Studies on Solid State Reaction Bonding of Metal and Ceramic (Report I)†

– Bonding of Al-Mg alloy to ZrO_2 ceramic –

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Abstract

The solid state reaction bonding of Al-Mg alloy (A5052) to ZrO_2 stabilized with CaO is studied. The reaction bonding was done by heating in vacuum (5×10^{-5} mm Hg) under a pressure ($0.3 - 70$ Kg/cm^2). The bonding is impossible at 500°C under no pressure even within 2 hr. At a pressure of 0.3 Kg/cm^2, the ZrO_2/A5052 joint bonded at 500°C for 2 hr showed a tensile strength of 6 Kg/mm^2. The strength of bonded joints between ZrO_2 and A5052 increased respectively with the increase of the pressure, bonding temperature and bonding time. The bonding was greatly influenced mutually by these bonding conditions.

Moreover, it is shown that the bonding process has three stages as follows;

(1) *Initial stage of breaking of oxide film on alloy surface and the filling of Al-Mg Alloy into pores in ZrO_2.*
(2) *Second stage of reaction of Mg, Al and SiO_2 in grain boundary part and the bond at grain boundary part.*
(3) *Third stage of reaction of Mg, Al and ZrO_2 grain and the bond at the grain.*

Also, the bonding of Al (A1050) to ZrO_2 and the bonding of A5052 to other oxide ceramics are done.

KEY WORDS: (Solid State) (Reaction Bonding) (Zirconia) (Al-Mg alloy) (Aluminum)

1. Introduction

With the development of industries today, the environment is getting stricter and severer for using metallic materials.

As a consequence, the applications for ceramics (Alumina, Zirconia, etc.), which are highly heat resistance, abrasive wear resistance and hot oxidation resistant and gas corrosion resistant, have remarkably expanded. However, due to the poor machinability of ceramics per se, the shortcomings of ceramic material (weakness against heat shock, etc.), the development of composite materials, using more than two materials as a composite, has become an urgent matter.

Under such circumstances, attention has been paid to bonding of ceramics to metallic materials. However, the bonding is different from bonding of metals themselves and is difficult. So, various methods are new devised[1]. The brazing, in which a liquid metal or glass filler wets the interface between metal and ceramic, is usually used. However, titanium-alloy fillers are used for forming bonds, as a conventional brazing alloy does not wet ceramics. In some cases, metallizing techniques[2),3)] for ceramic surface have been developed prior to a conventional brazing. The solid state reaction bonding occurs when certain metals and ceramic materials are held in intimate contact and heated. With base metals such as Ni, Fe, the reaction of macro-scopic spinel-type bond is formed between the metal oxide ceramic. Noble metals also undergo a similar bonding reaction, but the nature of the bond mechanism is not understood[4)].

In this paper, the solid state reaction bonding of Al-Mg alloy (A5052) to ZrO_2 stabilized with CaO is described. The reaction bonding was done by heating in vacuum (5×10^{-5} Torr) under a pressure ($0.3 \sim 70$ kg/cm^2).

2. Specimens and Experimental Procedure

Chemical compositions of oxide ceramics used are shown in **Table 1**. As Al-Mg alloy, A1050 and A5052 were used. The chemical compositions are shown in **Table 2**. **Fig. 1** shows schematic diagram of bonding apparatus and the setting method for bonding. The bonding specimen surfaces were polished with 1500 grade emery paper and degreased with acetone before bonding. The heating rate used for bonding was 50°C/min.

The reaction bonding was done by heating in vacuum (5×10^{-5} Torr) under a pressure ($0.3 \sim 70$ Kg/cm^2). The

Table 1 Chemical compositions of ceramics used

Materials	Chemical compositions (wt%)							
	SiO_2	TiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	Y_2O_3	ZrO_2
ZrO_2(CaO)	1.39	0.09	0.72	0.15	5.40	1.16	—	91.08
ZrO_2(Y_2O_3)	0.08	0.11	0.41	0.07	tr	tr	13.37	85.96
Al_2O_3	0.09	—	99.62	0.02	0.11	0.10	—	—
MgO	1.71	—	0.03	0.05	0.83	96.85	—	—

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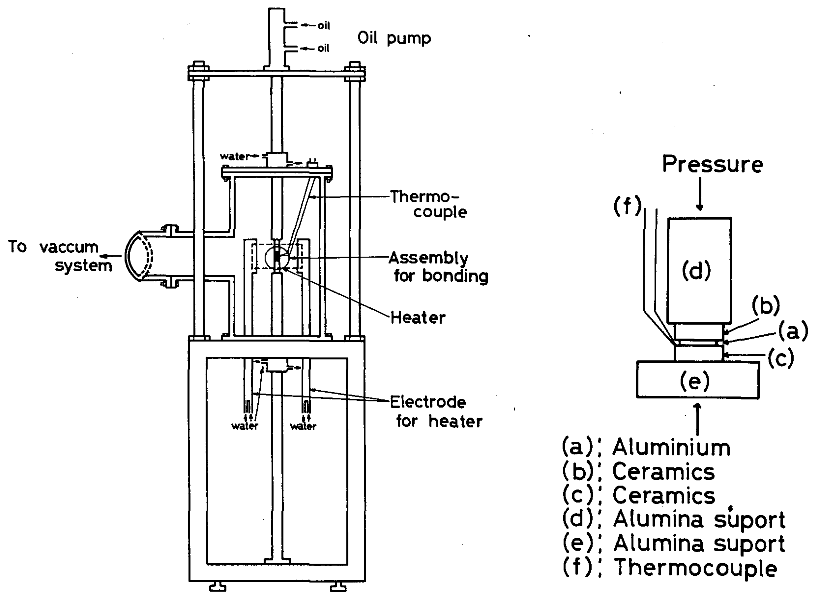

Fig. 1 Schematic diagram of bonding apparatus and setting method for bonding

Table 2 Chemical compositions of Al alloys used

Materials	Chemical compositions (wt%)								
	Cu	Si	Fe	Mn	Mg	Zn	Cr	Ti	Al
A1050	0.05	0.25	0.40	0.05	0.05	0.05	-	0.03	Bal.
A5052	0.01	1.0		0.10	2.2 -2.8	0.01	0.15 -0.35	-	Bal.

specimen size of ceramics was 13 mm diameter and 5 mm thickness, and the Al-Mg alloy size was 10 mm diameter and 0.5 mm thickness. The evaluation of mechanical strength of bonding joints was done by measuring tensile strength.

3. Results and Discussion

3.1 Bonding of A5052 and ZrO_2 ceramic

In order to clarify the bonding character of A5052 containing Mg and ZrO_2 stabilized with CaO, the solid state reaction bonding of A5052 and ZrO_2 was done at 500°C to know the influence of bonding pressure and time on the bonding. The results are shown in **Fig. 2.** O mark in this figure, shows the strength of above 0.2 Kg/mm^2 for bonding joints between A5052 and ZrO_2 and x mark shows the strength of below 0.2 Kg/mm^2. From this figure, the bonding is difficult at 500°C under no pressure even within 2 hr. At the bonding of short time for 3 min, the bonding is possible under 60 Kg/cm^2 pressure. The bonding for 10 min is possible even under 10 Kg/cm^2 pressure. **Figure 3** shows in detail the change in bonding strength with bonding pressure. As the bonding time is 10 min, the bonding strength increased with the increase of the pressure. However, in the case of 20 min, the bonding strength showed a constant value of about 6 Kg/mm^2 under above 30 Kg/cm^2 pressure. **Figure 4** shows the bonding temperature dependence of the bonding strength. From this figure the strength increased with the temper-

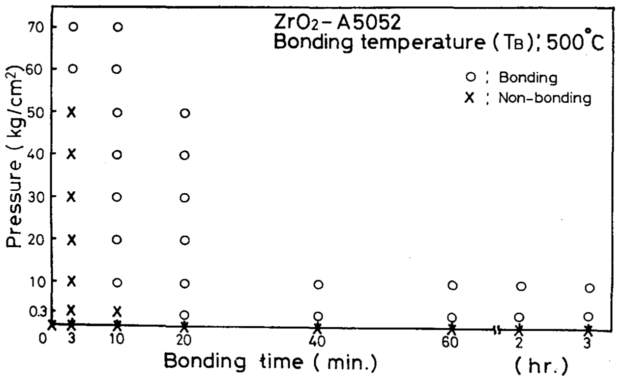

Fig. 2 Effect of bonding pressure and time on bonding of A5052 and ZrO_2 at 500°C

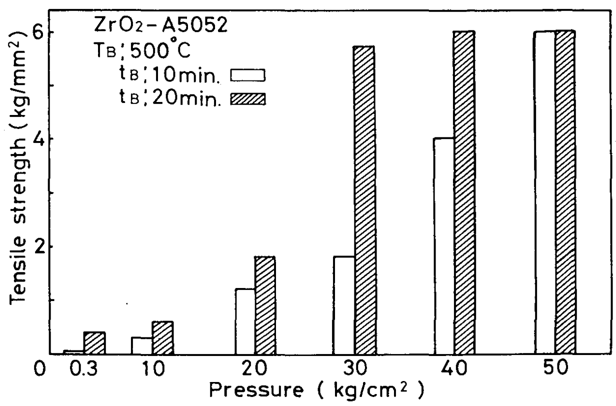

Fig. 3 Pressure dependence of bonding strength on bonding of A5052 and ZrO_2

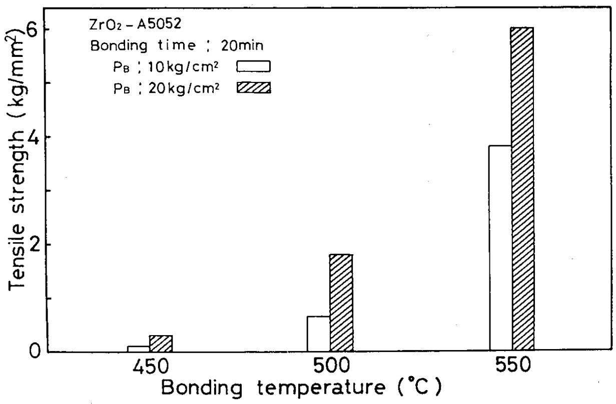

Fig. 4 Temperature dependence of bonding strength on bonding of A5052 and ZrO_2

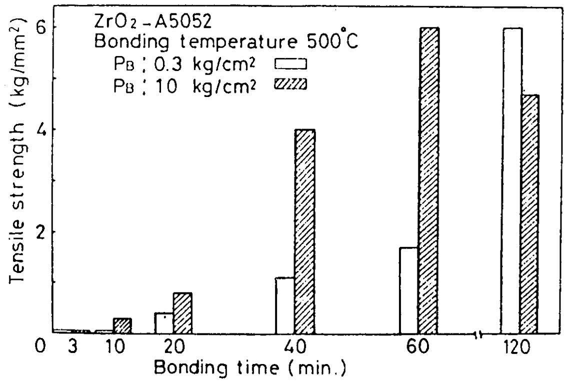

Fig. 5 Time dependence of bonding strength on bonding of A5052 and ZrO_2

ature under both 10 Kg/cm^2 and 20 Kg/cm^2. **Figure 5**

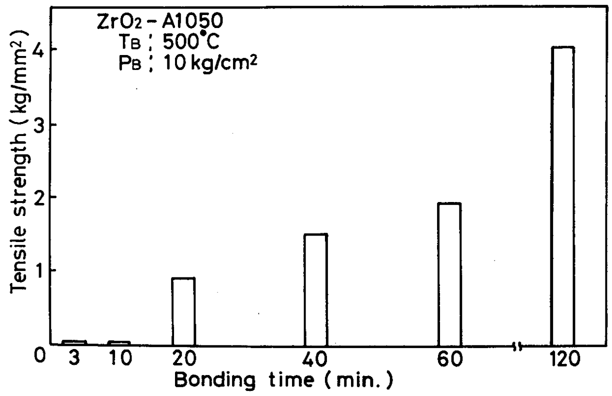

Fig. 6 Results of bonding of A1050 and ZrO_2

shows the bonding time dependence of the bonding strength.

From these figures, the strength increased respectively with the increase of bonding time and temperature.

To clarify the effect of Mg in the alloy on the bonding to ZrO_2, the bonding of A1050 containing no Mg and ZrO_2 was done at 500°C under 10 Kg/cm^2 pressure by changing the bonding time. The results are shown in **Fig. 6**. For A5052 containing Mg, the bonding is possible for 10 min under the same conditions as shown in Fig. 5. However, the bonding of ZrO_2 and A1050 is difficult under the same conditions as shown in Fig. 6. The strength of bonded joints increased with the increase of time also for A1050. The bonding difference between A5052 and A1050 may be due to the existence of Mg in

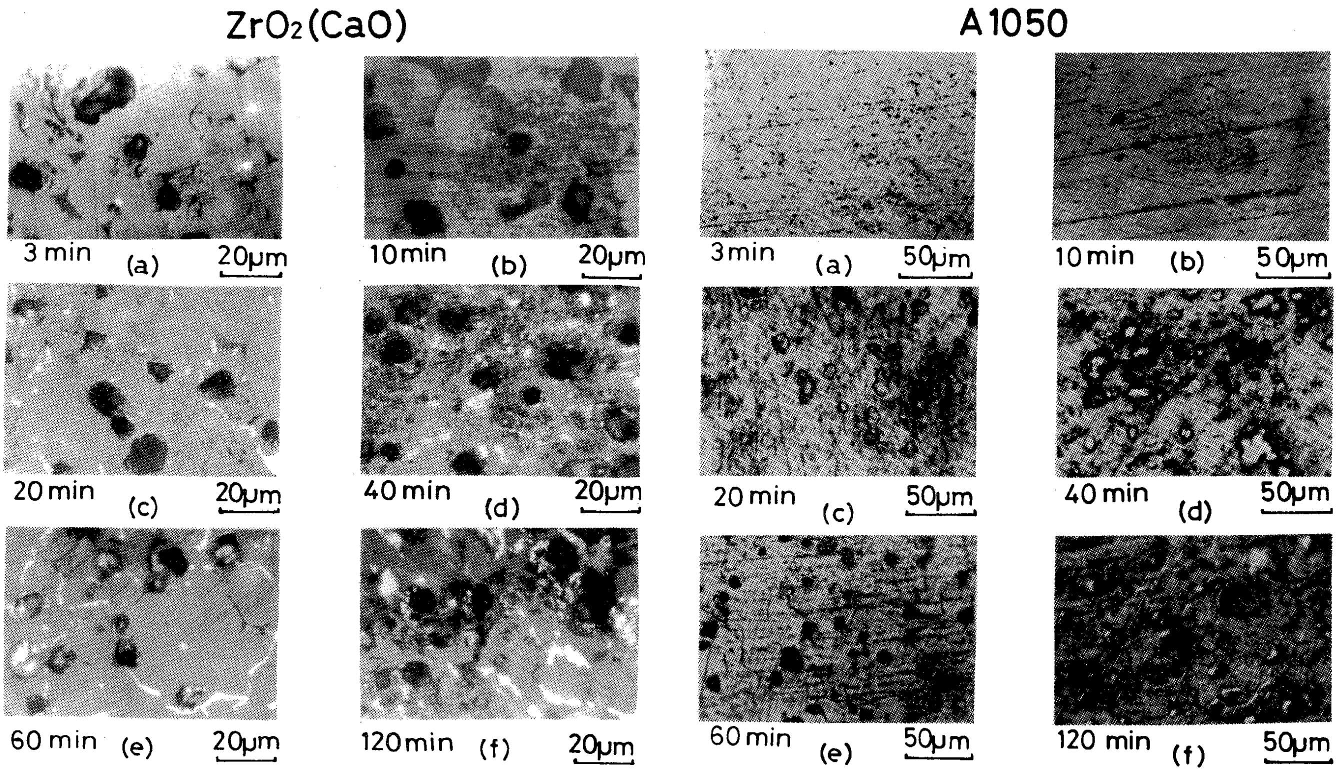

Fig. 7 Change in contact surfaces of both ZrO_2 and A1050 with time for bonding at 500°C under 10 kg/cm^2 pressure

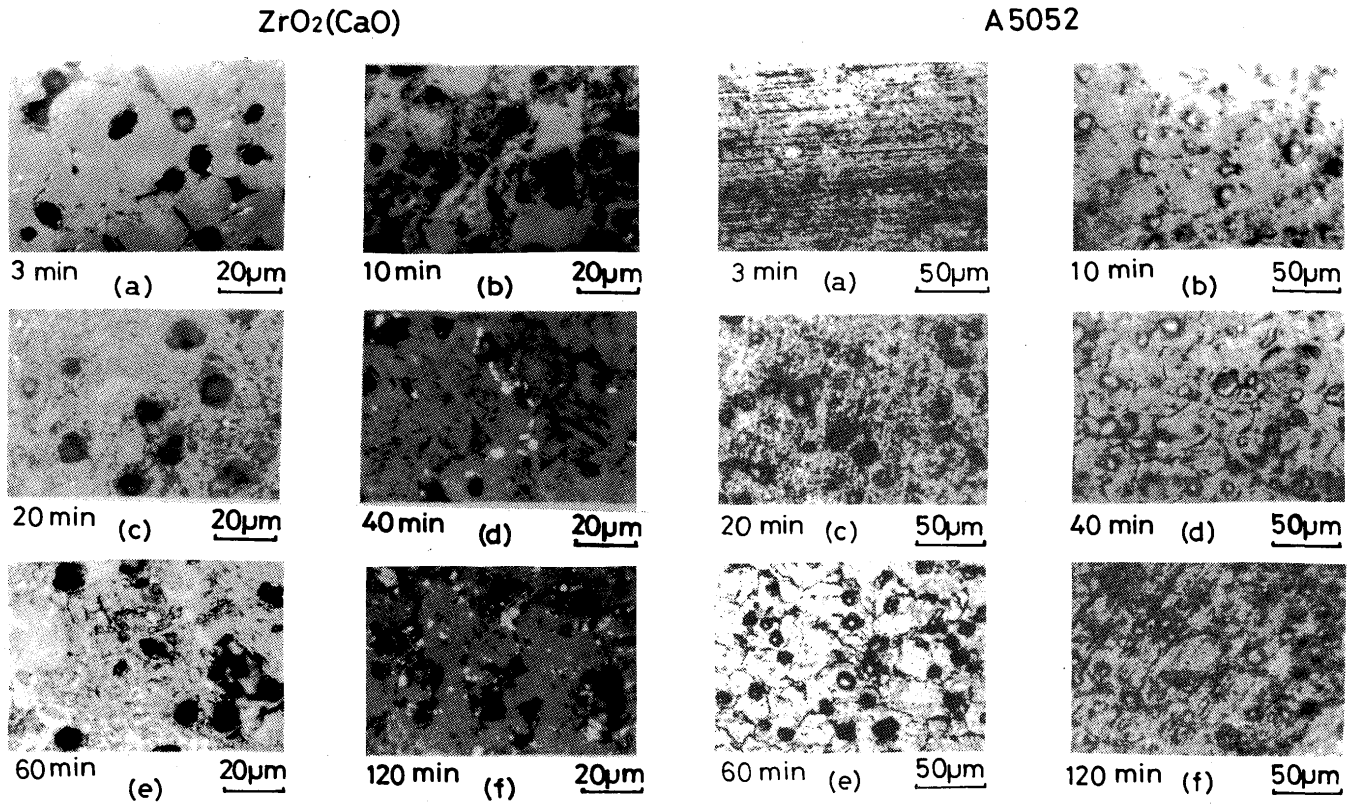

Fig. 8 Change in contact surfaces of both ZrO_2 and A5052 with time for bonding at 500°C under 10 kg/cm²

alloy.

3.2 Behavior of bonding surface and bonding part

The change in contact surfaces of both ZrO_2 and alloy with bonding time for bonding at 500°C under 10 kg/cm² pressure are shown in **Fig.** 7 for A1050 and in **Fig.** 8 for A5052.

As shown in these figures, the contact surfaces of both ZrO_2 and alloy changed little for 3 min, when bonding did not occur. For 10 min bonding of A5052, both surfaces changed greatly. However, for A1050, the similar surfaces after 10 min bonding was seen as the surface for 3 min bonding of A5052. The difference of contact surface may be due to the reaction of Mg and ZrO_2 ceramic at the interface.

Figure 9 shows the change in contact surfaces of ZrO_2 and A5052 with bonding pressure, when the bonding was done by heating at 550°C for 3 min. As shown in this figure, ZrO_2 and A5052 contact surface did not change under the pressure of 0.3 kg/cm². However, with increasing the pressure it was seen that the filling of the alloy into pores on ZrO_2 surface increased, being accompanied with forming holes on the alloy contact surface. At the high pressure of 60 kg/cm² and 70 kg/cm², the grain boundary and grain part of ZrO_2 surface were spoiled, as the alloy surface was done. Under such higher pressure,

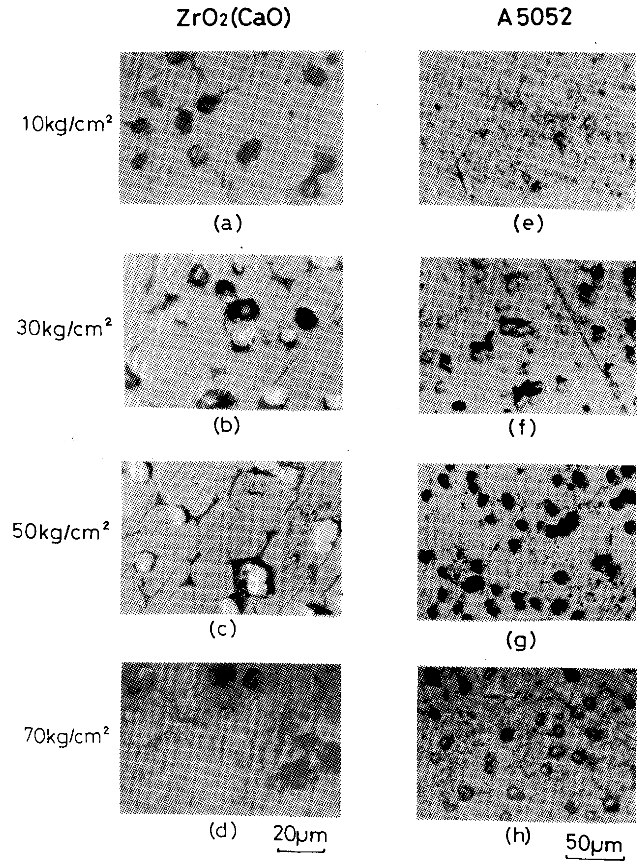

Fig. 9 Change in contact surfaces of ZrO_2 and A5052 with bonding pressure for bonding at 500°C for 3 min

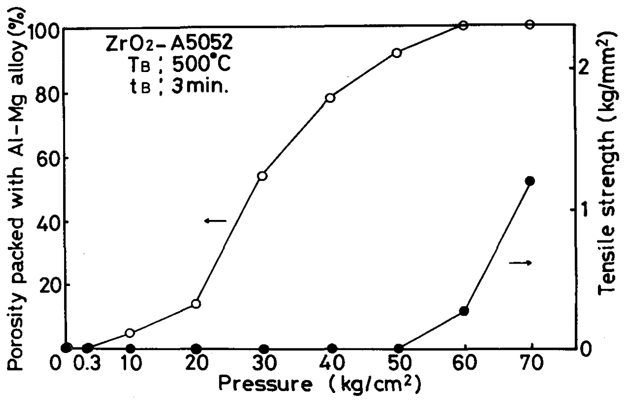

Fig. 10 Change in both filling ratio of A5052 in ZrO_2 pores and bonding strength with bonding pressure for bonding of A5052 and ZrO_2

even if the bonding time was a short time of 3 min, Al-Mg alloy reacted with ZrO_2 ceramic at the grain boundary and grain part at the contact interface between the alloy and ZrO_2. The bonding of Al-Mg alloy and ZrO_2 ceramic becomes possible. **Figure 10** shows both the change in the filling of the alloy in ZrO_2 pores and the bonding strength with bonding pressure for 3 min at 500°C. From this

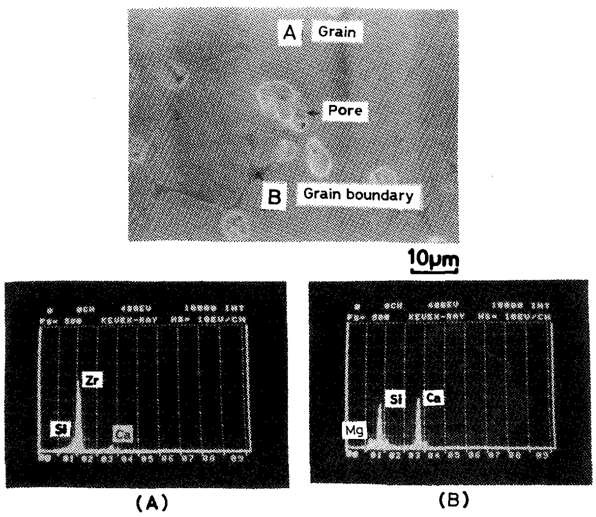

Fig. 11 SEM micrograph and DEX analysis results of ZrO_2 surface before bonding

figure, the bonding occurred, when the filling of A5052 in ZrO_2 pores becomes complete. **Figure 11** shows SEM and EDX analysis results of ZrO_2 ceramic surface before bonding. ZrO_2 ceramic was composed of ZrO_2 grain, pores and grain boundary containing mainly Si, Ca and Mg oxides. The fracture surface of ZrO_2 side of A5052/ZrO_2 joint bonded by heating for 3 min at 500°C under 60 kg/cm^2 pressure was observed by SEM. The results are shown in **Fig. 12.** It was seen that the pores of ZrO_2 ceramic were filled with Al-Mg alloy and Si existed in the alloy packed in the pore. **Figure 13** shows the change in bonding interface part with the bonding time, when A5052 was bonded with ZrO_2 at 500°C under 10 kg/cm^2 pressure.

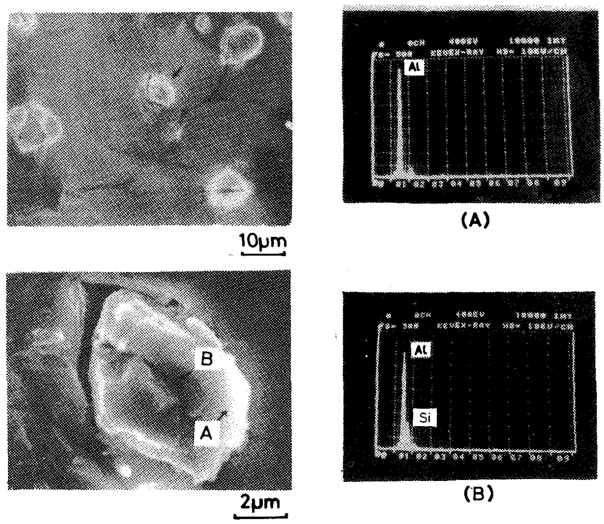

Fig. 12 Fracture surface of ZrO_2 side of A5052/ZrO_2 joint bonded by heating for 3 min at 500°C under 60 Kg/cm^2 pressure

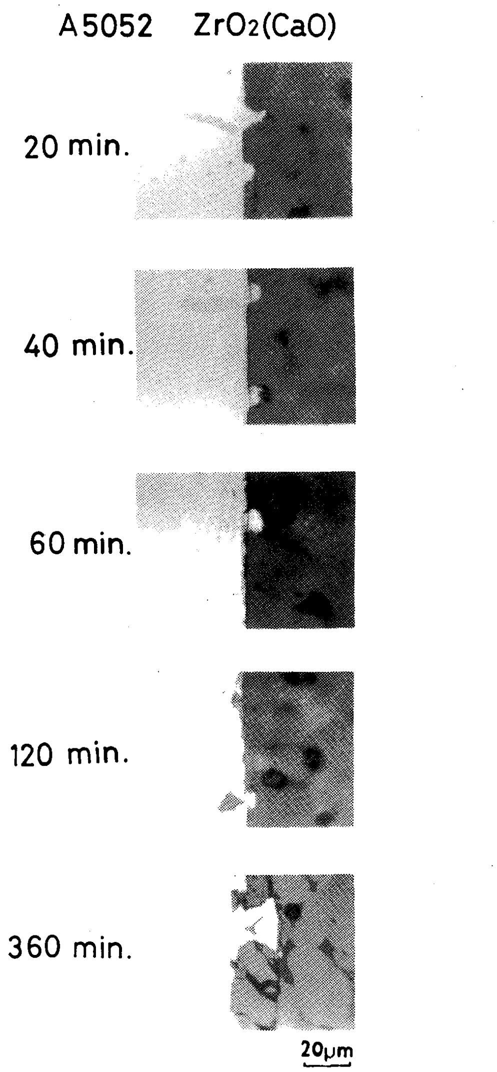

Fig. 13 Change in bonding interface part of A5052 and ZrO_2 with bonding time for bonding at 500°C under 10 Kg/cm^2 pressure

From this figure, it was seen that as the bonding time increased, the uneven parts increased at the interface of A5052 and ZrO_2.

This shows that the reaction of ZrO_2 and the alloy is acknowledged clearly with the lapse of bonding time. **Figure 14** shows the line analysis of EDX of bonding part of A5052 and ZrO_2 bonded by heating at 500°C for 20 min under 10 kg/cm^2 pressure. From these analysis results, it may be shown that the reaction of ZrO_2 and the alloy occurs.

From these results, it may be considered that during the bonding of ZrO_2 and A5052, at first, the filling of the alloy into ZrO_2 pores under the pressure occurred and then bonded part was expanded to the grain boundary and grain part under the higher pressure, so that the bonding became possible.

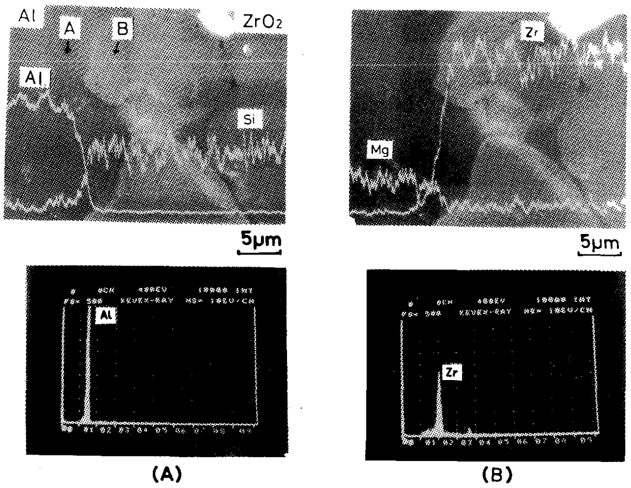

Fig. 14 SEM micrographs and EDX analysis of bonding part of A5052 and ZrO_2 boned by heating at 500°C for 20 min under 10 Kg/cm^2 pressure

3.3 Bonding of A5052 and other oxide ceramics

Solid state bonding was investigated for both combinations of A1050 to ZrO_2 ceramic and A5052 to ZrO_2 ceramic under a pressure. Bonds were formed under a bonding conditions by the reaction of ZrO_2 ceramic and Al or Al-Mg alloy. In this section, the bonding of A5052 with other oxide ceramics in Table 1 was done. The results are shown in **Fig. 15**. As shown in this figure, the bonding of A5052 and other ceramics was possible and the bonding of other ceramics may be due to the reaction of SiO_2, Al_2O_3 and ZrO_2 with Al-Mg alloy.

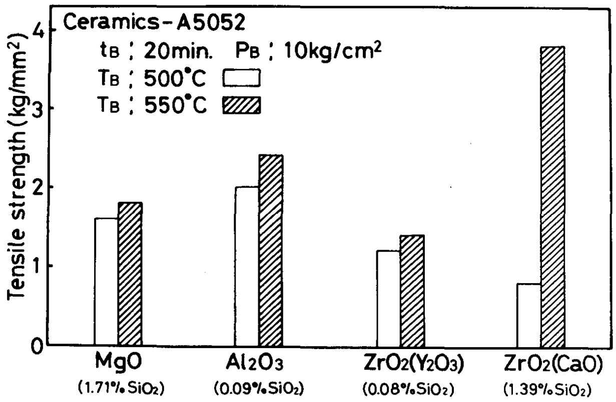

Fig. 15 Results of bonding of A5052 and other oxide ceramics

4. Conclusions

The obtained results in this experiment were summarized as follows;

(1) ZrO_2 (CaO) was bonded directly with A1050, A5052 by heating at 500°C under a pressure of 0.3 ~ 70 kg/cm^2. The bonding strength of bonded joint increased respectively with the increase of bonding time, temperature and pressure.

(2) The bonding process has three stages as follows;
   A) Initial stage of breaking of oxide film on the alloy surface and filling of the alloy into pores in ZrO_2 (CaO).
   B) Second stage of reaction of Mg, Al and SiO_2 in grain boundary part and the bond at grain boundary part.
   C) Third stage of reaction of Mg, Al and ZrO_2 grain and the bond at grain part.

(3) Bond formation between A5052 and other oxide ceramics such as MgO, Al_2O_3 was possible by heating at 500°C under 10 kg/cm^2 pressure.

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