



Title	Intermediary Layer of Titanium Oxide in Al <sub>2</sub> O <sub>3</sub> /Cu Joint Using Amorphous Cu-Ti Filler Metals
Author(s)	Naka, Masaaki; Asami, Katsuhiko; Okamoto, Ikuo et al.
Citation	Transactions of JWRI. 1983, 12(1), p. 145-148
Version Type	VoR
URL	<a href="https://doi.org/10.18910/4932">https://doi.org/10.18910/4932</a>
rights	
Note	

*The University of Osaka Institutional Knowledge Archive : OUKA*

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

The University of Osaka

## Intermediary Layer of Titanium Oxide in $\text{Al}_2\text{O}_3/\text{Cu}$ Joint Using Amorphous Cu-Ti Filler Metals<sup>†</sup>

Masaaki NAKA \*, Katsuhiko ASAMI\*\*\*, Ikuo OKAMOTO\*\* and Yoshiaki ARATA\*\*

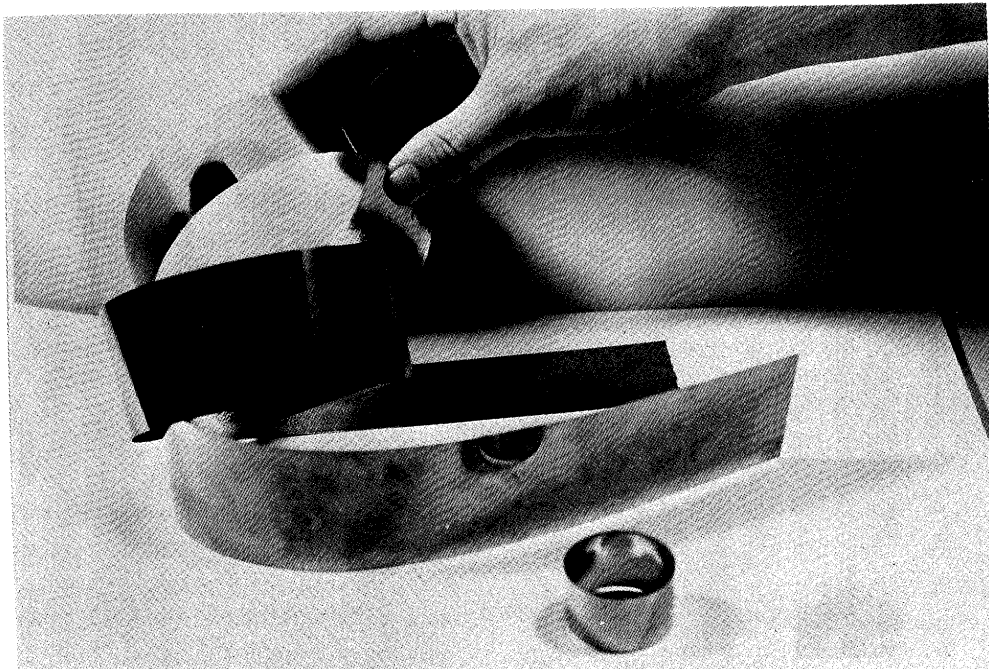
**KEY WORDS:** (Joining) (Ceramics) (Intermediary Oxide) (Titanium Oxide) ( $\text{Al}_2\text{O}_3$ ) (Copper) (Amorphous Filler Metals) (Copper-Titanium)

In the joining of metals and ceramics, the reactions occurring at the interface greatly affect the quality of the joint. Knowledge of the reactions involved is necessary to control the process conditions.

In joining alumina to steel using copper filler metal in a slightly oxidizing atmosphere<sup>1)</sup>, an intermediary layer compound  $\text{FeAl}_2\text{O}_4$  is formed at the bonding interface between alumina and steel. In the joining of ceramics to metals using Mo metallizing, an oxide is added to the

metal powder; and in the liquid state this oxide wets both the metal and the ceramic<sup>2),3)</sup>. In solid-state bonding of metals to ceramics, a direct interaction of the materials takes place<sup>4),5)</sup>. The present paper describes the presence of intermediate phases at the bonding interface between alumina and copper using amorphous Cu-Ti filler metal.

The materials used were high purity commercial alumina (99.5%) and tough pitch copper (0.03%O). The



**Fig. 1** Appearance of amorphous  $\text{Cu}_{50}\text{Ti}_{50}$  filler metal. This foil possesses an excellent flexibility as shown in the figure.

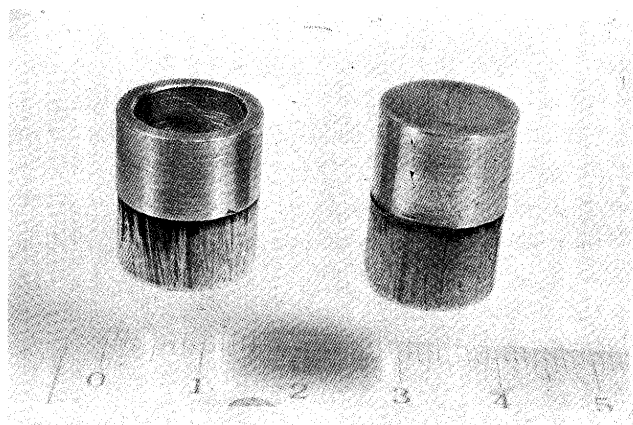
<sup>†</sup> Received on April 30, 1983

\* Associate Professor

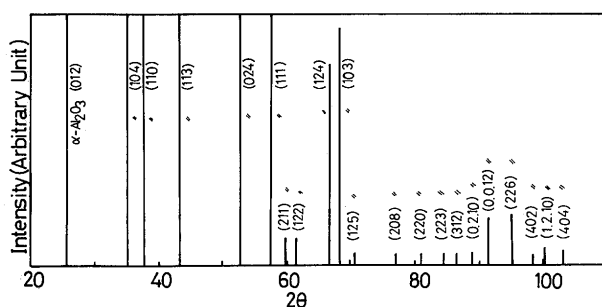
\*\* Professor

\*\*\* Research Instructor, The Research Institute for Iron, Steel and other Metals, Tohoku University, Sendai

amorphous Cu-Ti filler metals were ejected on a rotating wheel through a quartz orifice. The filler metals were 5 cm in width and 50  $\mu$ m in thickness. The amorphous structure of filler metal was confirmed by X-ray diffractometry. The composition range of Cu-Ti filler metal was 30 to 65 atomic percent titanium. **Figure 1** shows the appearance of filler metals. The joining was conducted in  $5 \times 10^{-5}$  torr at 1025°C for 30 min. The liquid filler metal wetted both alumina and copper during joining and hence a strong bond was formed after cooling down. Various types of joints were produced using amorphous Cu<sub>50</sub>Ti<sub>50</sub> filler metal. A butt joint of alumina rod with copper rod and a butt joint of alumina cylinder with copper cylinder are shown in **Fig. 2**.

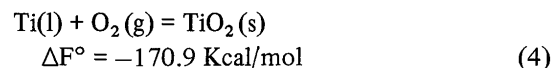
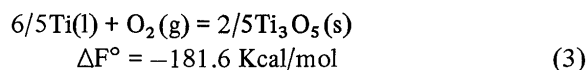
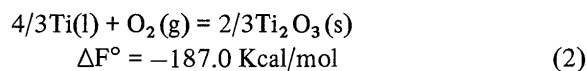
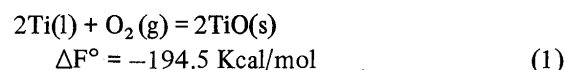


**Fig. 2** Butt joint of Al<sub>2</sub>O<sub>3</sub> rod with copper rod (right), and butt joint of Al<sub>2</sub>O<sub>3</sub> cylinder with copper cylinder (left). The upper and lower parts in the specimens are copper and Al<sub>2</sub>O<sub>3</sub>, respectively.



**Fig. 3** X-ray diffraction pattern of the revealed surface on Al<sub>2</sub>O<sub>3</sub> with Cu · K $\alpha$ , after dissolution of copper and filler metal from a Al<sub>2</sub>O<sub>3</sub>/copper joint.

In order to characterize the intermediary layer of joint, copper and filler metals were dissolved from the alumina/copper joint in concentrated hot HCl, and X-ray diffraction analysis with Cu · K $\alpha$  radiation and X-ray photoelectron spectroscopic examination with Mg · K $\alpha$  excitation were conducted to identify the intermediary compound. X-ray diffraction analysis indicated only the alumina, and did not identify the intermediary compound as shown in **Fig. 3**. The revealed surface of alumina was, therefore, analysed by X-ray photo-electron spectrometry. The Cls spectrum from the contaminant carbon was used for the calibration of binding energy of electrons. The Ti2p<sub>3/2</sub>, Ti2p<sub>1/2</sub>, Al2p and O1s electron spectra are given in **Figs. 4 to 6**, respectively. The binding energies of the Ti2p<sub>3/2</sub>, Ti2p<sub>1/2</sub> are taken as 458.50 and 464.30 eV, respectively. The spectra of Ti2p are attributed to TiO<sub>2</sub>. The thermodynamic calculation suggests the presence of titanium oxide containing lower oxygen content than that of TiO<sub>2</sub>. The free energies ( $\Delta F^\circ$ ) of the following reactions are considered at 1025°C<sup>6</sup>.



where the symbols of s, l and g denote the state of solid, liquid and gas, respectively. The comparison of  $\Delta F^\circ$  of Eqs. (1) – (4) indicates that the instabilities of titanium oxides increase in the sequence of TiO, Ti<sub>2</sub>O<sub>3</sub>, Ti<sub>3</sub>O<sub>5</sub> and TiO<sub>2</sub>.

The binding energy of the Al2p electron (74.65 eV) measured is very close to that of Al<sub>2</sub>O<sub>3</sub> (74.70 eV). The spectra of O1s are separated into O1s in Al<sub>2</sub>O<sub>3</sub> (531.25 eV) and O1s in TiO<sub>2</sub> (529.20 eV). The binding energy of O1s in Al<sub>2</sub>O<sub>3</sub> observed is different from that of Al<sub>2</sub>O<sub>3</sub> (531.55 eV). This fact indicates that titanium dissolves into alumina and forms (Al, Ti)<sub>2</sub>O<sub>3</sub> solid solution at the interface of the joint. Further, the isothermal solidification process during joining took place between copper metal and filler metal. During joining, first copper dissolves into liquid filler metal. Copper containing titanium, then, precipitates from the liquid filler metal. These results give the structure at the interface of joint of Al<sub>2</sub>O<sub>3</sub> with copper using Cu-Ti filler metal as shown in **Fig. 7**.

It can be concluded that the intermediary compounds

of titanium oxide  $\text{TiO}_x$  and  $(\text{Al}, \text{Ti})_2\text{O}_3$  are formed during joining using amorphous Cu-Ti filler metals.

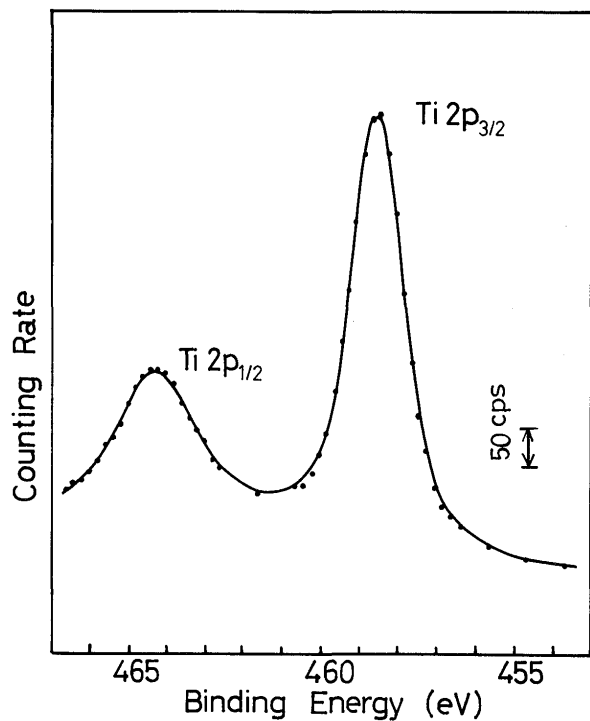


Fig. 4 Ti2p spectrum of the revealed surface on  $\text{Al}_2\text{O}_3$ , after dissolution of copper and filler metal from a  $\text{Al}_2\text{O}_3/\text{copper}$  joint.

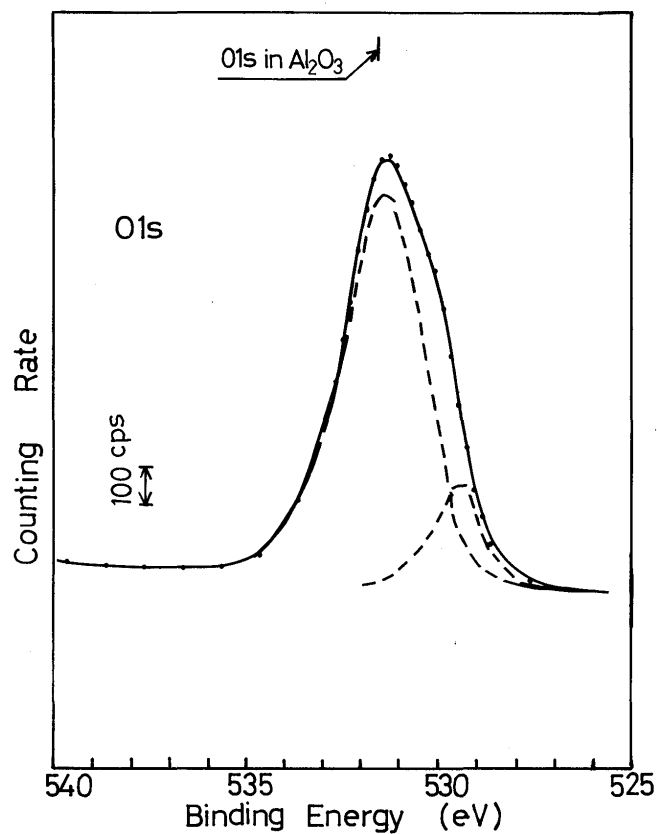


Fig. 6 O1s spectrum of the revealed surface on  $\text{Al}_2\text{O}_3$ , after dissolution of copper and filler metal from a  $\text{Al}_2\text{O}_3/\text{copper}$  joint.

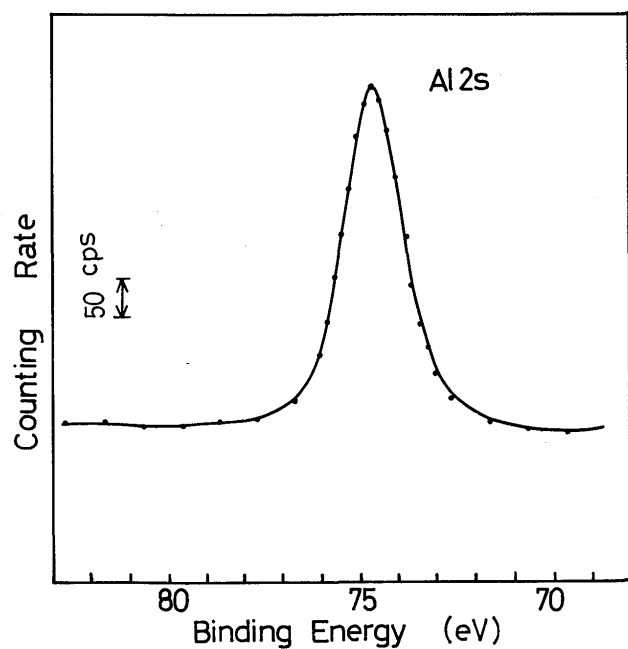


Fig. 5 Al2p spectrum of the revealed surface on  $\text{Al}_2\text{O}_3$ , after dissolution of copper and filler metal from a  $\text{Al}_2\text{O}_3/\text{copper}$  joint.

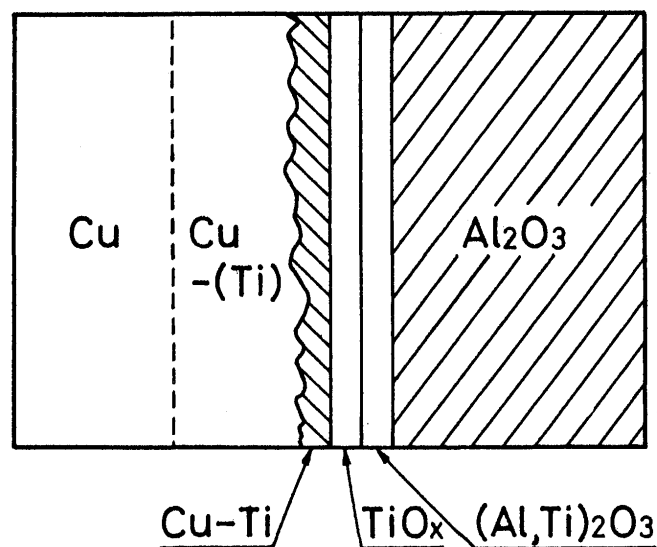


Fig. 7 Structure at the interface of  $\text{Al}_2\text{O}_3/\text{copper}$  joint using Cu-Ti filler metal.

## References

- 1) I. Okamoto, M. Naka, K. Asami and K. Hashimoto: Trans. JWRI, 11 (1982), No. 1, 133.
- 2) J.T. Klomp and Th. P.J. Botden: Br. Ceram. Res. Assoc., Spec. Publ., No. 48, (1965), 129.
- 3) R.M. Fulrath and E.M. Hollar: Am. Ceram. Soc. Bull., 47 (1968), 493.
- 4) W. Dawihl and E. Klinger: Ber. Dtsch. Keram. Ges., 46 (1969), 12.
- 5) H.J. de Brium, A.F. Moodie and C.E. Warbe: J. Aust. Ceram. Soc., 7 (1971), 57.
- 6) И.С. КУЛИКОВ: РАСКИСЛЕНИЕ МЕТАЛЛОВ, МОСКВА, 1975.