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Effect of Cooling Rate on Brazing Cu with $Pd_{40}Cu_{30}Ni_{10}P_{20}$ Filler

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Abstract

Metallic glass has unique properties such as high mechanical strength and high corrosion resistance, and therefore many industrial applications are expected. In the present preliminary study, we demonstrate brazing of Cu with $Pd_{40}Cu_{30}Ni_{10}P_{20}$ metallic glass filler. The brazing was performed at 873K for 1 minute and then the specimen was quenched. The brazing was succeeded except several voids were observed in the filler. When the cooling rate was 60K/s, $Pd_{40}Cu_{30}Ni_{10}P_{20}$ filler retained amorphous. Dendrites, which grew from Cu surface to the $Pd_{40}Cu_{30}Ni_{10}P_{20}$ filler, was observed over the Cu/$Pd_{40}Cu_{30}Ni_{10}P_{20}$ interface because the glass forming ability near the interface was decreased due to elution of Cu. Tensile fracture strength reached 250 MPa. In contrast, when the cooling rate was 5K/s, the filler was partially crystallized. Tensile strength of that was less than 100 MPa.

Keywords: Amorphous materials, Brazing, Metastable phase, Pd-based alloy, Metallic glass.

1. Introduction

Metallic glasses have many unique properties, such as high mechanical strength, high elastic limit, low elastic coefficient, small solidification shrinkage and high corrosion resistance associated with the atomic structure. These unique properties that can be rarely found in crystalline materials are very attractive for practical applications. However, in spite of these attractive properties, the application field of the metallic glass has not been readily expanded. This attributed to the narrow process window which must be controlled to avoid a crystallization of metallic glass during manufacturing procedure. Bulk metallic glass is ordinarily made from a molten base alloy by the melt spinning method or casting method. Some types of metallic glass, such as Mg- (A. Inoue, K. Ohtera, K. Kita and T. Masumoto, 1988), Zr- (A. Inoue, T. Zhang and T. Masumoto, 1990 and A. Peker, W. L. Johnson, 1993), Fe- (A. Inoue and J. S. Gook, 1995 and A. Inoue, T. Zhang, T. Itoi and A. Takeuchi, 1997), Pd- (A. Inoue, N. Nishiyama and T. Matsuda, 1996) and Ni-based (R. Akatsuka, T. Zhang, M. Koshiba and A. Inoue, 1999) alloys, have a low critical cooling rate, R_c. Especially, $Pd_{42.5}Cu_{30}Ni_{7.5}P_{20}$ has an R_c of 0.067 K/s (N. Nishiyama and A. Inoue, 2002). Such high glass forming ability enables the production of bulk amorphous alloys that are several centimeters in thickness. Up to now, the joining of metallic glasses has been attempted by using electron beam welding, explosive welding, pulse current welding, friction welding (Y. Kawamura, T. Shoji and Y. Ohno, 2003 and Y. Kawamura and Y. Ohno , 2001a and Y. Kawamura and Y. Ohno, 2001b) and laser welding (J. Kim, D. Lee, S shin and C. Lee, 2006 and B. Li, Z. Y. Li, J. G. Xiong, L. Xing, D. Wang and Y. Li, 2006). These studies have focused on joining metallic glasses each other. But very few attempts have been made to join metallic glasses to crystalline metals. Especially little is known about the interface microstructure between metallic glass and crystalline metal. The purpose of this study is to demonstrate the possibility of brazing Cu with $Pd_{40}Cu_{30}Ni_{10}P_{20}$ metallic glass filler.

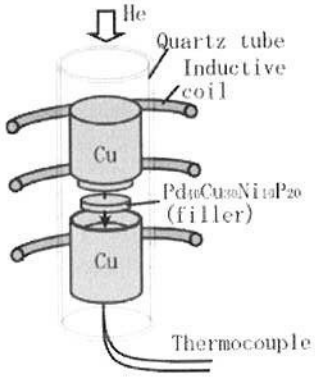


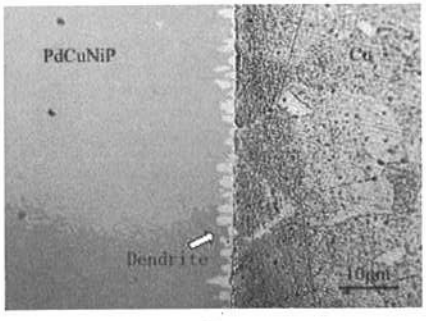
Fig.1 Schematic illustration of the apparatus used in this study.

2. Experimental

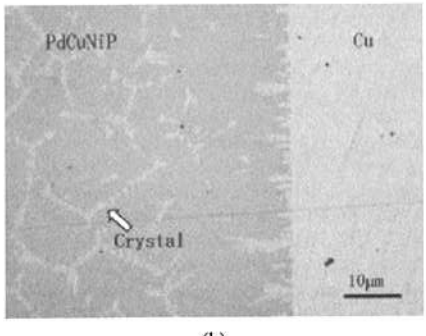
Two oxygen-free Cu rods (99.994%), ϕ 6 mm in diameter, were brazed with $Pd_{40}Cu_{30}Ni_{10}P_{20}$ in an inductive heating furnace. $Pd_{40}Cu_{30}Ni_{10}P_{20}$ filler was prepared by arc-melting a mixture of pre-alloyed PdP, Cu, Ni and Pd together with B_2O_3 flux under an Ar atmosphere. Flux treatment was repeated five times to remove impurities in the $Pd_{40}Cu_{30}Ni_{10}P_{20}$ alloy. The resulting alloy buttons were cast to form a metallic glass plate. The plate was cut into a disk shape ϕ5 mm in diameter and 1.5 mm in thickness. The glass transition temperature, crystallization temperature and melting temperature of the $Pd_{40}Cu_{30}Ni_{10}P_{20}$ were 574 K, 660 K and 808 K, respectively. A shallow hole, which was ϕ 5mm in diameter and 0.5 mm in depth, was machined at the center of the Cu surface as shown in **Fig.1**. $Pd_{40}Cu_{30}Ni_{10}P_{20}$ filler was set in this hole and joined to the other Cu rod with a flat surface. The brazing was carried out at 873 K for 1 minute in He and then the specimen was quenched at the rate of 60 K/s and 5K/s by blowing He. Tensile testing was carried out at a strain rate of 1×10^{-4}/s. Micro-focused x-ray diffraction (XRD) using Co Kα radiation with a spot size of ϕ50 μm in diameter was used to identify the crystallinity of the metallic glass filler. A cross section of the joint was polished and etched with 50% HNO_3 aq. at room temperature. The microstructure was analyzed by the optical microscope and energy dispersive X-ray (EDX) spectroscopy.

3. Results and discussion

The Cu specimens were successfully brazed with the $Pd_{40}Cu_{30}Ni_{10}P_{20}$ filler with thickness of 0.5 mm. Pressure was not applied to the metallic glass filler during brazing, and several voids were observed in the filler. **Fig. 2**(a) and (b) show optical micrographs of the cross section of the joint at the interface between Cu and $Pd_{40}Cu_{30}Ni_{10}P_{20}$ quenched at the rate of 60K/s and 5K/s, respectively. The filler quenched at a rate of 60K/s showed no contrast but the filler quenched at a rate of 5K/s showed partially clear contrast of crystal phases. Though the critical cooling rate of $Pd_{40}Cu_{30}Ni_{10}P_{20}$ is 0.1K/s, the stability of the supercooling state is sensitively affected by contamination from atmosphere and base material and so on. So the filler seemed to be crystallized. **Fig. 3** shows the micro-focused XRD patterns taken from the center of the cross section of $Pd_{40}Cu_{30}Ni_{10}P_{20}$ filler. A broad peak around 2θ=49°, which is characteristic of the amorphous, was observed in the filler quenched at the rate of 60K/s. The smallest six compounds such as the monoclinic structure, tetragonal Cu_3Pd-like structure, rhombohedral $Pd_{15}P_2$-like structure, Ni_2Pd_2P intermetallic compound, fcc-(Ni, Pd) solid solution and bct-Ni_3P-



(a)



(b)

Fig.2 Microstructures of the Cu/$Pd_{40}Cu_{30}Ni_{10}P_{20}$ interface. Cooling rates are (a) 60K/s and (b) 5K/s.

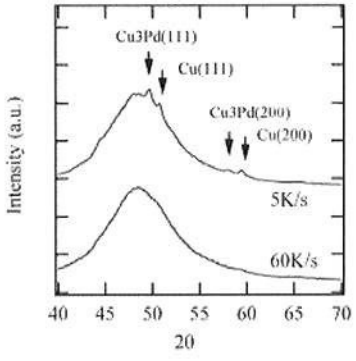


Fig.3 Micro-focused X-ray diffraction patterns taken from the center of the cross section of $Pd_{40}Cu_{30}Ni_{10}P_{20}$ filler quenched at the rate of 60K/s and 5K/s.

like structure, are known as crystal phases of $Pd_{40}Cu_{30}Ni_{10}P_{20}$ (A. Inoue and N. Nishiyama, 1997 and I. R. Lu, G. Wilde b, G.P. Gorler, R. Willnecker, 1999). The results showed that $Pd_{40}Cu_{30}Ni_{10}P_{20}$ filler exhibits amorphous and joined to Cu because the cooling rate was considerably faster than the critical cooling rate of the glass formation (R_c=0.1 K/s). In contrast, at 2θ=50° and 57°, characteristic of Cu_3Pd (111) and (200), were observed in the filler quenched at the rate of 5K/s. Furthermore, dendrites approximately 5 μm in length and 2 μm in width were observed. The elemental composition of the

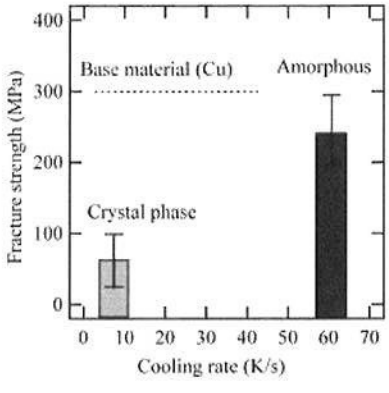


Fig.4 Tensile fracture strength of the Cu specimens brazed with $Pd_{40}Cu_{30}Ni_{10}P_{20}$ filler.

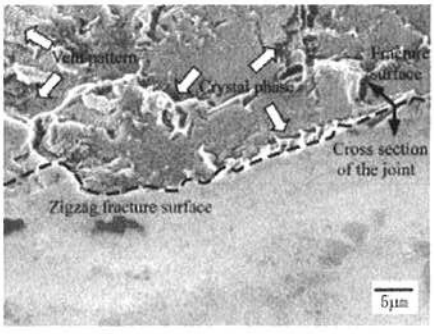


Fig.5 Fracture surface of the $Pd_{40}Cu_{30}Ni_{10}P_{20}$ filler quenched at the rate of 5K/s. (Tilte 45°)

filler analyzed by EDX was almost the same as that before brazing, but the Cu content in the dendrites was about 80%. It seemed that Cu eluted into $Pd_{40}Cu_{30}Ni_{10}P_{20}$ filler during brazing. When Cu exceeds more than 80% in the Pd-Cu-Ni-P alloy, the glass forming ability is greatly decreased compared to that of $Pd_{40}Cu_{30}Ni_{10}P_{20}$. Therefore, dendrites seemed to grow at the Cu/$Pd_{40}Cu_{30}Ni_{10}P_{20}$ interface during quenching. **Fig. 4** shows the tensile fracture strength of the brazed specimens. The tensile tests were repeated four times at the same conditions. Almost all of specimens were fractured in the filler. Tensile fracture strength of the specimen quenched at the rate of 60K/s reached 250 MPa, but that quenched at the rate of 5K/s was less than 100MPa. Crystal phases are generally composed of brittle intermetallic compounds, so that the mechanical strength of crystallized $Pd_{40}Cu_{30}Ni_{10}P_{20}$ filler was less than that of the amorphous one. SEM observation of the tensile fracture surface of the specimen quenched at the rate of 5K/s is shown in **Fig.5**. It is noted that there are two kinds of traces in the fractured surfaces; the zigzag surface and the vein-like pattern. It is known that the typical fracture surface of metallic glass shows a vein pattern because metallic glass fractures by transforming strain energy to thermal energy and simultaneously forming a pseudo-molten state. On the other hand, the precipitated crystals were seen on the zigzag fracture surfaces. That indicates that fracture took place in the crystals, and then crack propagated along the crystal and amorphous phases alternately.

4. Conclusion

Metallic glass brazing of Cu using $Pd_{40}Cu_{30}Ni_{10}P_{20}$ filler was successful. When the specimen was quenched at the rate of 60K/s, the

metallic glass filler was retained amorphous and was jointed to Cu. The tensile fracture strength reached 250 MPa. On the other hand, when the specimen was quenched at the rate of 5K/s, the metallic glass filler was partially crystallized. The tensile strength was less than 100 MPa. The fracture surface showed two kinds of traces; the vein pattern and the zigzag pattern on which the precipitated crystals were seen. That indicates that fracture took place in the crystals, and then crack propagated along the crystal and amorphous phases alternately.

Acknowledgments

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