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Strengthening behavior and thermal conductivity of Cu/Al composite with penetration architecture

Xiao-ling CHEN[1], Zhi-qing CHEN[1], Bo HU[1], Long YAN[2], Jing-ya WANG[1], Tao YING[1], Xiao-qin ZENG[1]

1. National Engineering Research Center of Light Alloy Net Forming, School of Materials Science and Engineering, Shanghai Jiao Tong University, Shanghai 200240, China;
2. AVIC Aeronautical Radio Electronics Research Institute, Shanghai 200233, China

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Abstract: To improve the strength of Al alloys without severely deteriorating the thermal conductivity, the Cu/Al bimetallic composite comprising penetration architecture was artificially designed and fabricated via the additive manufacturing combined with the squeeze casting. The composite exhibited a good balance of the strength (~340 MPa) and thermal conductivity (200 W/(m·K)), outperforming the traditional Al alloys. High thermal conduction is attributed to the geometrical Cu scaffold, which provides a rapid pathway for the electron conduction. Simultaneously, the good metallurgical bonding is attained by the formation of the Al_2Cu eutectic phase along the interfaces, which effectively enhances the strength of the Cu/Al composite.
Key words: Cu/Al composites; compressive strength; squeeze casting technology; thermal conductivity

1 Introduction

Cu/Al composites are extensively appealing for the electrical and thermal conductor components in the heat dissipation, aviation, communications and automotive applications [1−3]. They perfectly take advantage of the excellent thermal conductivity of Cu, combined with light weight and low cost of Al. The Cu/Al bimetallic composites can reduce 40% mass and 60% expense, yet offers the equivalent electrical and thermal conductivity [4].

Recently, numerous techniques have been developed to fabricate Cu/Al composites, including welding [5−7], extrusion [8], rolling [9,10] and compound casting [11−15]. However, most fabrication techniques are restricted to manufacturing Cu/Al composites with simple shapes, such as plate-shaped and circular pipes. This severely constrains the widespread application in complex components, such as heat dissipators in the 5G base station and heat sink of the thrust chamber in the engines, which are highly required in the specific industries, for instance, communication and aerospace. Another limitation of the traditional Cu/Al composite arises from the trade-off between mechanical strength and thermal conductivity, which are mutually exclusive properties. Generally, the traditional strengthening approaches, such as solid solution and precipitation, are inevitably detrimental to thermal conductivity. This originates from the intensive electron/phonon scattering at the lattice distortion that are induced by the solute atoms and phase interfaces [16]. For instance, KIM et al [17] found that the Al−50vol.%Cu composite exhibited high Vickers hardness of HV 151.3, while the thermal conductivity was only 130 W/(m·K), even 40% and 66% lower than that of pure Al (216 W/(m·K)) and Cu (385 W/(m·K)) [16], respectively. This was

Corresponding author: Jing-ya WANG, Tel: +86-15026426312, E-mail: jingya.wang@sjtu.edu.cn;
Tao YING, Tel: +86-13166360936, E-mail: yingtao85@sjtu.edu.cn
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mainly caused by the high content of intermetallic compound, which improved the strength, but dramatically reduced the thermal conductivity. LEE et al [18] fabricated Cu/Al composites by repeated hydrostatic extrusion and obtained an improved thermal conductivity of 322 W/(m·K), but with the tensile strength of 102 MPa, 47% lower than that of pure Cu (194 MPa) [16].

To address these issues, a novel strategy has been proposed that encompasses an artificially designed penetration architecture for the Cu reinforcement, accomplished by a feasible manufacturing approach. A thoroughly penetrated architecture is envisaged, which separates the pure Al matrix and pure Cu reinforcement. This makes the Al matrix fully surrounded by the outer Cu shell, expectedly forming a rapid pathway for the electron conduction. It is strongly inspired by the copper cladding aluminum structure [19] and the interpenetrating structure of Mg−NiTi composite with enhanced performance [20]. Such a geometrical structure with the penetration architecture is expected to exhibit high thermal conductivity along the penetration direction. Simultaneously, intermetallic compounds (IMCs) are introduced at the interfaces between the Cu reinforcement and Al matrix to provide the metallurgical bonding and strengthening effect, and thus high strength is expected for the entire composite. Additionally, a feasible fabrication process is designed, involving the additive manufacturing for the complex Cu scaffold and the squeeze casting under pressure for the entire Cu/Al composite. Additive manufacturing is a flexible and feasible approach for fabricating the scaffolds with complex architectures. Squeeze casting is generally conducted at high temperatures and pressures, which potentially contributes to the good interfacial bonding with the defect-free and uniform interface [12,21].

Hence, in the present work, a novel design concept is developed, where a good balance between mechanical strength and thermal conductivity is achieved by the geometrical structure and fabricated by the novel and flexible formation approach. A Cu/Al composite was fabricated, and the mechanical and thermal properties, as well as the interfacial characteristics, were systematically investigated in this work. The novel Cu/Al composite with the penetration architecture was expected to simultaneously obtain high strength and thermal conductivity along the penetrating direction. This could potentially extend the applications of the Cu/Al composites in the complex components required in the automobiles, heat dissipators and high-voltage cable joints.

2 Experimental

2.1 Fabrication

Figure 1 presents the formation process of the Cu/Al composite. A three-dimensional Cu scaffold, with the thoroughly penetrated square holes of 1.5 mm × 1.5 mm, was designed using the Solidworks software and fabricated by the selective laser melting (SLM). The original digital model was sliced and processed using the commercial Materialise Magics 21.0 software. The thickness of each layer was set to be 50 μm. Before printing, the pure Cu substrate was preheated to 200 °C to reduce the temperature difference with the melted powders, and ensure the precise dimensions of the manufactured scaffold. The laser power, diameter of the laser beam and the scanning width were selected to be 300 W, 0.1 mm and 0.05 mm, respectively. The dimension of the additively manufactured Cu (AM-Cu) scaffold was 20.5 mm × 20.5 mm × 40 mm and comprised 10×10 arrays of unit cells

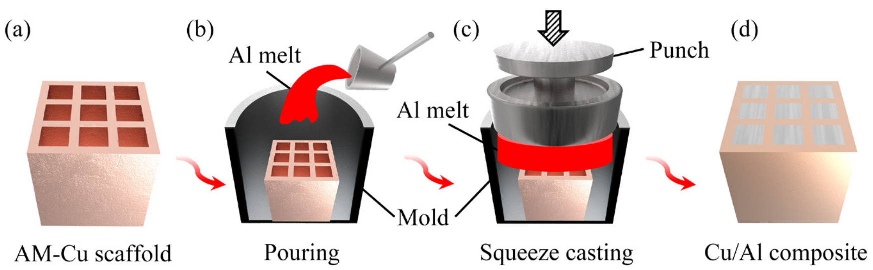

Fig. 1 Schematic of formation process of Cu/Al composite: (a) AM-Cu scaffold; (b) Pouring of Al melt; (c) Squeeze casting processing; (d) Fabricated Cu/Al composite

(Fig. 1(a)). Subsequently, pure Al was heated in an electrical furnace until completely melted and then poured into the AM-Cu scaffold at 720 °C under a pressure of ~100 MPa (Figs. 1(b, c)). Eventually, the Cu/Al composite was generated after the solidification, as illustrated in Fig. 1(d). The whole composite gives 43.75% volume faction for the Cu reinforcement.

2.2 Microstructure characterization

The sample with dimensions of 10 mm × 10 mm × 5 mm was cut from the fabricated ingots. The density of the Cu/Al composite was examined using the Archimedes method with the density kit ME-DNY−4. The phase composition was analyzed by the X-ray diffraction (XRD) using a SHIMADZU XRD−7000 with Cu K_{α} radiation at an accelerating voltage of 40 kV and a scanning speed of 4 (°)/min. The samples were carefully polished using W7 diamond powder and W3.5 MgO powder. The microstructure of the Cu/Al composite was characterized using ZEISS Axioscope5 optical microscope and TESCAN MIRA3 field-emission scanning electron microscope (SEM) under an accelerating voltage of 5 kV. Then, the composition distribution was analyzed by the electron dispersive spectrometer (EDS) using an Oxford spectrometer equipped with the microscope. The defects were characterized by the X-ray computed tomography (XCT) using a Xradia versa XRM−520 3D X-ray microscope with an accelerating voltage of 140 kV. The sample was rotated 360° around the normal axis of the X-ray source and detector, generating 1000 2D projection slices. The 3D volume renderings were automatically reconstructed using ORS Dragonfly software.

2.3 Mechanical and thermal tests

Vickers hardness tests were conducted at 4.9 N using the SCTMC HV−10 macroscopic Vickers hardness testing machine. Quasi-static uniaxial compression tests were performed at the ambient temperature under a strain rate of $1\times10^{-3}\ s^{-1}$ using the electromechanical universal testing machine (Shenzhen Sans Testing Machine Co., Ltd., China). Samples for the unidirectional compression were cut from the AM-Cu, casting Al and Cu/Al composite, respectively, with the diameter of 6 and 8 mm in height, as shown in Fig. 2(a). The compression specimen contains ~42% (volume faction) Cu reinforcement. Thermal analysis was conducted using the laser flash method in a NETZSCH LFA 467, with a spot size of 8.9 mm. Samples for the thermal analysis were cut from the fabricated Cu/Al composite and ground to thin chips with the diameter of 12 mm and the height of 2 mm, as depicted in Fig. 2(b). The sample comprised several units and the exposed surface area was ~113.1 mm^2, giving ~40% (volume faction) for Cu reinforcement, which is comparable with the Cu/Al composite and the specimens for compression tests. The thermal diffusivity was directly measured along the penetration direction of the composite. At least three samples were used in each experiment to ensure the repeatability.

3 Results and discussion

3.1 Microstructure

Figure 3 shows the typical microstructure of the Cu/Al composite. The Cu scaffold was perfectly filled with the Al matrix, as displayed in Fig. 3(a). Apparently, the composite comprised three distinct regions with different morphologies in Fig. 3(b). The interfacial microstructure without pores or microcracks was observed, indicating the formation of the good metallurgical bonding between the Cu scaffold and Al matrix. A single layer of intermetallic compound (IMC), with an average width of ~120 μm, was formed in the transition

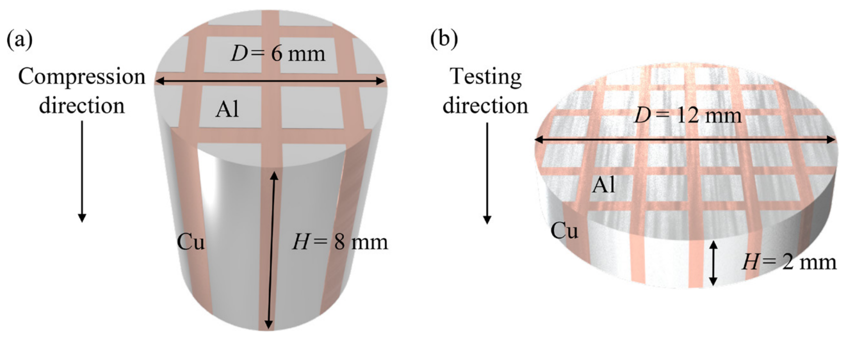

Fig. 2 Schematic diagrams of sample used for compression test (a) and thermal analysis (b)

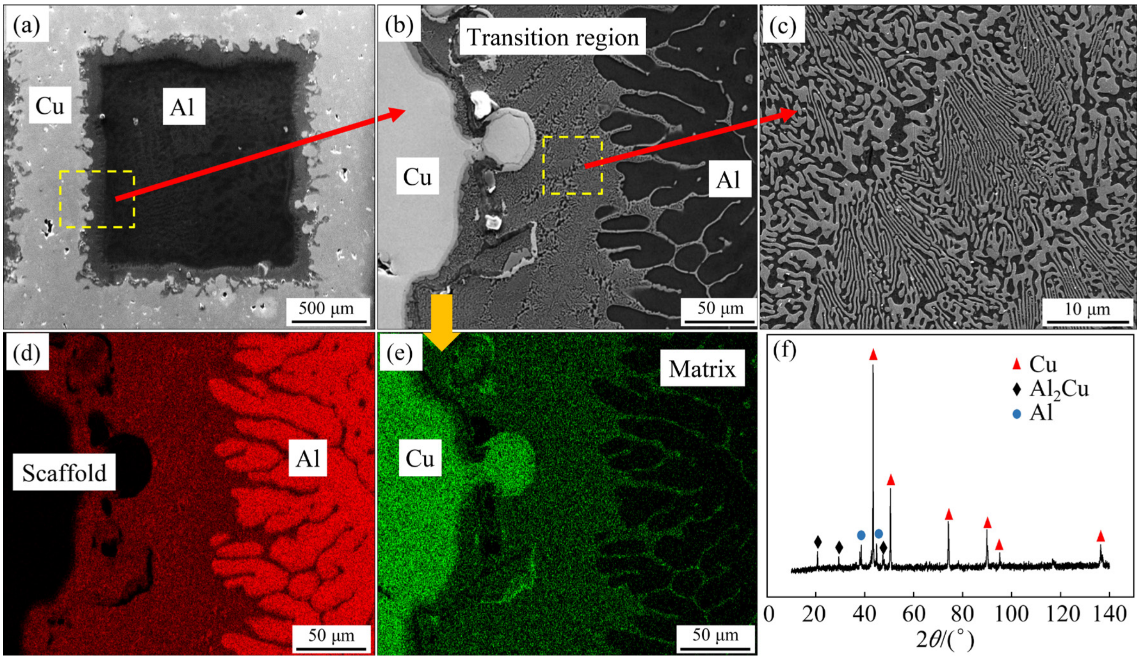

Fig. 3 SEM images (a−c), distribution of alloying elements (d, e) and XRD pattern (f) of Cu/Al composite

region, and exhibited a eutectic structure, as presented in Fig. 3(c). Pure Cu and pure Al were identified in the scaffold and matrix regions, respectively, by the alloying element distribution measured by EDS analysis (Figs. 3(d, e)). However, the transition region comprised both Cu and Al alloying elements, where the eutectic Al_2Cu phase was formed, as confirmed by the XRD results shown in Fig. 3(f). According to the Al−Cu phase diagram (Fig. 4) calculated using the Pandat software, the Al_2Cu phase was preferentially formed during the compound casting since the pouring temperature of the Al melt was higher than the eutectic temperature of 548 °C.

It was quite interesting to notice the formation of one-layer Al_2Cu eutectic phase in the interfacial region, which significantly differed from that of other Cu/Al composites containing several interaction layers [10,11] fabricated by the compound casting. At the beginning of the squeeze casting in this work, the Al melt was rapidly infiltrated into the Cu scaffold under high pouring temperature and quite high pressure. When the front edge of the Al melt reached the Cu scaffold, the Cu started to locally melt in the surface layer with the formation of the interfacial region, including the mixing melt of the Al and Cu liquid metals [12], and simultaneously the interdiffusion took place through the interface, matrix and scaffold. With

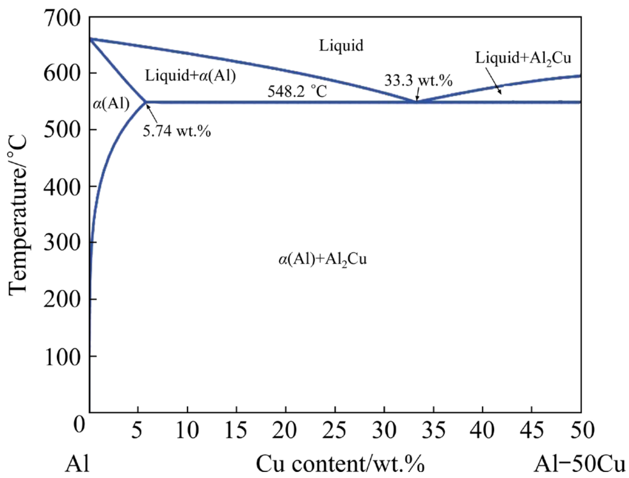

Fig. 4 Al−Cu phase diagram calculated by Pandat software

decreasing the temperature to the eutectic point, the eutectic Al_2Cu phase was primarily formed and grew up with the movement of the liquid/solid interface during the following solidification, eventually leading to the IMC layer. The formation and constitution of the IMC layers in the interfacial region were strongly depended on the diffusion process during the casting. However, the high external pressure was applied during the squeeze casting, thus giving the strongly accumulated pressure in the interface between the Al melt and Cu of every unit due to the constraint of the scaffold. The solidification under high pressure inevitably gave the most rapid heat transfer, which significantly increased the heat transfer coefficient and the cooling rate [22]. This would effectively suppress the diffusion of Cu and Al atoms through the interface, thus constraining the formation of multilayer IMCs. Besides, the applied pressure possessed a critical influence on changing the eutectic composition, which increased the Cu concentration to form Al−Cu eutectic phase [23]. This meant that more diffused Cu atoms would be preferably consumed in the formation of single-layer Al_2Cu eutectic phase. Thus, it could be concluded that the high pressure generated around the melt benefits for the suppression of the multi-layer IMC in the interfacial region. The single-layer IMC is expected to improve the interfacial bonding since only two interfaces exist and this decreases the possibility of forming cracks during deformation.

3.2 Formation quality

The defects in the Cu/Al composite were characterized via the XCT and are illustrated by the 3D reconstruction of a single unit. Figure 5(a) shows the defect distribution from the top view side. For clarification, the pores located within the AM-Cu and along the interface are shown in Figs. 5(b) and (c), respectively. Evidently, most defects were located in the AM-Cu scaffold, which was attributed to incomplete sintering or melting during the additive manufacturing. This gave the density of 8.347 g/cm^3 to the AM-Cu, lower than the theoretical value of 8.96 g/cm^3. Few defects were distributed along the interfaces (Fig. 5(c)), which indicated excellent metallurgical bonding without significant microcracks and defects. Additionally, limited pores and defects were observed in the Al matrix, which indicated that the solidification under pressure benefited for improving the formation quality. The defect diameters within the entire composite were statistically analyzed and represented using the equivalent pore radius in Fig. 5(d). The defect diameters were distributed in the range of 10−45 μm. Over 50% of the pores had an equivalent radius smaller than 15 μm, eventually conducting a porosity of ~0.57% and density of 5.218 g/cm^3.

Squeeze casting under relatively high external pressure is beneficial to reducing the gas and shrinkage porosities during solidification [24,25], which is inspired by the die casting and closed die forging [25]. Indeed, the low porosity attained in this study is even comparable to that of the A390

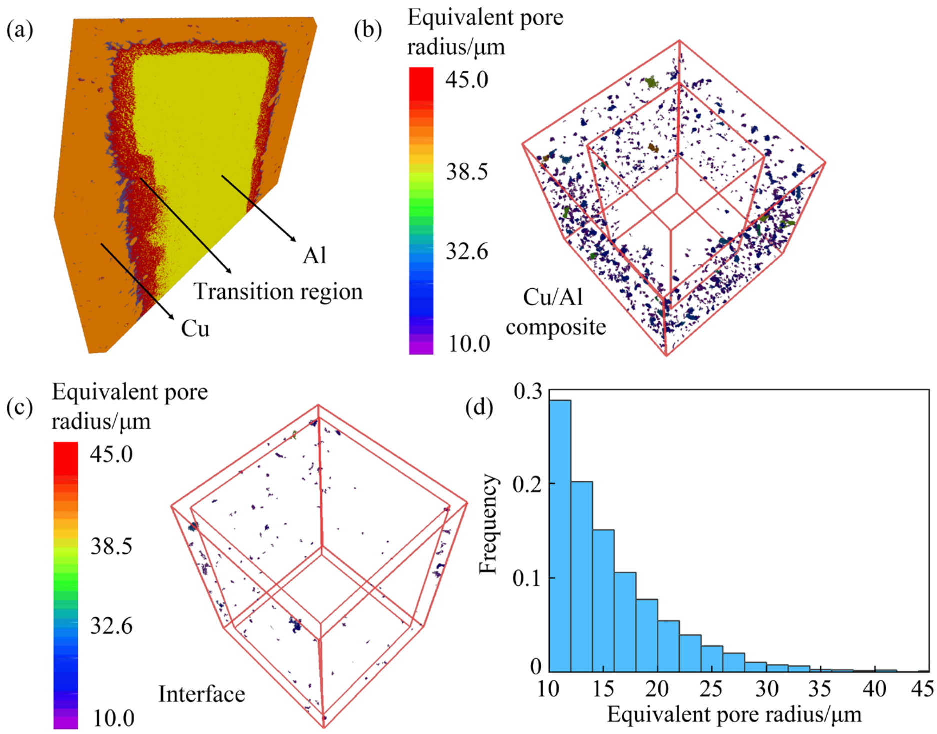

Fig. 5 Defect distribution from top view side (a), 3D visualization of defects in AM-Cu (b) and along interface (c), and equivalent pore radius distribution in composite (d)

alloy fabricated by the die casting [26]. Apart from that, squeeze casting helps to overcome the destructive effect of the oxide layer on metallurgical bonding. Generally, the oxide layers are prone to form during the compound casting, which easily generates the pores and microcracks at the interface region, and prevents the metallurgical bonding between the matrix and scaffold. Herein, surface treatments, such as coating [7,15], are usually adopted to remove the stable surface oxides and ensure the good bonding, which makes the formation process complex. However, the excellent metallurgical bonding with a thick IMC layer and reduced defects was formed in this work, even without any surface treatment. This was mainly attributed to the breakthrough of the hindrance from the oxide layer under the high pressure applied, which confirmed the sufficient reaction between the Cu scaffold and the Al melt. This significantly simplified the processing of the composite and reduced the overall cost, potentially expanding the applications of composites.

3.3 Mechanical and thermal properties

Figure 6(a) presents the variation in hardness in the different regions of the composite. The eutectic structure reached the highest hardness of HV (190±7.7), as shown in Fig. 6(a). The pure Cu scaffold and pure Al matrix exhibited hardness of HV (80.4±6.9) and (31.1±1.1), respectively. The difference in the hardness was obviously represented by the indent sizes in Fig. 6(b), where the indent size in the Al_2Cu eutectic phase was significantly smaller than that in the Al matrix and Cu scaffold. This is in good accordance with the previous work [12], indicating that the hardness of Al_2Cu phase was much higher than that of the Al substrate. The hardness result implied that the strong metallurgical bonding was achieved through the formation of the eutectic phase. Another moderate hardness of HV (53.7±16.2) was observed in the transition zone close to pure Al, which was attributed to the influence of the dendritic Al_2Cu phase with high hardness formed during the solidification process.

Figure 7 presents the compressive engineering stress–strain curves measured for the Cu/Al composite, AM-Cu and pure cast Al cut from the excess of the Cu/Al composite. The composite exhibited a superior compressive yield strength of 340 MPa, exceeding that of AM-Cu (205 MPa) and pure cast Al (35 MPa), even higher than the theoretical value (111 MPa) estimated by the rule-of-mixtures. This was largely ascribed to the strong metallurgical bonding due to the formation of IMC layer. The eutectic microstructure, containing Al_2Cu intermetallic compound, generally

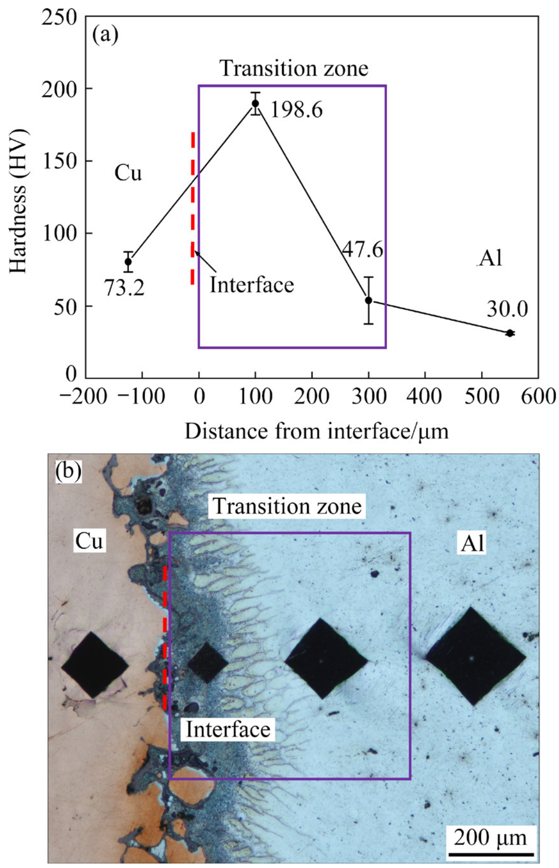

Fig. 6 Vickers hardness in various regions of composite (a), and corresponding indent imprints (b)

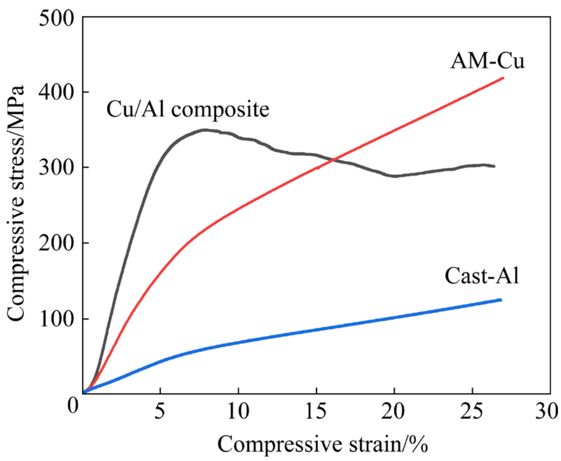

Fig. 7 Compressive engineering stress−strain curves of Cu/Al composite, AM-Cu and pure cast Al cut from excess of Cu/Al composite

exhibits high strength, as confirmed by the hardness in Fig. 6(a), which significantly helps for improving the strength of the composite. Interestingly, the Cu/Al composite demonstrated markedly higher stiffness with steeper slope in the elastic region than AM-Cu and cast-Al. This probably originates from the defects in the fabricated AM-Cu and cast-Al, especially the cast-Al taken from the excessive part of the composite.

The thermal characteristics of the composite were experimentally analyzed, and the thermal diffusivities were determined to be 70, 95 and 91 mm^2/s for the Cu/Al composite, AM-Cu and cast-Al, respectively. Then, thermal conductivity was estimated as follows:

$$\lambda=\alpha\rho c \tag{1}$$

where λ stands for the thermal conductivity; α is the thermal diffusivity and ρ is the density, which are experimentally determined; c is the specific heat capacity. The specific heat capacities of the pure Cu and pure Al are 24.40 and 24.44 J/(mol·K) [5], respectively. The specific heat capacity for Al_2Cu eutectic phase formed in the present composite is calculated to be 23.91 J/(mol·K) [27], quite close to that for pure metal. Thus, the theoretical specific heat capacity of Cu/Al composite was estimated to be 24.4 J/(mol·K), based on the rule of mixtures. On the basis of this, the thermal conductivity of the Cu/Al composite was calculated to be 200 W/(m·K), which was comparable to that of the cast Al (214 W/(m·K)), but lower than that of the AM-Cu (310 W/(m·K)). This is largely ascribed to the formation of the IMC layer with the Al_2Cu phase, which not only possesses a lower thermal conductivity (21.8 W/(m·K) [5]) but also introduces the interfaces that hinder the electron conduction. Nevertheless, the thermal properties of the novel Cu/Al composite significantly outperform those of conventional Al−Cu composites [17,28], as well as Al alloys [29,30]. The thermal conductivity in this Cu/Al composite (~44% Cu reinforcement) increases by over 50%, compared with the Al−50%Cu (particle reinforcement) composite with thermal conductivity of 130 W/(m·K) [17]. Additionally, the combination of the thermal and mechanical properties of the present Cu/Al composite significantly outperforms the novel cellular copper–aluminum composite materials with the thermal conductivity of 42 W/(m·K) and compressive yield strength of 42 MPa fabricated by SABERI and OVEISI [28] via the powder metallurgy and mechanical milling. It is interesting to notice that the same amount of Cu reinforcement gives a significant difference in the thermal conductivity, which suggests that the Cu/Al composite in this work sufficiently has the advantages of the thermal conductivity of pure Cu. The novel penetration architecture provides a rapid path for the electron conduction along the penetration direction, which sufficiently adopts the merits of high thermal conductivity of the pure metals, especially the outer pure Cu shell. This mitigates the negative influence of IMCs on the conductivity to some extent, thus enhancing the thermal conductivity along the penetration direction, since the Al_2Cu phase is only distributed between the pure Cu and Al within 100 μm.

The thermal conductivity and compressive strength of the Cu/Al composite were compared with those of other materials, as shown in Fig. 8. Since few studies have simultaneously focused on the thermal and compressive mechanical properties, the corresponding values were taken from the Cu and Al alloys for comparison [28,30,31]. A good balance between the thermal conductivity and strength was attained in the preset Cu/Al composite, breaking through the traditional trade-off in Al-based materials. This is attributed to the novel design strategy, which incorporates the advantages of the geometrical structure, formation approach and excellent metallurgical bonding. The designed penetration architecture significantly benefits for a comparable thermal conductivity along the penetration direction, through giving the smooth and rapid pathway for the motion of the electrons. Therefore, the adverse effect of the IMCs on the thermal conductivity can be lessened to some extents. The formation process under pressure effectively provides a good metallurgical bonding with the formation of single-layer Al_2Cu eutectic phase, and simultaneously ensures high formation quality for the composites with reduced defects and microcracks at the interface and the matrix. This further improves the thermal conductivity of the Cu/Al composite through reducing the electron scatter at the interfaces and amount of the IMCs with lower conductivity, compared with conventional Al-based composites with the multi-layer IMCs. In addition, the good metallurgical bonding and reduced brittle IMCs expectedly decrease the possibility of forming cracks during the deformation. Notably, this flexible and feasible formation approach, involving the additive manufacturing and squeeze casting, can contribute to fabricate the complex components, which potentially enables the Cu/Al composites to apply in extensive fields in the complex components.

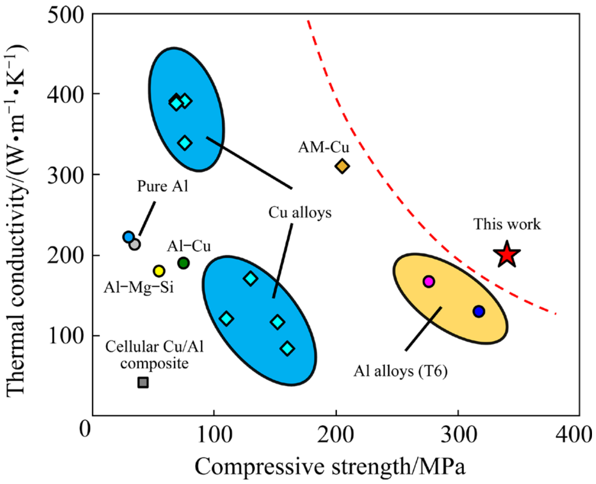

Fig. 8 Thermal conductivity versus compressive strength for Cu/Al composite, AM-Cu and cast Al measured in this work, compared with those from Cu alloys and Al alloys in literature [28,30,31]

However, it should be noted that IMCs normally behave brittleness and increasing IMC layer thickness generally reduces the mechanical strength of the composites [13,24]. However, the thickness of the interfacial reaction was not intentionally controlled in the present work. This caused the formation of a quite thick IMC layer. Herein, the advantageous approaches are highly required in the future to control the thickness of the IMC layer, for instance further increasing the cooling rate to reduce the reaction time and void the diffusion. Since the defects are detrimental to the electron conduction, the improved formation quality for the additive manufactured Cu scaffold is strongly demanded to reduce the defects, which potentially enhances the thermal conductivity of the composite. Apart from that, the thermal conductivity of Cu/Al composite is significantly related to the amount of the Cu reinforcement. Thus, it is highly required to regulate the volume fraction of the pure Cu for higher thermal conductivity by controlling the morphology and structure of the scaffold.

4 Conclusions

(1) The Cu/Al composite with an artificially designed penetration architecture was developed and fabricated by squeeze casting Al melt in the additive-manufactured Cu scaffold. Good metallurgical bonding was obtained without obvious defects and microcracks along the interface.

(2) An excellent balance between thermal conductivity (200 W/(m·K)) and compressive strength (~340 MPa) was achieved, which was attributed to the artificially designed structure and excellent metallurgical bonding, accomplished by the novel formation approach.

(3) This study offers a promising strategy for the design and fabrication of high-performance Cu/Al composites, utilizing the advantages of the geometrical structure and the feasible formation approach.

CRediT authorship contribution statement

Xiao-ling CHEN: Data curation, Writing – Original draft; **Zhi-qing CHEN:** Methodology, Investigation; **Bo HU:** Methodology; **Long YAN:** Data curation; **Jing-ya WANG:** Supervision, Writing – Review & editing; **Tao YING:** Supervision; **Xiao-qin ZENG:** Conceptualization, Editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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具有贯穿结构的铜/铝复合材料的强化及热导率

陈孝凌[1]，陈志青[1]，胡　波[1]，严　龙[2]，王静雅[1]，应　韬[1]，曾小勤[1]

1. 上海交通大学 材料科学与工程学院 轻合金精密成型国家工程研究中心，上海 200240；
2. 中国航空无线电电子研究所，上海 200233

摘　要：为了在提高铝合金强度的同时避免降低其热导率，设计了一种具有单向贯穿结构的新型铜/铝双金属复合材料，并采用增材制造结合挤压铸造的工艺实现制备成形。该复合材料表现出较好的强度(约 340 MPa)和热导率(200 W/(m·K))匹配性，综合性能超过传统铝合金。这种良好的导热性能归因于单向贯穿的 Cu 骨架增强体结构能够为电子传导提供快速通道。同时，界面处生成 Al_2Cu 共晶相，实现良好的界面冶金结合，有效改善复合材料的力学性能。

关键词：Cu/Al 复合材料；抗压强度；挤压铸造技术；热导率

(Edited by Bing YANG)