



Al/Cu复合材料 制备工艺及界面组织演变研究现状

关杰仁，王秋平

(江苏科技大学 海洋装备研究院增材制造研发中心，镇江 212003)

摘要：Al/Cu复合结构是将纯Al和纯Cu通过特定的机械或冶金方法结合在一起的典型的层状双金属复合材料，具有导电性能优异、质轻、价格低等特性，已成为广泛的研究热点。本文介绍了Al/Cu复合材料金属间化合物的种类及物理性质，阐述了界面扩散反应行为的影响因素。综述了国内外关于Al/Cu复合材料的制备工艺及界面组织性能的研究进展，并对复合界面研究方向和采用增材制造技术制备Al/Cu复合结构的改进方向进行了展望，为Al/Cu复合结构的制备提供关键的理论依据和技术支撑。

关键词：Al/Cu复合；扩散；金属间化合物；制备工艺；界面；激光增材制造

文章编号：1004-0609(2022)-09-2510-11

中图分类号：TB331

文献标志码：A

引文格式：关杰仁，王秋平. Al/Cu复合材料制备工艺及界面组织演变研究现状[J]. 中国有色金属学报, 2022, 32(9): 2510–2520. DOI: 10.11817/j.ysxb.1004.0609.2021-42264

GUAN Jie-ren, WANG Qiu-ping. Research progress of preparation technique and interface microstructure evolution of Al/Cu composite materials[J]. The Chinese Journal of Nonferrous Metals, 2022, 32(9): 2510–2520.
DOI: 10.11817/j.ysxb.1004.0609.2021-42264

随着Cu资源的消耗，铜制备材料价格昂贵，寻找理想的替代材料不仅能降低成本，而且还能获得更优异的性能^[1]。Al/Cu复合材料兼具铝较低的密度和铜较高的热导率、电导率以及可焊接性而被广泛研究，主要应用于电子、电力和汽车行业^[2]。Al/Cu界面结合是通过熔化和扩散两种结合方式，形成具有一定厚度界面层的冶金结合。相关研究表明，Al/Cu界面处存在多种金属间化合物^[3]，复合体的界面层是最容易发生断裂的位置^[4]，特别是金属间化合物的形成和宽化将促进裂纹生长，对结合强度产生不利影响^[5]，因此，对界面过渡层组织的研究至关重要，而Al/Cu界面处的金属间化合物的形成机制和互扩散行为主与加工工艺有关。传统

的成熟制备工艺包括电磁连铸法^[6]、离心浇铸法^[7]和拉拔法^[8]等，本文基于Al-Cu金属间化合物性质和界面扩散行为，综述了制备Al/Cu复合结构最新工艺及引起的界面组织和性能变化，为Al/Cu复合结构的进一步研究提供借鉴与参考。

1 Al/Cu金属间化合物

由Al-Cu二元相图可知^[9]，平衡状态下主要存在 $\theta(\text{Al}_2\text{Cu})$ 、 $\eta_2(\text{AlCu})$ 、 $\zeta_2(\text{Al}_3\text{Cu}_4)$ 、 $\gamma_1(\text{Al}_4\text{Cu}_9)$ 等相，其空间晶格结构如图1所示。实际上，加工过程中形成的金属间化合物组分取决于加工温度和时间，AO等^[10]在150 °C的工作温度下未发现金属间

基金项目：江苏省高等学校自然科学研究面上项目(20KJD430005)；江苏省高校高技术船舶协同创新中心自培育项目(HZ20200006)

收稿日期：2021-09-01；**修订日期：**2021-10-18

通信作者：王秋平，助理研究员，博士；电话：13333856825；E-mail：a21096@163.com

化合物, 而更高的温度和保温时间促进了元素扩散, 在界面处形成了 Al_2Cu 相、 AlCu 及 Al_4Cu_9 等金属间化合物。LIU等^[11]在不同保温时间范围内研究了Al-Cu扩散区域的组织及物相, 形成的四种金属间化合物 θ 相、 η_2 相、 ζ_2 相、 γ_1 相遵循抛物线式的增长模式, 其性能参数如表1所示, γ_1 相的弹性模量最大, 蠕变应力指数最小。CHEN等^[12]计算了三种Al-Cu金属间化合物的形成能, 结果表明 AlCu 是三种金属间化合物中最稳定的相, 形成能最小的 $\text{Al}_2\text{Cu}(\Delta H_f = -15.036 \text{ kJ/mol})$ 相首先形成, 后续向 $\text{AlCu}(\Delta H_f = -20.656 \text{ kJ/mol})$ 和 $\text{Al}_4\text{Cu}_9(\Delta H_f = -20.466 \text{ kJ/mol})$ 转变。

除了形成能外, 金属间化合物的形成机制还与扩散相关, 界面处原子的扩散能力遵循经典的动力学理论, 原子的扩散系数(D)由温度(T)和激活能(Q)决定, 即^[13]

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

式中: R 为摩尔气体常数($8.314 \text{ J/(mol}\cdot\text{K)}$)。

HUG等^[14]发现在Cu、Al扩散过程中沿径向存在浓度梯度, 易使界面附近的富Cu区产生柯肯达尔效应(Kirkendall effect), 形成空洞。由于Cu在Al中的扩散系数远大于Al在Cu中的扩散系数, 形成的界面反应层主要借助Cu向Al一侧的扩散^[15], 其

表1 Al/Cu金属间化合物的弹性模量(E)和硬度(H)^[11]

Table 1 Elastic modulus (E) and hardness (H) for different Al/Cu intermetallics^[11]

Al/Cu composite	H/GPa	E/GPa	Creep stress index
θ	6.88	112.45	14.94
η_2	11.74	227.91	17.09
ζ_2	11.94	228.73	11.38
γ_1	9.41	254.69	10.75

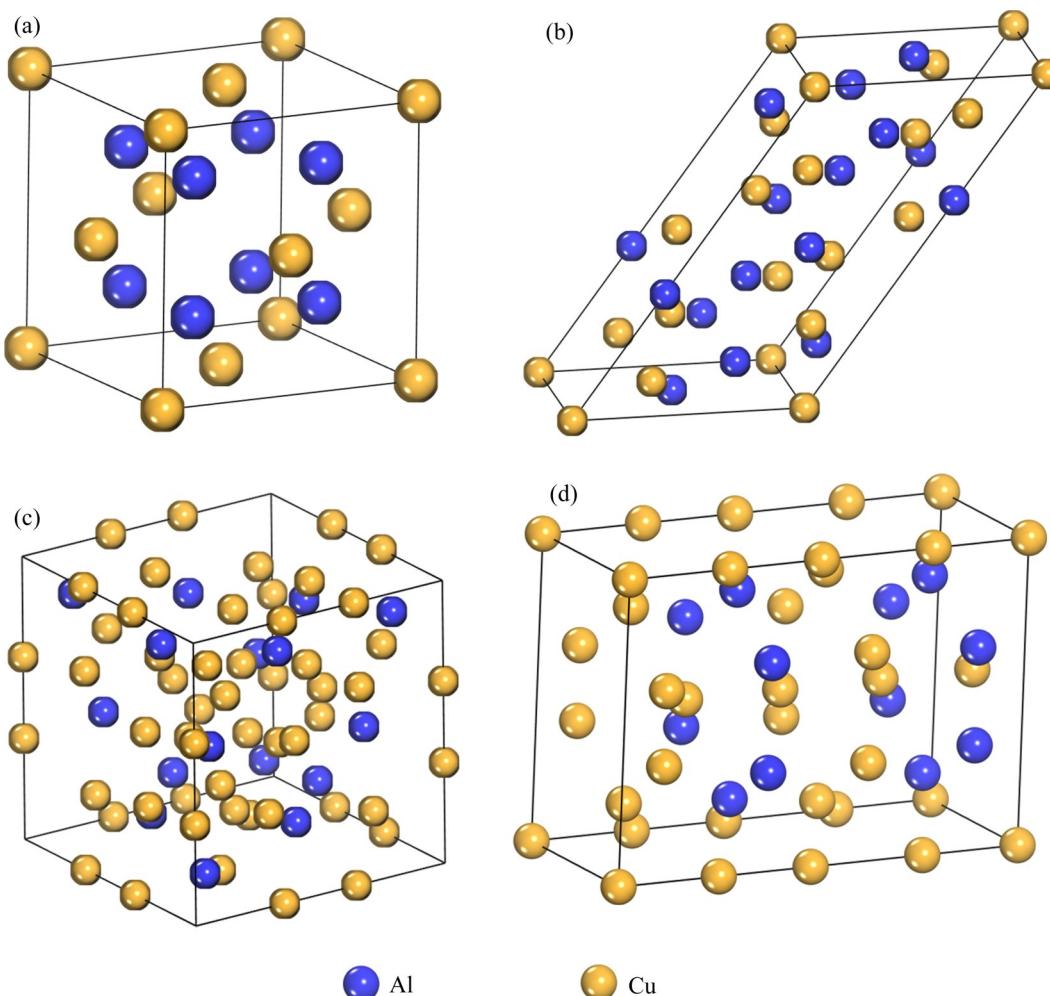


图1 Al-Cu金属间化合物空间结构

Fig. 1 Spacial structures of Al-Cu intermetallics: (a) Al_2Cu ; (b) AlCu ; (c) Al_4Cu_9 ; (d) Al_3Cu_4

扩散反应过程如图2所示,生成的界面反应层厚度遵循公式^[16]:

$$x = k_0 \sqrt{t} \exp\left(-\frac{Q}{RT}\right) \quad (2)$$

式中: k_0 为反应常数; t 为反应时间。界面反应时间越长,温度越高,越利于扩散层厚度增加。KIM等^[17]在340~500 °C区间范围内研究了Al/Cu界面扩散行为,当温度升至400 °C时,扩散层厚度显著增加, Al_4Cu_9 的增长速率高于 Al_2Cu ,低于400 °C烧结温度下,采用粉末冶金法合成的Al-Cu复合体具有更好的力学和热学性能。HAN等^[18]研究了等温550 °C条件下Al/Cu扩散结合对界面演变的影响,在真空和氩气的保护作用下,增加结合时间为15~25 min,界面区域生成 Al_4Cu_9 、CuAl、 Al_2Cu 等金属间化合物,相似的结论在ZARE等^[19]对Al/Cu的复合铸造研究中也有报道,且从Cu侧向Al侧依次为 Al_4Cu_9 、CuAl、 Al_2Cu ,在这些相的附近形成了 $\alpha(\text{Al})$ 和 Al_2Cu 的共晶组织。为了提高材料的混合性,低温短时内在界面处获得均匀的组织,ZAHERI等^[20]将Cu、Al(+Fe纳米颗粒)、Zn等粉末混合,过渡层区域获得了更薄、更均匀的金属间化合物层。ZHAO等^[21]采用浸镀的方式将镍镀在纯铝

基体上,在450~550 °C范围内用扩散复合的方法制备Al/Cu双金属材料,Ni中间层有效消除Al-Cu金属间化合物,Al/Ni界面由 Al_3Ni 和 Al_3Ni_2 组成,Ni/Cu界面处为Ni-Cu固溶体,复合材料的拉伸剪切强度有所提升。虽然,在Al、Cu界面结合处添加中间层能够抑制Al-Cu化合物的形成,但中间层的厚度、保温区间和保温时间对Al/Cu复合体力学性能的影响规律有待探究。

2 Al/Cu 复合结构工艺及界面组织性能

2.1 轧制法

轧制法是制备复合板材最常用的方法,受轧制力的影响,双金属产生塑性变形,导致金属接合表面氧化层破裂,新鲜的金属由裂口暴露出相互嵌入形成稳固接合,具有成本低、设备简单的优点。KIM等^[22]采用轧制的方法制备了Cu/Al/Cu层复合结构,Cu、Al层的相互作用变形促进界面强力结合,其伸长率高于纯铜板和纯铝板,相似的研究结论在LIU等^[23]的研究中也有报道,Cu和Al复合体

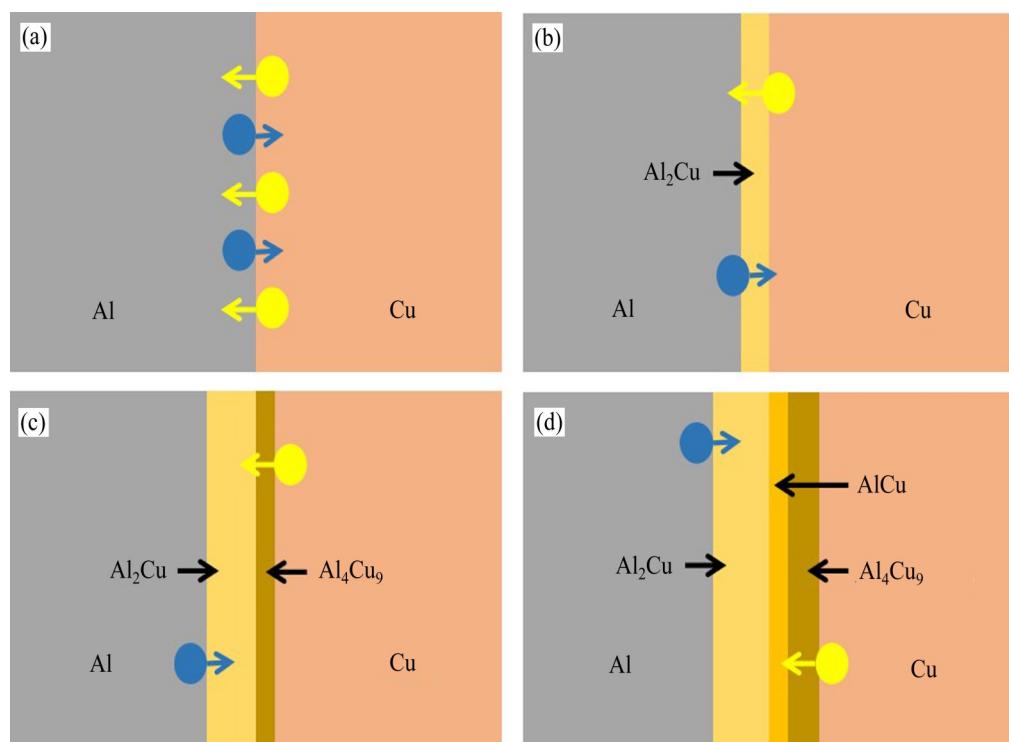


图2 Al/Cu界面扩散反应示意图

Fig. 2 Schematic diagram of Al/Cu interface diffusion reaction: (a) Atom interface; (b) $2\text{Al}+\text{Cu}\rightarrow\text{Al}_2\text{Cu}$; (c) $2\text{Al}_2\text{Cu}+7\text{Cu}\rightarrow\text{Al}_4\text{Cu}_9$; (d) $5\text{Al}_2\text{Cu}+\text{Al}_4\text{Cu}_9\rightarrow14\text{AlCu}$

的协同变形效应使得拉伸强度和伸长率均有升高。由于轧制过程中会出现加工硬化, 常需要进行退火热处理, 加速原子重排和减少晶格缺陷, 提高材料加工性能^[7]。CHEN等^[5]研究了冷轧双金属Al/Cu板的结合强度, 随着烧结温度和时间的增加, 结合强度先升高后降低, 扩散层呈现明显的层状分布, 主要有 Al_2Cu 、 AlCu 、 Al_3Cu_4 、 Al_4Cu_9 等相。LI等^[24]采用异步叠扎法制备Al/Cu/Al复合薄板, 350 °C退火后界面发生元素扩散, 受不同轧辊转速的影响, 累积的变形能使Cu/Al界面优先发生扩散, 400 °C退火后产生大量的金属间化合物, 三点弯曲变形过程中出现裂纹和分层。陈剑虹等^[15]采用累积叠扎结合多次退火处理的方式研究了Al/Cu界面反应的扩散机制, 由于Cu的扩散激活能高于Al, 退火过程中Cu原子向Al侧扩散, 热力学不稳定的界面从Al侧至Cu侧生成 $\text{Al}_2\text{Cu}\rightarrow\text{AlCu}\rightarrow\text{Al}_4\text{Cu}_9$, 叠扎道次增加使Al基体晶粒持续细化, 8次道间退火促进部分Cu原子固溶在Al晶格点阵中, 复合板的抗拉强度高达176.8 MPa, 但伸长率较差。

2.2 爆炸复合法

爆炸复合法于1944年由美国学者卡尔提出,

利用爆炸形成的能力使待复合的金属板发生高速碰撞, 导致金属板间发生相互挤压摩擦, 金属发生塑性变形和原子间发生扩散反应形成牢固的冶金结合。王乐等^[25]采用爆炸复合工艺制备了Al/Cu板材, 波形结合界面存在部分锯齿状结构。CARVALHO等^[26]研究了不同爆炸焊接条件下Al/Cu结合的可焊接性, 当采用Al作为飞板焊接时, 更容易发生界面连续熔化。KUZ'MIN等^[27]利用超声辅助爆炸焊的方法研究了Al/Cu接头的组织和性能, 熔融金属的含量显著减少, 界面呈现直线型轮廓, 焊接接头强度增加。爆炸复合法难以形成硬脆性金属间化合物, 不会产生明显的扩散, 但稳定性较差, 存在污染严重、安全性低等问题。

2.3 超声焊接法

相比于激光连接方法, 固态焊接法能够制备高质量的Al/Cu异质接头^[28-29]。作为一种固态焊接法, 超声焊接具有高效、变形小等优点, 可实现异种金属材料的连接^[30-31]。ZHANG等^[32]采用超声焊接方法研究了Al/Cu接头的界面组织和物相, 除了机械互锁结构外, 在约10 nm的过渡层中发现了非晶相和纳米晶相, 如图3所示, 超声振动引发的固

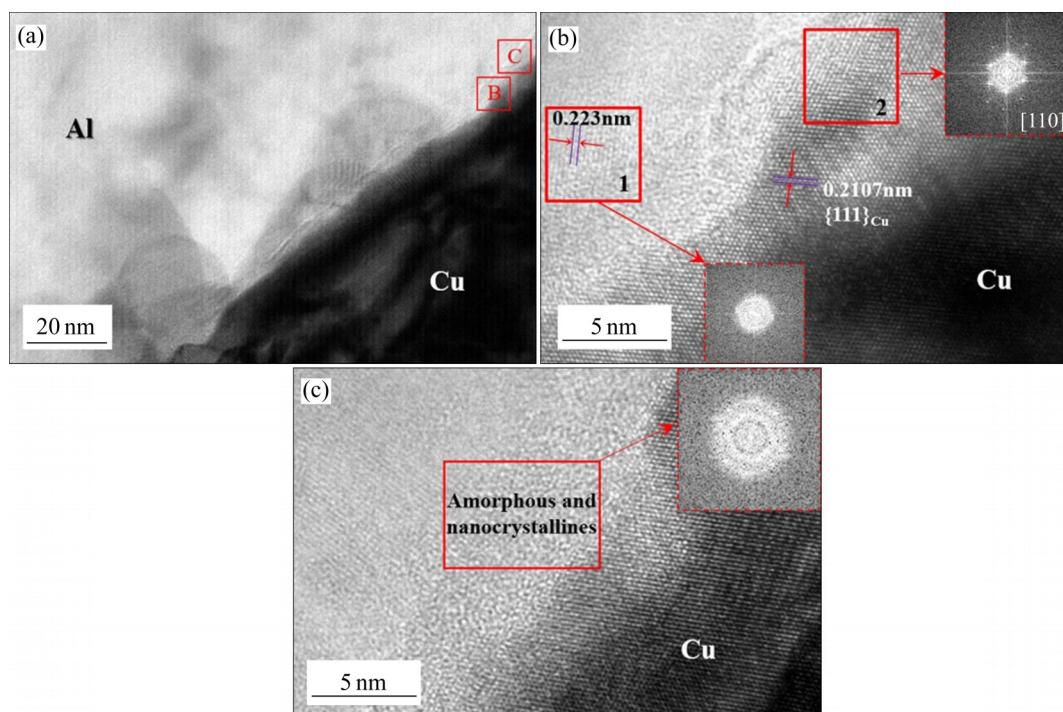


图3 Al/Cu过渡层低放大倍数的明场形貌^[32]

Fig. 3 Low magnification bright-field images of Al/Cu diffusion layer (a) and HRTEM images of transition layer taken from Fig. (a) ((b), (c))^[32]

态塑性变形增加了过渡层附近的位错和层错密度，加速了Al/Cu的元素互扩散。

MA等^[33]研究超声焊接Al-Cu接头的动态焊接演化过程，如图4所示，首先在Al侧高的表面凸起处形成微焊缝，随后在界面处的连续相对摩擦诱发剧烈的塑性变形，导致微焊缝交替的断裂和形成、微凸起的平整化、Al向Cu表面附着，当Cu侧表面凸起完全嵌入Al基体中，界面处主要为互锁模式。熊志林等^[34]对超声波焊接Cu/Al接头的界面反应行为开展研究，较小的焊接能量形成扩散反应区，焊接能力增大，Al/Cu界面处发生变形，形成高密度位错累积，元素快速扩散，逐渐析出Al₂Cu和Al₄Cu₉相，继续增大焊接能量使反应层变厚。LIU等^[35]采用电流辅助超声焊接的方法制备了Al/Cu异质材料，相比于超声焊接方法，沿着焊接界面处形成更薄的金属间化合物层，获得了更高的拉伸强度和断裂能，且界面过渡层中形成Al₂Cu相，与FUJII等^[36]的研究结论相似，在剧烈变形区域内，Cu向Al中的扩散速率增加。但界面处形成的连续金属间化合物会降低接头焊接强度，无法满足力学性能的要求^[10, 37]。LI等^[38]使用Zn箔片作为中间媒介，采用超声点焊的方式在Al/Cu接头处获得Cu₅Zn₈、Al₂Cu两种金属间化合物，抑制了其它Cu/Al金属间化合物的形成，轴承加载能力显著提高。

上述工艺方法虽然能实现Al/Cu材料的复合成型，然而存在坯料制备过程复杂、工艺流程长、界面结合性能差等问题。为了实现Al/Cu复合材料大规模、高效率、低成本的生产需求，北京科技大学谢建新课题组^[39-40]采用连续定向凝固技术制备Cu-12%Al合金线材，已实现工业化生产，同时指出，

连续铸造工艺是生产复合板的主流方向^[41]。WANG等^[42]采用水平连续复合铸造的技术制备了Cu-Al双金属复合板，如图5所示，并研究了轧制变形行为和界面演化，为了避免分层开裂，冷轧条件下第一道次要求压下率超过60%，当温度升至200~300℃时，压下率可降至55%，最终拉伸强度达到180~191 MPa、伸长率达到8%~12%。然而，传统工艺只能实现简单结构的复合制备，对于复合材料三维空间复杂结构的制造受到严重制约。

2.4 激光增材制造法

增材制造技术具有强大的构建功能，在空间中，具有选择性地放置多材料的能力。作为增材制造技术中的一种，以激光为热源、粉末为原材料的粉床熔融技术能够直接成型出无需后处理、高致密度、表面质量优异、尺寸精度高的结构件，由于凝固速率高达10⁶~10⁸ °C/s^[43]，获得的组织细小，层间结合性强，力学性能与锻件相当^[44]，金属粉末原材料可以回收再利用，节省成本，对于复杂结构具有高度制造柔性，根据规划路径选择性熔融，能够快速制造空间复杂结构，包括多孔、栅格、薄壁等^[45]。激光粉床熔融技术能够实现钛合金^[46-47]、高温合金^[48-49]、铝合金^[50-51]及铜合金^[52-53]的高性能一体化成型，已成功应用于航空航天、船舶海工、生物医疗、汽车工业等高精尖领域。随着不同材料成型工艺的日趋成熟，基于零件整体性能优化提升为目的，突破传统制造技术材料复杂性、层次复杂性和功能复杂性上的瓶颈，近年来研究广泛。新布伦瑞克大学的SHAKERIN等^[54]将热作模具钢块固定在成形基板上，采用EOSM290设备在模具钢(H13)

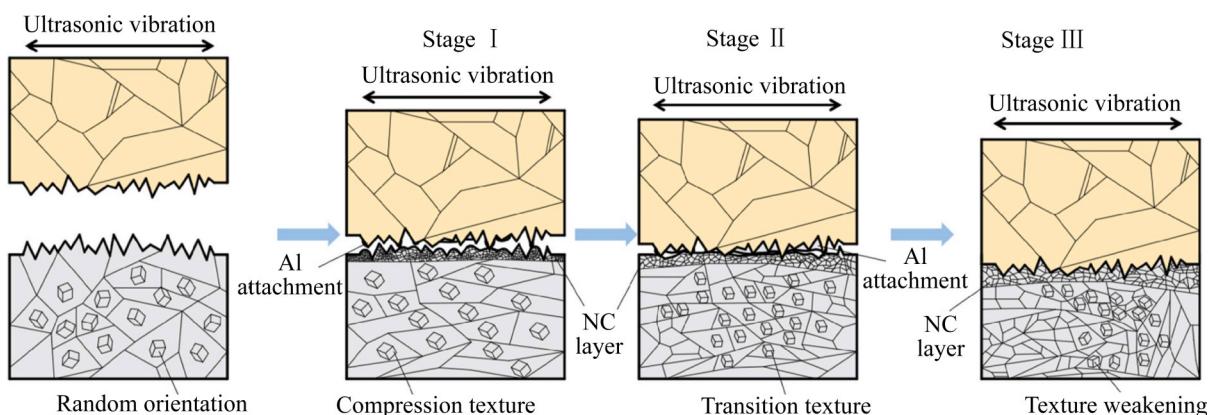


图4 超声焊接Al-Cu接头焊缝动态演变示意图^[33]

Fig. 4 Schematic diagram of dynamic weld evolution of ultrasonically welded Al-Cu joints^[11]

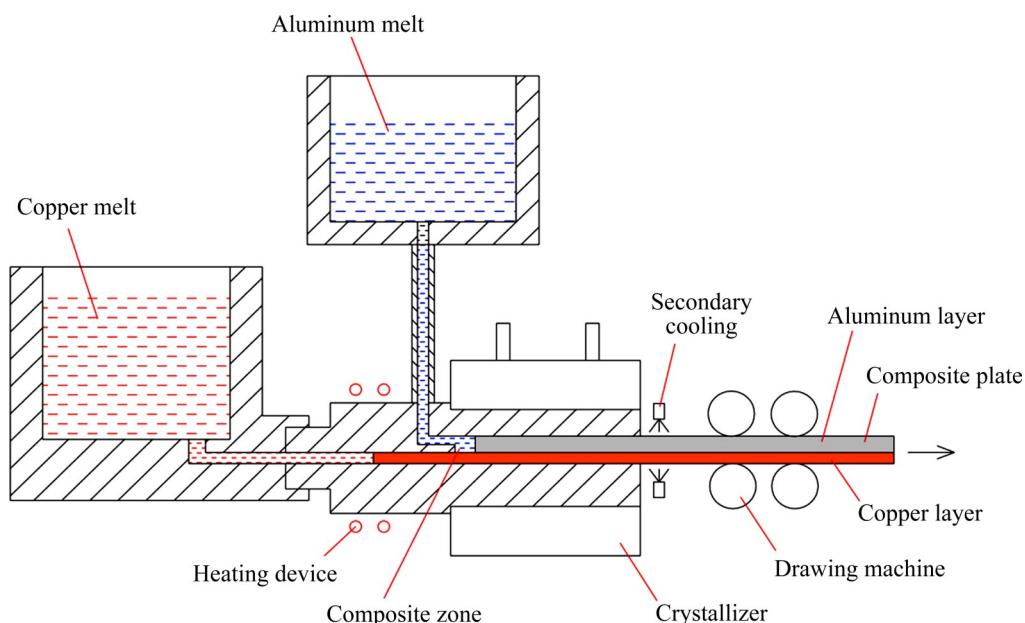


图5 水平连续复合铸造制备Al-Cu复合板的示意图^[42]

Fig. 5 Schematic diagram of horizontal continuous composite casting for fabricating Al-Cu composite plate

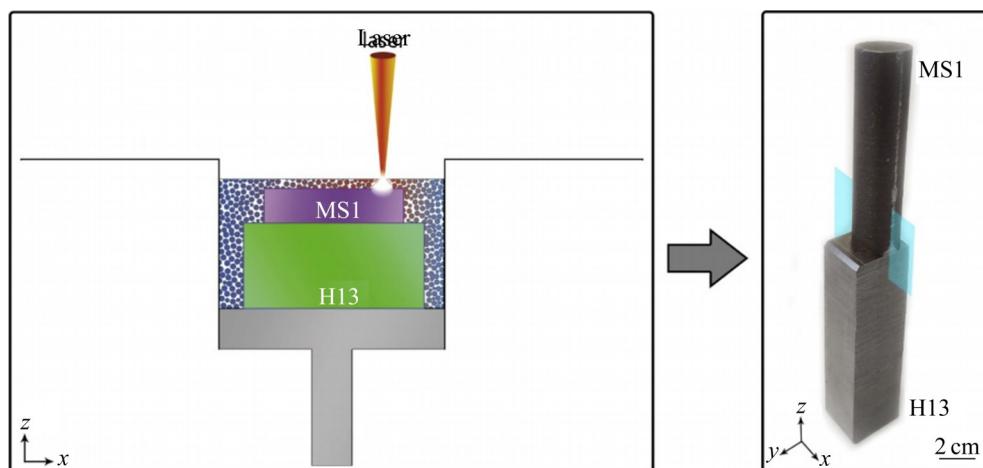


图6 LPBF技术制备MS1-H13双金属结构示意图^[54]

Fig. 6 Schematic diagram of MS1-H13 bimetals manufactured by LPBF^[54]

表面成形马氏体钢(MS1)粉末,如图6所示,界面处无裂纹且连续,证明了激光粉床熔融技术成形多种材料的可行性。英国曼彻斯特大学WEI等^[55]基于LPBF和LENS技术原理,发展一种以超声振动为基础的精密送粉装置,如图7所示,已成功制备Cu/钢异质合金结构。然而,该装置成型空间小,对于大型尺寸零件的加工存在耗时长、剩余粉末无法分离回收等问题。

近年来,王迪等^[56]致力于采用激光粉床熔融技术成型多金属材料,通过工艺调控实现异质材料界

面的性能控制。SING等^[57]采用粉床熔融技术制备了Al/Cu复合结构,界面处形成Al₂Cu金属间化合物,未发现其他相,但由于两种材料的热膨胀系数不同,快速凝固过程中在界面处产生裂纹,受较高的反射率和热导率的影响,铜侧形成一定量的气孔。因此,需要采取合适的工艺调控途径降低缺陷。GUAN等^[58]通过有限元模拟结果和基于粉床熔融成形工艺优化途径,制备了界面处无裂纹的Al/Cu双金属结构,如图8(a)~(d)所示,并结合柯肯达尔效应解释了元素间的扩散行为。基于超声焊接工

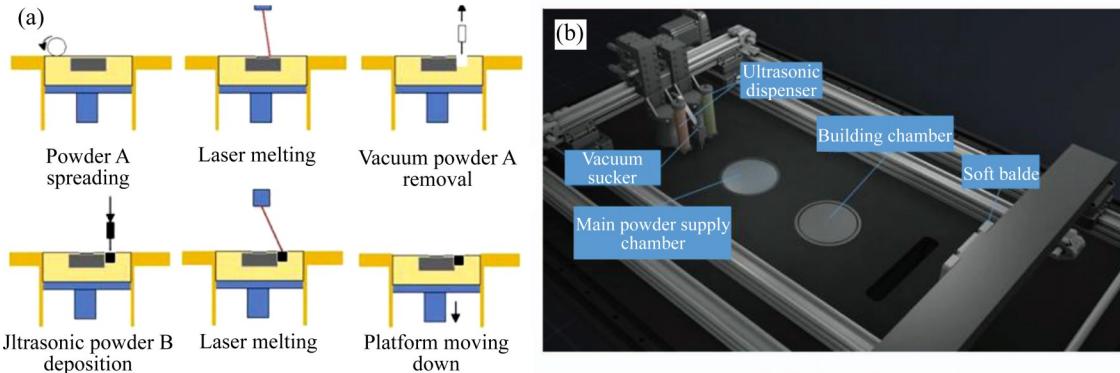


图7 多材料成型过程示意图以及装置结构示意图^[55]

Fig. 7 Schematic diagram of multi-materials process(a) and relevant schematic diagram of setup(b)^[55]

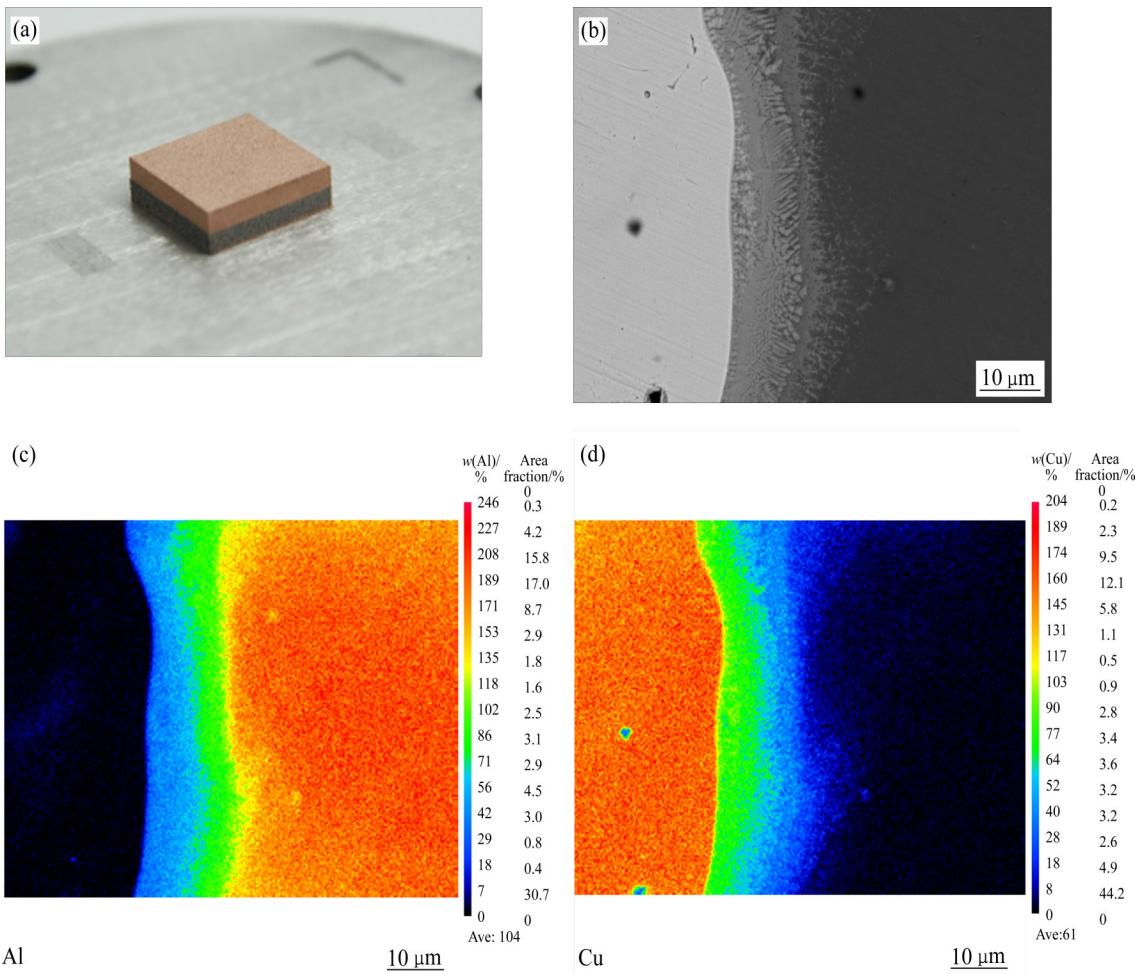


图8 采用激光粉床熔融技术制备的Al/Cu双金属复合结构^[58]

Fig. 8 Al/Cu bimetals composite fabricated by laser powder bed fusion technique^[58]: (a) Macro appearance; (b) Interface microstructure; (c) Al element distribution; (d) Cu element distribution

艺原理，美国航天局(NASA)喷气推进实验室(JPL)提出“超声波增材制造”，连续焊接金属片，结合增材和减材处理能力，实现铝-铜多材料一体化制造，并已用于探索如何生产高效的热交换器。

3 未来研究方向

1) 目前，基于Al/Cu复合结构制备工艺的探索

主要集中于讨论扩散引起的金属间化合物形成、界面层厚度及力学性能变化等方面, 缺乏对Al/Cu复合材料界面微域变形行为的深入研究。CHEN等^[59]采用分子动力学方法研究了压缩加载条件下Al/Al₂Cu/Cu的界面特性和塑性变形机制, 在Cu(111)/Al₂Cu(110)界面产生的位错阻碍了变形过程, 导致界面构型呈现韧性断裂特征。实际上, 不同保温时间下将生成其他Al-Cu金属间化合物, 从原子层次上考虑, 不同物相晶格晶面的相互作用对于变形行为影响的研究有待系统阐述。

2) 从抑制金属间化合物生成、控制界面层厚度角度出发, 在不引入外部干预和中间层媒介的条件下, 采用激光增材制造技术可以获得最少的Al-Cu相, 且能满足零件结构形状、尺寸精度的要求。但受Al、Cu热物理性能的影响, 需要严格控制成型工艺, 降低界面处形成的缺陷, 保证良好的冶金结合。

3) 现阶段, 国内基于粉床熔融技术的设备只能成型同一种粉末材料, 一旦开舱换粉将影响成型的连续性, 不利于性能的保障。因此, 厥需开发多材料的成型装备, 合理规划软硬件的协同加工方式, 为Al/Cu复合结构的制备提供新技术支持并奠定新理论基础。

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Research progress of preparation technique and interface microstructure evolution of Al/Cu composite materials

GUAN Jie-ren, WANG Qiu-ping

(Research Center of Additive Manufacturing, Marine Equipment and Technology Institute, Jiangsu University of Science and Technology, Zhenjiang 212003, China)

Abstract: Cu/Al composite structure is typical bimetallic composite material that combine pure Al and pure Cu mechanically or metallurgically by specific method. Due to the characteristics of excellent conductivity, light weight and low price, it has become the focus of extensive research. The physical properties and kinds of intermetallic compounds in Al/Cu composites were introduced, and the effect factors of interface diffusion reaction behavior were illustrated. The preparation, interface microstructure and performances of Al/Cu composite were reviewed. The research direction of composite interface and the improvement direction of Al/Cu composite structure prepared by additive manufacturing were prospected. It provides key theoretical basis and technical support for the preparation of Al/Cu composite structure.

Key words: Al/Cu composite; diffusion; intermetallic compounds; preparation technique; interface; laser additive manufacturing

Foundation item: Project(20KJD430005) supported by the Natural Sience Foundation for Colleges and Universities of Jiangsu Province, China; Project(HZ20200006) supported by the Self-cultivation Project of Jiangsu University High-tech Ship Collaborative Innovation Center, China

Received date: 2021-09-01; **Accepted date:** 2021-10-18

Corresponding author: WANG Qiu-ping; Tel: +86-13333856825; E-mail: a21096@163.com

(编辑 李艳红)