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# Cu-based metallic glass surfacemodified with Cu for soldering[†]

—Productions and applications of Cu clad metallic glass—

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**KEY WORDS**: (Cu-based metallic glass) (Clad material) (Soldering) (Wettability) (Quench) (passive oxide film) (Surface modification)

## 1. Introduction

It is necessary to develop joining techniques for metallic glasses because they are required to build in to manufactured products. One of the serious problems with welding metallic glass is crystallization in the molten zone. If the cooling rate from the liquidus temperature to the glass transition temperature is slower than the critical value, metallic glass rapidly transforms into intermetallic compounds. Formation of the intermetallic compounds is not preferred because most of them are brittle and of low weld strength compared. Recently, there have been some attempts to join pieces of Zr-based metallic glass using electron beam welding, explosive welding, pulse current welding, friction welding [1-3], laser welding [4-7] and ultrasonic bonding [8].However, weldability is strictly limited by the specimen size, the glass forming ability and the heat source.

Soldering is one of candidate techniques for joining metallic glass. Soldering can be processed approximately below 600K, which is far below the transition temperatures of metallic glasses. The low processing temperature doesn't induce a structural relaxation of atomic order and thus enables joining without any crystallization. Furthermore, soldering enables us to join metallic glass with dissimilar materials such as crystalline alloys, and surface-metalized ceramics.

However, a serious problem in soldering metallic glass is poor wettability. Although Pd-based metallic glass exhibits a good wettability to Pb free solder, Zr-Cu based metallic glasses doesn't wet at all because of a strong and homogeneous passive oxide film, which mainly consists of $ZrO_x$, and protects the surface. Some attempts have already been reported to remove the passive oxide film. A. Imai et al. removed the surface oxide film of the $Cu_{60}Zr_{30}Ti_{10}$ metallic glass by Ar ion beam, and then deposited Ag thin layer on it in an ultra high vacuum environment [9]. The Ag-deposited $Cu_{60}Zr_{30}Ti_{10}$ surface showed a resistivity to the reoxidation in air, and also good wettability to Sn-Ag-Cu solder. S. Tamura et al. studied wetting behavior of Zr-based metallic glass in a Sn-Cu-Ni solder bath by ultrasonic cavitation in air [10]. They found that the surface morphology of the metallic glass was roughed when cavitation bubbles collapsed on the surface, and this phenomenon promoted the wetting between the metallic glass and the solder. The application of ultrasonic cavitation is useful for the immediate removal of passive

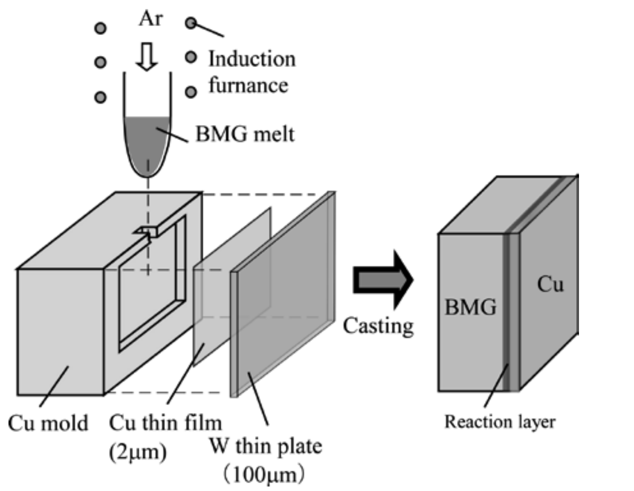


**Fig. 1** Production of Cu thin film cladding bulk metallic glass (BMG).

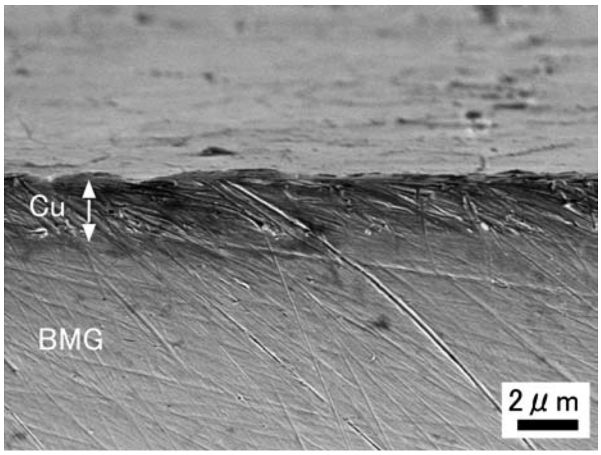


**Fig. 2** Cross-section image of Cu-cladding bulk metallic glass $Cu_{36}Zr_{48}Al_8Ag_8$.

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oxide film on the metallic glass. However, the removable area was limited around the ultrasonic-transducer.

In this paper, we report a development of Cu thin film clad Cu-based metallic glass $Cu_{36}Zr_{48}Al_8Ag_8$ for the aim of improving solder wetting. By cladding Cu thin film over the Cu-based metallic glass surface, the formation of passive surface oxide film was intercepted. Microstructure at the $Cu/Cu_{36}Zr_{48}Al_8Ag_8$ interface and the solder wettability were discussed.

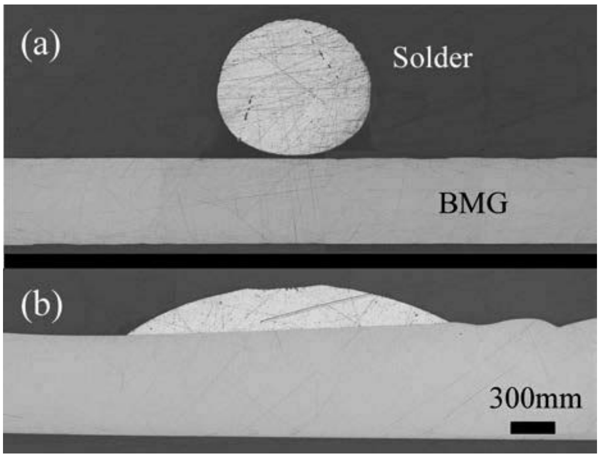


**Fig. 3** Wettability of Sn-3Ag-0.5Cu solder on (a) monolithic metallic glass and (b) Cu cladding metallic glass.

## 2. Experimental

A pre-alloy of $Cu_{36}Zr_{48}Al_8Ag_8$ (in atom%) was prepared by arc-melting a mixture of pure Cu (99.99%), Zr (99.99%), Al (99.99%) and Ag (99.999%) metals in an oxygen-gettered argon atmosphere. **Figure 1** shows schematic illustration of the procedure to produce Cu thin film clad Cu-based metallic glass. A rectangular hollow with dimension of $^{H}25x^{W}25x^{D}2$mm was machined on the side of a Cu mold. The rectangular hollow was covered with a tungsten thin plate with thickness of 100 μm, on which Cu thin film with thickness of 2 μm was adhered. The pre-alloy ($Cu_{36}Zr_{48}Al_8Ag_8$) was melted in a quartz glass nozzle at 1473 K by induction heating, and then the melt was cast into the gap of the hollow. Just after casting, the Cu thin film was semi-molten by a residual heat of the melt and was welded to it. Then the melt and Cu thin film were quenched together by the Cu mold. As a result of the process, Cu thin film was clad on a surface of $Cu_{36}Zr_{48}Al_8Ag_8$ metallic glass..

The thermal stability associated with the glass transition and crystallization was analyzed using a differential scanning calorimeter (DSC) at a heating rate of 20K/s. The phase identification was conducted by micro-focused X-ray diffraction (XRD) analysis using Co Kα radiation. The microstructure of the interface between Cu thin film and metallic glass was observed with an optical microscope (OM), a scanning electron microscope (SEM), and a transmission electron microscope (TEM). The Surface oxide layer was analyzed by auger electron spectroscopy (AES). Solder wettability was evaluated by dipping Sn-3Ag-0.5Cu solder on the Cu thin film clad metallic glass at 553K for 60 s.

## 3. Results and Discussion

Cu thin film with thickness of 2 μm was successfully welded to amorphous $Cu_{36}Zr_{48}Al_8Ag_8$. The cross-section image is shown in **Fig. 2**. From microstructure analysis, it was found that reaction layer such as $Cu_5Zr$ and $Cu_{10}Zr_7$ was formed at the interface between Cu and $Cu_{36}Zr_{48}Al_8Ag_8$. AES results show that the formation of a surface oxide layer was suppressed by the cladding Cu thin film.

Wettability of Sn-3Ag-0.5Cu solder on (a) monolithic metallic glass and (b) Cu cladding metallic glass are shown in **Fig. 3**. Solder was not wet on monolithic metallic glass at all, however, was wet on Cu thin film clad metallic glass. The improvement of the solder wetting originated from interception of the oxide film formation.

## 4. Conclusions

Amorphous $Cu_{36}Zr_{48}Al_8Ag_8$ was successfully clad by Cu thin film. Solder wetting was drastically improved by cladding Cu thin film because formation of a native oxide film was intercepted by the Cu layer.

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