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Formation of intermetallic compounds of Cu/Al multilayer foils during cold rolling

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In metallurgical theory, the generation of new phases requires high temperature diffusion over a long period of time. Here, we report an experimental study of the generation of new phases using large mechanical deformations and high pressure, rather than heat. A 4.83-mm-thick 23-layered Cu/Al sandwich strip was formed by cold rolling. When the Cu/Al sandwich was further rolled to a 0.13-mm-thick foil on a four-high micro-foil mini mill at room temperature, some rectangular-shaped phases appeared at the Cu/Al interfaces. The scanning electron microscopy, transmission electron microscopy, and X-ray diffraction analyses reveal that the phases are a mixture of Al_2Cu/AlCu/Al_4Cu_9, which indicates that chemical reactions occur at the Cu/Al interfaces during cold deformation. This study provides new insights into the design and development of composite materials for various applications.

INTRODUCTION

The microstructure and properties of metals can be changed by the application of heat and/or force. Though the role of thermal activation in diffusion and phase transformation has been established, the function of force has only recently been investigated as a useful method for controlling the microstructure and properties of metal materials [1,2]. Severe plastic deformation processes based on the force effect theory, such as equal-channel angular extrusion/pressing (ECAE/P) [3–5], high-pressure torsion [6,7], and accumulative roll bonding (ARB) [8,9], have been rapidly developed. Initially, the processes were only applied to mono-materials, such as steel [10,11], aluminum [12], copper [13], nickel [14], and titanium [15]. However, these processes have recently been used to fabricate multi-layered composites with dissimilar metals, such as Al/Mg [16–19], Al/Zn [20], Al/Steel [21], Al/Ni [22], Cu/Nb [23,24], Cu/Ni [25], Ti/Ni [26], Al/Ni/Cu [27], and Ti/Al/Nb [28].

After the ARB process was successfully applied to obtain a super fine grain, multi-layered dissimilar metal composites are observed. Although a significant reduction in the thickness can be achieved through ARB, work hardening may prevent severe plastic deformation. Therefore, understanding whether chemical reactions can occur and new phases can be formed during producing multi-layered composites by severe plastic deformation is important. However, these behaviors are not well understood. Accordingly, the occurrence of chemical reactions in heavily strained dissimilar metals at room temperature must be demonstrated.

We want to determine the behavior of a laminated-metal composite when pressure is applied. A four-high micro-foil mini mill (3M mill) was previously developed to roll extremely thin strips [29]. Using the 3M mill, a 0.13-mm-thick Cu/Al composite foil was obtained at room temperature without annealing. Surprisingly, some new phases were observed at the interfaces of the roll-bonded Cu/Al composite foil. Transmission electron microscopy/selected area eltrecon diffraction (TEM/SAED) and scanning electron microscopy/energy-dispersive X-ray spectrometry (SEM/EDS) reveal that the new phases are composed of Al_2Cu/AlCu/Al_4Cu_9. Thus, room-temperature chemical reactions conclusively occur under force effects.

EXPERIMENTAL SECTION

Special rolling process

Pure copper T2 (99.8% purity) with size 100 mm×30 mm×0.21 mm (length×width×thickness), and pure aluminum A1100 (99.7% purity) with size 100 mm×30 mm×0.20 mm were selected as initial materials. To obtain good bonding between the metal layers, aluminum and copper sheets were annealed at 500°C and 600°C for 1 h, respectively, and were polished using a stainless steel brush to remove surface oxidation films. After ultrasonic washing in acetone and hot air drying, 23 layers were alternately stacked (11 layers Al and 12 layers Cu). The sample was cold rolled at room temperature using six passes. The first two passes were performed on a two-high mill with a roll diameter

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of 180 mm and rolling speed of 0.224 m s^{-1}. The remaining four passes were performed on a 3M mill (see Fig. S1) with a working roll diameter of 50 mm and rolling speed of 0.04 m s^{-1}. Finally, a 0.13-mm-thick Cu/Al composite foil sample was obtained. We emphasize that no intermediate annealing occurs during the rolling processes. The rolling schedule is shown in Table S1.

Specimen preparation for microstructure observation

After rolling, the metallographic samples were prepared by mechanical grinding and polishing. The microstructure of the longitudinal section of the Cu/Al composite foil (without etching) was observed with an Olympus GX51 optical microscope. The phase composition and microstructure of the samples were investigated with SEM equipped with EDS, and were further confirmed by TEM/SAED and TEM/EDS.

X-ray diffraction (XRD)

Phase identification was performed using XRD (D/MAX-RC) with rotating copper anode (CuKα) radiation (wavelength 0.15405 nm). The X-ray beam size on the specimen is about 0.185 mm, and the scattered radiation is registered by four different position-sensitive imaging detectors located at 300–600 mm from the base material. The scattered radiation covers about 2θ = 10–100° of the angular range of diffractions.

RESULTS

Cu/Al composite strip and its structural evolution

Fig. 1a shows the picture of the primary sandwich, which is 0.9 mm in thickness after two roll-bonding passes, and Fig. 1b shows the macrograph of the Cu/Al composite strip, which is 0.13 mm in thickness after six passes. As shown in Fig. 2a, it can be observed in the longitudinal section that the Cu and Al layers are flat and continuous and are nearly parallel to each other within the Cu/Al laminated strip. In addition, the Cu/Al interfaces are well bonded, and defects such as holes or gaps at the joints are not observed at the interfaces, which indicates that two passes of rolling with heavy reduction (ε_1 = 55.07%; ε_2 = 58.53%) result in good mechanical bonding between the Cu and Al layers.

After the primary sandwich was torn along the bonding

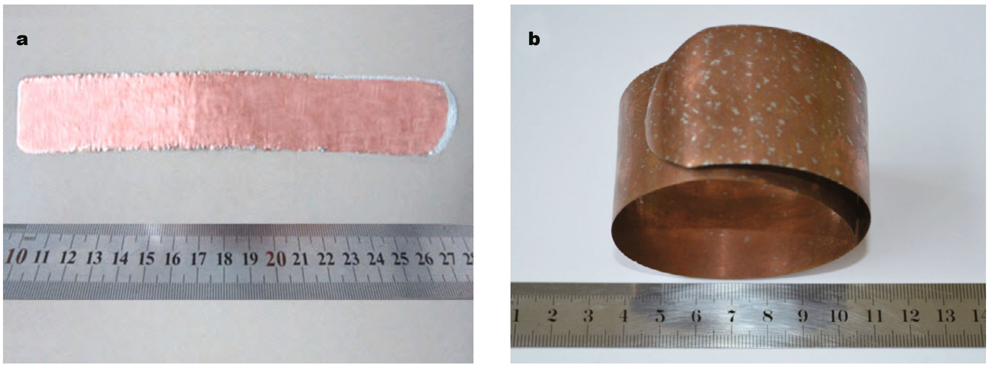

Figure 1 Macrographs of the 23-layered Cu/Al composite strip at different passes: (a) primary sandwich of the Cu/Al strip after two passes and (b) the Cu/Al composite strip after six passes.

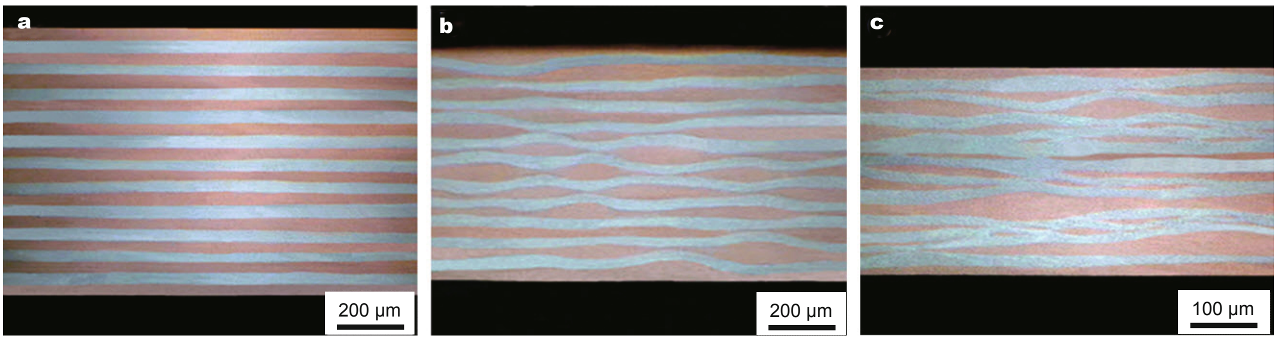

Figure 2 Optical micrographs of the structural evolution in the longitudinal section of the Cu/Al composite strip (a) after two passes, (b) after three passes, and (c) after four passes.

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interface, optical microscopy revealed that the surfaces of Cu and Al were uneven, as shown in Fig. 3. During cold rolling, the rupture of the work-hardening layers of Cu and Al surfaces results in the exposure of fresh metals, so the clean surfaces are in close contact. The close contact between the layers causes primary bonding to form, and the bonding between the Cu and Al layers makes the surfaces uneven.

After three passes, the initially flat Cu layers bend and neck down (Fig. 2b), indicating that inhomogeneous extensibility occurred. Fig. 2c shows the micrograph of the longitudinal section of Cu/Al composite strip after four passes. The interfaces between Cu and Al layers are destroyed, and a local structure of Cu surrounded by Al appears. The hardness of the annealed Cu and Al before rolling was measured to be 70 and 32 HV, respectively. The values of the work-hardening exponents of Cu and Al are known to be approximately 0.33 and 0.20, respectively. During the co-deformation of the Cu/Al laminated strip, the differences in the mechanical properties and flow properties resulted in their inhomogeneous deformation. These differences explain why necking, fracture, and departing occurred earlier in Cu layers.

Formation of new phases

After five passes, the Cu/Al composite strip was rolled to 0.2 mm, and the accumulated reduction ratio reached 95.86%. At this stage, some rectangular-shaped phases were observed at the Cu/Al interfaces, as shown in Fig. 4. These phases are mostly found at the upper and lower borders of the oval-shaped interfaces, as shown in Fig. 4a. After six passes, the rectangular-shaped phases can be found at any oval-shaped interfaces site, including the upper and lower borders of the oval-shaped interfaces, as shown in Fig. 4b. Fig. 4c shows the residual Cu surrounded by the interfacial-reaction products, which indicates that the Cu/Al interfacial reactions have not completely finished. The residual Cu would continue to transform with repeated rolling. Consequently, Fig. 4c reveals that the interfacial reactions occurred under the action of force. Moreover, from Fig. 4b we see that when the Cu/Al interfaces were straight (no bending), new phases are not formed even though the reduction in thickness is unchanged.

Optical microscopy reveals that the observed phases are a mixture of two different phases. One phase beside the Al layer is light grey, and the other phase beside the Cu layer is dark grey. In fact, SEM observation reveals that the mixture

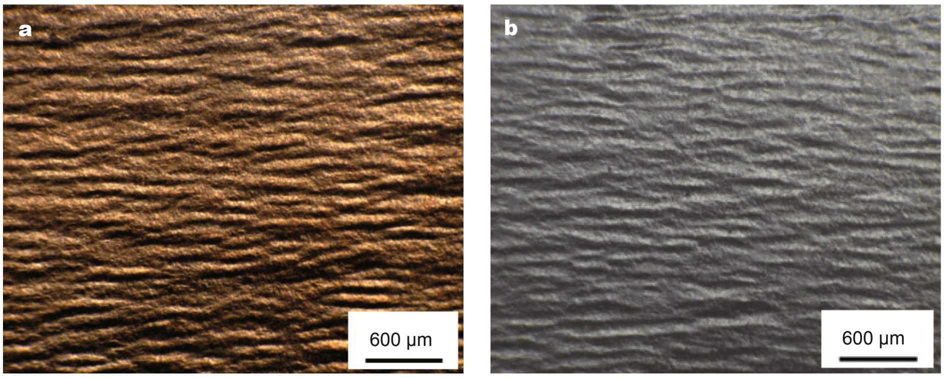

Figure 3 Micrographs of (a) Cu and (b) Al surfaces after two passes.

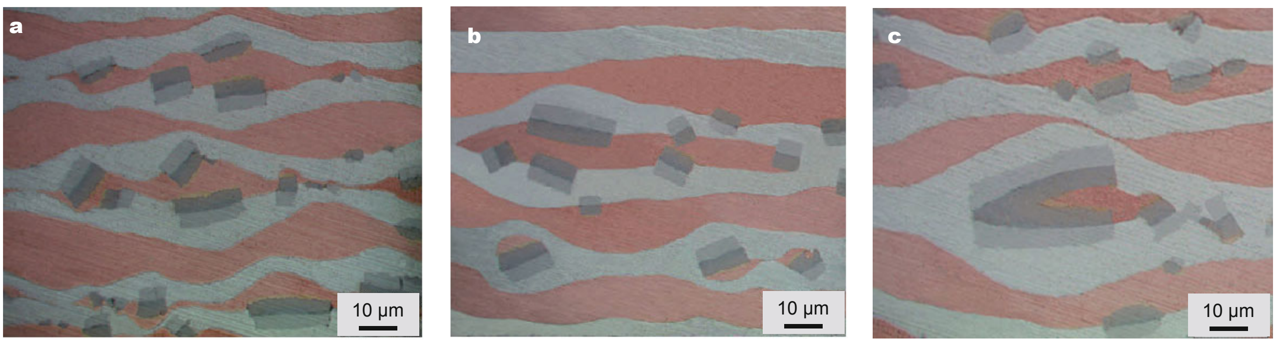

Figure 4 (a) After five passes, new phases located at the upper and lower borders of the oval-shaped interfaces, (b) after six passes, the phases located at any oval-shaped interface site, and (c) interfacial reactions occurring under the action of force.

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is composed of three different phases, as shown in Fig. 5. To identify these phases, TEM was used, and the resulting micrograph of the phase beside the Al layer is shown in Fig. 6a. Both the SAED pattern and the TEM/EDS spectra indicate that the phase beside the Al layer is the intermetallic compound Al_2Cu instead of a supersaturated Al–Cu solid solution. In Fig. 7, the TEM micrograph, SAED pattern, and TEM/EDS spectra indicate that the phase beside the Cu layer is the intermetallic compound Al_4Cu_9. Finally, SEM/EDS spectra suggest that the phase located between the other two phases is the intermetallic compound AlCu, as shown in Fig. 8. Thus, the new phases shown in Fig. 5 are arranged in the following order: Al/Al_2Cu/AlCu/Al_4Cu_9/Cu.

To further identify the phases, XRD was performed. The

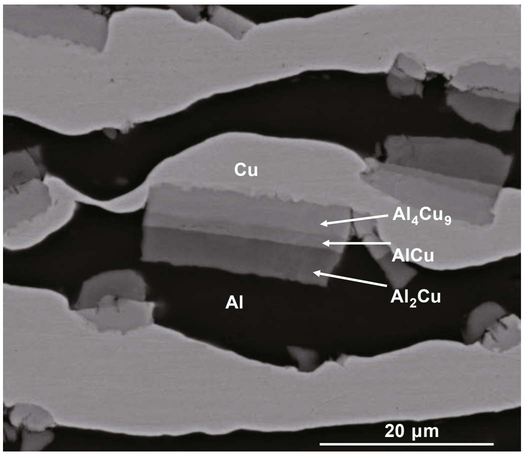

Figure 5 New phases arranged in the order Al/Al_2Cu/AlCu/Al_4Cu_9/Cu.

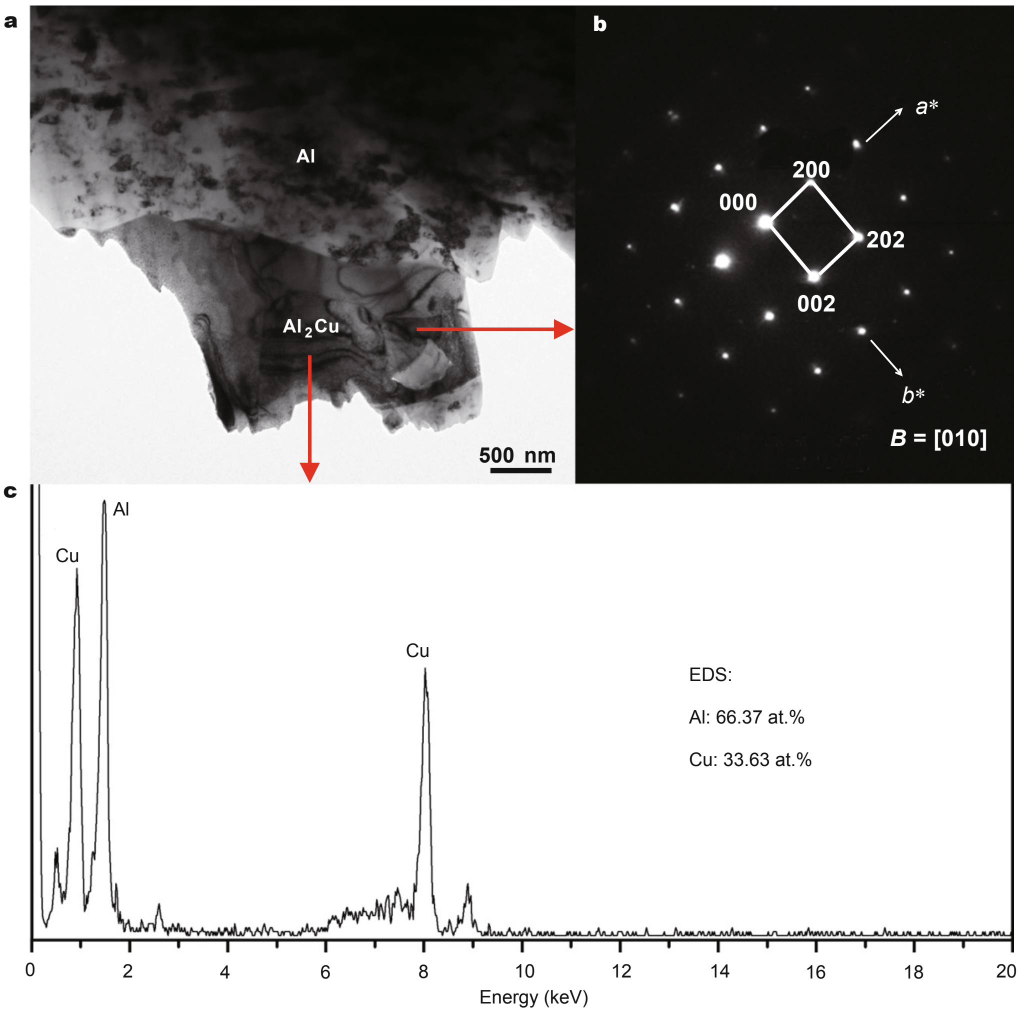

Figure 6 (a) TEM images of Al layer and the phase beside Al layer, (b) the SAED pattern corresponding to the phase beside Al layer, and (c) the EDS results corresponding to the phase beside the Al layer.

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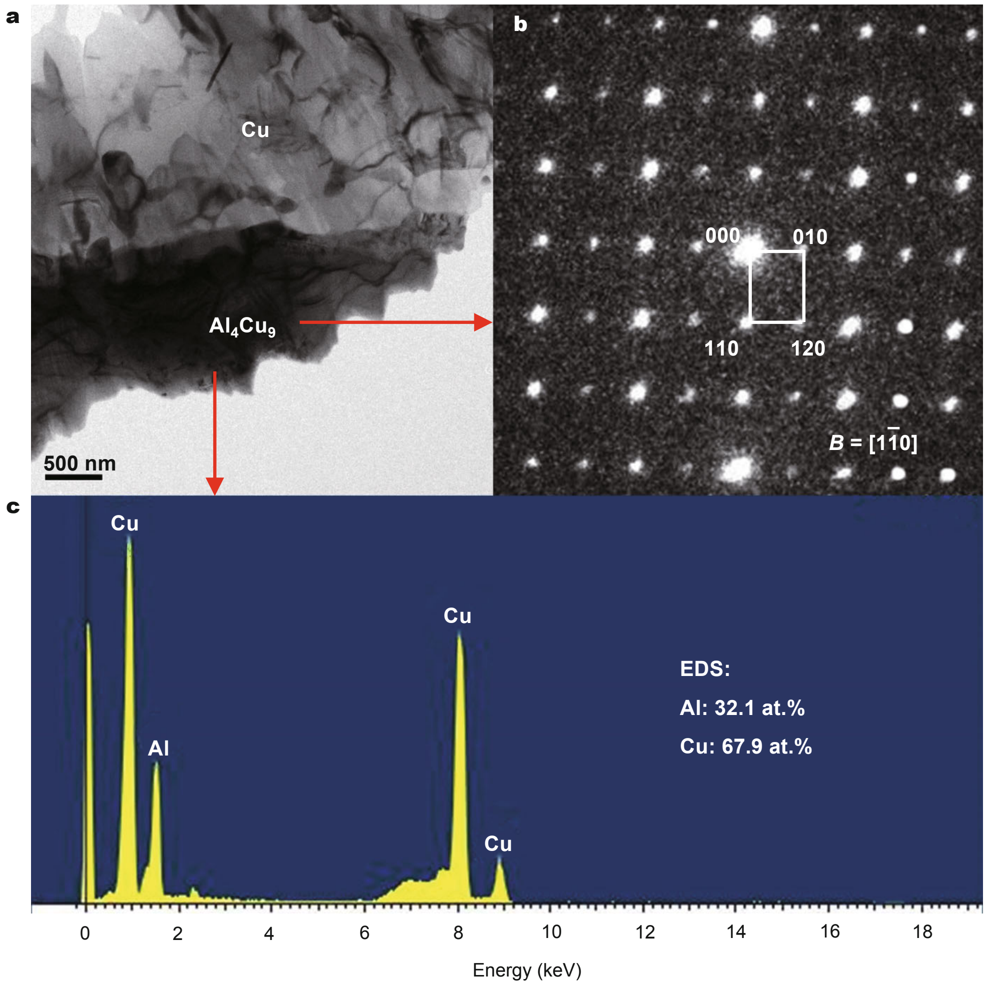

Figure 7 (a) TEM images of Cu layer and the phase beside Cu layer, (b) the SAED pattern corresponding to the phase beside the Cu layer, and (c) the EDS results corresponding to the phase beside the Cu layer.

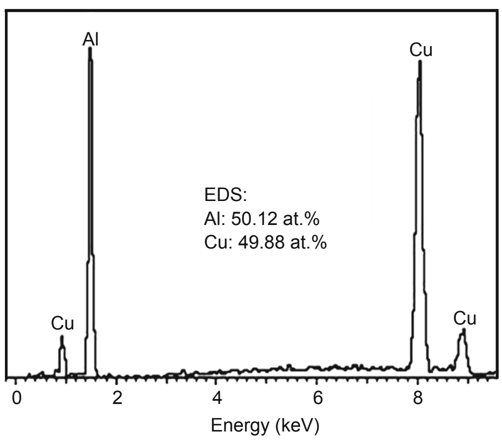

Figure 8 SEM/EDS spectra, showing that the phase located in the middle of three phases is the intermetallic compound AlCu.

XRD results also demonstrate the presence of Al_2Cu, AlCu, and Al_4Cu_9, as shown in Fig. 9. Furthermore, the identified phases agree well with the SEM/EDS spectra, TEM/EDS spectra, and SAED pattern results. Therefore, these results confirm the occurrence of Cu/Al interfacial reactions at room temperature by force effects, and the chemical-reaction products are composed of Al_2Cu, AlCu, and Al_4Cu_9.

DISCUSSION

Initially, we believe that the formation of new phases is related to the increase in temperature of Cu/Al composite during repeated cold rolling. Moreover, the test results show that the Cu/Al composite temperature was increased to 60°C because of the significant absolute reduction in thickness (1.27–2.4 mm per pass) during the cold rolling of the first two passes. However, after two passes, there are no new phases formed, as shown in Fig. 2a. During the next

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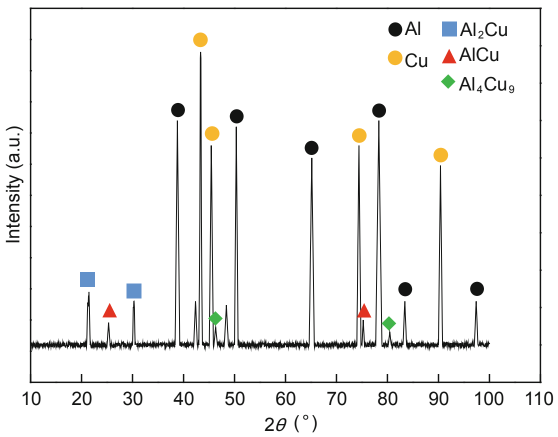

Figure 9 XRD results of the Cu/Al multilayer composite.

four passes performed on the 3M mill, the test results show that the temperature of Cu/Al composite is nearly as low as room temperature because the absolute reduction is low (only 0.07–0.35 mm per pass) and the Cu/Al composite is thin (0.2–0.9 mm). Therefore, we can exclude the possibility that high temperature facilitates the formation of new phases. We should determine which mechanism causes the new phase formation.

From metallography theory, the formation of the intermetallic compound requires high-temperature diffusion over a long period of time. However, it is not known why cold rolling can increase the diffusion rates of metal atoms. Based on the Arrhenius Equation, the diffusivity D for an integrated crystal lattice can be expressed as

$$D = D_0 \exp\left(-\frac{Q}{RT}\right), \tag{1}$$

where D_0 is the diffusion constant, Q is the activation energy for diffusion through the integrated crystal lattice, R is the gas constant, and T is the temperature in Kelvin.

Therefore, the diffusivity D can be increased by increasing the temperature or by reducing the activation energy for diffusion. To increase the diffusion rate of metals, high temperature heating is widely used because of its simple application. In contrast, the activation energy for diffusion through an integrated crystal lattice is high, so reducing the activation energy to increase the diffusion rate of metals is seldom used.

However, in our experiment, high-density dislocations were formed in Cu and Al layers after heavy deformation, and thus, Cu and Al atoms at both sides of Cu/Al interface can be diffused along dislocations. Additionally, it is easier for atoms to diffuse across Cu/Al interfaces along dislocations than through a defect-free lattice (Fig. 10) because the activation energy for diffusion along dislocations is much less than that through a defect-free lattice. Consequently, the diffusivity for Cu/Al composite strip could be rewritten as

$$D = D_0 \exp\left(-\frac{Q-\Delta E}{RT}\right), \tag{2}$$

where D_0 and R are the diffusion and gas constants, respectively, Q is the activation energy for diffusion through the integrated crystal lattice, and T is the temperature in Kelvin, and ΔE is the lattice-distortion energy caused by the dislocations.

From Equation (2), the lattice distortion induced by dislocations results in a decrease of the activation energy for diffusion, which contributes to an increase in atomic diffusion along the dislocation. In addition, the diffusivity D not only follows an exponential relationship with the thermodynamic temperature T, but also follows an exponential relationship with $Q-\Delta E$. Both $Q-\Delta E$ and T play important roles in the diffusivity. For the cold-rolled Cu/Al composite foil, when the thickness of the sample was reduced below 0.20 mm from the initial thickness of 4.83 mm (cumulative working ratio reached 95.86%), the heavy reduction in thickness led to an increase of the dislocation density of Cu and Al layers, increasing the amount of dislocation pipes and enhancing the diffusivity at room temperature. Thus, both $Q-\Delta E$ and T provide necessary conditions for the formation of new phases at room temperature.

Nevertheless, the interfacial-reaction products are not distributed continuously and uniformly at Cu/Al interfaces, as shown in Figs 4a–c. We notice that the formation of the interfacial-reaction products is controlled by the shapes of the Cu/Al interfaces. When the Cu/Al interface has the

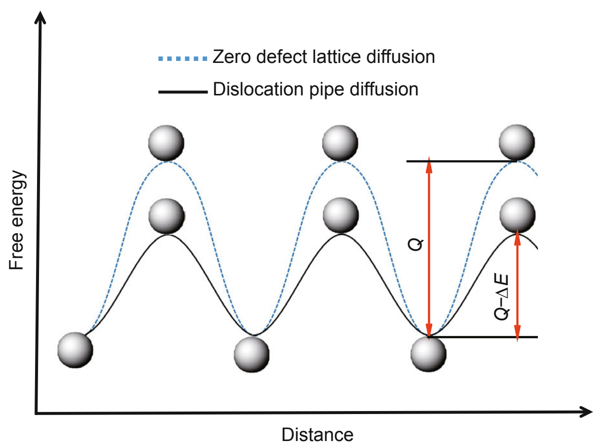

Figure 10 Comparison between the diffusion activation energy along dislocations and that through a defect-free lattice.

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shape of a semi-closed ellipse (Fig. 4a), the interfacial-reaction products are only distributed at the upper and lower vertexes (Fig. 11a, *a* and *b* sites) of the oval-shaped interfaces and are not observed elsewhere. When the Cu/Al interface has the shape of a fully closed ellipse (Fig. 4b), the interfacial-reaction products could be distributed at any site along the Cu/Al interfaces (Fig. 11c) and are not restricted to the upper and lower vertexes of the oval-shaped interfaces.

To explain this new phenomenon, we propose the idea shown in Fig. 11b. After heavy deformation (over 95% in our experiment), there are many dislocations in the grains. These dislocations can be regarded as "short-circuit" paths. The atomic diameter of Al is 3.64 Å and that of copper is 3.14 Å. The width of the dislocation pipe is several times that of atoms [30], so it is wide enough to accommodate copper or aluminum atoms. If there are dissimilar-metal atoms at the entrances of the paths, they can be forced into the dislocation pipes under rolling pressure, as schematically shown in Fig. 11d. Simultaneously, because the atomic positions are distorted in the dislocation core region, the interatomic forces that restrict atomic movement in the region become weaker than those in equilibrium positions. Thus, the dislocations are high-diffusivity paths allowing dissimilar atoms to rapidly migrate.

By further analysis, we find in Fig. 11a (semi-closed region) that when metals flow along the rolling direction, the metals at the *c* and *d* sites are subjected to maximum resistance flowing along the rolling direction and those at the *a* and *b* sites are subjected to a much smaller resistance. This smaller resistance explains why the interfacial-reaction products were found at the *a* and *b* sites but not at the *c* and *d* sites. However, when the foil was rolled to be sufficiently thin, the fully closed region was formed where the copper atoms were surrounded by neighboring aluminum atoms (Fig. 4c). Thus, the metals at the *c* and *d* sites were also

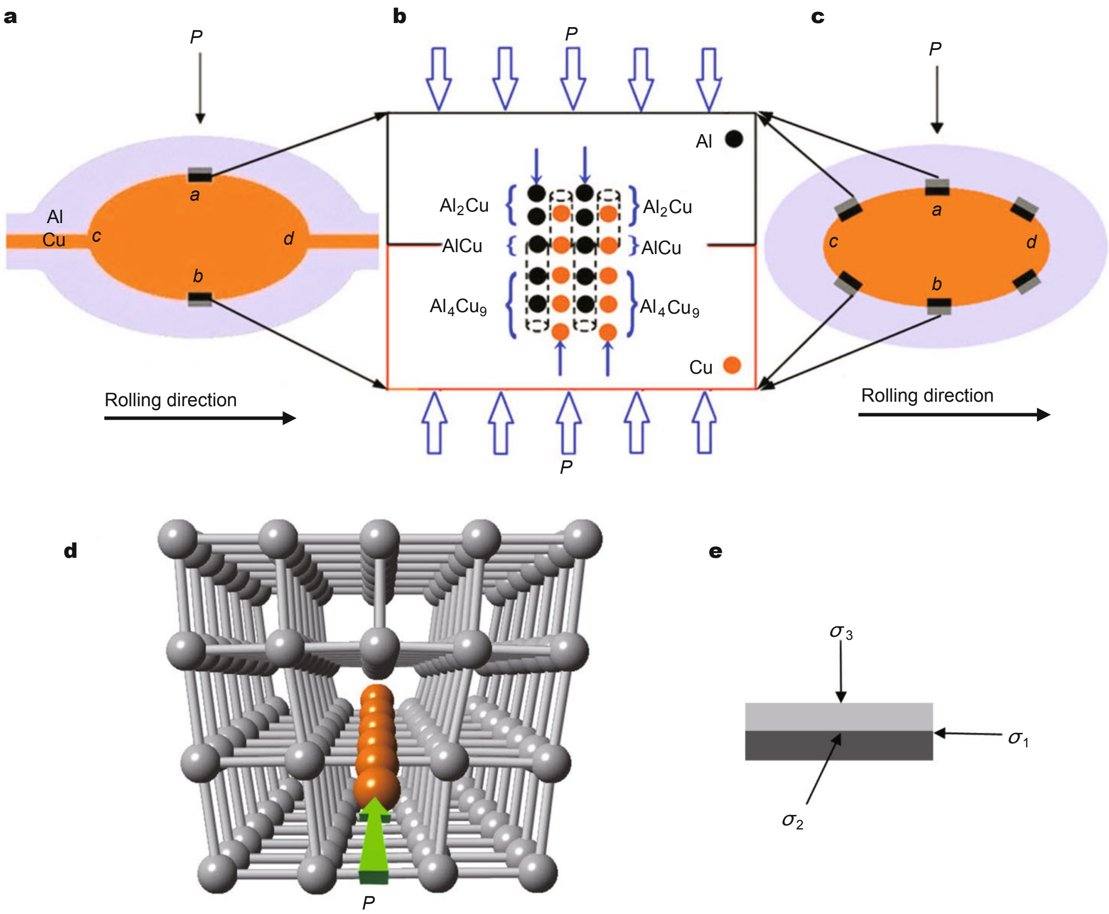

Figure 11 Schematic illustrations for the formation of intermetallic compounds under the action of force: (a) semi-closed region, (b) two-dimensional schematic for diffusion along dislocations, (c) fully closed region, (d) three-dimensional schematic for diffusion along dislocations, and (e) the formation of a rectangular-shaped phase subjected to three-dimensional compressive stresses.

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subjected to a large resistance like those at the *a* and *b* sites. Therefore, the interfacial-reaction products were found not only at the *a* and *b* sites but also at the *c* and *d* sites.

If the copper and aluminum atoms met via pipe diffusion along dislocations (Fig. 11b), a supersaturated solid solution was formed first and then intermetallic compounds were formed as more atoms entered the pipe dislocations. For a cold-rolled foil of dissimilar-metal layers, if interfacial reactions occur, the following conditions must be satisfied: (1) high-density dislocations, which provide high-diffusivity paths, allow dissimilar-metal atoms to diffuse and increase the diffusivity as a result of heavy reduction; (2) fully closed or semi-closed regions, in which high pressure acts as a driving force for atomic diffusion, are formed when the laminated-metal composite foil is rolled sufficiently thin to form a structure like that in Fig. 4. Additionally, the generation of new phases results from high mechanical deformation and high pressure. In particular, the new phases are subjected to three-dimensional compressive stresses (σ_1, σ_2, and σ_3), so they appear rectangular in shape, as shown in Fig. 11e.

We can use these results to understand other results. For example, the ARB technique was used by Eizadjou *et al*. to produce Cu/Al composite strips [31]. Although the reduction ratio was sufficiently large, there were no fully closed or semi-closed regions, and thus, no interfacial reactions occurred. Similarly, Min *et al*. [32] used the ARB technique to prepare a multilayered Al/Ni composite strip. Before each ARB pass, the samples were held at 523 K for 8 min to enhance the diffusion bonding between the two layers. However, the annealing process resulted in a decrease in the dislocation density for Al and Ni layers. In such case, no interfacial reactions occurred even though closed Al/Ni interfaces had been formed during the ARB process.

CONCLUSIONS

Using the 3M mill, we showed that intermetallic compounds (Al_2Cu/AlCu/Al_4Cu_9) were formed during the cold rolling of the 23-layered Cu/Al composite strip. Moreover, the room-temperature chemical reactions that formed these compounds were a result of high mechanical deformation and high pressure, rather than deformation heat. This technique can be used for obtaining new intermetallic compounds and new composite materials, and it was applied here to produce Al/Al_xCu_y/Cu composites. This result reveals the possibility of obtaining combined properties such as high conductivity properties like that of copper, low density properties like that of aluminum, and excellent corrosion resistance like that of intermetallic compounds.

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Author contributions Yu Q and Liu X designed and directed the overall study. Rolling experiments were carried out by Liu X. Yu Q wrote the manuscript and discussed the results and analyzed the data with Liu X. Sun Y was responsible for OM and TEM.

Conflict of interest The authors declare that they have no conflict of interest.

Supplementary information Supporting data are available in the online version of the paper.

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中文摘要 金属学理论认为新相的形成需要高温且长时间. 然而, 本研究发现, 在没有热作用的前提下, 大的机械变形和大的压力也可以使一些新相产生. 实验中首先采用冷轧方法制备出4.83 mm厚的三明治Cu/Al复合带(23层), 然后在四辊微成型轧机上(3M轧机)进行冷轧. 当Cu/Al复合带被轧到厚度为0.13 mm时, 发现在Cu/Al界面处有矩形状的相出现. 采用SEM、TEM和XRD分析得出, 这些相是由三种不同的金属间化合物Al_2Cu/AlCu/Al_4Cu_9所组成, 这表明冷变形过程中Cu/Al界面之间发生了化学反应. 这一发现为设计和研制具有特殊性能的复合材料提供了新思路.

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