

Title	Interfacial Phenomena During Field Assisted Bonding of Zirconia to Metals
Author(s)	Arata, Yoshiaki; Ohmori, Akira; Sano, Saburo
Citation	Transactions of JWRI. 15(2) P.387-P.390
Issue Date	1986-12
Text Version	publisher
URL	http://hdl.handle.net/11094/10883
DOI	
rights	本文データはCiNiiから複製したものである
Note	

Osaka University Knowledge Archive : OUKA

<https://ir.library.osaka-u.ac.jp/>

Osaka University

Interfacial Phenomena During Field Assisted Bonding of Zirconia to Metals†

Yoshiaki ARATA*, Akira OHMORI** and Saburo SANO***

KEY WORDS: (Ceramics) (Zirconia) (Metal) (Field Assisted Bonding)

Field assisted bonding is a method to make a direct bonding between metals and solid electrolyte ceramics by applying an electric field. This method has been reported for glasses¹⁾ and beta-alumina²⁾ in which the mobile ion is Na⁺. Other works also have been done for the effect of external pressure,³⁾ for the effect of surface imperfection,⁴⁾ and so on. In this paper, field assisted bonding is extended to zirconia, which is an O²⁻ ion conducting solid electrolyte ceramic.

Zirconia used in this work is stabilized with 15 mol% MgO, and the size used for field assisted bonding is 13 mm in diameter and 4 mm in thickness. Industrial grade pure metals (>99.5%), Cu, Ni and Al are used for both anode and cathode electrodes. The size of a metal plate is 15 mm X 25 mm and 2 mm in thickness. Both surfaces of a zirconia disk and each surface of two metal plates which directly contact with the zirconia disk are polished with a #1500 emery paper. After polishing, the specimens are degreased in acetone with an ultrasonic cleaner. Then the specimens are assembled as a zirconia disk is sandwiched between two metal plates. For applying an electric field,

lead wires are percussion welded to the metal plates. The assembly is setted in a vacuum furnace as shown in Fig. 1. Then, the assembly is heated in a vacuum (7×10^{-3} Pa) to a required temperature at the heating rate of 50 K/min. When the temperature reaches a required temperature, an external pressure and a dc current are applied to the specimen. In this work, all experiments were done below 1073K, and the maximum applied field was 100 V and 2A. A slight external pressure of 10 MPa was applied to make an intimate contact between a zirconia disk and electrode plates. After field assisted bonding, the assembly is cooled at the rate of 10 K/min.

Under high applied electric field, current across zirconia increases with time.⁵⁾ Same result was recognized in this work. As shown in Fig. 2, a constant voltage was applied until current reached a required constant current, and then the constant current was kept. By changing the value of constant current and the time of applying electric field, the total amount of electricity across zirconia was controlled.

Figure 3 shows interface parts of both anode side and

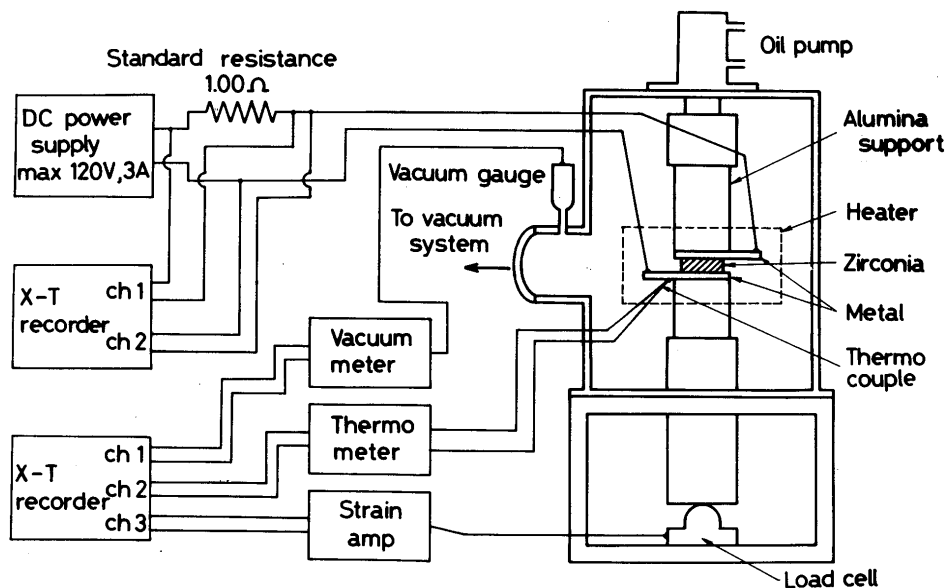


Fig. 1 Schematic diagram of bonding apparatus and circuit for applying electric field.

† Received on Nov. 5, 1986

* Professor

** Associate Professor

*** Graduate Student

cathode side of Cu (anode)/Zirconia/Cu (cathode) joint bonded at 1073 K for 20 min. The applied field was 50 V, 1 A and the total amount of electricity was 900 coulombs. As shown in Fig. 3, a layer was produced at interfaces of both anode side and cathode side. The layer produced at the anode interface was identified as Cu₂O by X-ray diffraction. A possibility of bonding between zirconia and metals by making an oxide layer at the anode interface was mentioned,⁶⁾ and the bonding strength will depend on the nature of oxide.²⁾ In this case, Cu₂O layer was thick and brittle, so the joint broke easily in the layer. Strong bonding was achieved at cathode side for the bonding of zirconia to Cu. By pin contact Pt cathode electrode, segregation of zirconia itself did not occur.⁵⁾ But, by

face contact Cu cathode electrode, a layer which has metallic luster was produced by segregation of zirconia at the cathode side interface as shown in Fig. 3. Figure 4 shows a SEM photograph and EDX element analysis results of the cathode side bonding part. The layer contains both Cu and Zr as shown in Fig. 4. If an electric field was not applied, both anode side and cathode side layers were not produced. The detail of the layer formation will be discussed in future reports.

Similar result was obtained for the joint of Ni (anode)/Zirconia/Ni (cathode), but clear layer was not seen at the anode side interface as shown in Fig. 5. At the cathode side a layer is produced, and the layer contains Ni and Zr as shown in Fig. 6. Though the bonding strength of anode side was 0.5 MPa, the bonding strength of cathode side was 5 MPa. The specimen was broken in zirconia itself when the cathode side bonding strength was measured. It was thought that since zirconia became brittle under a high electric field⁷⁾ and some cracks were induced by a

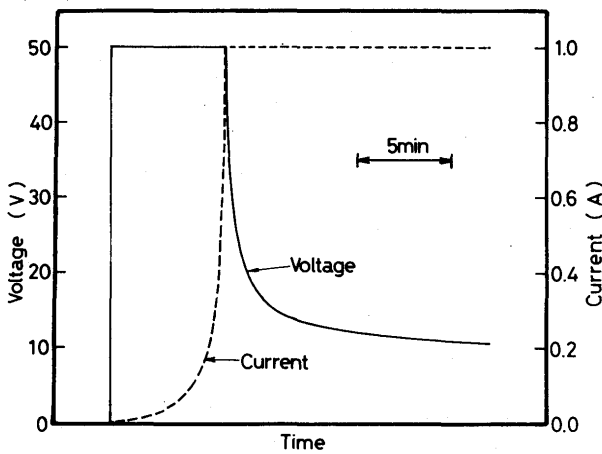


Fig. 2 Voltage and current changes during bonding of Ni (anode)/Zirconia/Ni (cathode) at 1073 K.

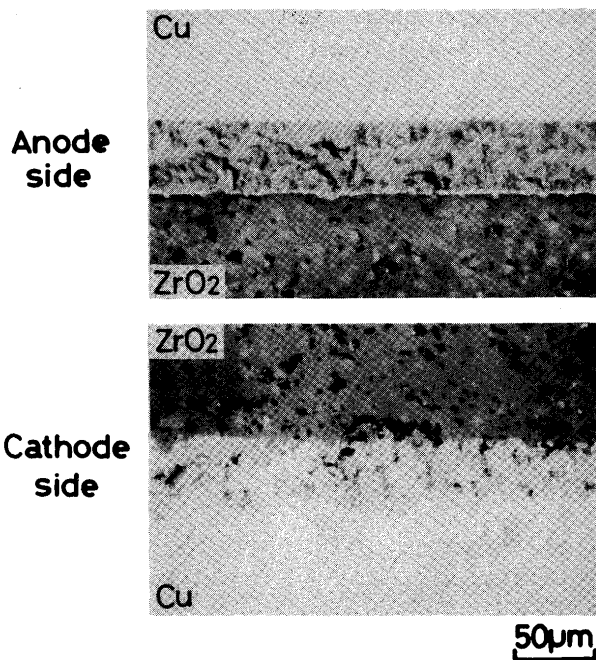


Fig. 3 Microphotographs of both anode and cathode side cross-sections of Cu (anode)/Zirconia/Cu (cathode) joint bonded at 1073 K for 20 min under electric field of 50 V and 1 A.

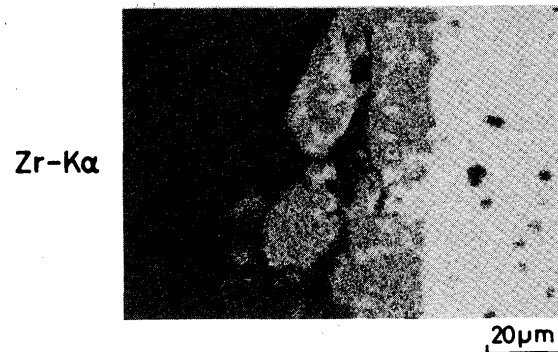
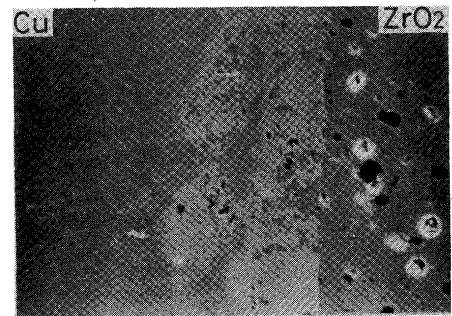


Fig. 4 SEM photograph and EDX element analysis results of cathode side interface of Cu(anode)/Zirconia/Cu(cathode) joint bonded at 1073 K for 20 min under electric field of 50 V and 1 A.

thermal stress.

Several bonding conditions and results are summarized in Table 1. Since current or voltage changes with time, the quantities in Table 1 reflect maximum values as shown in Fig. 2. From Table 1, Cu, Ni and Al make a layer at cathode side interface with zirconia under an electric field. By making a cathode side layer, zirconia bonds strongly with metals. As shown in Table 1, if an electric field was not applied the cathode side layer was not produced. For Cu, a cathode side layer was not produced at the bonding temperature of 873 K, though the total amount of elec-

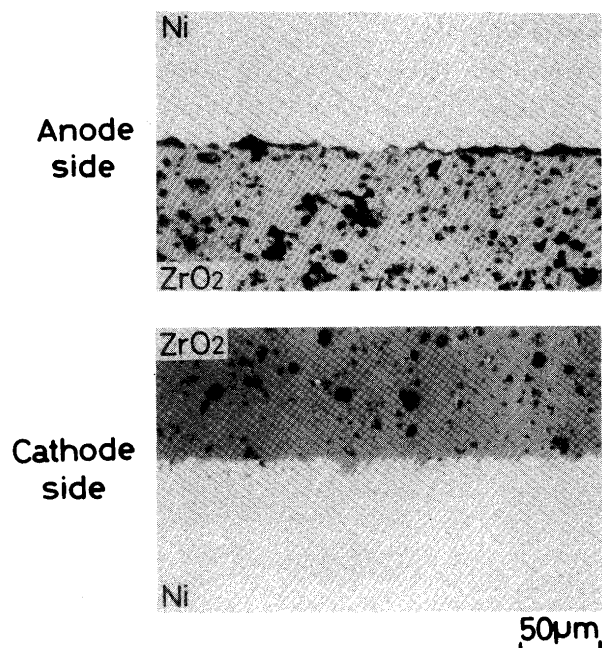


Fig. 5 Microphotographs of both anode and cathode side cross-sections of Ni (anode)/Zirconia/Ni (cathode) joint bonded at 1073 K for 60min under electric field of 100V and 1 A.

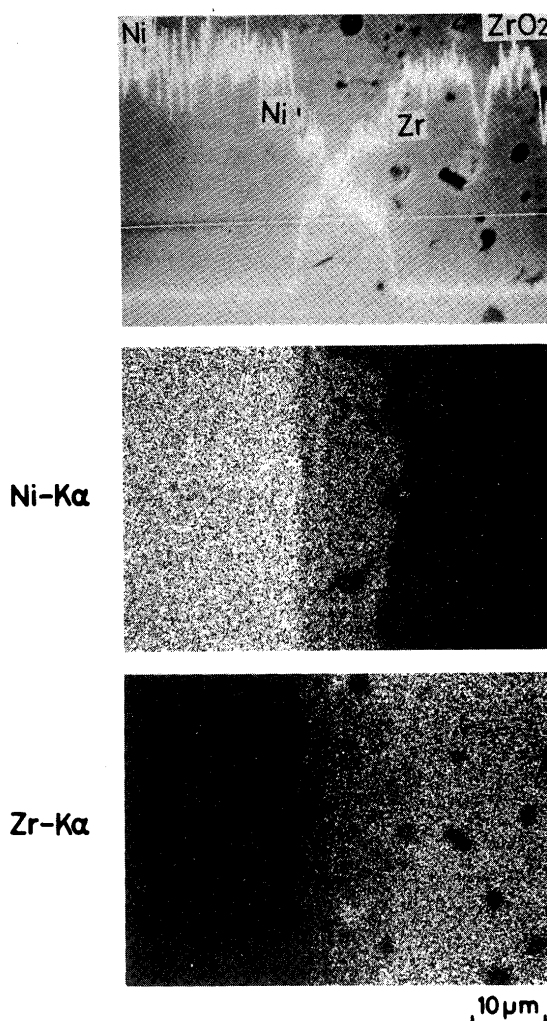


Fig. 6 SEM photograph and EDX element analysis results of cathode side interface of Ni (anode)/Zirconia/Ni (cathode) joint bonded at 1073 K for 60min under electric field of 100V and 1 A.

Table 1 Bonding conditions and results of field assisted bonding of zirconia to metals.

No	Metals	Bonding conditions					Formation of layer [*]	
		Temp. (K)	Time (min)	Voltage (V)	Current (mA)	Amount of electricity (C)	Anode side	Cathode side
1	Cu	873	60	100	1000	1500	○	×
2	Cu	973	60	50	2000	5100	○	○
3	Cu	973	60	50	1000	1600	○	○
4	Cu	1073	20	50	1000	900	○	○
5	Cu	1073	20	0	0	0	×	×
6	Ni	873	60	100	1000	1050	×	○
7	Ni	973	30	50	1000	900	×	○
8	Ni	1073	60	50	1000	3400	×	○
9	Ni	1073	60	0	0	0	×	×
10	Al	873	40	100	90	80	×	○

* ○:Clear layer was produced ×:Clear layer was not produced

tricity was 1500 coulombs. So, applying an electric field and heating to a temperature is necessary for making a cathode side layer. When glass or beta-alumina was field assisted bonded to metals, bonding was achieved at anode side by moving of Na^+ ions to cathode.^{1,2)} But, for the bonding of zirconia to Cu, oxide layer was produced at anode side by moving of O^{2-} ions to anode under an electric field. As above mentioned, the oxide layer produced at the zirconia/Cu interface was weak, so a strong bonding was not obtained at anode side. The bonding strength at anode side will depend on the nature of oxide layer. However, the most interest point is that strong bonding was achieved by mutual diffusion of Zr and metals at the cathode side interface between zirconia and metals under an electric field. The detail of this bonding process will be discussed in future reports.

References

- 1) G. wallis and D. I. Pomerantz, "Field-Assisted Glass-Metal Sealing", J. Appl. Phys., Vol. 40 No. 10 (1969) p 3946.
- 2) B. Dunn, "Field-Assisted Bonding of Beta-Alumina to Metals", J. Am. Ceram. Soc., Vol. 62 No. 11-12 (1979) p 545.
- 3) Y. Arata, A. Ohmori, S. Sano and I. Okamoto, "Pressure and Field-Assisted bonding of Glass to Aluminum", Trans. JWRI, Vol. 13 No. 1 (1984) p 35.
- 4) T. R. Anthony, "Anodic Bonding of Imperfect Surfaces", J. Appl. Phys., Vol 54 No. 5 (1983) p 2419.
- 5) H. Yanagida and T. Kandachi, "Voltage-Current-Time characteristic of Stabilized Zirconia with Pin Contact Platinum Electrode in Argon Ambient", Yogyou-Kyoukai-Shi, Vol. 84 No. 7 (1976) p 338.
- 6) S. Sano, Master Thesis, OSAKA UNIV., (1982).
- 7) G. B. Barbi, "Alternations of Conductivity and Structure of Zirconia Base Electrolytes Under High Electric Fields", Commission of the European Communities, (1984).