MANUFACTURING TECHNOLOGIES OF Cu-Al BIMETALLIC COMPOSITES (REVIEW)

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Copper-aluminium composites are widely used in the power transmission, telecommunications, aviation, automotive and electronics industries. The review is devoted to the analysis of manufacturing technologies of bimetallic copper-aluminium composites. It is shown that two metallurgical methods for the production of Cu–Al composite blanks have been used — continuous casting with vertical core filling and continuous casting (VCFC) with horizontal core filling (HCFC). The last of the methods is most suitable for industrialization. The features of phase transformations at the Cu/ Al interface during casting of bimetallic composites, their thermomechanical deformation, and welding are considered. It is shown that the interface between the Al-core and the Cu-shell is a multilayer structure consisting of layers of phases γ_1 (Cu₃Al₄), δ (Cu₃Al₂), η_2 (CuAl), ε_2 (Cu₃Al₂+x), θ (CuAl₂), and eutectic α (Al)/ θ (CuAl₂) layers. For the production of copper-clad aluminium (CCA) wire from bimetallic blanks, methods of rolling, drawing and welding are used. The final goal of the review is to determine the initial data and technical solutions for the development of an effective technology for butt welding of copper-aluminium blanks in the production of CCA composites. 32 Ref., 1 Tabl., 6 Fig.

Keywords: copper-aluminum composite, copper-clad aluminum, continuous casting, Cu/Al interface, intermetallic layer, eutectic

Introduction. The review is devoted to the analysis of metallurgical technologies for the production of CCA composites, the study of diffusion processes, the microstructure formation, and phase transformation of the Cu/Al al interfacial boundary during production process, heating, deformation, and welding. With the development and distribution of CCA composites, the problem arose of reliable welding of CCA blanks before the operation of drawing the wire billets to a given diameter. Particularly urgent problem is the need to weld billets of Cu–Al composite metals during CCA wire production – on cold drawing lines, where the welded joint is subjected to high mechanical stress and plastic deformation. The complexity of reliable welding of CCA composites is associated with the possibility of delamination of the aluminium core and the copper cladding layer, as well as the fact that when heated, aluminium and copper enter into a chemical reaction with the formation of brittle intermetallic layers. The final goal of the review is determination of the initial data and key technical solutions for the development of efficient technology and equipment for welding of copper-aluminium composite cable joints.

A significant number of publications are devoted to the study of technologies for the production of CCA composites, as well as, the study of its properties and structure, diffusion processes at the "copper-aluminium" interface during thermal, deformation and welding. These works provide information on various aspects of the problem of obtaining CCA composites, including plates, rods, and wires, in particular:

1. Analysis of the effectiveness of various (metallurgical and thermo-mechanical) technologies for the production of CCA-composites;

2. Study of diffusion processes and the formation of the Cu/Al interfacial boundary in the bimetal under various temperature-time conditions for the production of heat treatment and operation of the CCA;

3. Numerical modelling of the processes of formation of intermetallic compounds (IMC) during rolling and annealing of CCA-composites;

4. Study of the process of formation of the Cu/Al interface during welding and subsequent heat treatment of CCA composites.

Let's take a look at some of these publications.

CCA-composites have been widely used in the fields of power transmission, signal transportation and energy transfer, in aviation, petroleum, chemical, shipbuilding, automobile, and electronics [1‒8], and other industrial fields. Standard [1] specifies the dimensions, electrical characteristics and mechanical characteristics of aluminium — based and copper-clad aluminium conductors, for lightweight aircraft electrical cables and aerospace applications. It applies to

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stranded conductors over the nominal cross-sectional area range 0.22 mm^2 to 107 mm^2 inclusive. Standard [2] covers bare round copper-clad aluminium wire for electrical applications.

At present, one of the most common method of production of layered metal composites is a roll casting composite method. In addition, some other methods, such as the electromagnetic continuous rolling [8, 9], friction pressure welding [10], ultrasonic welding [11], and overlay welding method [12], and others, have also been studied and used in some development applications. However, for Cu/Al composite panels, the roll casting method has more advantages because of its high security and low energy cost. Wide, semi-solid, and whole roll casting can be achieved through roll casting method for Cu/Al composite panels, without any change in the original good electrical conductivity of the copper and aluminium, and the Cu/Al composite panels are a metallurgical combination, which can further improve the bonding strength of Cu/Al composite materials.

There are two main roll casting composite methods — a vertical core-filling continuous casting (VCFC) [7] technology and horizontal core-filling continuous casting (HCFC) [9‒12] technology. HCFC technology is not only convenient for obtaining an impurity-free interface but is also suitable for industrialization. When preparing bimetallic composites by VCFC or HCFC technology, the dissimilar liquid metals were cast and bonded simultaneously. The bonding process was realized when liquid core metal filled into the presolidified metal sheath and solidified. Therefore, HCFC technologies can be classified into the category of liquid-solid bonding technology.

In paper [7] CCA rods with a diameter of 12 mm and a sheath thickness of 2 mm were fabricated by self-developed VCFC device (fig. 1). Fig. 2 shows the macro-morphology of a CCA rod fabricated by VCFC. It can be seen that Cu coating with dense

Fig. 1. Schematic diagram of vertical core-filling continuous casting for CCA composites: *1* — graphite crucible for aluminium; *2* — liquid aluminium; *3* — runner; *4* — graphite crucible for copper; *5* — liquid copper; *6* — heat insulating material; *7* crystallizer; *8* — secondary cooler; *9* — pinch rolls; *10* — CCA composites

structure and uniform thickness was obtained by this technology. Based on the theoretical analysis and the previous experimental results, the main processing parameter ranges for the experiments were determined as follows: copper casting temperature $T_{\text{Cu}} = 1250 \text{ °C}$, continuous casting speed $v = 60$ mm/min, secondary cooling located at 75 cm beneath the crystallizer.

The phase compositions of the fracture surface and Cu/Al interface were identified using X-ray diffractometry (XRD) and transmission electron microscopy (TEM), respectively. Fig. 3 shows the SEM image of interfacial microstructure of CCA composite. It can be seen that there was a 250 μm-thick interface between the Cu sheath and Al core. Along the radial direction from the Cu sheath to the Al core, the interface can be

Fig. 2. Macro-morphology of CCA rod

ISSN 2415-8445 СУЧАСНА ЕЛЕКТРОМЕТАЛУРГІЯ, № 3, 2023 49

Fig. 3. SEM image of interface in CCA composites

divided into three sublayers with different thicknesses and morphologies. The contents of Cu and Al at the interface were measured by EDS analysis. The results are tabulated in Table.

The interfacial mechanism of formation of aluminium composites with a copper sheath was studied. The results showed that the interfacial structure of Cu/ Al was mainly composed of layered γ_1 (Cu₉Al₄), cellular θ (CuAl₂), and α (Al)+ θ (CuAl₂) phases. Sublayer I with little thickness was predicted to be layered γ_1 (Cu₉Al₄) phase. Sublayer II with large thickness was predicted to be cellular θ (CuAl₂) phase. In sublayer II, acicular compounds were predicted to be residual ε_2 (Cu₃Al₂+*x*) phase corresponding to the composition analysis result. Sublayer III had a distinct feature of lamellar eutectic morphology.

Fig. 4, *а* shows the relation that Cu/Al interface temperature varies with time. It can be seen that the cooling rate of Cu/Al interface was very fast. Therefore, the interfacial bonding of Cu/Al was a continuous cooling solid−liquid reaction process, and the formation of Cu/Al interfacial layer was a result of the comprehensive effect of interfacial reaction-diffusion and rapid solidification.

Results of EDS analysis of interface in CCA composites

Position	x (Cu), %	x(Al), %	Predicted phase
A	99.43	0.57	α (Cu)
B	62.88	37.12	γ_1 (Cu ₉ Al ₄)
\mathcal{C}	53.36	46.64	ε_2 (Cu ₃ Al _{2+x})
D	35.24	64.76	θ (CuAl ₂)
E	21.96	78.04	α (Al)+CuAl,
F	1.79	98.21	α (Al)

Fig. 4, *b* shows the Cu−Al binary phase diagram. *T*1 is the initial contact temperature between solid Cu and liquid Al (about 700 °C), *T*2 is the melting point of Al (660.452 °C), *T*3 is the temperature of peritectic reaction $L + \varepsilon_2 = \eta_1 (624 \text{ °C})$ and 74 is the temperature of peritectic reaction of $L + \eta_1 = \theta$ (592 °C). Liquid Al flowed from the Al honeycomb duct to contact with the inner wall of solid Cu tube at *T*1.

The diffusion of Al atoms to Cu matrix led to a rapid dissolution of Cu, forming a thin liquid diffusion layer (DL) at the Cu/Al interface. Combined with the binary phase diagram of Cu−Al (fig. 4, *b*), the phases of ε_2 (Cu₃Al₂+x), γ_1 (Cu₉Al₄) and β (Cu₃Al) could be generated at the temperature *T*1. Due to the slow growth rate of β phase and the short contact reaction time of Cu/Al, β (Cu₃Al) phase could be ignored. Moreover, residual acicular ε_2 (Cu₃Al₂+x) phase was observed at the Cu/Al interface.

Based on the studies carried out in [7], the conclusions:

1) According to the analysis results of SEM, XRD and TEM, the interfacial structure of CCA composite contained three sublayers: Sublayer I was layered $\gamma_1({\rm Cu}_9{\rm Al}_4)$ phase, Sublayer II was cellular θ (CuAl₂) and acicular residual ε ₂(Cu₃Al₂+x)

Fig. 4. Relationship between temperature of Cu/Al interface and time (*а*) and Cu−Al binary phase diagram (*b*)

Fig. 5. Schematic diagram of the HCFC device for preparing CCA rod: (*a*) device and (*b*) composite mold: *1* — molten aluminum holding furnace; 2 — molten copper holding furnace; 3 — runner; 4 — composite mold holding furnace; 5 — composite mold; 6 secondary cooling; *7* — pinch rolls; *8* — CCA rod; *9* — liquid aluminium; *10* — mandrel; *11* — liquid copper; *12* — solidification front of liquid copper; *13* — solidification frontof liquid aluminum

phases, and Sublayer III was $α(AI)+θ(CuAI₂)$ pseudo eutectic structure.

2) The nucleation driving force of ε_2 (Cu₃Al₂+x) was greater than that of γ_1 (Cu₉Al₄), which indicated that ε_2 (Cu₃Al₂+x) formed firstly at the Cu side interface.

3) The γ_1 (Cu₉Al₄) phase formed through solid diffusion reaction between ε_2 (Cu₃Al₂+x) and solid Cu. The θ (CuAl₂) and θ (CuAl₂)+ α (Al) phases mainly formed by peritectic reaction and eutectic reaction, respectively.

In paper [9] CCA rods with a diameter of 30 mm and a sheath thickness of 3 mm were fabricated by horizontal core-filling continuous casting (HCFC) technology. Fig. 5 shows the schematic diagram of the HCFC device and the detailed structure of the composite mold.

The microstructure and morphology, distribution of chemical components, and phase composition of the interface between Cu and Al were characterized by scanning electron microscope (SEM), transmission electron microscope (TEM), and energy dispersive spectrometer (EDS). The formation mechanism of the interface and the effects of key processing parameters, e.g., aluminium casting temperature, secondary cooling intensity, and mean withdrawing speed on the interfacial microstructure and bonding strength were investigated. The results show that the CCA rod has a multilayered interface, which is composed of three sublayers — sublayer I is $Cu₉Al₄$ layer, sublayer II is CuAl₂ layer, and sublayer III is composed of α -Al/ CuAl₂ pseudo eutectic. The thickness of sublayer III, which occupies 92 to 99 pct of the total thickness of the interface, is much larger than the thicknesses of sublayers I and II. However, the interfacial bonding strength is dominated by the thicknesses of sublayers I and II; i.e., the bonding strength decreases with the rise of the thicknesses of sublayers I and II.

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The CCA rod with the largest interfacial bonding strength of 67.9 ± 0.5 MPa was fabricated under such processing parameters as copper casting temperature 1503 K (1230 °C), aluminum casting temperature 1063 K (790 °C), primary cooling water flux 600 L/h, secondary cooling water flux 700 L/h, and mean withdrawing speed 87 mm/min. The total thickness of the interface of the CCA rod fabricated under the preceding processing parameters is about 75 lm, while the thicknesses of sublayers I and II are about 1.1 and 0.1 lm, respectively.

In paper [10] Al–17 at.% Cu alloy, which had the same eutectic structure as the eutectic microstructure layer on the interface of Cu–Al composite plate, was prepared by changing the cooling rate of ingot solidification to substitute the eutectic microstructure layer on the interface of Cu–Al composite plate (fig. 6), and the compression deformation behavior was investigated.

The conclusions are as follows: (1) when the deformation temperature ranges from 300 to 400 °C, the softening effect of dynamic recrystallization of α-Al in the eutectic microstructure layer is greater than the hardening effect, and then the uniform plastic deformation of eutectic microstructure is caused; (2) according to the true stress–strain curves of the eutectic microstructure layer in different deformation conditions, the relationships between true strain and material parameters were obtained by polynomial fitting based on the Arrhenius hyperbolic sinusoid model, and the constitutive equation of flow stress in the eutectic microstructure layer was established.

In paper [11] copper-aluminium (Cu–Al) based lamellar composites were prepared using a solid-liquid compound casting (SLCC) technology. Characterization results showed that the Cu–Al composites were fully-sintered at 700 °C under an argon atmosphere using the SLCC technology. Cu-Al interfacial

Fig. 6. Microscopic results and element distribution of the composite interface: a, b — microstructure of the interface; c, d — EDS line sweep analysis results of *A*–*B* in (*a*) and *C*–*D* in (*b*)

bonding was uniform with a well-defined transitional and inter-diffusion region. Intermetallic compounds and solid solutions of CuAl₂, CuAl, Cu₉Al₄, CuAl₃ and Cu_3Al_2 were detected at the interfacial region. With the increase of annealing temperature, the width of the Cu‒Al interfacial region was increased, and the interfacial bonding strength was also increased, whereas the types of the intermediate phases were changed. With the increase of dwelling time at a given annealing temperature, the width of Cu–Al interfacial region was increased, the interfacial bonding strength was decreased and the mesophases were changed. The bonding strength of the as-prepared composite was 30 MPa, whereas those of specimens annealed at 200 °C for 2 h, 300 °C for 2 h, 400 °C for 2 h, 300 °C for 30 min and 300 °C for 1 h were 59, 39, 74, 56, and 49 MPa, respectively. The Cu-Al interfacial bonding mechanisms were identified to be rapid inter-diffusion of copper and aluminium and formation of interfacial and graded microstructures. The formation of copper-aluminium interface is a combined result of inter-atomic diffusion and interfacial chemical reactions, the latter of which is more dominant in the diffusion process.

In paper [12] the copper/aluminium (Cu/Al) clad sheets were produced on a horizontal twin-roll caster and then were multi-pass rolled and annealed. The thickness of the as-cast clad sheet was 8 mm. Rolling was performed with total reductions of 12.5 %, 25 %, 37.5 %, 50 %, and 62.5 %, separately. The effects of the rolling and annealing processes on the interface and peel strength of the Cu/Al clad sheets were investigated. The evolution of the interface and crack propagation were studied. The interface thickness of the as-cast clad sheet reached 9 μ m to 10 μ m and the interface was composed of three IMC layers including Al_2Cu , AlCu, and Al_4Cu_9 . The average peel strength (APS) was only 9 N/mm. After multi-pass rolling, the peel strength first slightly increased and then gradually decreased with the increase of the rolling pass number. After rolling to 7 mm and annealing at 350 °C for 2 h. After rolling to 7 mm and annealing at 350 °C for 2 h, the clad sheet had the highest APS of 25 N/mm. This process can be used as the method to improve the peel strength of the Cu/Al clad sheets The improvement in the peel strength was due to the following three factors: (1) mechanical locking formed in the Cu/Al direct contact region after rolling, (2) the region of the Al matrix fracture, and (3) mechanical biting from the Cu/Al direct contact region. In the process of multi-stage rolling, the multilayer structure of intermetallic phases and eutectic

is destroyed and dispersed, forming a Cu/Al interface without a continuous brittle layer with dispersed inclusions of intermetallic particles.

In paper [13] the solid-liquid method was used to prepare the continuous casting of copper cladding aluminium by liquid aluminium alloy and solid copper, and the interfacial phase formation of Al-Cu bimetal at different pouring temperatures (700, 750, 800 °C) was investigated by means of metallograph, scanning electron microscopy (SEM) and energy dispersive spectrometry (EDS) methods. The results showed that the pouring temperature of aluminium melt had an important influence on the element diffusion of Cu from the solid Cu to Al alloy melt and the reactions between Al and Cu, as well as the morphology of the Al–Cu interface. When the pouring temperature was 800 °C, there were abundant Al–Cu intermetallic compounds (IMC) near the interface. However, a lower pouring temperature (700 °C) resulted in the formation of cavities which was detrimental to the bonding and mechanical properties. Under the conditions in this study, the good metallurgical bonding of Al–Cu was achieved at a pouring temperature of 750° C.

In paper [14] an innovative horizontal continuous casting method was developed and successfully used to prepare copper-clad aluminium (CCA) rods with a diameter of 85 mm and a sheath thickness of 16 mm. The solidification structure and element distribution near the interface of the CCA ingots were investigated by means of a scanning electron microscope, an energy dispersive spectrometer, and an electron probe X-ray microanalyzer. The results showed that the proposed process can lead to a good metallurgical bond between Cu and Al. The interface between Cu and Al was a multilayered structure with a thickness of 200 μm, consisting of Cu₉Al₄, CuAl₂, α-Al/CuAl₂ eutectic, and α -Al + α -Al/CuAl₂ eutectic layers from the Cu side to the Al side. The mean tensile-shear strength of the CCA sample was 45 MPa, which fulfills the requirements for the further extrusion process. The bonding and diffusion mechanisms are also discussed in this paper.

In paper $[15]$ it is noted that high performance Cu– Al composites have widely applied in aviation, aerospace and other fields, at the same time the continuous casting as one of composite forming technologies has been also developed in recent years. Obviously, it is an effective and cheap way to numerically simulate the solidification process of short process continuous casting for manufacturing Cu-Al composites before fabricating them. To meet the need of simulation, in this work, a numerical method for theoretically describing the Cu–Al composite forming in continuous casting processes was proposed. The vertical continuous casting of copper clad aluminium bar billet

ISSN 2415-8445 СУЧАСНА ЕЛЕКТРОМЕТАЛУРГІЯ, № 3, 2023 —————————————————————————————53

and the horizontal continuous casting of copper and aluminium composite plate were performed. Based on this method, the steady state temperature fields in solidification processes in the above two kinds of casting technologies were numerically simulated by using pro-CAST software package.

In this work the effects of the theoretical parameters on the steady state temperature fields and then on the performance of Cu–Al composites fabricated by using the above two casting technologies were carefully discussed. It is found that the experimental and simulated results are in good agreement.

In high power automotive electronics [16] copper wire bonding is regarded as the most promising alternative for gold wire bonding in $1st$ level interconnects. In the Cu–Al ball bond interface the growth of intermetallic compounds can deteriorate the electrical and mechanical properties of the interconnection. A summary of the thermo-mechanical properties of Cu-Al intermetallic compounds is given. Delamination experiments were performed to study interfacial cracking in the Cu–Al system. Interfacial delamination initiates in the Al-rich intermetallics (CuAl, CuAl₂) and propagates easily into other intermetallic layers. The $Cu₉Al₄-Cu_{s.s.}$ is also found to be susceptible for delamination fracture. To quantify the corrosion and oxidation sensitivity of Cu–Al intermetallics 8 different aging experiments under various conditions (Δ*t*, AT, air or halogen-rich solution or combined) were performed. In all cases the oxidation/corrosion attacks at the non-equilibrium Cu_{ss} — interface with the Cu–Al intermetallics is more severe than of a homogeneous bulk solid solution of aluminium in copper (with more than $5 \text{ wt.} \%$ Al).

In paper [17] the copper wire bonded chip samples were annealed at the temperature range from 150 °C to 300 °C for 2 to 250 h, respectively. The formation of Cu/Al IMC was observed and the activation energy of Cu/Al IMC growth was obtained from an Arrhenius plot (ln(growth rate) versus 1/T). The obtained activation energy was 26 Kcal/mol and the behaviour of IMC growth was very sensitive to the annealing temperature. Determined that the reaction rate of Cu/ Al IMC formation is 100 times slower than that of Au/ Al IMC formation.

The phase diagram of Cu–Al system shown in fig. 4 identifies the possible IMCs formed between copper and aluminium. Cu/Al IMC phases which formed at the temperature range between 150 to 300 are as follows: γ₂-phase (Cu₉Al₄) — 69.2 at.% Cu; δ-phase $(Cu_3 Al_2)$ — at.% Cu; ζ_2 -phase $(Cu_4 Al_3)$ — 57.1 at.% Cu; $η_2$ -phase (CuAl) — 50.0 at.% Cu; θ-phase $(CuAl₂)$ — 33.3 at.% Cu.

To investigate the effects of IMC formation on the copper wire bondability on Al pad, ball shear tests were performed on annealed samples. For as-bonded samples, ball shear strength ranged from 240–260 gf, and ball shear strength changed as a function of annealing times. For annealed samples, fracture mode changed from adhesive failure at Cu/Al interface to IMC layer or Cu wire itself. The IMC growth and the diffusion rate of aluminium and copper were closely related to failure mode changes. Micro-XRD was performed on fractured pads and balls to identify the phases of IMC and their effects on the ball bonding strength. From XRD results, it was confirmed that the major IMC was $Cu₉Al₄$ and it provided a strong bondability.

It is known that in the production of dissimilar metals composites and their joining, pressure welding methods can also be used, namely: friction welding [18-21], resistance butt welding [22, 23], flash butt welding [24, 25], hybrid butt welding, involving a combination of resistance heating, flashing, and upsetting [26, 27]. To obtain dissimilar joints, explosion welding, and magnetic pulse welding are also used [28–32]. Analysis of the features of these methods in the production of copper-aluminium composites is planned in the next review.

Conclusions

1. The production of copper-aluminium composites is based on a combination of several technologies, namely: composite continuous casting of liquid aluminium on solid copper, multi-stage rolling, drawing, and heat treatment.

2. There are two main roll casting composite methods — a vertical core-filling continuous casting technology and horizontal core-filling continuous casting (HCFC) technology. HCFC technology is not only convenient for obtaining an impurity-free interface but is also suitable for industrialization.

3. HCFC technology allows ensuring good metallurgical bond between copper and aluminium. The interface between Al-core and Cu-sheath is a multilayered structure, consisting of γ_1 (Cu₉Al₄), δ (C u_3 Al₂), η_{2} (CuAl) ε_2 (Cu₃Al₂+*x*), θ (CuAl₂), and eutectic α (Al)/ θ (CuAl₂) layers.

4. In the process of multi-stage rolling, the multilayer structure of intermetallic phases and eutectic is destroyed and dispersed, forming a Cu/Al interface without a continuous brittle layer with dispersed inclusions of intermetallic particles.

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тЕХНологіЇ вИготовлЕННЯ БіМЕталіЧНИХ КоМПоЗИтів Cu‒Al (оглЯД) I.B. Зяхор¹, A.O. Наконєчний¹, Wang Qichen², Linyu Fu³, B.B. Кольцов⁴

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Мідно-алюмінієві композити широко використовуються у галузях передачі електроенергії, телекомунікацій, в авіації, автомобільній промисловості та електроніці. огляд присвячено аналізу технологій виготовлення біметалічних мідно-алюмінієвих композитів. Показано, що переважно використовуються два металургійні способи виробництва заготовок Cu–Al композитів — безперервне литво з вертикальним заповненням серцевини (VCFC) та безперервне литво з горизонтальним заповненням серцевини (HCFC). останній із способів найбільше підходить для індустріалізації. Розглянуто особливості фазових перетворень на міжфазній границі Cu/Al під час литва біметалічних композитів, їх термомеханічної деформації, зварювання. Показано, що границя розділу між Al-серцевиною і Cu-оболонкою є багатошаровою структурою, що складається з шарів фаз $\gamma_1({\rm Cu}_9{\rm Al}_2)$, $\delta({\rm Cu}_3{\rm Al}_2)$, η₂(CuAl), ε₂(Cu₃Al₂+x), θ(CuAl₂) і евтектики α(Al)/θ(CuAl₂). Для виробництва із біметалічних заготовок покритого міддю алюмінієвого дроту (copper-clad aluminium (CCA) wire) використовують способи прокатки, витяжки і зварювання. Кінцева мета огляду — визначення вихідних даних і технічних рішень для розробки ефективної технології стикового зварювання мідно-алюмінієвих заготовок при виробництві сса композитів. Бібліогр. 32, табл. 1, рис. 6.

Ключові слова: мідно-алюмінієвий композит, покритий міддю алюміній, безперервне литво, Cu/Al міжфазна границя, інтерметалідний шар, евтектика

Надійшла до редакції 31.08.2023