

Microstructure and X-Ray Diffraction analysis of Al-Cu Couples Diffused at High Temperature

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Abstract

By employing a thermal field of 813 K, couples of Al with Cu were welded as consequence of the atomic diffusion. The periods of permanency inside the field were 200, 320 and 480 hours. After the periods of test, SEM observations show the formation of two contiguous plate likes (~15 to 30 micrometres, width) that they are in normal position to the concentration gradient. By utilizing elemental analyses and X-ray diffraction techniques one plate like was identified as the η phase (CuAl) and the other one as the θ phase (CuAl₂) of the aluminium-copper system. Immediate to the θ plate like it is expanded a zone formed by irregular θ grains surrounded by a network of α Al. Finally, far from the welding line, the saturated θ grains decompose and transform into pearlite according to the reaction $\alpha\text{Al} + \theta \rightarrow \alpha\text{Al} + \alpha\text{Cu}$.

Keyword: Al-Cu Couples; η phase plate like; θ phase plate like; Welding line; Weld bead.

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Introduction

Aluminium and copper are electric conductors by excellence. Occasionally, when these metals are in contact “hot points” could be formed if a high electric current is passing through them and the contact is faulty. Corrosive atmospheres also cause the heating of the union by the formation of electrochemical cells at the interface. The mass transport through that interface is one of the main items to analyse as well as the modification of the granular structure in both two metals. Based on the fact of that the crystal structure (face centred cubic, FCC) are of the same type, the inter-diffusion among these metals could be carried out without any problem. However, the difference of its metallic radii⁽¹⁾ as well as that of the respective diffusion Coefficients⁽²⁾ can get to different equilibrium states that those foreseen by the classic

theory (Fick's law)⁽³⁾ Considering the facts above mentioned the aim of this work is to study the modification of contact physical state after a process of extreme heating under temperatures up to 813 K by long periods

Materials and Experimental Procedures

Materials

In order to carry out the diffusion process, cylindrical samples (19 mm diameter and 10 mm length) of aluminium and copper (both commercially pure) were machined and, a number of couples of aluminium with copper were formed by making contact through its basis. Later on, the couples were heated with the aid of an electric furnace; the work temperature was established at 813 K and the periods of permanency inside the furnace were 200, 320 and 480 hours. In all the cases the couples were cooled in air. At the end of the mentioned periods, the diffused couples were diametrically cut and polished; after polish, the

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welding among both metals was clearly observed. Later on, for observing the resulting microstructure, a number of SEM micrographs were taken. Additionally, the mentioned sections were irradiated with X-ray in selected area mode.

Equipment for characterization

A temperature controlled furnace was used to apply the temperature for the established periods; the electronic circuit of the utilized furnace is able to control variations in the temperature of ± 5 K. For the microstructure observation and the elemental analysis a JEOL40 KV scanning electron microscope (SEM) was utilized. The study of X-ray diffraction was performed with the aid of a Bruker D8 Advance device; a wavelength of 1.5406 angstrom was used. The hardness measures were carried out with a hardness meter OPL-Made In France-No. 034 and for imprints a weight of 50 gram was employed

Results and Discussion

Microstructure

Figure 1 shows the metallurgical structure of the welding line; in the image, at right side of the Cu it can be observed a plate like of 15 to 33 micrometres in thickness composed by the η_2 phase (CuAl).⁽⁴⁾ Likewise, it can be observed that this plate like presents smooth interfacial lines to both sides. On the other side and stuck to η_2 phase plate like another plate like is formed whose chemical composition corresponds to the η_2 phase (CuAl_2)⁽⁴⁾ Contrary to the former case, this θ phase plate like is presenting an irregular contour at the right side and it is not possible to estimate a defined thickness. To complete the analysis, in the right part of the image field, the microstructure consisted of η_2 phase irregular grains and granular pearlite; at the same time, some θ grains are registering the presence of the η_2 phase in form of precipitate.

The micrograph depicted in Figure 2 shows the microstructure of the intermediate zone (the zone between the welding line and the aluminium). In the observed optical field, the zone is mainly constituted by lamellar pearlite, something of granular pearlite and diluting platelets of primary θ phase; for the case of lamellar pearlite, fine pearlite and coarse pearlite can be seen. In detail, fine pearlite is in the platelet's boundaries and surrounding the platelet

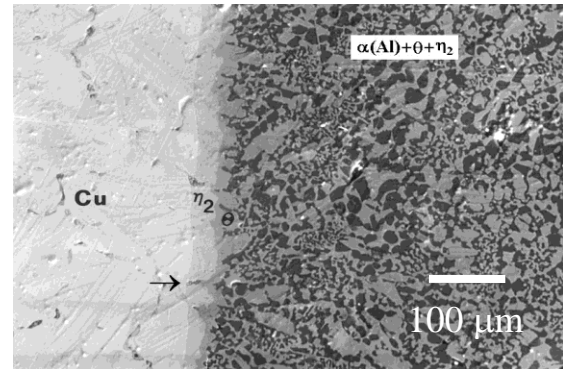


Figure 1. Microstructure of the welding line of Al-Cu couple. 813 K, 480 h.

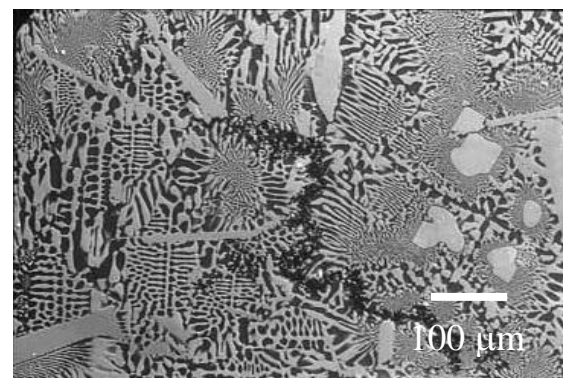


Figure 2. Microstructure of the zone between the welding line and the aluminium.

Figure 3 shows the manner in that the copper diffuses in the aluminium. As can be seen in the image, the diffusion is performed through the grains boundary. In detail, in the clear zone it can be seen the formation of copper solution micro crystals.

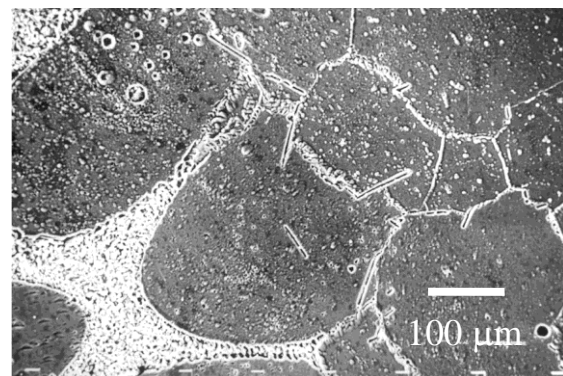


Figure 3. Microstructure of aluminium extreme.

X-Ray diffraction

The first zone for x-ray analysis was the welding line; the spectrum corresponding to this zone (upper spectrum) is shown in Figure 4. Based on this analysis the peaks observed show the presence of the θ_2 , θ intermetallic phases and Cu, being the reflection at 43.2° that of higher intensity corresponding to the close-packed (111) plane of

the Cu. The other two reflections are well defined; the peak observed at 38.4° is produced by the (300) and (211) planes of the η_2 and η_2 phases respectively, meanwhile, the peak observed at 50.3° is produced by the (200) plane of Cu. For this spectrum, the 2θ angle position and relative intensity (I/I₀) of each band are summarized in Table 1.

Table 1. Peak Positions with its respective values of d-spacing, relative intensity, intermetallic phase with the diffraction plane.

| Welding line | | | |
|---------------------|----------------------|------------------|---|
| 2 θ (degree) | d-spacing (angstrom) | I/I ₀ | Intermetallic phase (diffraction plane) |
| 38.4249 | 2.34083 | 0.0489 | θ (211), η_2 (300) |
| 43.2442 | 2.09046 | 1 | Cu(111) |
| 50.3399 | 1.81117 | 0.3143 | Cu(200) |
| Intermediate zone | | | |
| 38.4044 | 2.34203 | 0.3351 | θ (211), η_2 (300) |
| 42.4649 | 2.12700 | 1 | α Cu(111) |
| 44.5977 | 2.03011 | 0.2624 | η_2 (310) |
| 47.2432 | 1.92241 | 0.3204 | θ (202)(310) |
| 65.0234 | 1.43319 | 0.2072 | α Al(220) |
| 69.1000 | 1.35825 | 0.2597 | θ (420) |
| Cu | | | |
| 43.1827 | 2.09330 | 1 | Cu(111) |
| 50.3194 | 1.81186 | 0.4659 | Cu(200) |

In the intermediate zone the more prominent peaks show the presence of α Al, θ , η_2 and α Cu phases (Figure 4). In the image, peaks appearing at 38.4, 47.2 and 69.1 degrees are reflections produced by parallel crystal planes of the θ phase. Besides the band observed at 42.4° corresponds to the close-packed (111) plane of the copper solution, since, the introduction of aluminium atoms in the Cu lattice increases the separation among the (111) planes (metallic radii: Al, 1.43 angstrom; Cu, 1.28 angstrom); (1) therefore, the shift to the left of the band produced by the (111) plane of the copper solution in the lower spectrum could be explained in this way. To complete the analysis, the peak observed at 65° corresponds to the aluminium solution as mentioned and, as it is marked in the spectrum image, the peak is the reflection of the (220) plane (see Table 1).

The diffraction pattern shown in the Figure 5 corresponds to the Cu side. Two well defined bands at 43.1 and 50.3 degrees are due to the presence of Cu phase alone. The band with the highest intensity is produced by the reflection of

the close-packed (111) plane of the Cu phase, meanwhile, the second band is produced by the (200) crystal plane from the same phase.

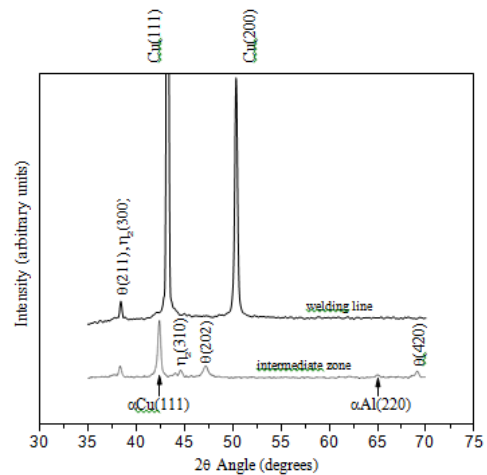


Figure 4. X-Ray diffraction patterns that correspond to: the welding line; the intermediate zone.

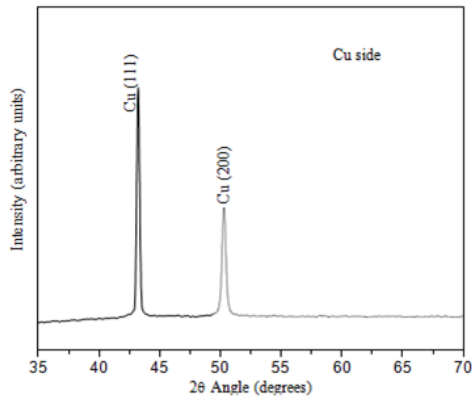


Figure 5. X-Ray diffraction spectrum corresponding to the Cu side after the diffusion process.

Elemental analysis

Based on the results by the Electron Probe Micro Analyser, the Cu phase zone did not show any aluminium content in the whole zone. This result is in agreement with the X-ray diffraction performed in the Cu zone. In the welding line, a plate like of 15 to 33 micrometres in thickness extend parallel to the welding line with an aluminium content of 27.7 wt.%; thus, according to the phase diagram, this plate like is formed by the η_2 intermetallic phase. Immediately and together to the previous plate like appears a second plate like with a thickness similar to the first one with an aluminium content of 51.5 wt.%, this second plate like was identified as the θ intermetallic phase (see Figure 1). In the zone between the welding line and the aluminium (see Figure 2), the analysis of aluminium content changed in an irregular way, which is congruent with the metallographic structure obtained after the diffusion process.

Hardness

It is relevant to do mention that the profiles of hardness, throughout the different periods, did not show any difference as for the values of hardness in each zone. So, the Vickers hardness (HV) obtained on the η_2 intermetallic phase plate like is 591 ± 19 ; meanwhile, the corresponding HV to θ intermetallic phase plate like is 513 ± 12 . Inside the zone between the welding line and the aluminium the reading of hardness was of 185 (on the average). In the side where copper is dominant, the hardness average registered is of 45; this last hardness had very little change within this side being able to consider as constant.

Discussion on Graphical Analysis

From the analysis of the results of the thermal process applied to the selected metals, several aspects deserve an adequate explanation. In the case that occupies us, at the extreme conditions in the temperature of test (813 K) the welding line was carried to a different equilibrium state to that one predicted by the classic theory (Fick's law).⁽³⁾

In the welding line the weld bead is composed by two parallel plate likes (η_2, θ) as described, with the η_2 plate like joined to copper. In the work of Yasuhiro Funamizu and co-worker (1971)⁽⁵⁾ it is established the formation of the Cu, γ_2 , δ , ζ_2 , η_2, θ and Al solid solution phases in this order, with a test temperature of 808 K by periods of 80 hours. But, in the dispositive employed by Yasuhiro Funamizu, the Cu plate was sandwiched between two discs of Al and placing aluminium oxide powder between the metals. Therefore, in our case, the employed temperature (813 K) and the disposition of the two metal parts could be used to explain the differences observed.

In this work, the parallel plate likes η_2 and θ advance into the copper and, as can be seen in Figure 1 two types of diffusion from the Al towards the Cu are observed: bulk diffusion; intergranular diffusion. Also, in Figure 1 it can be seen that the primary θ plate like is divided into segments that give origin to the irregular shape grains at right afterwards; at the same time, this process of segmentation are producing the breaking of the η_2 plate like in some points (marked \rightarrow), this situation causes that some part of η_2 phase results inside θ grains. This observation is in agreement with the analyses of chemical elements and of diffraction.

In the intermediate zone (Figure 2), the pearlitic structure observed is the result of a four-phase transformation $\alpha\text{Al} + \theta \rightarrow \alpha\text{Al} + \alpha\text{Cu}$ ⁽⁶⁻⁸⁾ (before transformation the θ grains are surrounded by a network of αAl); in detail, at the boundary of θ platelets fine pearlite is forming as a result of the mentioned reaction. Later on, fine pearlite transform into coarse pearlite after aging just as it is observed in the image of Figure 2. Therefore, in the intermediate zone, the phases that are present correspond to the same ones detected by diffraction (lower spectrum in Figure 4).

In this work, the mechanism of diffusion observed is different to the one reported by Yasuhiro Funamizu. Also, this mechanism differs from the one predicted by the Fick's law.⁽³⁾ At the beginning of the thermal process it is possible that the formation of plate likes is given one such as was found by Yasuhiro Funamizu, but later, with the temperature level employed and the short interval of composition of the absent phases, it was enough to propitiate the metastable structure shown here. It is evident that the five additional degrees to the test temperature here initiate the mentioned reaction that took place. Given the above- mentioned, it can be supposed that the execution of the Fick's law is limited to certain range of temperatures for each particular metallic couple. Finally, in the literature are found similar results in other metallic systems that corroborate the no fulfilment of the mentioned law^(9,10).

Conclusions

1. The aluminium-copper joints that are immersed in thermal processes at temperatures up 813 K or bigger present the formation of two parallel thinplate likes (~15 micrometres) in the welding line. The plate like joined to the copper has the composition of the η_2 phase and the other one the composition of the θ phase according to aluminium-copper system.
2. The continuous transport of mass through the welding line cause the segmentation of the θ plate like by the penetration of the α Al on certain line sites, mainly; later on, the resulting segments are subdivided and form a structure of θ grains of irregular shape that they are surrounded by a network of aluminium solid solution. In some points of the welding line the penetration can continue until the η_2 plate like, what give place to the separation of two-phase grains. The two types of grains observed are thermodynamically metastable.
3. In the separate zone of the welding line (i.e. intermediate zone), θ grains decompose through the reaction $\alpha\text{Al} + \theta \rightarrow \alpha\text{Al} + \alpha\text{Cu}$. As a result of such decomposition fine pearlite is formed directly at the boundaries of θ platelets. Finally, primary pearlite is transformed into coarse pearlite.

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