

Direct Tungsten/Copper Bonding for Divertor Application

Keisuke YAMAUCHI, Naoki SANO and Katsuaki TANABE

Department of Chemical Engineering, Kyoto University, Nishikyo, Kyoto 615-8510, Japan

(Received 21 February 2022 / Accepted 30 March 2022)

We carried out a fundamental investigation of a uniaxial direct W-to-Cu bonding at relatively low temperatures in ambient air, which would potentially allow for simple preparation and maintenance of divertor wall components. W/Cu bonds formed at 500°C with a bonding pressure of 0.1 MPa, but the mechanical interfacial strength was about 1 MPa, significantly lower than the state-of-the-art values for bonding around at 1000°C in vacuum. Higher degree of interfacial oxidation and atomic interdiffusion were observed for higher bonding temperature, through x-ray photoelectron spectroscopy. The electrical conductivity across the bonded W/Cu interface, an indicator of thermal conductance, was measured to be lower for higher bonding temperature, presumably due to the interfacial oxidation.

© 2022 The Japan Society of Plasma Science and Nuclear Fusion Research

Keywords: nuclear fusion reactor, plasma-facing wall, divertor, armor, heatsink, bonding, brazing, tungsten, copper

DOI: 10.1585/pfr.17.1405049

1. Introduction

In magnetic-confinement nuclear fusion reactors, the divertor is positioned as an important component for multiple functions, such as particle exhaust, heat removal, and plasma confinement. The divertor plate comprises a bonded structure of an armor segment on the plasma side and a heatsink segment on the cooling tube side. The candidate materials for the armor include tungsten (W) [1], silicon carbide [2], and carbon fiber composites [3]. W has the highest melting point among elementary metals. In addition, W is strong against physical sputtering by the plasmas owing to the high bonding energies of atoms for its large atomic number. W can also minimize the chemical sputtering effect due to its small reactivity with hydrogen. For these advantages, W is one of the most promising candidate for the armor material of divertors. The ITER project plans to employ the so-called tungsten monoblock divertor structure. Whilst, the candidate materials for the heat sink include highly thermally conducting copper (Cu) [4], Cu-based alloys (CuCrZr, etc.) [5], and reduced-activation ferrite steels (F82H, etc.) [6].

For the bond between the armor and heatsink segments, various techniques have been investigated, such as brazing [7, 8], electron beam welding [9, 10], explosive welding [11, 12], and friction stir welding [13, 14]. At present, W/Cu brazing at temperatures around 1000°C in vacuum is thought as the first candidate [7, 8]. Meanwhile, W/Cu direct bonding has been studied also at around 1000°C in hydrogen atmosphere [15]. Reference [15] constructed the interface between W and Cu by the diffusion bonding method at temperatures close to the melting point of Cu, 1085°C. In the present study, we conduct a series of fundamental bonding experiments for W/Cu direct dif-

fusion bonding and analyze the interfacial characteristics, to explore the possibility of the realization of a simple, direct bonding technique in ambient air at temperatures significantly lower than the melting point of Cu. Such an approach may enable for low-cost, high-throughput production of reactor components. For instance, processes of higher temperatures simply consume larger supplied electric power and typically employ large, laborious apparatuses.

For practical nuclear fusion reactors in the future, the armor segment may be required to be frequently replaced due to its damage, degradation, and radioactivation by plasma and neutron irradiation. Therefore, the view of industrialization such as production efficacy becomes important; low-cost, high-throughput manufacture and maintenance of the components are to be highly demanded. Particularly, sole replacement of the armor segment in a shorter cycle would be desirable because material degradation typically occurs on the armor, but not other segments, in practical reactors. If realized, simple direct bonding at lower temperature in ambient air would enable, for instance, local installation of the W armor blocks by using a handy heating and pressing tool like a clothes iron, without replacement of the whole divertor cassette. In addition, direct bonding may have advantages such as cleanliness and high thermal conductance, relative to the bonding methods mediated by interfacial agents. Furthermore, detachability could be even realized by properly adjusting the mechanical interfacial bonding strength. Despite the outstanding thermal and mechanical properties, the difficulty in processing and bonding of W has limited its application. In addition to the nuclear fusion application, the versatile bonding technique could benefit the fields of heating facilities, machining tools, high-temperature electrodes, x-ray

author's e-mail: tanabe@cheme.kyoto-u.ac.jp

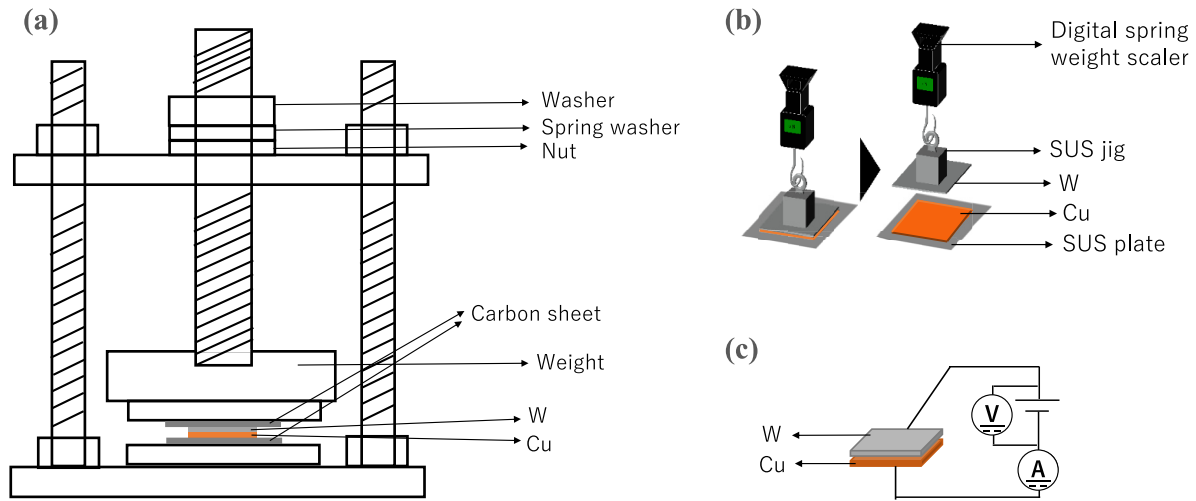


Fig. 1 Schematic illustrations of (a) the bonding method, (b) the mechanical measurement, and (c) the electrical measurement.

shields, etc.

2. Experimental Methods

We carried out the whole experimental processes of this study in a non-cleanroom, regular experimental room with a particle density of approximately 5 million m^{-3} , which we measured with a regular particle counter. We used W plates with dimensions of $1 \text{ cm} \times 1 \text{ cm} \times 0.2 \text{ mm}$ (99.95% purity, W-463386, Nilaco Corp.) and Cu plates of $1 \text{ cm} \times 1 \text{ cm} \times 1 \text{ mm}$ (99.99% purity, CU-113513, Nilaco Corp.) for the experiments in this study. The roughness level of the W and Cu surface to be bonded was both about 100 nm (R_a), as we observed in scanning electron microscopy. Firstly, the metal plates were submerged in acetone for 5 min to degrease the metal surfaces to be bonded. The W plate was then bonded to the Cu plate under a uniaxial pressure of 0.1 or 0.5 MPa at various temperatures in the range of 300–700°C in ambient air for 1, 3, or 7 h. The heating and cooling rates were approximately $10^\circ\text{C}/\text{min}$. Figure 1 depicts the experimental methods.

After the bonding, detachment normal stresses were measured for the bonded samples to represent bonded interfacial mechanical strengths. We connected an outer surface of the bonded sample to a digital spring weight scaler via a solid wire that was firmly attached to the sample surface using a household adhesive glue. Then, we pulled the scaler outward in a direction normal to the sample die until the bonded sample was debonded, while the weight scaler recorded the maximum force at the point of delamination. This delamination force was simply divided by the area of the W and Cu plates, 1 cm^2 , to determine the bonding strength. In addition, as a relative indicator of the thermal conductance at the bonded interface, we evaluated the interfacial electrical conductivities by measuring the current-voltage characteristics across the bonded interfaces. To analyze the elementary evolution at the bonded

interfaces, x-ray photoelectron spectroscopy was also carried out for the delaminated W and Cu surfaces.

3. Results and Discussion

For the bonding temperature of 300°C , the W and Cu plates did not bond to each other. At 400°C , bonds formed, but the interfacial mechanical bonding strength was quite weak and the samples easily debonded while being handled. Firm bonds were for the bonding temperatures of 500°C and over. Softening of Cu and atomic interdiffusion of W and Cu at relatively high temperatures might contribute to the bond formation. W and Cu are immiscible, not to form a stable alloy. Nevertheless, in an entropic point of view, impurity-level atomic diffusion may occur, to be experimentally observed in a later section. However, accounting for our significantly lower bonding temperature and strength than those for Ref. [15], we cannot be assertive on the diffusion-dominant bonding mechanism at the present stage. Figure 2 presents the dependence of the interfacial mechanical bonding strength on the bonding time for the W/Cu samples bonded at 500 and 700°C at 0.1 and 0.5 MPa. In Fig. 2, the interfacial mechanical strength is generally observed to become higher for longer bonding time. However, for the bonding for longer duration, we observed apparent surface oxidation of W, occasionally even with layer-by-layer delamination. Due to such surface corrosion, particularly in ambient air, bonding at too high pressure for long duration may rather weaken the mechanical bonding strength. We have thus obtained W/Cu bonding in ambient air at a temperature of 500°C , significantly lower than those of earlier studies [7, 8, 15], about 1000°C . The mechanical bonding strength of our samples, about 1 MPa, was however far lower than the state-of-the-art reported values, over 100 MPa. This weakness issue is to be resolved. Nevertheless, a delamination force of 1 MPa corresponds to a virtual situation that 1 ton ($= 1 \text{ m}^3$)

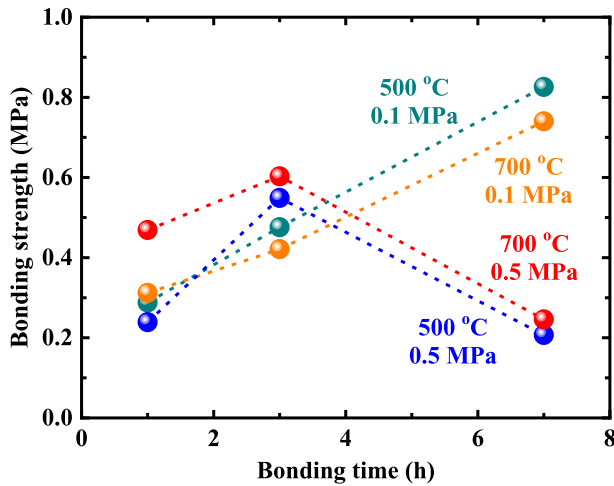


Fig. 2 Interfacial mechanical bonding strength in dependence on bonding time for the W/Cu samples bonded at 500 and 700 °C at 0.1 and 0.5 MPa.

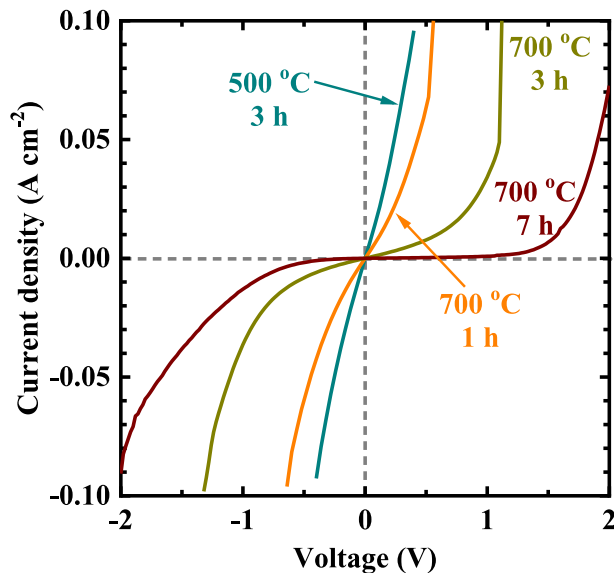


Fig. 3 Typical current-voltage characteristics of the W/Cu samples bonded at 0.1 MPa for various conditions.

of water is hung down from a 100-cm² tile. Our bonding strength thus might be still sufficient in most practical situations. For a referential bonding experiment in a nitrogen ambient furnace with an oxygen concentration of 0.8% at 700 °C, 0.1 MPa for 3 h, the resulted bonding strength was 2.0 MPa. The reduced oxygen ambient is thus thought to suppress the abovementioned corrosion of W.

Figure 3 presents typical current-voltage characteristics of the bonded W/Cu samples bonded at 0.1 MPa for various conditions. The electrical conduction property is observed to exhibit ohmic characteristics, represented by straight current-voltage lines, or Schottky barrier-like characteristics, represented by rectified current-voltage curves, depending on the bonding conditions. The electrical con-

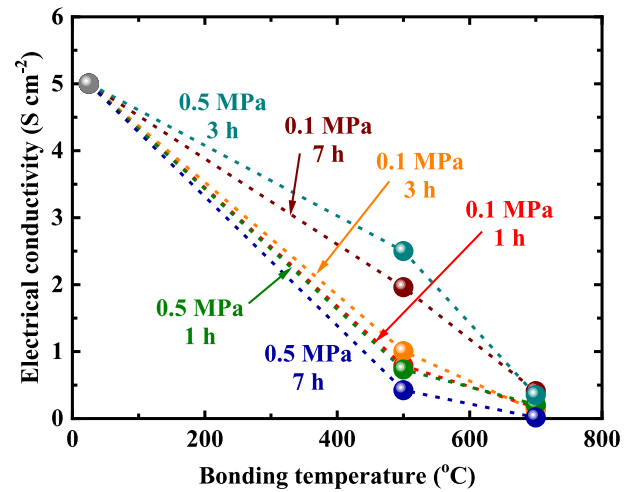


Fig. 4 Electrical conductivities of the W/Cu samples in dependence on bonding temperature for various conditions. The plot at room temperature is a referential data for a W/Cu sample not bonded but simply contacted with each other.

ductivity is generally observed to become lower for higher bonding temperature and longer bonding time. This trend is presumably because of the oxide formation at the surfaces and bonded interfaces of the metal plates. Here, we extract the representative electrical conductivity from the slope of the current-voltage curve at the origin (voltage = 0, current = 0), for each sample of bonding condition. Note that the current-voltage characteristics and thus the consecutive electrical resistivity is nevertheless for the whole bonded sample, including the bulk and surface parts of the metal plates, not solely for the bonded interface. The presented interfacial electrical conductivities are therefore not rigorously accurate but underestimated. Figure 4 presents the dependence of the electrical conductivities of the bonded W/Cu samples on the bonding temperature for various bonding conditions. As a reference, we additionally plotted the electrical conductivity data taken for a W/Cu sample not bonded but simply contacted with each other at room temperature. For bonding at higher temperatures, oxidation of the metal surfaces and bonded interface is thought to severely increase the electrical resistivity. As observed in the plots, the bonding temperature, rather than the bonding pressure or duration, dominantly influences the conductivity, presumably due again to the oxide formation in ambient air. Such a side effect has to be accounted for in practical applications, in view of thermal conductance.

To analyze the interfacial oxidation and atomic inter-diffusion between the W and Cu plates during the bonding process, x-ray photoelectron spectroscopy was taken for the W and Cu surface delaminated after the bonding experiments. Figure 5 presents the dependence of the elementary peak area of x-ray photoelectron spectroscopy on

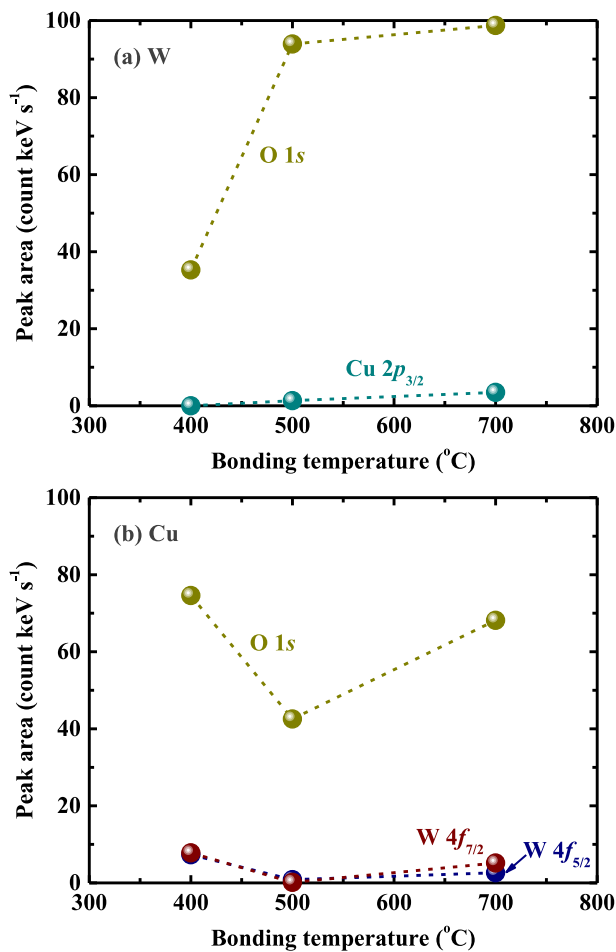


Fig. 5 Elementary peak area of x-ray photoelectron spectroscopy in dependence of bonding temperature for (a) W and (b) Cu surface delaminated after bonding at 0.1 MPa for 3 h.

the bonding temperature, measured for the samples bonded at 0.1 MPa for 3 h. On both of the W and Cu plates, the opponent elements are detected, and thus elementary interdiffusion of the metal atoms in the bonding process is observed. Oxidation of the bonded interfaces is also observed as the relatively high counts of the oxygen signals on both W and Cu. For the W side, the amount of interdiffusion and oxidation are observed larger for higher bonding temperatures. This trend for oxidation well explains the result of the electrical conductivity in Fig. 4. Whilst, these characteristics are seemingly not very sensitive to the bonding temperature for the Cu side.

Overall, although W/Cu bonds formed even at a temperature as low as 500°C and a bonding pressure as low as 0.1 MPa, the resulted interfacial mechanical strength is quite low relative to those obtained by the state-of-the-art techniques such as high-temperature brazing [7, 8]. While bonding in ambient air is practically convenient, the technical trade-off with the oxidation issue has to be accounted for, depending on the requirement of each situation. Although we carried out a quite simple bonding process in

the present study, technical additives, such as surface planarization, chemical surface pretreatments, and particulate control [16], may improve the bonding outcome towards the realization of further lower bonding temperature and higher interfacial mechanical strength.

4. Conclusions

In this study, we carried out a fundamental investigation of a uniaxial direct W-to-Cu bonding at relatively low temperatures in ambient air, which would potentially allow for simple preparation and maintenance of divertor wall components. W/Cu bonds formed at 500°C with a bonding pressure of 0.1 MPa, but the mechanical interfacial strength was about 1 MPa, significantly lower than the state-of-the-art values for bonding around at 1000°C in vacuum. Higher degree of interfacial oxidation and atomic interdiffusion were observed for higher bonding temperature, through x-ray photoelectron spectroscopy. However, accounting for our significantly lower bonding temperature and strength than those for Ref. [15], we cannot be assertive on the diffusion-dominant bonding mechanism at the present stage. The electrical conductivity across the bonded W/Cu interface, an indicator of thermal conductance, was measured to be lower for higher bonding temperature, presumably due to the interfacial oxidation. For bonding in ambient air at relatively high temperature, the technical trade-off with the oxidation issue thus has to be accounted for, depending on the requirement in each practical application.

The authors would like to thank Masayuki Tokitani, Hiroyuki Noto, and Takuya Nagasaka of the National Institute for Fusion Science for discussions. We also thank Miho Kageyama, Taisuke Maki, and Atsuhiko Inomata of Kyoto University for their support in the scanning electron microscopy, x-ray photoelectron spectroscopy, and oxygen concentration calibration, respectively. This study was financially supported, in part, by the National Institute for Fusion Science and the Japan Society for the Promotion of Science.

- [1] P. Rindt, S.Q. Korving, T.W. Morgan and N.J.L. Cardozo, Nucl. Fusion **61**, 066026 (2021).
- [2] T. Abrams, S. Bringuier, D.M. Thomas, G. Sinclair, S. Gonderman, L. Holland, D.L. Rudakov, R.S. Wilcox, E.A. Unterberg and F. Scotti, Nucl. Fusion **61**, 066005 (2021).
- [3] C. Stan-Sion, N. Bekris, G. Kizane, M. Enachescu, J. Likonen, M. Halitovs and A. Petre, Nucl. Fusion **56**, 046015 (2016).
- [4] H. Greuner, B. Boswirth, J. Boscary, P. Chaudhuri, J. Schlosser, T. Friedrich, A. Plankensteiner and R. Tivey, J. Nucl. Mater. **386**, 772 (2009).
- [5] C.S. Wiggins, A. Cabral and L.B. Carasik, Fusion Sci. Technol. **77**, 710 (2021).
- [6] K. Ezato, S. Suzuki, M. Dairaku and A. Akiba, Fusion Eng. Des. **75**, 313 (2005).
- [7] M. Tokitani, S. Masuzaki, Y. Hiraoka, H. Noto, H. Tamura, T. Tanaka, T. Muroga and A. Sagara, Plasma Fusion Res.

- 10**, 3405035 (2015).
- [8] M. Tokitani, Y. Hamaji, Y. Hiraoka, S. Masuzaki, H. Tamura, H. Noto, T. Tanaka, T. Tsuneyoshi, Y. Tsuji, T. Muroga and A. Sagara, *Nucl. Fusion* **61**, 046016 (2021).
- [9] N. Hara, S. Nogami, T. Nagasaka, A. Hasegawa, H. Tanigawa and T. Muroga, *Fusion Sci. Technol.* **56**, 318 (2009).
- [10] H. Fu, T. Nagasaka, T. Tanaka, A. Sagara, H. Serizawa and Y. Satou, *Fusion Sci. Technol.* **72**, 680 (2017).
- [11] S. Jiang, J. Shen, T. Nagasaka, T. Muroga, A. Sagara, S. Ohnuki, K. Hokamoto, S. Tanaka, D. Inao, Y. Morizono, R. Kasada and P. Zheng, *J. Nucl. Mater.* **539**, 152322 (2020).
- [12] X. Wu, S. Kondo, H. Yu, Y. Okuno, M. Ando, H. Kurotaki, S. Tanaka, K. Hakamoto, R. Ochiai, S. Konishi and R. Kasada, *Mater. Sci. Eng. A* **826**, 141995 (2021).
- [13] K. Yabuuchi, N. Tsuda, A. Kimura, Y. Morisada, H. Fujii, H. Serizawa, S. Nogami, A. Hasegawa and T. Nagasaka, *Mater. Sci. Eng. A* **595**, 291 (2014).
- [14] H. Serizawa, H. Ogura, Y. Morisada, H. Fujii, H. Mori and T. Nagasaka, *Nucl. Mater. Energy* **15**, 43 (2018).
- [15] J. Zhang, Y. Huang, Y. Liu and Z. Wang, *Mater. Des.* **137**, 473 (2018).
- [16] R. Inoue, N. Takehara, T. Naito and K. Tanabe, *ACS Appl. Electron. Mater.* **1**, 936 (2019).