

## Article

# A Novel Technique for Controllable Fabrication of Multilayer Copper/Brass Block

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**Abstract:** Fabricating a dissimilar-metal block with micro/nano-multilayered structures is usually used by engineers and scientists because of their excellent mechanical properties. In the current work, multilayered copper/brass blocks were effectively fabricated by a synthetical DWFR technique, which includes the processes of diffusion welding, forging and rolling. Diffusion welding was used as the first operation to metallurgically bond the copper and brass sheets, with a Zn diffusion transition layer (thickness of ~100 μm), which can guarantee the bonding strength of copper/brass interfaces during the subsequent forging and rolling processes. After diffusion welding, the original copper/brass blocks were required to be forged, with its total thickness reduced to ~10 mm. This can further restrain the delamination of copper and brass layers during the final rolling process. Rolling was utilized as the ideal operation that can precisely tune the thickness of copper/brass laminate. This novel DWFR technique can easily tune the multilayered copper/brass blocks with controllable layer thickness (from ~250 to ~800 nm). The copper/brass interfaces were well-bonded, and the utilization efficiency of raw materials was very high (>95%).

**Keywords:** diffusion welding; forging; rolling; copper/brass block; layer thickness



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## 1. Introduction

Multilayered dissimilar-metal blocks have always attracted attention due to their superior mechanical properties, such as high strength, good ductility and excellent impact toughness [1–5]. As investigated in many previous studies, the multilayered dissimilar-metal blocks can be prepared using the techniques of physical vapour deposition (PVD) [6,7], high pressure torsion (HPT) [8], accumulative rolling bonding (ARB) [9,10] and diffusion welding + rolling (DWR) [2,4,11], etc. Generally, all of the above-mentioned techniques have their weaknesses. The PVD technique can precisely fabricate the multilayered dissimilar-metal blocks with nanolayers, although its low production efficiency may limit its application for the preparation of large-size products. Similarly, it was also difficult for HPT technique to fabricate materials large enough for industrial applications because of the small size of raw materials required for HPT. The shape of the raw material of HPT is usually a disk and the diameter of the disk is determined by the dimension of the extrusion die, which is usually less than 20 mm [12]. The ARB technique was believed to be useful and highly efficient for the preparation of a multilayered dissimilar-metal block with a large size sample and a controllable micro/nano-thickness of each layer. However, some

concerns have arisen related to the low utilization efficiency of raw materials and low deformation bonding strength, which often result from the inevitable micro-cracks, oxygen and contaminants during repeated ARB operations [9,10,13]. Recently, Li et al. [2,4] indicated that multilayered dissimilar-metal blocks can be constructed using the DWR technique. The DWR-processed samples were of large sizes, well-bonded transition interfaces and demonstrated a high utilization efficiency of raw materials (~100%). Unfortunately, it was difficult to decrease the layer thickness to the value of <math><100\ \mu\text{m}</math>, which was ascribed to the delamination of the multilayered block with a larger original thickness during the rolling operation. As reported by Ma et al. [14] and Huang et al. [3], a higher strength and ductility were more likely to be obtained for multilayered blocks with layer thicknesses of several micrometers or nanometers. This was also reported for other materials, for which preparing nanostructures can usually achieve superior physical and chemical properties [15–20]. Hence, efforts should be made to further decrease the final layer thicknesses for dissimilar-metal blocks for DWR technique. There is also an urgent need to identify an appropriate processing technique that can fabricate multilayered dissimilar-metal blocks with the large production sizes, well-bonded interfaces, a high utilization efficiency of raw materials and a controllable layer thickness of the micro/nanometers.

Copper/brass blocks with micro/nano-multilayered structures are expected to have a high strength, according to previous literatures [3,4], thereby meeting the requirements for the structural components. In addition, pure Cu and Cu alloys had good thermal/electrical conductivities [21–24]. Thus, a copper/brass block can be used as a structural material in the field of thermal/electrical conductivity applications.

In this study, a combined processing technique of diffusion welding, forging and rolling (DWFR technique) is reported for the first time. Multilayered copper/brass blocks were effectively fabricated using the DWFR technique. This DWFR technique may provide technical guidance for fabricating other multilayered dissimilar-metal blocks.

## 2. Experimental Procedure

### 2.1. Materials and DWFR/ARB Processing

Commercial brass (Cu-30 wt.% Zn, ASTM-C26000, Tongling Nonferrous Metals Group Holdings Co., Ltd, Tongling, China) and pure copper (99.9 wt.% Cu, ASTM-C11000, Tongling Nonferrous Metals Group Holdings Co., Ltd, Tongling, China) sheets were used to fabricate the multilayered copper/brass block. Before the DWFR fabrication process, the copper sheets and brass sheets with an original size of 80 mm  $\times$  100 mm  $\times$  1/0.5 mm and 80 mm  $\times$  100 mm  $\times$  0.8/0.4 mm were mechanically polished by 1500# SiC paper (the grit size of ~10  $\mu\text{m}$ ), and then washed in an acetone solution for 15 min, to achieve the clean surfaces. The fabrication processing procedure that was followed is detailed in Figure 1. It includes the following four steps: (I) 20/50 pieces of copper sheets and 20/50 pieces of brass sheets are alternately stacked with total thickness of ~36 (40 layers) or ~45 mm (100 layers); (II) the diffusion welding technique is used to obtain an original multilayered copper/brass block with well-bonded copper/brass interfaces. The static pressure, heating temperature and holding time are 2 MPa, 920  $^{\circ}\text{C}$  and 2 h, respectively; (III) after diffusion welding treatment, the origin copper/brass laminate suffers forging deformation with its thickness reduced from ~36/~45 to ~10 mm at room temperature; (IV) synchronous rolling treatment is further utilized to tune the layer thickness of the copper/brass laminate at room temperature. The roll dimension is  $\Phi$ 120 mm  $\times$  250 mm, the velocity of roll is ~65 mm/s, and the thickness reduction is ~0.2 mm per pass at room temperature.

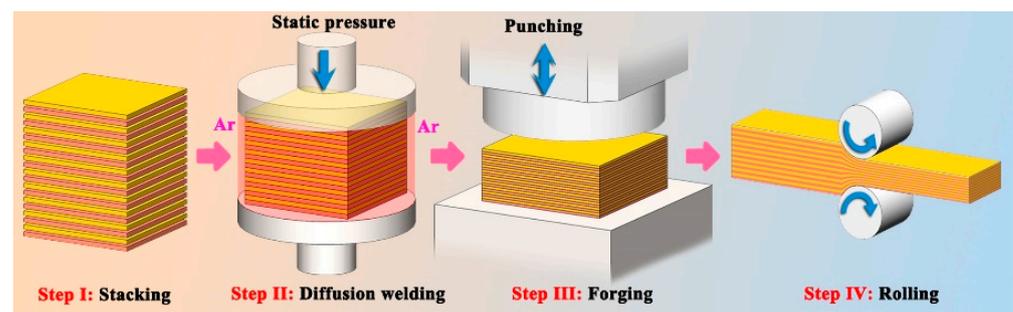


Figure 1. Schematic illustration of the procedure for the DWFR processing.

Before ARB fabrication processing, the copper sheets and brass sheets with the original size of  $80 \text{ mm} \times 25 \text{ mm} \times 1 \text{ mm}$  and  $80 \text{ mm} \times 25 \text{ mm} \times 0.8 \text{ mm}$  were also mechanically polished by 1500# SiC paper, and then washed in an acetone solution for 15 min, to achieve the clean surfaces. The roll dimension is  $\Phi 120 \text{ mm} \times 250 \text{ mm}$ , the velocity of roll is  $\sim 65 \text{ mm/s}$ , and the rolling strain is 50% per pass at room temperature.

## 2.2. Calculation of the Utilization Efficiency of Raw Materials

The utilization efficiency ( $\eta$ ) of raw materials in the present work were calculated using the following equation:

$$\eta = 1 - (m_1 + m_2 + m_3) / m_0 \quad (1)$$

where the  $m_0$  was the total mass of as-received copper and brass sheets,  $m_1$ ,  $m_2$  and  $m_3$  were the loss masses during the diffusion, forging and rolling processes, usually caused by cutting, cracking and delamination, etc.

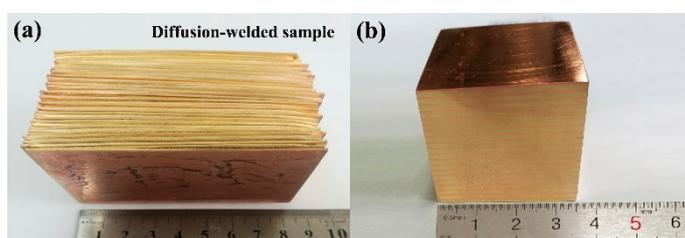
## 2.3. Characterizations

An optical microscope (OM, Olympus BX41M, OLYMPUS, Tokyo, Japan) was utilized to characterize the cross-sectional structure of the multilayered copper/brass block. The chemical element distribution and morphologies of the copper/brass interfaces were captured by a field emission scanning electron microscope (SEM, Quant 250 FEG, FEI, Hillsboro, OR, USA) equipped with X-ray energy dispersive spectroscopy (EDS, Oxford, Abingdon-on-Thames, Britain).

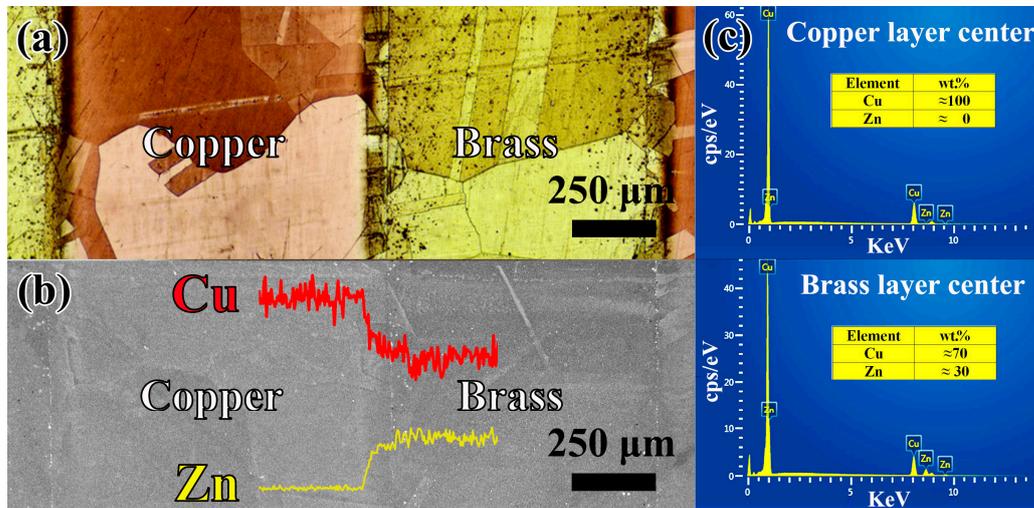
## 3. Results and Discussion

The diffusion welding technique is an effectively method that can connect dissimilar metals through the metallurgical bonding of the dissimilar-metal surfaces. The pressure is usually 2 MPa and the high processing temperature is  $920 \text{ }^\circ\text{C}$ . Figure 2 shows the multilayered copper/brass block prepared using a diffusion welding technique with 40 layers. The copper and brass sheets are successfully bonded to form a block. As presented in Figure 3a, both the copper layer and brass layer consist of an equiaxed coarse-grained structure embedded with some  $\Sigma 3$  twins. This is a typical annealed structure for pure copper and its alloys [3,4,25]. Figure 3b,c indicate that the copper layer and brass layer in the as-diffusion-welded copper/brass block are bonded with a Zn diffusion transition layer (thickness of  $\sim 100 \text{ }\mu\text{m}$ ) while the centers of copper and brass layer still show the original chemical compositions. The diffusion activity of Zn can improve the bonding strength and impede delamination of the copper/brass interface during the deformation process [4]. Thus, the Zn diffusion transition layer present in the copper/brass block is believed to guarantee the bonding strength during subsequent deformation operations. In order to further decrease the layer thickness and achieve a layer thickness of several micrometers or nanometers, an appropriate deformation technique should be used. In fact, rolling is hardly ever competent. This is attributed to the fact that high shear stress and strain are easily caused by the large thickness of the total copper/brass block during

the rolling operation [26,27]. In the current work, the 11-layered copper/brass block (total thickness of ~10 mm) and 18-layered block (total thickness of ~16 mm) have been cut from the original 40-layered diffusion-welded block. As displayed in Figure 4, a larger original thickness is more likely to result in the delamination of the copper/brass interfaces during the rolling process, which may indicate that ~10 mm may approach the critical value of thickness that restrains the delamination of the copper/brass interfaces. In order to obtain a layer thickness of several micrometers or nanometers, the original copper/brass block should firstly, be deformed with the thickness reduced to ~10 mm using an appropriate deformation operation. As analyzed for the stress state of uniaxial forging deformation, only the vertical punch load is generated by the hammer, which may cause a low shear stress between dissimilar layers and effectively shape metals without cracking. Thus, conventional forging is utilized as the first deformation operation to punch the copper/brass laminate, with the total thickness reduced from ~36 to ~10 mm.



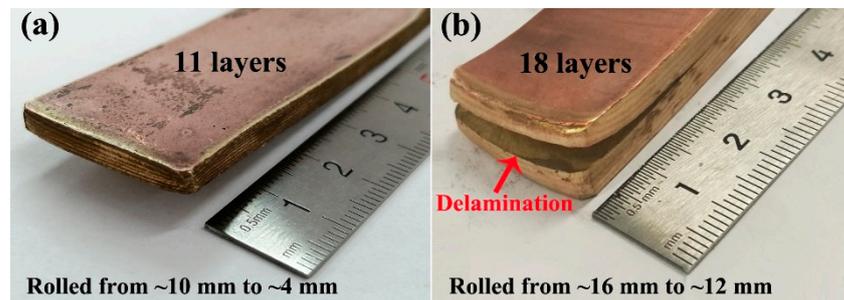
**Figure 2.** (a) The optical image of the diffusion-welded sample with 40 layers; (b) The optical image of the diffusion-welded sample with the dimension of 35 mm × 35 mm × 36 mm, which is cut from the diffusion-weld sample in (a).



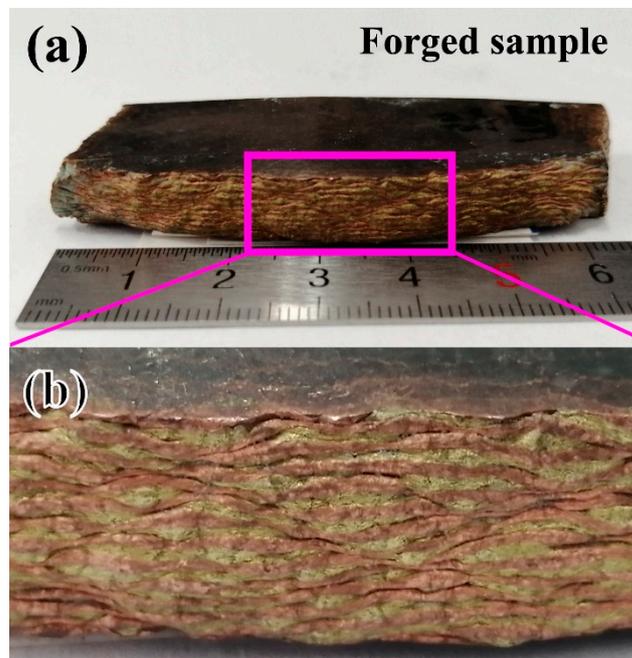
**Figure 3.** (a) Cross-sectional microstructures of the copper and brass layer after diffusion welding; (b) EDS line maps around copper/brass interface after diffusion welding; (c) EDS results of the copper layer center and brass layer center after diffusion welding.

As presented in Figure 5, no micro-crack or delamination is observed between the copper/brass interfaces for the forged sample, which can guarantee the high utilization efficiency of raw materials. Following the forging operation, rolling is selected as the second deformation operation to tune the thickness of copper/brass laminate because of its higher precision and higher surface flatness [28,29]. In the current work, multilayered copper/brass blocks with various layer thicknesses (250, 100, 50 and 5 μm) are effectively fabricated by our DWFR technique, clearly displayed in Figure 6. It is noted that no macroscopic delamination (well-bonded interfaces) or waste of the deformed copper/brass blocks during rolling operations occurred. The present DWFR technique is superior to the ARB

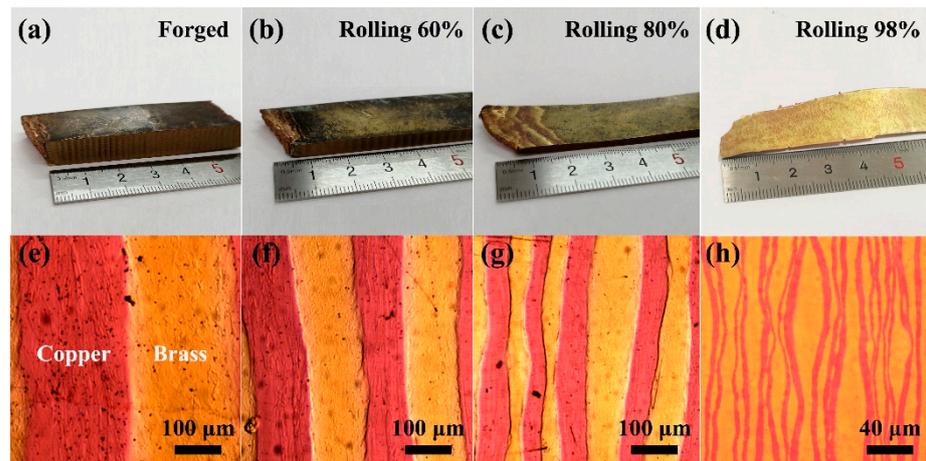
technique for processing copper and brass sheets with similar sizes at room temperature, which is confirmed by Figures 6 and 7, illustrating the occurrence of delamination during ARB processing. Some efforts are devoted to further reduce the thickness value of copper and brass layer down to  $<1000$  nm. Figure 8 exhibits the DWFR-processed sample with an average layer thickness of  $\sim 800$  nm and a total thickness of  $\sim 80$   $\mu\text{m}$ . We calculated that the utilization efficiency of raw materials for DWFR-processed copper/brass blocks is more than 95%, which is comparable to that of the DWR technique [4]. Although a nano-thickness layer of  $<100$  nm is not exhibited in the present work, it is believed that it can be easily obtained by increasing the number of original copper/brass sheets or reducing the final thickness of the rolled sample. In fact, this study provides only a brief overview of the DWRF technique, and the authors will conduct further investigations using different kinds of metals.



**Figure 4.** (a) The optical image of the 11-layered diffusion-welded sample with rolling strain of  $\sim 60\%$  (thickness reduced from  $\sim 10$  to  $\sim 4$  mm); (b) The optical image of the 18-layered diffusion-welded sample with rolling strain of  $\sim 25\%$  (thickness reduced from  $\sim 16$  to  $\sim 12$  mm). It is noted that the mentioned samples are both cut from the original diffusion-welded sample (40-layers).



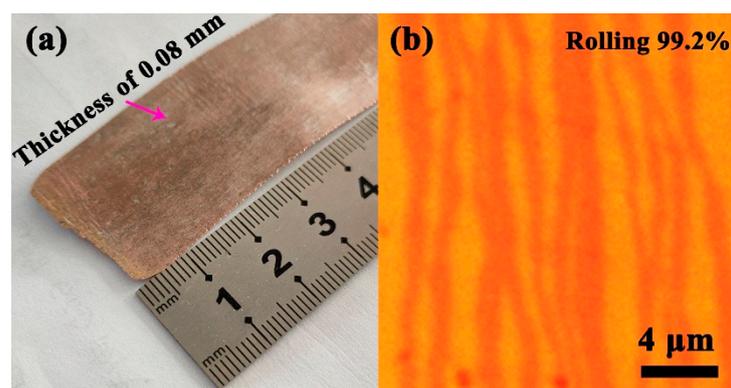
**Figure 5.** (a) The optical image of the forged sample with 40 layers; (b) The enlarged image of the cross-section of the forged sample.



**Figure 6.** (a–d) are optical images of the forged sample, rolling 60% sample, rolling 80% sample, rolling 98% sample (40 layers), respectively; (e–h) are the corresponding microstructures of cross-sections of the forged sample, rolling 60% sample, rolling 80% sample, rolling 98% sample, respectively.



**Figure 7.** The optical image of the ARB-processed copper/brass block. The original thicknesses of copper sheet and brass sheet are 1 and 0.8 mm, respectively.



**Figure 8.** (a) is the optical image of the DWFR-processed sample (100 layers); (b) is the corresponding microstructures of (a).

#### 4. Conclusions

In summary, the present work first reported a synthetic DWFR technique, including the process of diffusion welding, forging and rolling, which can effectively achieve the fabrication of multilayered copper/brass blocks. Some conclusions can be drawn, as follows:

- The DWFR technique can easily tune the multilayered copper/brass block with a controllable layer thickness, from ~250  $\mu\text{m}$  to ~800 nm.
- Well-bonded transition interfaces can be obtained via a diffusion welding treatment, which can suppress the delamination between copper and brass layers during severe deformation, caused by forging and rolling operations.
- DWFR and ARB products are of similar sizes, and can meet the size requirements for industrial products.
- The DWFR technique has a high utilization efficiency of raw materials (>95%), which greatly reduces the costs of raw materials.
- This DWFR technique also provides technical guidance for fabricating other multilayered dissimilar-metal blocks.

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