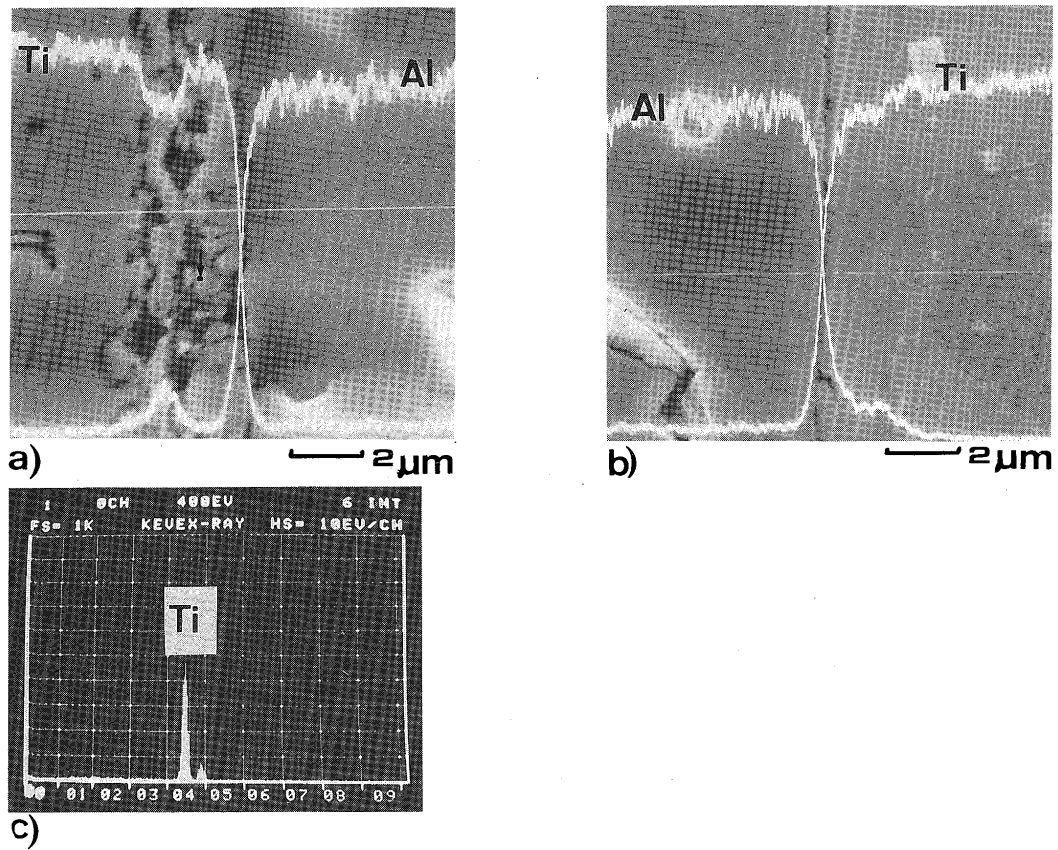


Fig. 7 a) Line analysis of bond from Fig. 6; arrow indicates place of point analysis (c)
b) Line analysis of corresponding direct bond

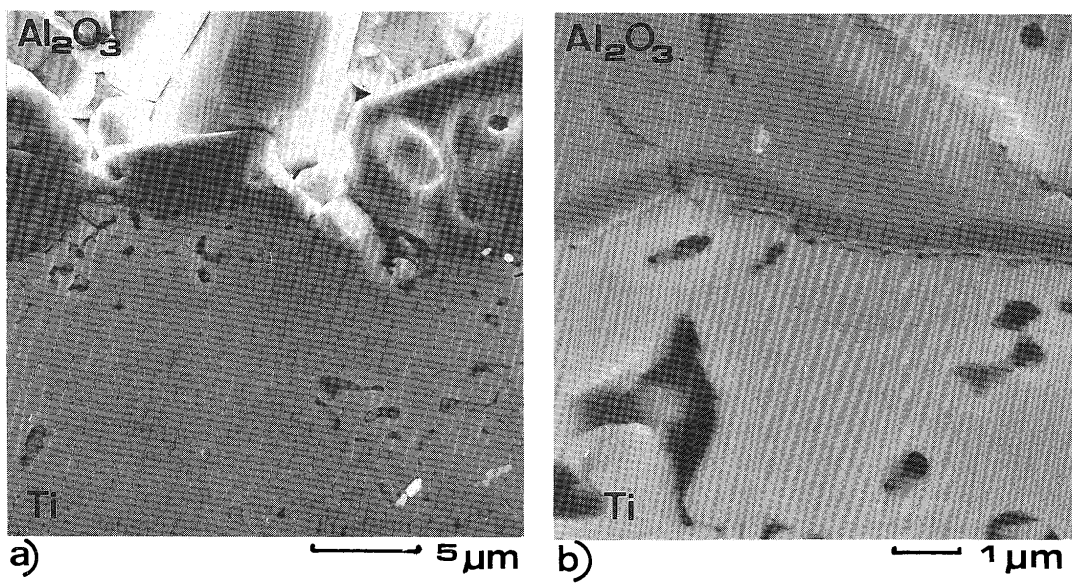


Fig. 8 Microstructures of bond between sintered from TiH_2 powder layer and Al_2O_3 ceramic (1173 K, 2400 s)

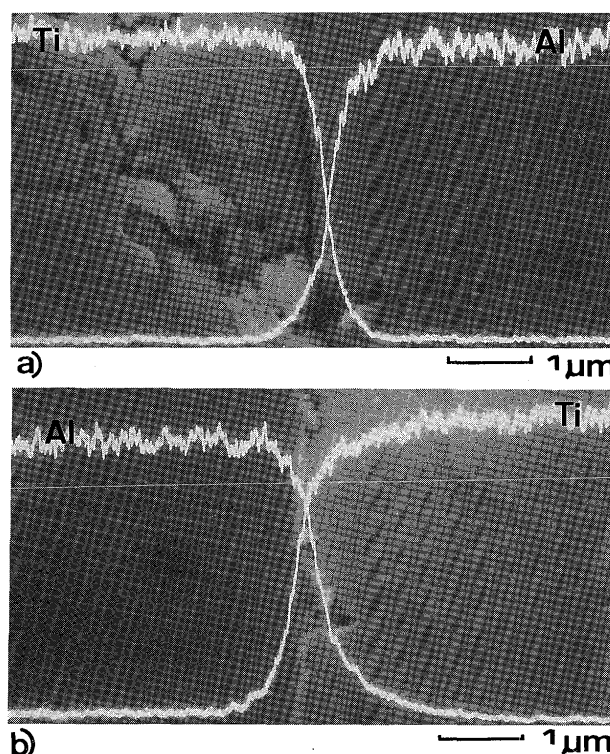


Fig. 9 a) Line analysis of bond between sintered from TiH_2 layer and Al_2O_3 (1173 K, 2400 s)
b) Line analysis of corresponding direct bond

real contact. **Figure 10** shows some examples of microstructures of bonds obtained by direct diffusion joining and bonds obtained with the application of TiH_2 powder interlayer. Although bonds were produced respectively in the same conditions it is clearly visible that the powder interlayer has the ability to fill up the irregularities on the ceramic surface. If the solid metal had to behave in such a manner, the pressure exerted during bonding should be increased significantly.

Figure 11 shows the results of 4-point bending test. Application of a powder interlayer by the given parameters of process (mainly not high pressure) increases bending strength from 105 MPa to 126 MPa but the strength of joint is significantly weaker than the strength of ceramic itself. The fracture occurred through ceramic and interface. The shift of maximum strength towards shorter bonding times results from much more active behaviour of Ti powder from TiH_2 decomposition than the behaviour of Ti in a solid state form. The increase in strength may be associated with the increase of the area of real contact and with the compensation of some amount of residual stresses.

The second assumption was backed up by the results obtained from specimens which had been spontaneously cooled down from the bonding to the room temperature in the vacuum chamber. Large cracks occurred in directly bonded specimens. Such cracks were not visible in speci-

mens bonded with the sintered interlayer. Also in the specimen of type A, but with dimensions of type B or C, the visible cracks occurred only at the direct bond. **Figures 12a** and **12b** show the macrophotographs of directly bonded specimens spontaneously cooled down from temperature of 1173 K. **Figure 12c** presents the specimen in which two types of bonds were used. Arrows indicate visible cracks.

Figure 13 shows fractured surfaces of a broken specimen. The fracture occurred through ceramic and interface. Some single grains of ceramic were torn out. **Figure 14** shows the results of point analysis of the fractured surface. It was also examined by X-ray diffraction method (using cobalt radiation). The XRD patterns revealed clearly the presence of AlTi_2 compound (**Fig. 15**).

4. Conclusions

Application of TiH_2 powder interlayer, sintered simultaneously during process of Al_2O_3 -Ti diffusion bonding, increased strength of obtained joints in comparison with joints obtained in the same conditions by direct diffusion bonding. This results mainly from three factors:

- increase of the area of real contact between metal and ceramic components,
- change of the value of diffusion coefficients,
- compensation of some amount of residual stresses.

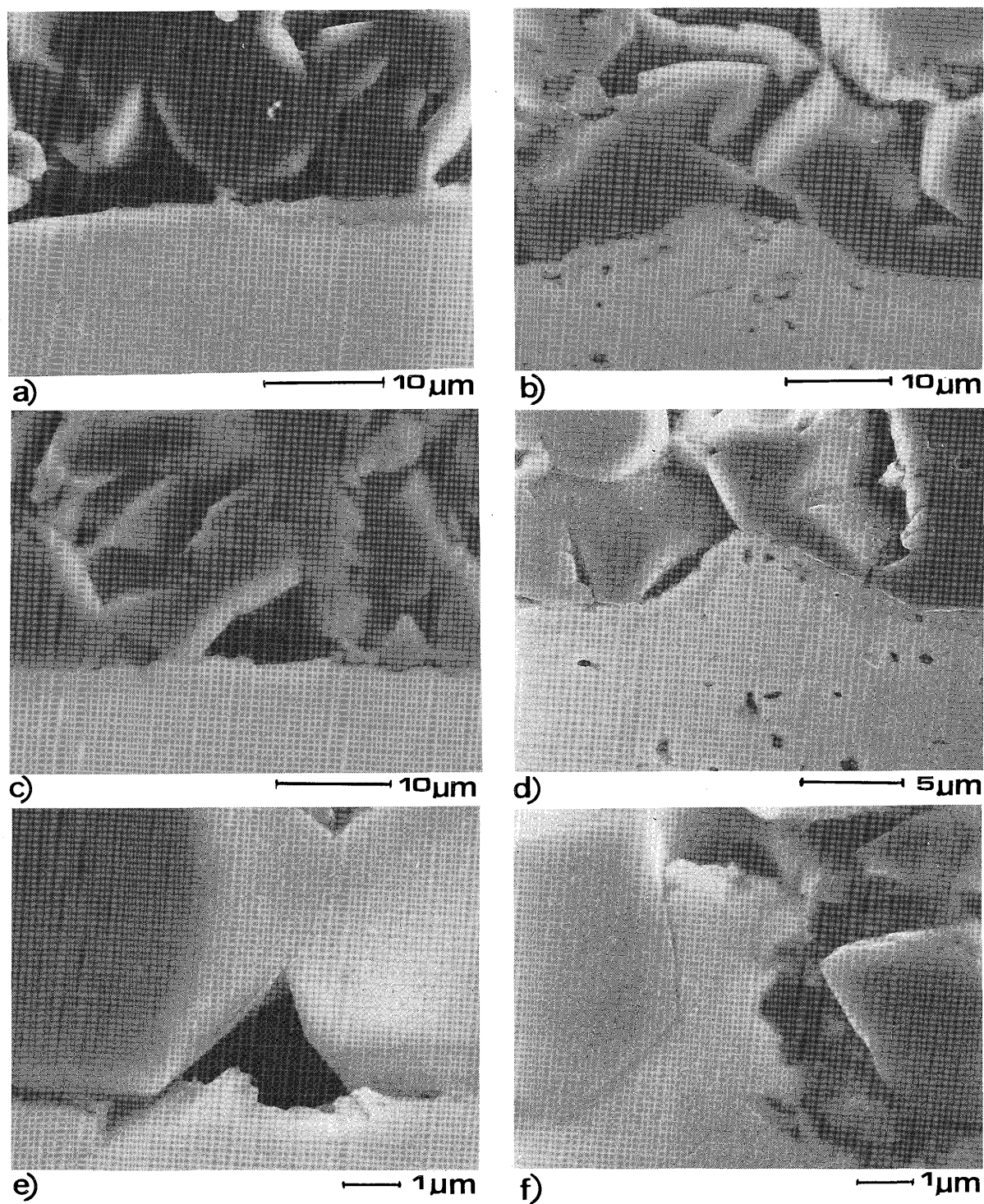


Fig. 10 Microstructures of direct Al_2O_3 -Ti bonds and corresponding bonds with sintered interlayer;
a, b – 1123 K, 1800 s; c, d – 1123 K, 1800 s; e, f – 1173 K, 2400 s

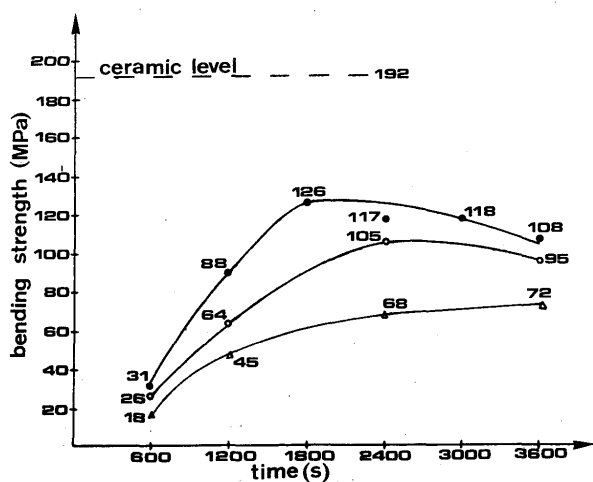
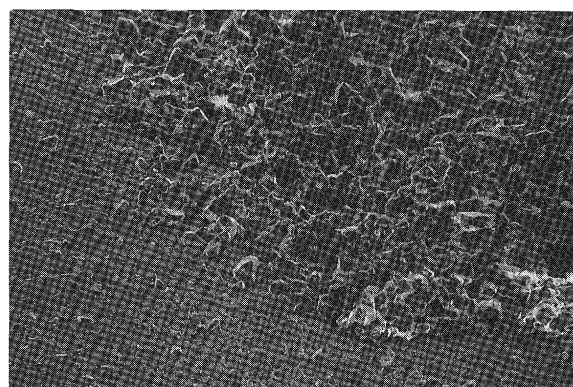


Fig. 11 Bending strength of joints in bonding time function
 Δ – direct diffusion bonding (1123 K, 1225 N)
 \circ – direct diffusion bonding (1173 K, 785 N)
 \bullet – diffusion bonding through TiH_2 interlayer (1173 K, 785 N)

This method seems to be useful also for different material combinations. To avoid difficulties with preparation of interlayers in presented above form it is advisable to use them in form of previously cold pressed powders or to cover the surfaces to be bonded with powder layers by for example plasma spraying method.

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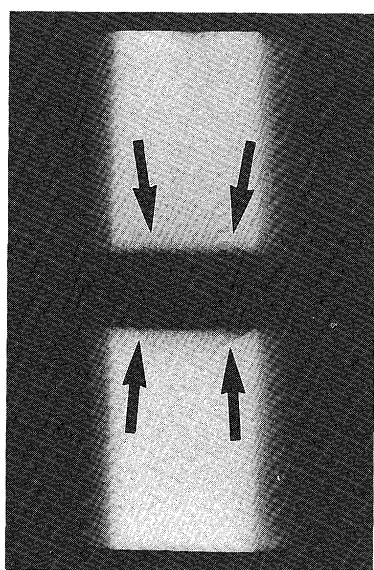


a)

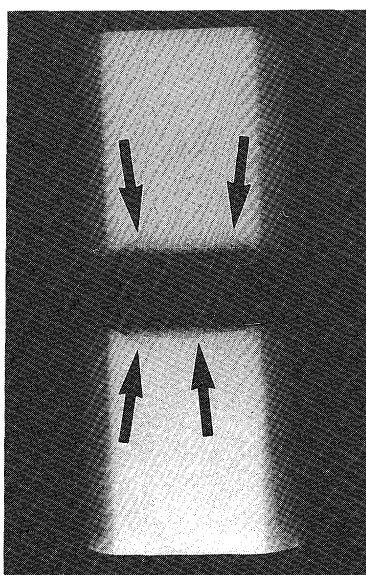


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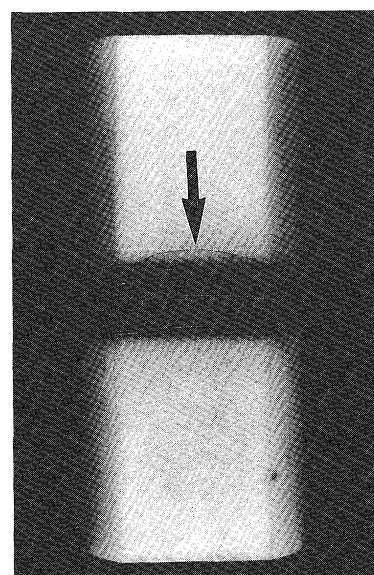
Fig. 13 Fractured surface of broken specimen bonded through TiH_2 interlayer (1173 K, 1800 s)



a)



b)



c)

Fig. 12 Macrophotographs of longitudinal sections of specimens spontaneously cooled down from 1173 K to room temperature; arrows indicate cracks
a, b) both bonds of direct type
c) upper bond of direct type, lower bond – with sintered interlayer

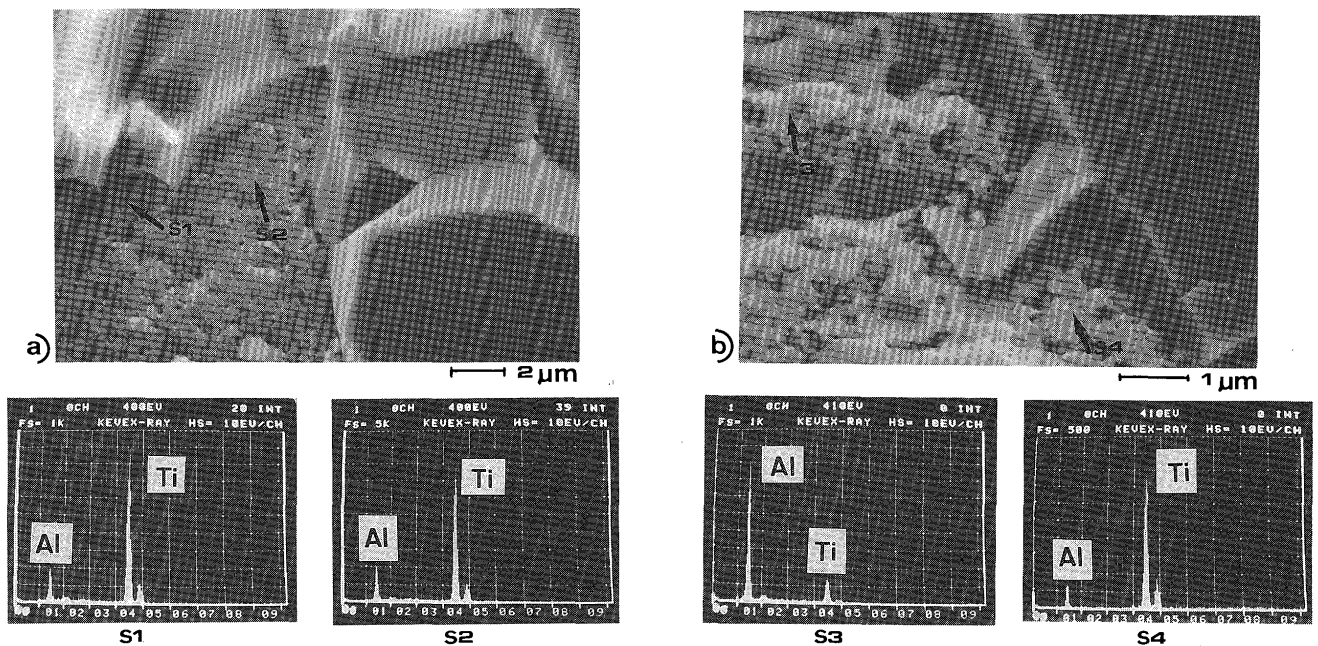


Fig. 14 Microstructure of a fractured surface (TiH₂ layer) and point analysis:
a) 1173 K, 1800 s, b) 1173 K, 2400 s

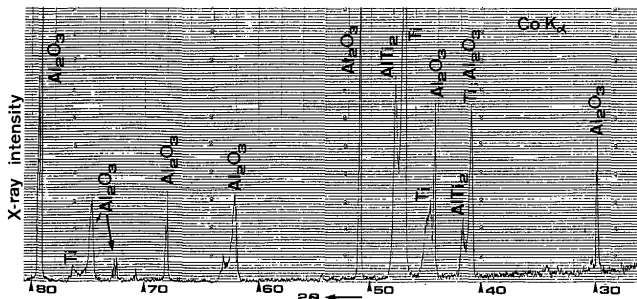


Fig. 15 XRD pattern of a fractured surface (TiH₂ layer, 1173 K, 1800 s)

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