

# Study by nanoindentation of the influence of the manufacturing process on the mechanical properties of Copper-Clad Aluminum wires

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**Abstract.** We studied the influence of the architecture of Copper-Clad Aluminum (CCA) wires produced by cold-drawing process, on the mechanical properties of materials, and the development of intermetallic compounds at the interface after annealing. Simple and architected-CCA (A\_CCA) rods, fabricated by stacking and re-drawing of original CCA wires, have been characterized before and after a thermal treatment. Nanoindentation tests, performed to map the hardness and the elastic modulus at the microscale, allow to well catch the recrystallization phenomenon, which takes place in both the Cu and the Al subjected to severe plastic deformation, as well as the formation of three intermetallic compounds (IMC) at the interface. Our measures underline that the mechanical properties of all the components involved in the wires, i.e., Cu, Al, and eventually the IMC developing after annealing, are only slightly influenced by the structuration of the wires. IMC developing at the Cu/Al interfaces present a high hardness, and might deteriorate the mechanical properties of the wires, even more for A\_CCA wires, which exhibit more interfaces than the traditional CCA samples.

**Keywords:** copper-aluminum bimetallic wires / architecture / intermetallic compounds / nanoindentation

## 1 Introduction

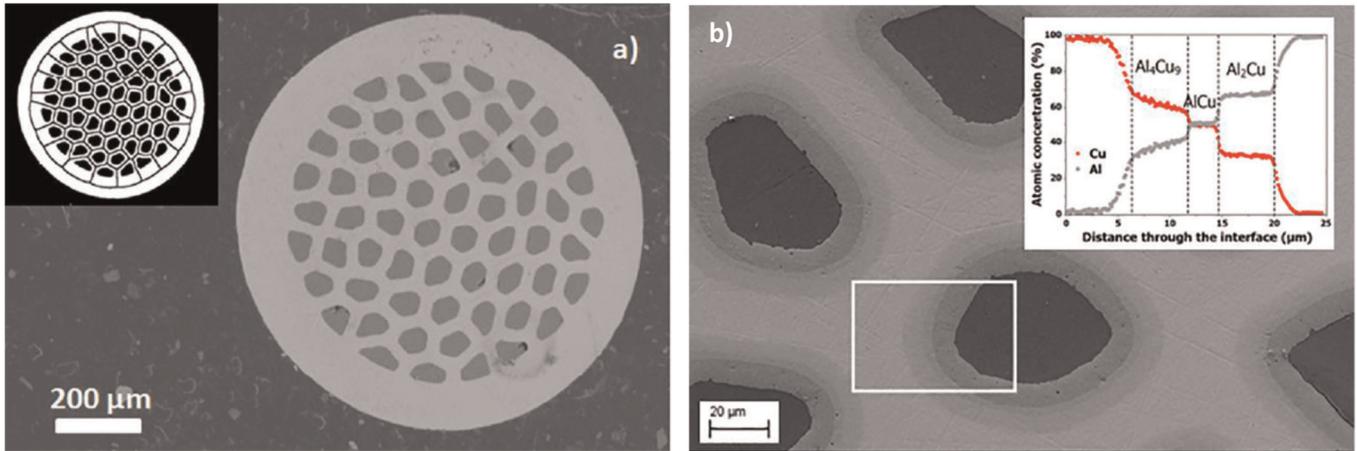
Metal-metal composites consist of two different materials bonded together at adjoining interfaces, where they mutually interact. Such innovative materials offer the possibility to achieve optimized combinations of properties, and thus are gaining increasing popularity in the aeronautics, automotive, marine industry, thermal engineering, electrotechnics, medicine, and many more industrial and commercial branches [1–3]. Among the various metal combinations that have been considered in the literature, Al–Cu bimetallic composites already found applications in electrotechnics [4–6]. Indeed, most of the electro-conductive wires in households and vehicles are made from copper, which is relatively expensive and heavy. Replacing copper wires by Copper-Clad Aluminum (CCA) composites, consisting in an aluminum core and a copper sleeve, allows to save high conductivity, mechanical strength and corrosion resistance, while reducing weight and cost of about 40% and 30%, respectively [7].

The forming technologies used for CCA composites mainly included co-rolling [8], hot hydrostatic extrusion [9], overlay welding [10], and rotary swaging [11]. It has been shown that processing technologies involving intensive

plastic deformation, performed preferably under cold conditions, ensure bonding of the components by introducing shear strain and produce clad composites and hybrid materials with favourable properties [11–13]. However, production under cold conditions supports significant increase in strength, but also invokes decrease in material formability (gradual exhaustion of plasticity). An annealing treatment after deep drawing is traditionally performed in the wire industry, in order to increase ductility of metals. The diffusion between copper and aluminum that takes place, also allows increasing the metallurgical bonding at the interface, and repairing cracks and holes that can form at the interface after a rolling process [13,14]. On the other hand, annealing also induces the development of various forms of intermetallic compounds (IMC) at the interfaces, that can negatively influence the physical properties, especially the electrical conductivity and the mechanical strength of the final product, as IMC are supposed to be less conductive and more brittle than copper and aluminum [15–17]. Nevertheless, it was shown that optimum mechanical properties of CCA thin wires can be reached after an appropriate heat treatment, keeping the thickness of the IMC layer as low as it allows improving the bonding between copper and aluminum at the interface and prevents the cleavage fracture mechanisms [18].

More recently, in order to improve the mechanical properties and the electrical conduction at lower frequencies, fibrous Al–Cu microcomposites wires, consisting in a

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**Fig. 1.** (a) Light microscopy observation of the radial view of the A\_CCA after drawing until a diameter of 1 mm. In inset is the corresponding thresholded picture. (b) SEM picture of the 1 mm diameter ACCA wire after an annealing treatment at 400 °C for 6 h (cross sectional view). In inset, atomic concentration profile of Al and Cu element through the interface.  $\text{Al}_2\text{Cu}$ ,  $\text{AlCu}$  and  $\text{Al}_4\text{Cu}_9$  can be well-identified [21].

bundle of Al wires in a Cu tube, were manufactured by cold rotary swaging [12,19] and restacking cold-drawing [20]. Indeed, the multiplication of the bonding interfaces and the creation of Cu paths inside the structure are expected to improve the mechanical and electrical properties as compared to CCA wire. On the other hand, increasing of the proportion of interfaces leads to an increase in the nucleation sites of IMC, and thus a proportion and a repartition on IMC inside the structure that might have a great influence on physical properties of the final products.

The objective of this work is to investigate the impact of the forming technics on the mechanical behavior of architected CCA wires, produced by cold-drawing of restacked CCA wires and subsequent annealing. We used nanoindentation to quantify the influence of the forming conditions of the wires on the mechanical properties of copper and aluminum, as a function of the plastic deformation applied. Secondly, we studied the impact of the IMC, which develop at the copper-aluminum interfaces of the wires during post-elaboration annealing treatments, on the mechanical behavior of the wires.

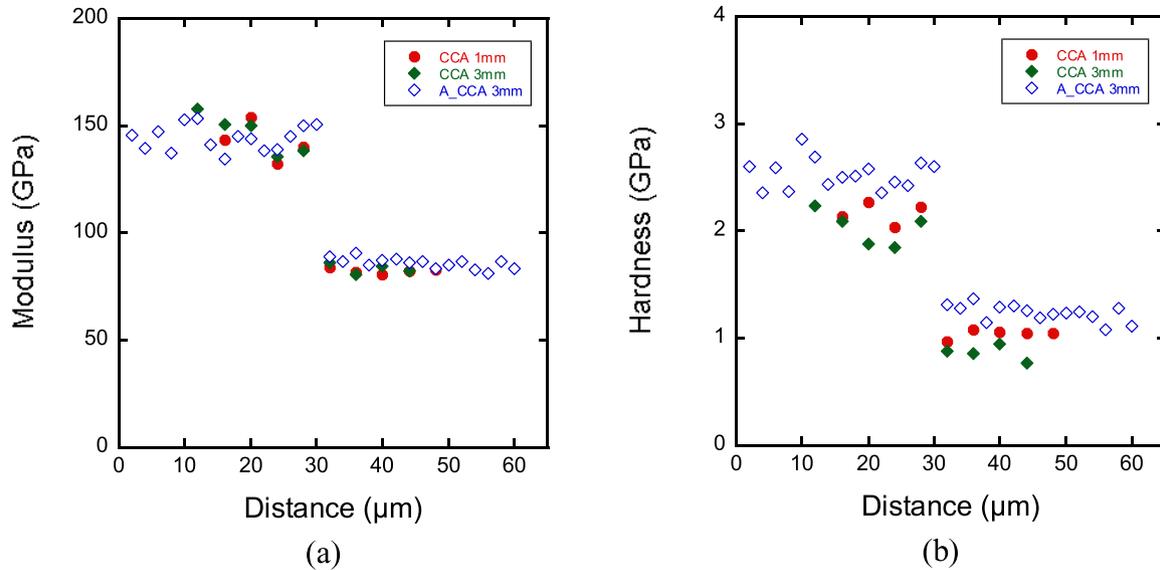
## 2 Materials and methods

The studied materials consist of CCA rods of 1 and 3 mm diameter, obtained by first introducing a 99.5% purity aluminum rod of 7.5 mm in diameter into a 12 mm diameter and 1 or 2 mm thick 99.97% purity OFHC (Oxygen Free High Conductivity) copper tube. This preform is then drawn down to the targeted 1 and 3 mm diameters (up to 34 passes in different intermediate dies, with a section reduction of 10 to 20% at each pass). The “architected copper-clad aluminum” rods (referenced as A\_CCA in this paper) were then produced using a single 1 mm diameter copper-clad aluminum wire. This wire was cut into 250 mm length wires, 61 of these 250 mm wires were spliced to form a cable, which has been introduced inside a 1 mm-thick copper tube. The produced set was finally drawn down to a diameter of 1 and 3 mm, through the same 34 dies

(see Fig. 1a). For both the CCA and the A\_CCA samples, the volume ratio between copper and aluminum remains constant (roughly 72/28) whatever the diameter. The effect of the diameter of the wire is considered through the drawing ratio,  $\eta = \ln(S_0/S)$ , where  $S_0$  represents the initial section before the beginning of the drawing process of the first CCA (i.e., 12 mm), and  $S$  is the section of the final CCA wire, or of the CCA wire inside the A\_CCA.

In order to study the influence of the IMC on the mechanical properties of both CCA and A\_CCA, a post-annealing treatment was performed at a temperature of 400 °C for 16 h under secondary vacuum. Details of the fabrication of the different samples are given in [21]. At this temperature, four IMC developing at the Cu/Al interface:  $\text{Al}_2\text{Cu}$ ,  $\text{AlCu}$ ,  $\text{Al}_3\text{Cu}_4$  and  $\text{Al}_4\text{Cu}_9$  [15,22] (see Fig. 1b) were observed.

The mechanical properties of the samples were studied using nanoindentation. Indeed this method is indicated since the tested material volume is scalable with respect to the thickness of the IMC layers that develop at the copper/aluminum interfaces in the samples. This technique allows to extract the mechanical characteristics of material, such as the hardness and the elastic modulus, as well as to investigate the deformation mechanisms. Tests were conducted using a MTS NanoXP system (MTS Systems Corporation, USA), with a contact module loading head equipped with a three-sided pyramidal Berkovich diamond indenter with a nominal tip radius of curvature of 50 nm. The load and displacement of the indenter were monitored all along the indentation, with a force resolution of about 75 nN and displacement resolution of about 0.1 nm, and the displacement data were corrected assuming a drift rate, which is reevaluated before each test. Penetration depth was set at 300 nm, in order to keep the plastic deformation developing underneath the indenter small enough, to avoid important interactions between the plastic zones of indents performed closely to each other, and prevent for significant impact on the measured values. Indentations lines of 20 to 50 indents were performed at room temperature, positioned over the Cu/Al interfaces, with a spacing of 1 or 2 μm



**Fig. 2.** Evolution of (a) the modulus and (b) the hardness across a Cu/Al interface in non-annealed CCA samples of 1 and 3 mm in diameter and in A\_CCA sample of 3 mm in diameter.

between adjacent indents, i.e., sufficiently close to each other to probe each IMC, if present. The loading curves were analyzed and the indentation hardness, defined as the material resistance to the plastic deformation during indentation, and the elastic modulus were extracted from indentation data, according to the classical Oliver and Pharr method [23]. To exclude the errors while measuring the  $E$  and  $H$  parameters at the nanoscale, a series of calibration experiments on the same sample of fused quartz was preliminary implemented. Moreover, as aluminum and copper are known, as soft materials, to be sensitive to the indentation size effect (ISE), their hardness depend on the indentation depth [24]. Consequently, tests were performed in displacement control, by employing the continuous stiffness measurement (CSM) technique, which allows the continuous evaluation of the hardness and the elastic modulus during the indentation loading period. This feature entails superimposing a small cyclic load on top of the main load during the loading segment of the sample. Loading and unloading penetration rates were set at 5 nm/s, the CSM signal was set to an amplitude of 2 nm at a frequency of 45 Hz. Furthermore, the calculated hardness values can only be compared when determined under the same test conditions, so that the depth-dependent hardness, and consequently the modulus, were averaged for a penetration depth between 80–90 nm for all tests performed, using the Analyst<sup>®</sup> software. Samples used for the nanoindentation tests consist of slides cut in the different CCA and A\_CCA of 1 and 3 mm of diameter, not annealed and annealed at 400 °C for duration of 16 h.

### 3 Results and discussion

#### 3.1 Characterization of the as-drawn CCA and A\_CCA wires

In order to study the impact of the architecture of the wires on the mechanical properties of copper and aluminum,

nanonindentation measurements have been performed on as-drawn A\_CCA samples of 1 and 3 mm in diameter, and the values of the modulus and the hardness were compared to the results of tests performed on not-annealed CCA samples of same diameters. According to the fact that the samples considered in this section do not have been thermally post-treated after drawing, IMC are not expected to be observed at Cu/Al interfaces.

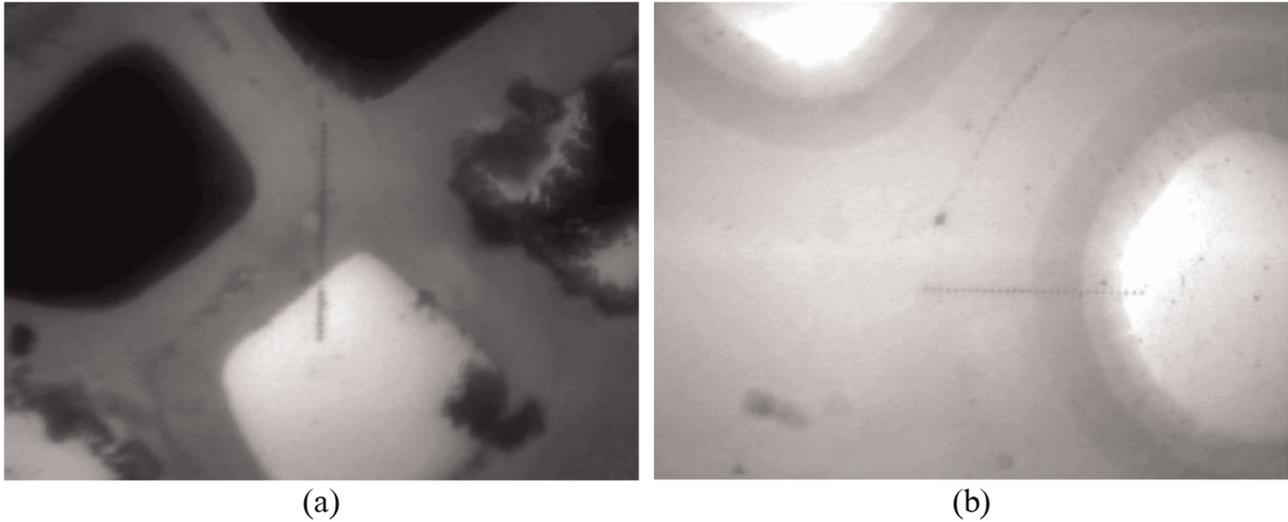
The Figure 2 presents the evolution of the modulus and the hardness for nanoindentation experiments performed across Cu/Al interfaces in non-annealed CCA samples of 1 and 3 mm in diameter and a non-annealed A\_CCA sample of 3 mm in diameter. The sudden decrease of the mechanical parameters between the test 15 (in the Cu) and 16 (in the Al) testifies for a clear transition between Cu (first measure points) and Al (further indents) so that no (or very few – very thin layer) IMC are present at the Cu/Al interface, either for the CCA or for the A\_CCA samples (see Fig. 3).

CCA wires consist in an aluminum rod surrounded by an copper tube, drawn down to a diameter of 3 and 1 mm. Accordingly, the plastic deformation applied to the samples with the smaller diameter, defined using the drawing ratio  $\eta$ , is higher, and thus the strain hardening of the materials. This hardening leads to a slight increase of the hardness of both the copper and the aluminum in samples of 1 mm in diameter, compared to the samples of 3 mm in diameter, while the modulus remains constant with the diameter decrease (Tab. 1, see also Fig. 2), suggesting no phase change or damage in both copper and aluminum due to the applied severe plastic deformation. The obtained values of the hardness of both the copper and the aluminum are somewhat higher than values reported in the literature for pure copper and aluminum [18,22,25], but could be explained by the even high plastic deformation applied to the materials, even for the largest diameter of wire.

On the other hand, several studies report about a dynamic recovering/recrystallization of the copper, and also to a lesser extent of the aluminum, due to its high

**Table 1.** Influence of the diameter on the modulus and the hardness of the copper and the aluminum in non-annealed CCA samples.

	Cu		Al	
	E (GPa)	H (GPa)	E (GPa)	H (GPa)
CCA 3 mm $\eta = 2.77$	$144 \pm 8$	$2.01 \pm 0.15$	$82 \pm 2$	$0.86 \pm 0.07$
CCA 1 mm $\eta = 4.97$	$149 \pm 3$	$2.21 \pm 0.11$	$84 \pm 3$	$1.07 \pm 0.07$

**Fig. 3.** Optical micrographs (magnification x40) of lines of indents performed across the interface in (a) a non-annealed and (b) a thermally treated A\_CCA sample of 1 mm in diameter, revealing the presence of IMC at the Cu/Al interface.

stacking fault energy, during high strain drawing at room temperature. In consequence, the authors note a decrease in the hardness, linked to a decrease of the dislocation density. Indeed, hardness is closely related to the yield stress, which depends on dislocation density. In the meantime, recrystallization leads to a decrease of the dislocation density, correlated to the occurrence of new undeformed grains, and thus results in a decrease of the hardness of the material. This recrystallization phenomenon has also been observed in previous studies on our samples, using transmission electron microscopy, for Cu at high deformation rates (for  $\eta > 5$ ) [18,21]. This threshold for the occurrence of the recovery of the copper suggests that the increase in the hardness observed for the CCA samples with the decrease of the diameter can be explained by an increase of the dislocation density in the material, correlated to the formation of large dislocation cells [21], due to the applied plastic deformation.

The Figure 4 presents the loading curves within copper and aluminum on either side of the interface (tests 15 and 16). We note an elasto-plastic behavior of both the Cu and the Al, correlated to an important residual deformation after unloading, of about 90%. Moreover, it could be observed discontinuities, so called pop-ins, on the loading curve, at the first times of the load application (see insert on Fig. 4). Pop-ins correspond to an absorption or a release of

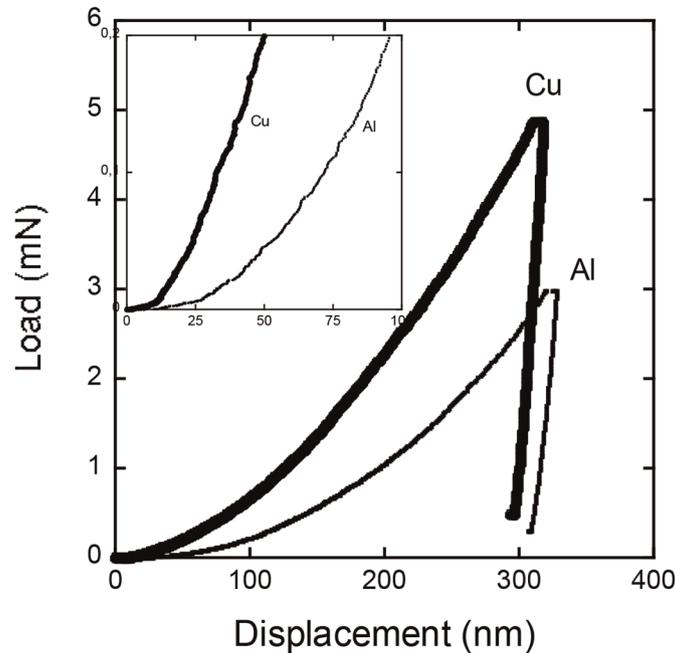
energy underneath the indenter tip, and are correlated with different physical phenomena, such as micro-cracking, phases transformations, deformation localization in sliding band, or dislocations motion. Pop-ins are frequently observed in FCC structure materials, such as Cu and Al, and linked to the plastic deformation process: after a purely elastic deformation of the material in the first step of indentation, the deformation behavior becomes elastic-plastic above the critical load at which the first pop-in event occurs, assuming slip being the mechanisms mainly responsible for the plastic deformation response of the material. These pop-ins are numerous and pronounced in the aluminum, revealing for a highly plastic behavior of Al immediately after the beginning of loading (smaller yield strength of Al than Cu). No (or even few) pop-in is observed on the loading curve of the copper.

A\_CCA rods of 1 and 3 mm diameter have been obtained by the restacking cold drawing method. The structure of these samples consists in a copper outer shell and a stack composed of 61 CCA (aluminum rods surrounded by copper tubes, as described previously).

The evolution of the modulus and the hardness for nanoindentation experiments performed across a Cu/Al interface in a non-annealed A\_CCA sample of 3 mm in diameter are presented on the Figure 2. As for the non-annealed CCA samples, the mechanical properties of the

**Table 2.** Influence of the diameter and the position of the measure on the modulus and the hardness of the copper and the aluminum in non-annealed A\_CCA samples.

		Cu		Al	
		E (GPa)	H (GPa)	E (GPa)	H (GPa)
A_CCA 3 mm $\eta = 7.37$	Outer ring	$140 \pm 8$	$2.28 \pm 0.15$	$86 \pm 2$	$1.23 \pm 0.08$
	Center	$144 \pm 6$	$2.53 \pm 0.14$		
A_CCA 1 mm $\eta = 9.57$	Outer ring	$135 \pm 9$	$2.29 \pm 0.21$	$85 \pm 5$	$1.24 \pm 0.04$
	Center	$130 \pm 11$	$2.55 \pm 0.19$		

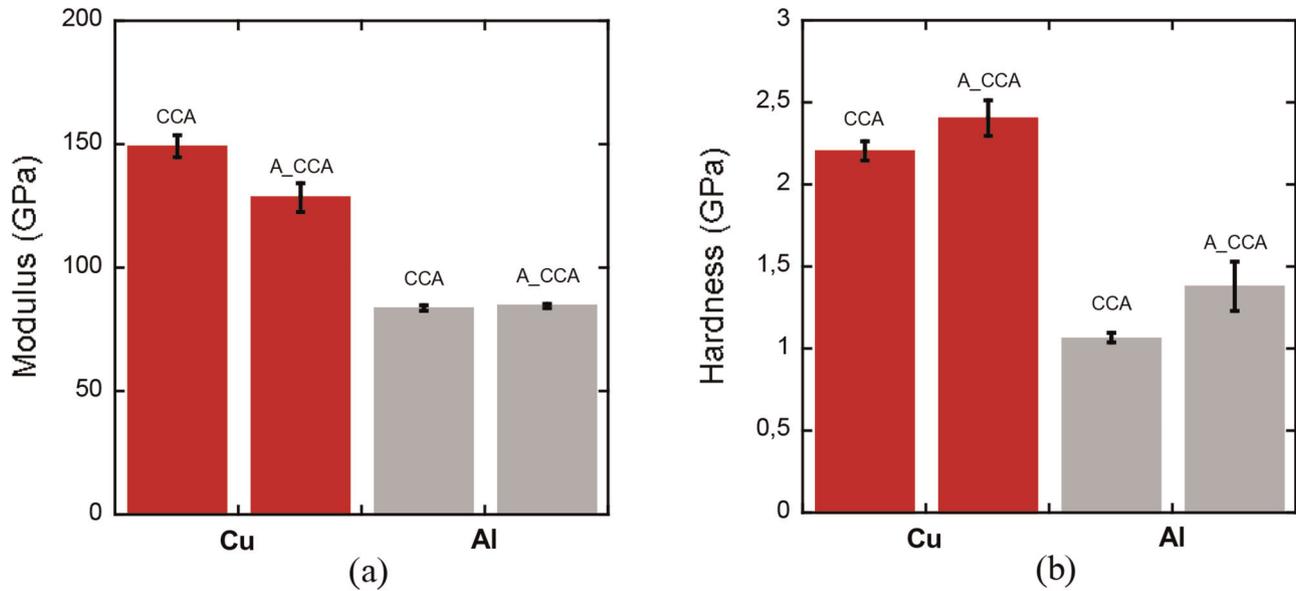
**Fig. 4.** Loading curves by nanoindentation on either side of the copper/aluminum interface in a non-annealed CCA sample of 3 mm in diameter (tests 15 and 16), with highlight of the pop-ins in insert.

A\_CCA show a clear drop at the interface between Cu and Al, and do not reveal thus the presence of IMC at the Cu/Al interface. The loading curves of the non-annealed A\_CCA samples are similar to those of the CCA wires, with an important residual deformation after unloading, and the presence of pop-ins on the loading segment, immediately after the application of the load.

To study in details the effect of the microstructure of the A\_CCA samples on their mechanical properties, nanoindentation measurements were deployed at the different parts of these composites. By using this technique, it is possible to study local hardness effects near phase boundaries and according to the overall position in the wire. Table 2 shows the values of the modulus and the hardness of copper and aluminum as a function of the diameter of the A\_CCA sample, and for the copper, as a function of the position of the measurement on the sample, i.e., in the external ring, to which the CCA cuts have been inserted, or inside the microstructure, in the pieces of CCA (aluminum cells surrounded by copper paths).

According to the mechanical parameters of Cu depending on the location, no significant difference was observed for the modulus for both non-annealed A\_CCA samples of 1 and 3 mm in diameter. However, the value of the hardness of the copper is clearly smaller on the outer ring than at the center of the same specimen. This could be explained by the fact that the copper paths of the pieces of CCA were restacked and redrawn to produce the A\_CCA samples, so that have been drawn twice, and so might be more hardened than the copper of the outer ring, which has been drawn only one time. Such “double-hardening” of the copper paths in the center of the sample could result in an increase of the hardness.

Compared to the original CCA wires (Fig. 5), we note an effect of the specific design of the A\_CCA samples on the values of the mechanical parameters for both the copper and the aluminum, with mainly an increase of the hardness with the structuring. This increase is even significant for the aluminum, and might be explained by the fact that the Al parts in the A\_CCA are located in the



**Fig. 5.** Variation of (a) the modulus and (b) the hardness of Cu and Al for the non-annealed CCA and A\_CCA samples of 1 mm in diameter.

center of the sample, so in strongly deformed areas, compared to the copper outer ring. However, it was observed in a previous study [21] that for the deformation rates related to the A\_CCA fabrication (i.e.,  $\eta > 5$ ) a dynamic recrystallization phenomenon, which should lead to relaxation processes in the materials, and thus to a decrease of the hardness. The fact that we measured larger values of the hardness for the A\_CCA samples, i.e., for even higher deformation rates, could indicate that the specific architecture of the A\_CCA samples prevents the recrystallization phenomenon in the materials. Moreover, it was observed an imperfect consolidation between the stacked CCA wires in the non-annealed A\_CCA samples [21,26], that might lead to a pill-up effect of the dislocations created by plastic deformation at the interfaces between the wires, and thus to an increase in the hardness.

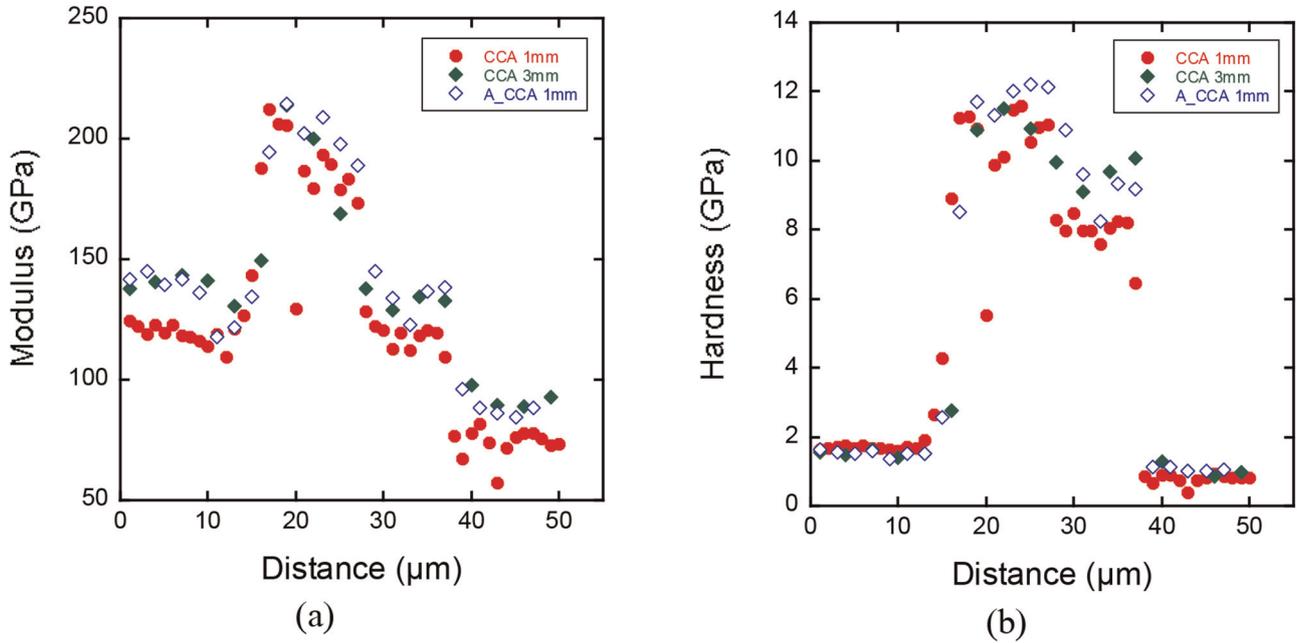
On the other hand, and contrary to the observations done on the CCA wires, it appears that the reduction of diameter of the A\_CCA samples from 3 to 1 mm through additional passes in dies results in a decrease of the modulus of the Cu, whatever the location. This result has to be carefully considered as it is well known that pill-up or sink-in are likely to develop as a change in the hardening coefficient of the material occurs [27], resulting in an error in the determination of the modulus. Nevertheless, the modulus of the aluminum remains stable whatever the drawing ratio, while the hardness of the aluminum increases as well as the one of the copper. Another hypothesis to explain the decrease of the modulus of the Cu might be damage process taking place as the copper undergoes an increasing plastic deformation (up to a drawing ratio close to 10 for the A\_CCA of 1 mm in diameter). In this case, the constant value of the modulus of the Al would suggest that the Al does not suffer any damage during its plastic deformation.

### 3.2 Influence of the IMC on the mechanical properties of the architected wires

Post-elaboration annealing treatments are traditionally performed on CCA rods in order to increase the ductility of metals and to consolidate by diffusion the interface between copper and aluminum, allowing for a hard bonding between both metals, and so a strengthening of the wires. However, diffusion mechanisms between Cu and Al lead to the formation of intermetallic compounds, which can reduce the wires performance, as IMC are known to present a brittle behavior. CCA and A\_CCA samples considered in the study were annealed under vacuum at 400 °C during 16 h, so that IMC develop at the Cu/Al interface (see differences of contrast on the optical micrograph on Fig. 3b). Nano-indentation tests then were performed both in the copper and aluminum to determine the effects of the annealing on their mechanical behavior, and at the Cu/Al interfaces, including IMC, to quantify the impact of these compounds on the mechanical behavior of the annealed CCA and A\_CCA wires.

The Figure 6 shows the variation of the modulus and the hardness across the Cu/Al interface in annealed CCA samples of 1 and 3 mm in diameter, and in an annealed A\_CCA sample of 1 mm in diameter. It is well-known that the intermetallic compounds generally possess higher hardness values than those of corresponding base metals, here of aluminum and copper. The increase of both the modulus and the hardness at the interface in these samples, in comparison to the sudden drop observed on non-annealed samples, clearly demonstrates the formation of diffusion products at the interface between copper and aluminum, because of the thermal treatment.

Moreover, we can note on the curves the same profile, for both the annealed CCA and A\_CCA samples, whatever the diameter. This might suggest the presence



**Fig. 6.** Evolution of (a) the modulus and (b) the hardness across a Cu/Al interface in annealed CCA samples of 1 and 3 mm in diameter and in A\_CCA sample of 1 mm in diameter (annealing at 400 °C for 16 h).

**Table 3.** Values of the modulus and the hardness of the different IMC present in CCA and A\_CCA samples after annealing.

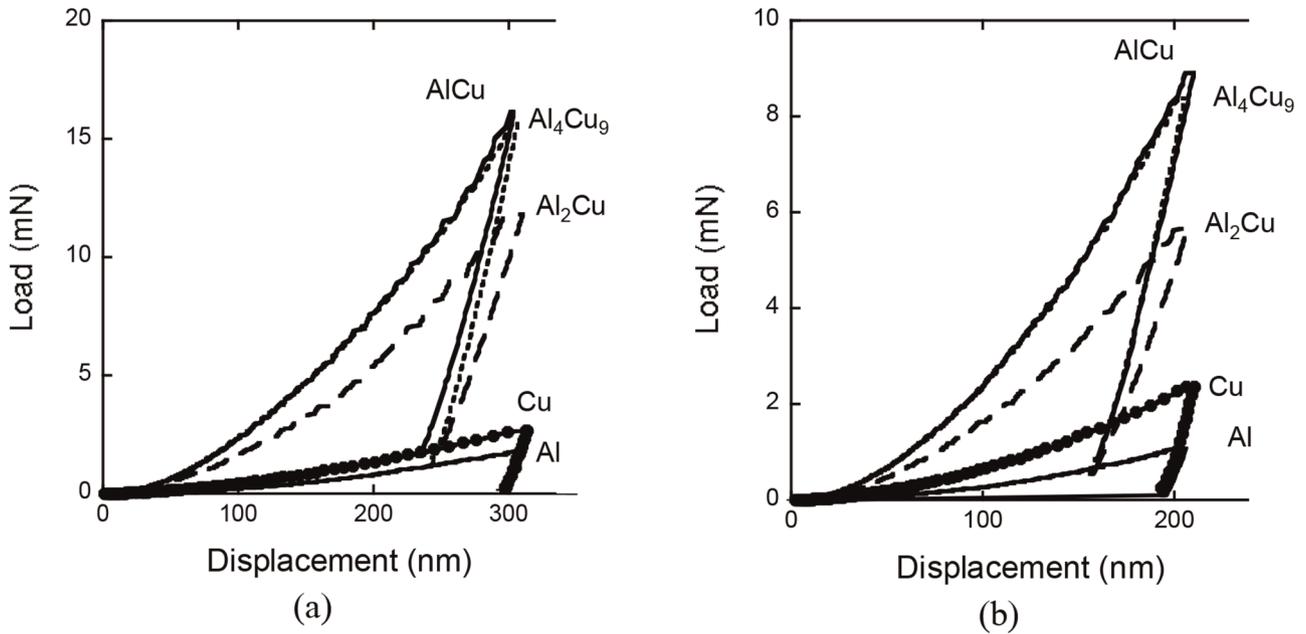
	$\text{Al}_4\text{Cu}_9$		$\text{AlCu}$		$\text{Al}_2\text{Cu}$	
	E (GPa)	H (GPa)	E (GPa)	H (GPa)	E (GPa)	H (GPa)
CCA 3 mm $\eta = 2.77$	$192 \pm 8$	$10.46 \pm 0.42$	$181 \pm 3$	$11.69 \pm 0.50$	$127 \pm 5$	$8.39 \pm 0.43$
CCA 1 mm $\eta = 4.97$	$206 \pm 4$	$10.50 \pm 0.17$	$190 \pm 7$	$10.80 \pm 0.44$	$128 \pm 7$	$8.42 \pm 0.42$
A_CCA 1 mm $\eta = 9.57$	$206 \pm 7$	$10.10 \pm 0.49$	$201 \pm 15$	$11.61 \pm 0.46$	$133 \pm 4$	$8.90 \pm 0.52$

of the same compounds at the interface, and over the same thickness, for both the samples of 1 and 3 mm in diameter, meaning that the diameter of the sample does not influence the thickness of the IMC formed at the Cu/Al interface during the annealing process. Considering that the space between two adjacent indents is of 1  $\mu\text{m}$ , we could evaluate the total thickness of the IMC layers to about 23  $\mu\text{m}$ , which is in good agreement with the thickness measured on SEM micrographs on the same samples [21]. Accordingly, in this paper, the authors underline the fact that the growth of the IMC at the Cu/Al interface is independent of the diameter of the sample.

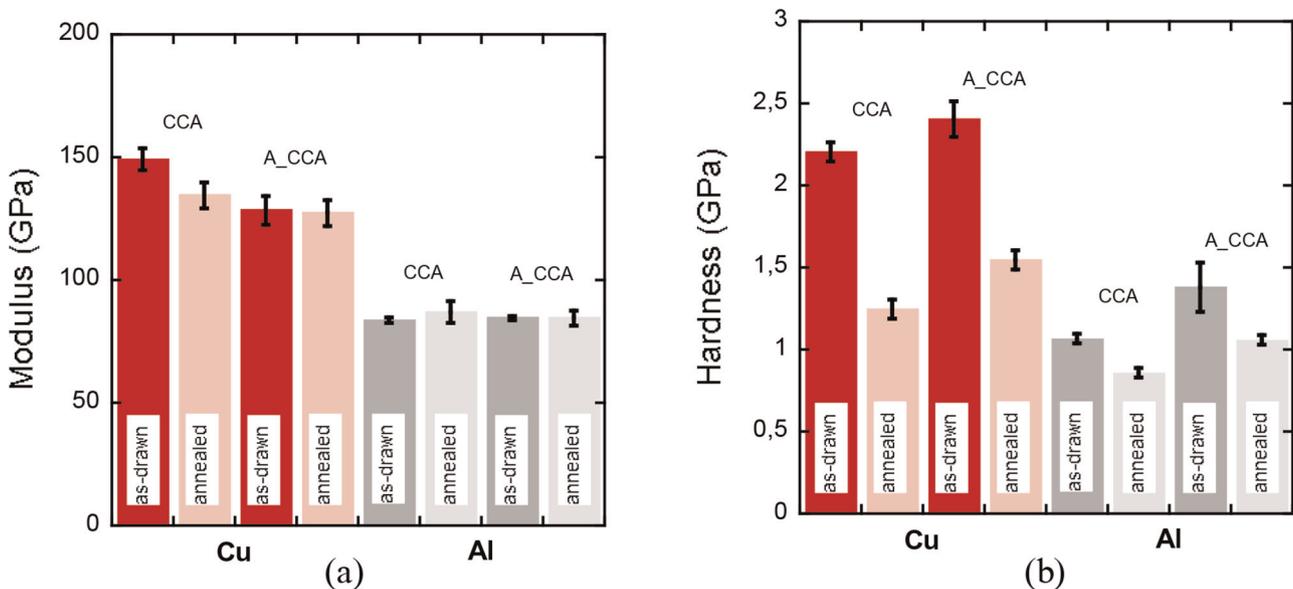
The Figure 6 clearly reveals the presence of at least three different compounds. Once more this result correlates what has been observed in previous works [18,21], reporting about the observation of three IMC through Cu/Al interfaces. These compounds were identified by EDXS analysis as  $\text{Al}_4\text{Cu}_9$ ,  $\text{AlCu}$  and  $\text{Al}_2\text{Cu}$ , over the interface from the Cu to the Al material (see insert in Fig. 1b). Thus it has been possible for us to link the loading curves and mechanical parameters obtained by nanonindentation to

the different compounds and their localization at the interface. Moreover,  $\text{AlCu}$  is described as presenting a higher hardness than the other phases, the hardness of  $\text{Al}_4\text{Cu}_9$  is close to the one of  $\text{AlCu}$ , but slightly smaller, and finally the hardness of  $\text{Al}_2\text{Cu}$  is intermediate between the one of  $\text{AlCu}$  and of the Cu and Al base materials [18,22]. This evolution of the hardness over the Cu/Al interface is well observed on the Figure 6b, for both the annealed CCA and A\_CCA samples. Accordingly, it is possible to estimate the thickness of each IMC layer, to about 7  $\mu\text{m}$  for the  $\text{Al}_4\text{Cu}_9$ , 5  $\mu\text{m}$  for the  $\text{AlCu}$  and 10  $\mu\text{m}$  for the  $\text{Al}_2\text{Cu}$ . Those results are also in good agreement with what was reported in [21] for the same samples.

The values of the modulus and the hardness of the different IMC layer are presented in the Table 3. As expected, the modulus and the hardness of the different IMC involved at the Cu/Al interfaces of our samples present very high values, up to more than 10 GPa for the hardness of  $\text{Al}_4\text{Cu}_9$  and  $\text{AlCu}$ . The comparison of the results obtained on the annealed CCA and A\_CCA samples reveals that both the evaluated mechanical



**Fig. 7.** Loading curves by nanoindentation on each compounds present at the copper/aluminum interface in (a) an annealed CCA sample and (b) an annealed A\_CCA sample, both of 1 mm in diameter.



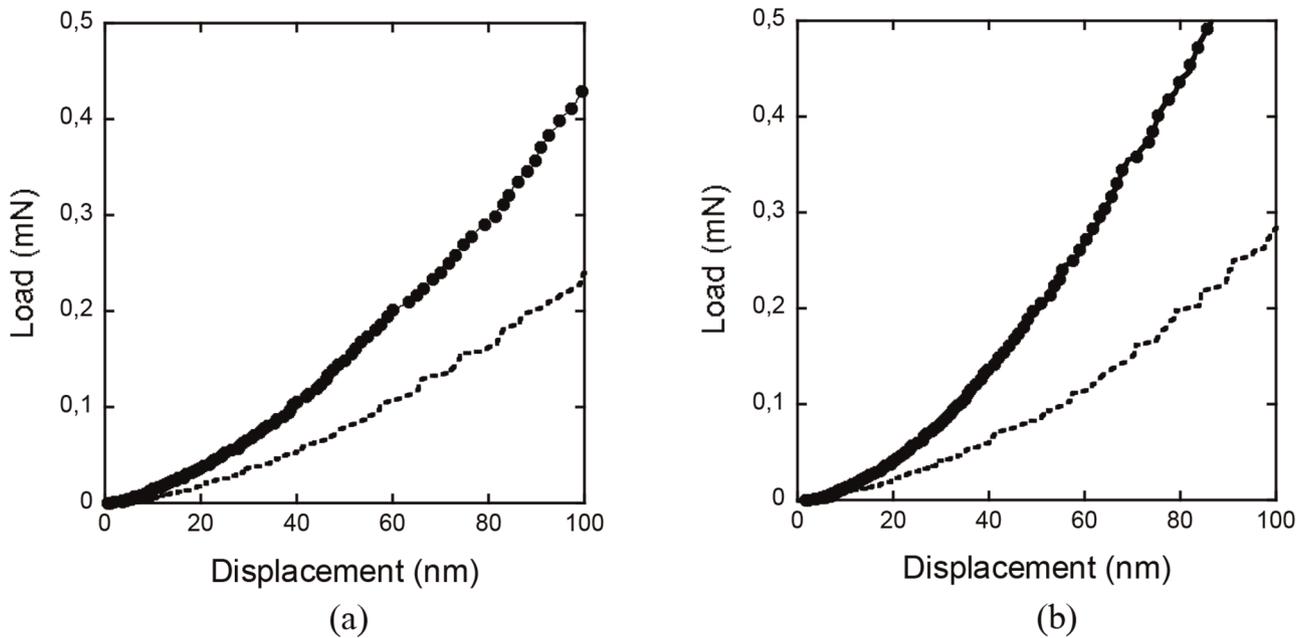
**Fig. 8.** Comparison of (a) the modulus and (b) the hardness of Cu and Al for the non-annealed and annealed CCA and A\_CCA samples of 1 mm in diameter.

parameters are very similar in the produced samples, suggesting that the architecture process applied to produce A\_CCA wires does not affect the mechanical behavior, and thus the structure of the IMC created during the thermal treatment.

The Figure 7 presents the loading curves by nanoindentation of the different components present in the annealed samples. The IMC compounds created during the thermal treatment also present an elasto-plastic behavior, with an important residual deformation. However, the plastic response of the IMC is smaller than the one of the copper and the aluminum, so that the presence of the IMC

at the Cu/Al interface in annealed CCA and A\_CCA samples results in a reduction of the ductility of the wires. According to the measurements of the modulus and the hardness on the annealed samples, the AlCu and the Al<sub>4</sub>Cu<sub>9</sub> require the highest loads compared to the Al<sub>2</sub>Cu.

The Figure 8 compares the values of the modulus and the hardness of the Cu and Al based materials before and after the annealing treatment. The modulus of both the copper and the aluminum remains quite constant after the annealing (except a slight decrease for the CCA sample), suggesting that no damage have been created in the base materials during the thermal treatment, or that it is



**Fig. 9.** Highlight of the pop-ins on the loading curves by nanoinindentation of Cu and Al in (a) an annealed CCA sample and (b) an annealed A\_CCA sample, both of 1 mm in diameter.

compensated by a thermally-activated recovery phenomenon. On the other hand, we observe for all the samples, whatever the architecture, a significant decrease in the hardness of both Cu and Al after the annealing, of about 40% for the Cu, and 20% for the Al. Such a decrease in the hardness might be explained by the recrystallization of the material. Indeed, in previous works on CCA and A\_CCA samples [15,21], it has been clearly demonstrated that recrystallization occurs in both copper and aluminum during annealing.

Another confirmation of the recrystallization phenomenon is given by the study of pop-ins, whose presence is correlated to the probability to trigger dislocation sources underneath the indenter tip. Therefore, the higher the dislocation density in the tested material is, the lower the probability to generate pop-in. The Figure 9 despite a zoom of the first steps of the loading curve of copper and aluminum during the indenter penetration. Compared to the loading curve of a non-annealed CCA sample (see insert on Fig. 3), we note after annealing the apparition of pop-in on the loading curve of the copper, related to a dislocation density reduction, due to a recrystallization phenomenon.

## 4 Conclusions

In this work, we investigate the influence of the architecture of copper-clad aluminum composites by cold-drawing process, and the drawing ratio, on the mechanical properties of both the base materials, and the intermetallic compounds that develop at the interface after annealing. Architected-CCA rods, fabricated by stacking and re-drawing of original CCA wires, are considered because the multiplication of Cu/Al interfaces and of Cu paths inside the final wires is expected to enhance their mechanical and electrical performances. For this study, simple CCA and

architected A\_CCA wires were tested before and after a thermal treatment at 400 °C for 16 h, leading to the diffusion of copper and aluminum to form ICM at the interfaces. Nanoindentation tests were performed on the different samples, to measure the hardness, which is related to the yield strength, and the elastic modulus at the microscale, allowing to map mechanical properties gradient in relation with the microstructure. It was shown that the recrystallization phenomenon, which takes place in both the Cu and the Al subjected to severe plastic deformation, can be well catch by nanoindentation, as well as the formation of the three intermetallic compounds at the interface. On the other hand, our measures underline that the mechanical properties of all the components involved in the wires, i.e., Cu, Al, and eventually the IMC developing after annealing, are only slightly influenced by the structuration of the wires. IMC developing at the Cu/Al interfaces present a high hardness, much higher than both the base copper and aluminum, and might thus deteriorate the mechanical properties of the wires, even more A\_CCA wires, which exhibit more interfaces than the traditional CCA samples. Accordingly, special attention has to be paid to determinate the annealing procedure of the architected wires that would allow to improve their electrical performances without degrading their mechanical resistance.

## References

1. T.P.D. Rajan, B.C. Pai, Developments in processing of functionally gradient metals and metal-ceramic composites: a review, *Acta Metall. Sin.* **27**, 825–838 (2014)
2. S.C. Tjong, Y.-W. Mai, Processing-structure-property aspects of particulate- and whisker-reinforced titanium matrix composites, *Compos. Sci. Technol.* **68**, 583–601 (2008)

3. M.U. Saeed, B. Li, Z. Chen, et al., A novel processing approach to produce microchannel embedded carbon-epoxy composites, *J. Manuf. Process.* **22**, 26–33 (2015)
4. I.-K. Kim, S.I. Hong, Effect of heat treatment on the bending behavior of tri-layered Cu/Al/Cu composite plates, *Mater. Des.* **47**, 590–598 (2013)
5. L. Kunčická, R. Kocich, Deformation behaviour of Cu-Al clad composites produced by rotary swaging, *IOP Conf. Ser. Mater. Sci. Eng.* **369**, 1–8 (2018)
6. M. Aravind, P. Yu, M.Y. Yau, et al., Formation of Al<sub>2</sub>Cu and AlCu intermetallics in Al(Cu) alloy matrix composites by reaction sintering, *Mater. Sci. Eng. A.* **380**, 384–393 (2004)
7. T. Jin, G. Li, Y. Cao, et al., Experimental research on applying the copper-clad aluminum tube as connecting tubes of air conditioners, *Energy Build.* **97**, 1–5 (2015)
8. N. Kazuyuki, M. Kazuo, H. Chihiro, Development of manufacturing process of clad bar by rotary rolling, *ISIJ Int.* **37**, 899–905 (1997)
9. K.Y. Rhee, W.Y. Han, H.J. Park, et al., Fabrication of aluminum/copper clad composite using hot hydrostatic extrusion process and its material characteristics, *Mater. Sci. Eng. A* **384**, 70–76 (2004)
10. M. Zebardast, A.K. Taheri, The cold welding of copper to aluminum using equal channel angular extrusion (ECAE) process, *J. Mater. Process. Technol.* **211**, 1034–1043 (2011)
11. R. Kocich, A. Macháčková, L. Kunčická, et al., Fabrication and characterization of cold-swaged multilayered Al-Cu clad composites, *Mater. Des.* **71**, 36–47 (2015)
12. R. Kocich, L. Kunčická, C.F. Davis, et al., Deformation behavior of multilayered Al-Cu clad composite during cold-swaging, *Mater. Des.* **90**, 379–388 (2016)
13. X. Li, G. Zu, M. Ding, et al., Interfacial microstructure and mechanical properties of Cu/Al clad sheet fabricated by asymmetrical roll bonding and annealing, *Mater. Sci. Eng. A* **529**, 485–491 (2011)
14. L.Y. Sheng, F. Yang, T.F. Xi, et al., Influence of heat treatment on interface of Cu/Al bimetal composites fabricated by cold rolling, *Compo. Part B Eng.* **42**, 1468–1473 (2011)
15. A. Gueydan, B. Domengès, E. Hug, Study of the intermetallic growth in copper-clad aluminum wires after thermal aging, *Intermetallics* **50**, 34–42 (2014)
16. M. Abbasi, A. Karimi Taheri, M.T. Salehi, Growth rate of intermetallic compounds in Al/Cu bimetal produced by cold roll welding process, *J. Alloys Compd.* **319**, 233–241 (2001)
17. W.-B. Lee, K.-S. Bang, S.-B. Jung, Effects of intermetallic compound on the electrical and mechanical properties of friction welded Cu/Al bimetallic joints during annealing, *J. Alloys Compd.* **390**, 212–219 (2005)
18. E. Hug, N. Bellido, Brittleness study of intermetallic (Cu, Al) layers in copper-clad aluminium thin wires, *Mater. Sci. Eng. A* **528**, 7103–7106 (2011)
19. L. Kunčická, R. Kocich, T.C. Lowe, Advances in metals and alloys for joint replacement, *Prog. Mater. Sci.* **88**, 232–280 (2017)
20. W. Głuchowski, Z. Rdzawski, J. Domagała-Dubiel, et al., Microstructure and properties of multifibre composites, *Arch. Metall. Mater.* **61**, 911–916 (2016)
21. F. Moisy, A. Gueydan, X. Sauvage, et al., Influence of intermetallic compounds on the electrical resistivity of architecture copper clad aluminum composites elaborated by a restacking drawing method, *Mater. Des.* **155**, 366–374 (2018)
22. C.Y. Chen, W.S. Hwang, Effect of annealing on the interfacial structure of aluminum-copper joints, *Mater. Trans.* **47**, 1938–1947 (2007)
23. W.C. Oliver, G.M. Pharr, An improved technique for determining hardness and elastic-modulus using load and displacement sensing indentation experiments, *J. Mater. Res.* **7**, 1564–1583 (1992)
24. W. Nix, H. Gao, Indentation size effects in crystalline materials: A law for strain gradient plasticity, *J. Mech. Phys. Solids* **46**, 411–425 (1998)
25. R. Kocich, L. Kunčická, A. Macháčková, et al., Improvement of mechanical and electrical properties of rotary swaged Al-Cu clad composites, *Mater. Des.* **123**, 137–146 (2017)
26. F. Moisy, X. Sauvage, E. Hug, Investigation of the early stage of reactive interdiffusion in the Cu–Al system by in-situ transmission electron microscopy, *Materialia* **9**, 100663 (2020)
27. X. Hernot, O. Bartier, Y. Bekouche, et al., Influence of penetration depth and mechanical properties on contact radius determination for spherical indentation, *Int. J. Solids Struct.* **43**, 4136–4153 (2006)

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