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Investigation on contact melting of Cu/Al laminated composite

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ABSTRACT: The study presents investigation of chemical composition, microhardness and electrical conductivity of Cu/Al laminated metal composite after heat treatment at temperatures higher than Cu–Al eutectic melting point. The Cu/Al bimetal was obtained via explosion welding. Chemical composition of the material after heat treatments was identified using EDS analysis. Eddy current testing was applied to investigate electrical conductivity of the composite's components. Strain-hardened zones were identified in the explosion welded composite. The experimental value of electrical conductivity of explosion welded composite was in good coherence with calculated by additivity rule results. Heat treatments resulted in the formation of multiple interlayers which had high microhardness value and had intermetallics in composition. The electrical conductivity of the identified interlayers was significantly lower than of Cu and Al.

KEYWORDS: Al; Contact melting; Cu; Electrical conductivity; Explosion welding; Intermetallics

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RESUMEN: *Investigación sobre fusión de contacto en materiales compuestos laminados Cu/Al.* Se presentan los resultados de composición química, microdureza y conductividad eléctrica obtenidos en materiales compuestos laminados Cu/Al, tratados térmicamente a temperaturas superiores a las del eutéctico Cu - Al. El bimetal Cu/Al fue obtenido por soldadura por explosión. La composición química del material tras los tratamientos térmicos se analizó por medio de análisis EDS (Espectroscopia de Energías Dispersiva de Rayos X). La conductividad eléctrica se midió mediante ensayos con corrientes Eddy. Las zonas endurecidas en el material soldado por explosión fueron identificadas. El valor experimental obtenido de la conductividad eléctrica está en concordancia con el calculado por la regla de las mezclas. Los tratamientos térmicos dan lugar a la formación de múltiples intercaras de alta dureza, compuestos de intermetálicos. La conductividad eléctrica de las intercaras identificadas es significativamente menor que las correspondientes al Cu y al Al.

PALABRAS CLAVE: Aluminio; Cobre; Conductividad eléctrica; Intermetálicos; Fusión de contacto; Soldadura por explosión

1. INTRODUCTION

Clad metals are widely applied in a range of various industries. One of the main advantages of such materials is the ability to combine properties of their components. Often laminated metal composites have unique properties not represented in their components. Such features can usually be achieved by special design described for example by Gurevich *et al.* (2014) or the formation of new compounds, which are generally presented by intermetallics that are receiving more attention over the past decades (Morris and Muñoz-Morris, 2005). According to the research of Li *et al.* (2011) there are many techniques of joining of different metals, which can be classified into four main groups, however the most commonly applied cladding methods are explosion welding, roll bonding and laser welding.

Al/Cu clad metals are usually used as conductors of electricity and heat (Veerkamp, 1995). When such materials are subjected to heat, intermetallic compound interlayers can occur in the bond area of the composite. The impact and properties of intermetallic compound interlayers have been widely explored in previous studies. The decrease in strength properties as a result of intermetallic formation was reported by Braunovic and Alexandrov (1994); Lee *et al.* (2005); Liu *et al.* (2011); Sheng *et al.* (2011); Kim and Hong (2013); Uscinowicz (2013). Lee *et al.* (2005) have reported the decline of electrical conductivity caused by the emergence of intermetallics in Al/Cu bimetals however in the study of Acarer (2012) no significant decrease in electrical conductivity was observed after the formation of small quantities of $CuAl_2$ phase between metals. According to the study of Trykov *et al.* (2014) the decrease in thermal conductivity value is observed with the formation of intermetallics. The kinetics of diffusion processes, emerging in the composite under heat treatment have been widely investigated at a range of different temperatures by Abbasi *et al.* (2010); Guo *et al.* (2010); Wang *et al.* (2014); however the previous research usually considered temperatures of solid state diffusion processes, while the impact of higher temperatures on the bond area is insufficiently explored. The influence of high temperatures can result in the contact melting effect in the bond area. The effect can be represented by the following steps: when the material is heat treated above 548.2 °C, the solid state diffusion processes occur in thin adjacent to the bond areas of the composite. In this case the solid state diffusion continues until the solubility limit (2.48 at.% Cu) is reached, which leads to the formation of liquid phase. Subsequent processes are related to dissolution of solid components in liquid. According to the Al-Cu phase diagram, the contact melting effect in this system is observed when L $\leftrightarrow$ eutectic [(Al)+Θ]. Recent investigations on Al/Cu contact melting were reported by Bystrenko and Kartuzov (2014); Korotkov *et al.* (2014).

The contact melting significantly accelerates the interaction of components in the bond. The speed of contact melting is larger when one component is highly soluble in another. The relation of contact melting speed to temperature is given by:

$$V_{CM} = L \cdot e^{\frac{E_{CM}}{R \cdot T}} \tag{1}$$

where E_{CM} – denotes the contact melting activation energy, L – the coefficient, depending on experiment conditions, R – gas constant.

The aim of this study is to investigate the properties of Al/Cu composites treated at temperature of contact melting.

2. MATERIALS AND EXPERIMENTS

Laminated metal Al/Cu composite was obtained via *explosion welding* (EW). The flyer Al plate was in parallel arrangement with the base Cu plate. The stand-off distance between plates and explosive thickness were 1 mm and 30 mm respectively. To protect the outer surface of Al from the impact of detonation products the rubber buffer strip was placed between Al plate and explosive. The thicknesses of Al and Cu plates were 5.5 and 6.5 mm respectively. Chemical composition of heat treated before EW alloys used in this study is presented in Table 1. The detonation velocity of explosive material used in this study was 2500–2700 $m.s^{-1}$.

The explosion welded specimens were subsequently heat treated at 570 °C for a range between 0.25 and 45 h. To avoid oxidation and outflow of molten metal, the specimen were coated with a mixture of sodium silicate (70%) and talc powder (30%).

The microstructure of the composite was studied using optical microscope Olympus BX-61 and scanning electron microscope Versa-3d. Hardness measurements were carried out using standard microhardness tester PMT-3 with the Vickers indenter test with loads of 10–50 g for 15 s.

EDS Analysis was used to determine chemical composition of the bond area after heat treatments.

TABLE 1. Chemical composition of materials used in this study

	Composition (wt.%)							
Al plate	Fe <0.3	Si <0.3	Mn <0.025	Ti <0.15	Zn <0.1	Cu <0.05	Mg <0.05	Al >99.3
Cu plate	Fe <0.005	Ni <0.002	Zn <0.004	Sn <0.002	S <0.004	Pb <0.005	O <0.05	Cu >99.3

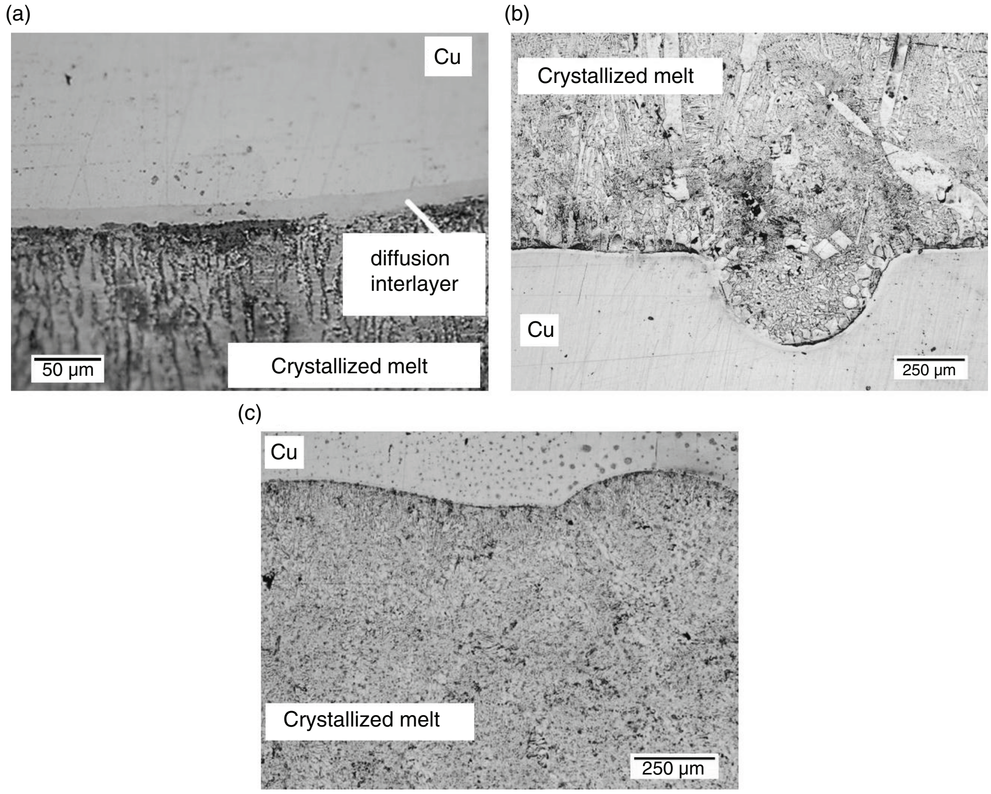

FIGURE 1. Optical images of the bond area of explosion welded and heat treated at 570 °C for (a) 0.25 h, (b) 0.5 h and (c) 2 h Al/Cu composite.

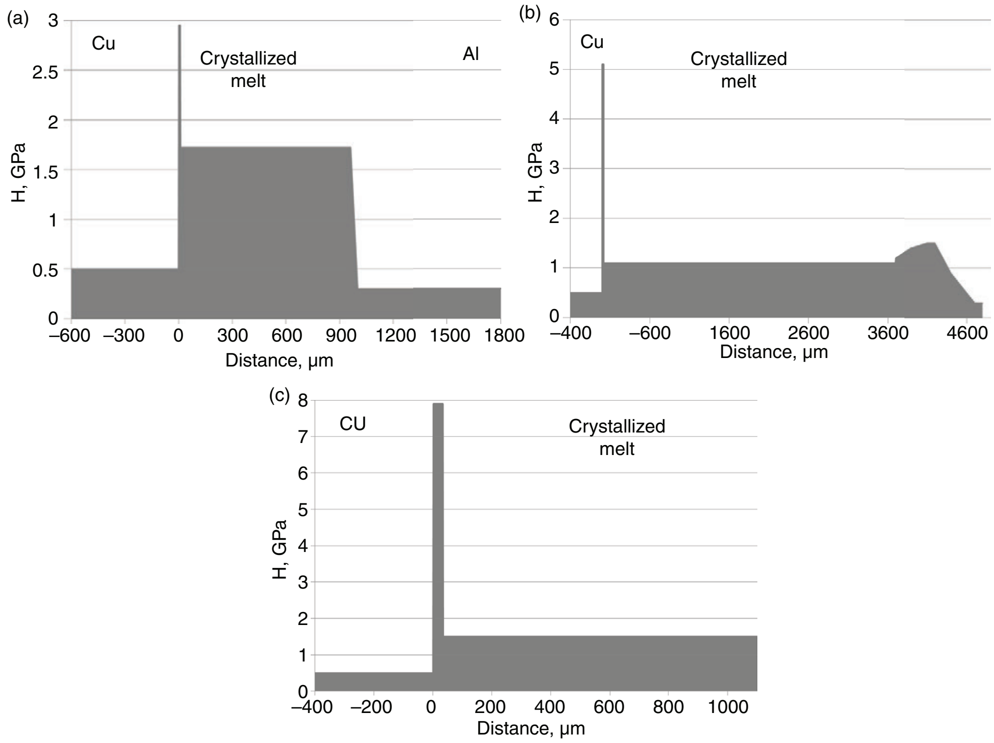

FIGURE 2. Microhardness distributions of the bond area of explosion welded and heat treated at 570 °C for (a) 0.25 h, (b) 0.5 h and (c) 2 h Al/Cu composite.

Eddy-current testing was applied to investigate the electrical conductivity of the composite. Electrical conductivity measurement device Vihr-1M was used.

3. RESULTS AND DISCUSSION

Microhardness distributions identified strain-hardened zones both in Al and copper layers of 50 and 30 µm thickness respectively. Microhardness value of Cu and Al outside of strain-hardened zones was 0.5 and 0.3 GPa respectively.

Heat treatment at 570 °C for 0.25 h contributed to the formation of two interlayers as shown in Fig. 1a. The interlayer adjacent to copper was homogeneous, its width and microhardness value were 10 µm and 2.9–3 GPa. According to the investigated properties it can be proposed the interlayer composition consists of intermetallics. Another observed interlayer consisted of crystallized melt and had dendritic structure. The thickness and microhardness of the interlayer were measured to be 960 µm and 1.7–1.75 GPa (Fig. 2a). The growth of dendrite's size was observed with the increase in distance from the Cu layer. The size of the most distant dendrites reached 10 µm.

The increase in heat treatment duration up to 0.5 h caused the growth of the thickness value of both interlayers up to 21 µm and 3675 µm for Cu and Al side interlayers respectively (Fig. 1b). As shown in Fig. 2b the hardness value of Cu side interlayer increased up to 5.0–5.25 GPa and decreased to 1.0–1.1 GPa for Al side interlayer. The crystallized melt dendrites, the distances of which from Cu side interlayer did not exceed 200 µm, were aligned perpendicular to the bond. Dendrites located in the remaining area did not have any special alignments. The increased hardness value of approximately 2.5 GPa was observed in the 500 µm thick area adjacent to Al (Fig. 2b). Such change in properties can be explained by the growth of intermetallic compound in the investigated material with apparently high fraction of Al (Θ-phase is an example of such phase, $CuAl_2$ - 67% of Al).

Heat treatment for 2 h terminated the contact melting and resulted in the conversion of the whole Al layer into crystallized melt. As shown in Fig. 1c after the treatment the composite had 2 layers apart from Cu:

- The 30-35 µm thick layer adjacent to Cu with 7.8–8.0 GPa microhardness value (Fig. 2b). The layer had apparently different kinds of intermetallic compound.
- Highly heterogeneous crystallized melt zone with arbitrary aligned dendrites. Microhardness value of the zone was measured to be 1.4–1.5 GPa (Fig. 2c).

Heat treatment for 45 h resulted in the conversion of most of the Cu layer volume into diffusion zone. As shown in Fig. 3 five individual layers were observed during metallographic investigations in the diffusion zone between Cu and crystallized melt. The thickness of the first and third layer was 2–5 µm and therefore it was not possible to determine the microhardness value of the layers, while the thickness a microhardness value of the second, fourth and fifth layer was 50–55 µm, 3.8–4.2 GPa;170–185 µm, 4.9-5.1 GPa and 360–380 µm, 8.4–8.5 GPa respectively.

To determine the Al/Cu ratio in the obtained layers EDS chemical mapping analysis was applied across the line presented on Fig. 4a. The line intersects Cu, diffusion zone and crystallized melt layers of the material. The analysis presented in Fig. 4b revealed the presence of 5 individual layers in the diffusion zone. According to Al-Cu phase diagram

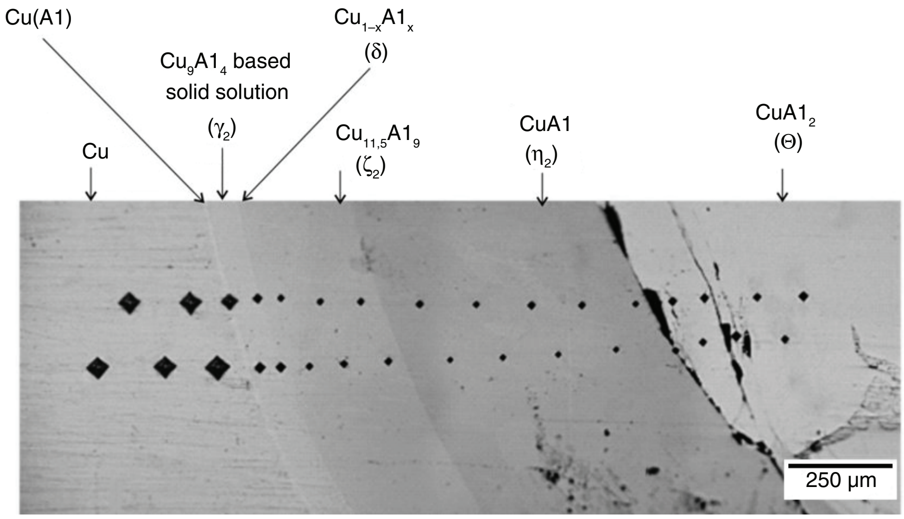

FIGURE 3. Optical image of explosion welded and heat treated at 570 °C for 45 h Al/Cu composite.

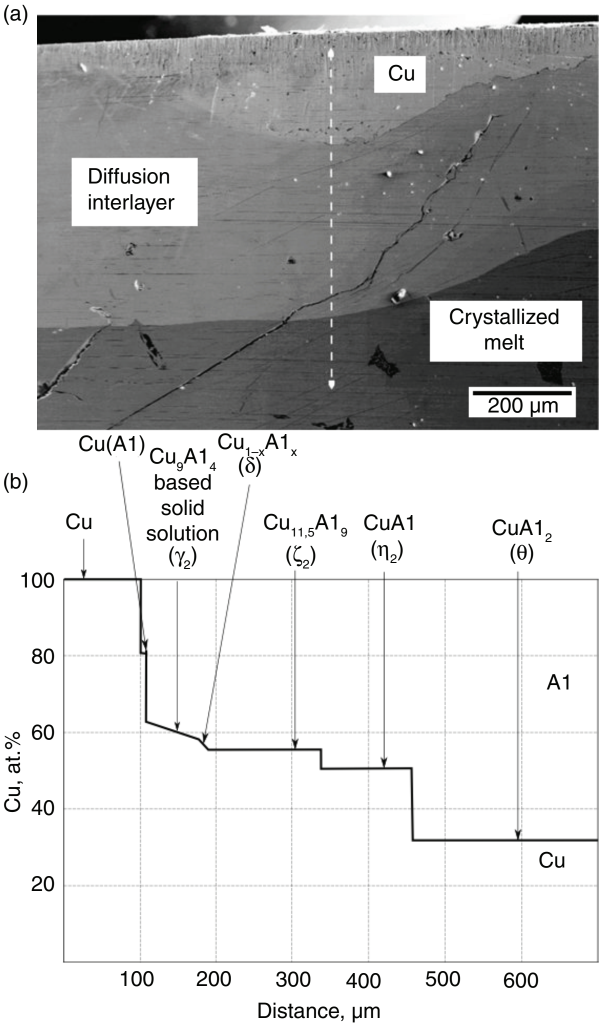

FIGURE 4. Explosion welded and heat treated at 570 °C for 45 h Al/Cu composite: (a) SEM image and (b) EDS map of Al/Cu ratio distribution across the white dotted line.

phase composition of layers, presented in the diffusion zone can be identified as:

1st layer - solid solution of Al in Cu Cu(Al)

2nd layer - solid solution of variable composition with aluminide Cu_9Al_4 base (γ_2-phase)

3rd layer - aluminide Cu_1-xAl_x (δ-phase)

4th layer - aluminide $Cu_{11.5}Al_9$ (ζ_2-phase)

5th layer - aluminide CuAl (η_2-phase)

The subsequent investigation of crystallized melt revealed that it had a mixture of $CuAl_2$ intermetallic and Al-base solid solution in its composition (Fig. 5). The size of intermetallic inclusions reached up to 100 μm.

The formation of the identified layers can be explained by following mechanisms:

At the beginning of the contact melting process solid state diffusion leads to the formation of Al(Cu) and $CuAl_2$ phases in adjacent to Al/Cu interface areas of aluminum layer and copper layer respectively. The formation of $CuAl_2$ is apparently accompanied by the formation of aluminides which exist at 570 °C (η_1-phase, ζ_2-phase, δ-phase, γ_2-phase and possibly β-phase). The thickness of the identified layers depends on the phase enthalpy of formation and solubility range. When the contact between Al(Cu) phase and $CuAl_2$ phase is reached, free energy in the contact zone is higher than the eutectic melt energy. Thus liquid interlayer starts to form. The adjacent to the liquid interlayer phases dissolve in the interlayer while maintaining its eutectic composition. The solid state diffusion of Al and Cu atoms in copper and aluminum layer respectively contributes to the formation of various intermetallic layers as well as solid solution Al(Cu) layer. During the cooling process liquid solidifies into eutectic with $CuAl_2$ crystals (Fig. 5b), while η_1-phase turns into η_2-phase and β-phase turns into a mixture of γ_2 and Cu(Al) phases.

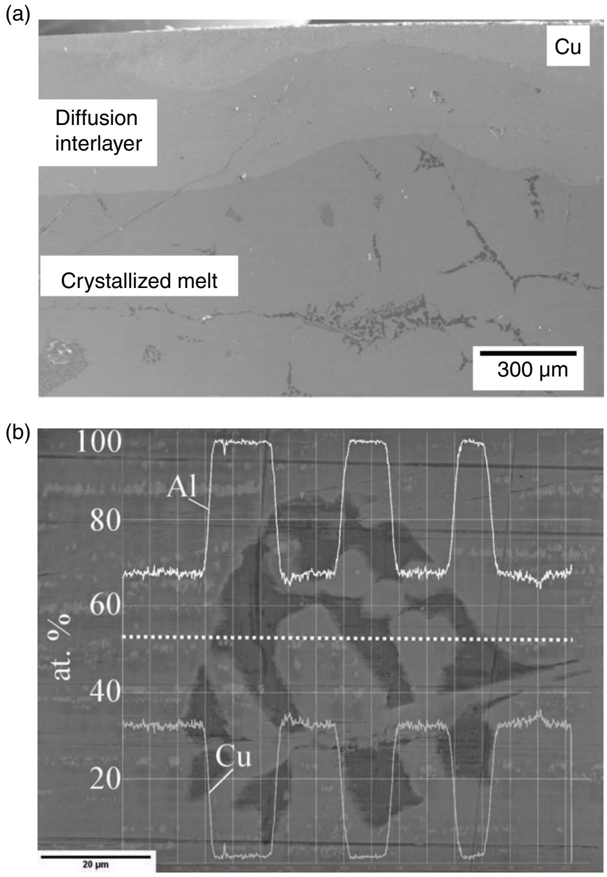

FIGURE 5. Explosion welded and heat treated at 570 °C for 45 h Al/Cu composite: (a) and (b) SEM image with EDS map of Al/Cu ratio distribution in crystallized melt inclusions across the white dotted line.

Three processes should occur in order for various intermetallic compounds to form: the penetration of Al atoms in existing intermetallic layer, transition of Al atoms across the layer, intermetallic formation reaction. During the contact melting process the

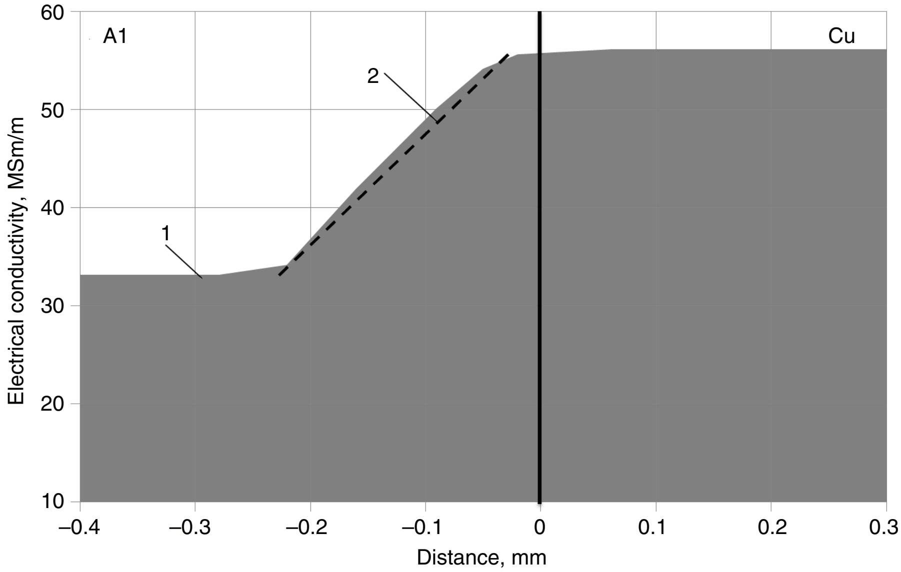

FIGURE 6. Electrical conductivity distribution across the bond area of explosion welded Al/Cu composite: 1 – experimental data, 2 – calculated via (3) data.

thickness values of existing intermetallic layers are lower than those formed during solid state diffusion. Thus diffusion processes are accelerated compared to solid state diffusion.

Due to the possible applications of obtained materials in electrotechnical devices, which require electrical conductivity of the composite to have appropriate values, the electrical conductivity measurement techniques were developed for the investigated materials.

Eddy current method was applied to determine the electrical conductivity of the composite. The probe which contained two solenoids was placed on Al side of the composite. Then AC flowing through one of the solenoids induced magnetic field in the thin upper adjacent to the probe layer of the specimen, which contributed to eddy currents flow in the layer. The eddy currents induced magnetic field which affected the field induced by the solenoid. The change in the magnetic field can be registered by another solenoid. The eddy currents value depends on the conductivity of the investigated material and thus assessment of the conductivity was carried out. During the investigation thin layers of the material on Al side were successively removed as the measurements were repeated. The approach allowed creating distribution of the electrical conductivity value across the composite.

The depth of penetration is given by:

$$\delta = \frac{1}{\sqrt{\sigma \cdot \pi \cdot f \cdot \mu}} \tag{2}$$

where σ – denotes electrical conductivity (Sm.m^{-1}), π – pi constant, f – test frequency (Hz), μ – magnetic permeability (H.m^{-1}).

The value of f for Vihr-1M device is 109–110 kHz, which corresponds to the depth of penetration of 190–210 µm and 240–260 µm for Cu and Al alloys used in this study respectively. The values of the depth of penetration correspond with the experimentally obtained results of the electrical conductivity distribution across the composite before heat treatments and are presented on Fig. 6. On the Al side the increase in the obtained conductivity value is observed 200–220 µm from Cu side. The equivalent conductivity value of the zone can be calculated using additivity rule:

$$\sigma = n \cdot \sigma_{Cu} + m \cdot \sigma_{Al} \tag{3}$$

where σ_{Cu}, σ_{Al} and n, m – denote electrical conductivity and percentage of Cu and Al respectively.

Pronichev *et al.* (2012) reported that the diffusion layers, formed during heat treatment at 530 °C for 30 hours contributed to a significant change in the conductivity distribution across the material. After heat treatment the heat conductivity distribution had two linear regions as shown in Fig. 7a. On the first region as the diffusion layer was involved into the eddy current testing the conductivity value decreased down to 14–16 MSm.m^{-1}. The impact of intermetallics was observed in approximately 400 µm thick area adjacent to diffusion zone. The value corresponds with the calculated via (2) depth of penetration, which in this case is 350–400 µm. The increase in conductivity value up to 59–60 MSm.m^{-1} was observed on the second linear region which was due to the involvement of Cu layer into testing.

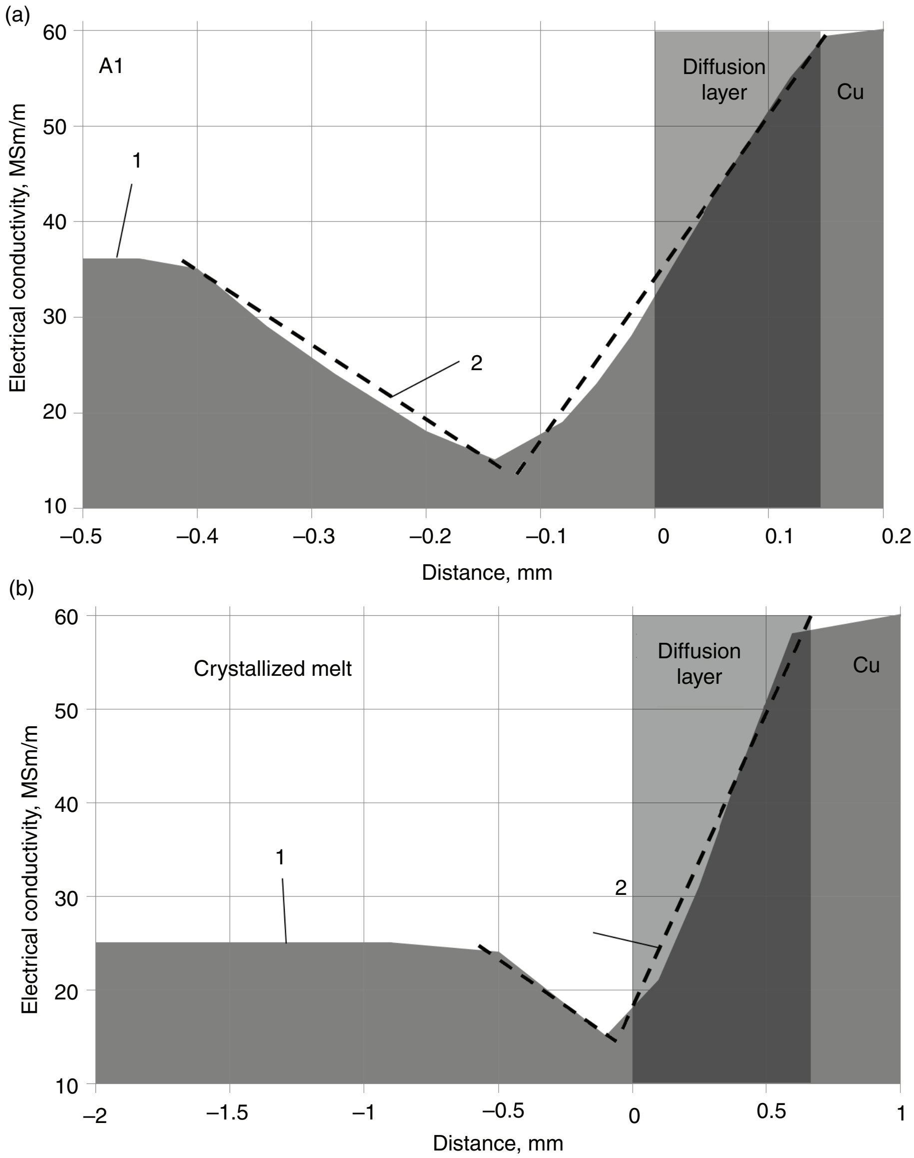

FIGURE 7. Electrical conductivity distribution across the bond area of explosion welded and heat treat: (a) at 530 °C for 30 h and (b) at 570 °C for 45 h Al/Cu composite; 1 – experimental data, 2 – calculated via (3) data.

Heat treatment at 570 °C and subsequent cooling to 20 °C resulted in the crystallized melt and diffusion layer emergence. The crystallized melt had a mix of Al(Cu) and Θ-phase in its composition, while diffusion layer consisted of γ_2, δ, ζ_2 and η_2 phases. As reported by Braunovic and Alexandrov (1994) and Pfeifer *et al.* (2012), the conductivity of Θ-phase is 12–16 $MSm.m^{-1}$ (Fig. 7b), so the depth of penetration for the area where Θ-phase is presented is 300–320 μm. The conductivity of γ_2, δ, ζ_2 and η_2 phases is 14–16 $MSm.m^{-1}$ and the depth of penetration for layers of intermetallics from these phases is 470–490 μm.

The regions of electrical conductivity distribution related to the emergence of crystallized melt and diffusion layers are linear and are in coherence with the additivity rule.

4. CONCLUSIONS

- Heat treatment at 570 °C of explosion welded Al/Cu bimetal leads to contact melting in the bond area of the composite. The diffusion processes are significantly accelerated compared to solid state diffusion.
- Crystallized melt formed during contact melting is a highly heterogeneous mix of $CuAl_2$ intermetallic (85%) and Al(Cu) (15%) grains. The reactive diffusion processes in the bond between Cu and crystallized melt result in the formation of Cu_9Al_4, Cu1-xAl_x, Cu11,5Al9 and CuAl intermetallics which were identified during EDS analysis.

- The method of the electrical conductivity of the composite's components measurement was proposed. The electrical conductivity of intermetallics and crystallized melt was measured to be 14–16 $MS.m^{-1}$ and 25–27 $MS.m^{-1}$ respectively.

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