MICROSTRUCTURE AND PROPERTIES OF COPPER DEFORMED BY ACCUMULATIVE ROLL-BONDING

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ABSTRACT

Accumulative Roll Bonding (ARB), which is a process of severe plastic deformation (SPD), was applied to commercially pure copper 99, 97 % by performing up to 7 ARB cycles at room temperature (RT) conditions without lubrication, at cryogenic temperature (CT) conditions, and high temperature (HT) conditions, respectively. Microstructural characterizations were performed by optical microscopy (OM) and scanning electron microscopy (SEM). Observations revealed that refined grains were produce in Cu after ARB and, the continuous recrystallization microstructure was observed. Vickers hardness (VH) measurements were performed on the deformed samples, showing a gradual increase of hardness showing a gradual increasing of hardness with increasing strain/decreasing grain size at first, but then a decrease of hardness with further increasing strain.
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### GLOSSARY

### ABREVIATIONS

<table>
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<th>Abbreviation</th>
<th>Description</th>
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<tr>
<td>ARB</td>
<td>Accumulative Roll-Bonding</td>
</tr>
<tr>
<td>ASTM</td>
<td>American Society for Testing and Materials</td>
</tr>
<tr>
<td>CT</td>
<td>Cryogenic Temperature</td>
</tr>
<tr>
<td>FCC</td>
<td>Face Centred Cubic</td>
</tr>
<tr>
<td>HT</td>
<td>High Temperature</td>
</tr>
<tr>
<td>HV</td>
<td>Hardness Vickers</td>
</tr>
<tr>
<td>IACS</td>
<td>International Annealed Copper Standard</td>
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<tr>
<td>OM</td>
<td>Optical Microscopy</td>
</tr>
<tr>
<td>RT</td>
<td>Room Temperature</td>
</tr>
<tr>
<td>SPD</td>
<td>Severe Plastic Deformation</td>
</tr>
<tr>
<td>SEM</td>
<td>Scanning Electron Microscopy</td>
</tr>
<tr>
<td>UFG</td>
<td>Ultra-Fine Grain</td>
</tr>
<tr>
<td>UNE-EN</td>
<td>European Standard</td>
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SYMBOLOGY

$A$  Surface
$C_{v}^{SP}$  Specific heat at constant volume
$c_v$  Thermic capacity for unity mass
d  Average grain diameter.
d$_s$  Grain size
$\Delta T$  Temperature gradient
$\Delta x$  Distance gradient
F  Load
k  Constant of extent measure of failure dislocations in front the barriers.
$K$  Thermal conductivity of a solid
$K_L$  Phonon thermal conductivity
$K_S$  Phonon thermal conductivity of crystals
H  Normal direction
L  Length
$q$  Thermal energy
$R_1$  Resistance electric Up
$R_2$  Resistance electric down
$r_1$  Total reduction
\( \sigma_y \)  \hspace{1cm} \text{Yield stress.}

\( \sigma_0 \)  \hspace{1cm} \text{Friction stress}

\( T \)  \hspace{1cm} \text{Temperature}

\( t \)  \hspace{1cm} \text{Time}

\( t_0 \)  \hspace{1cm} \text{Initial thickness strip}

\( \nu \)  \hspace{1cm} \text{Average rate of the particles}
1. – INTRODUCTION

1.1.- MOTIVATION AND AIM

Accumulative Roll Bonding ARB is a relatively new method of severe plastic deformation, as proposed by Saito et al [1]. The goal is to impose an extremely high tension in the material through a grain refinement and thus obtain an increased strength of improved mechanical properties without changing the dimensions. Being able to obtain a material with improved mechanical properties without the addition of additives or alloys is very interesting because copper has higher conductivity than alloyed copper, with a difference of 10-15% IACS (International Annealed Copper Standard). Altering the chemical composition of the material also alters the electrical and thermal properties. Through the process of ARB we can improve the material’s electrical and thermal properties without altering the dimensions. Therefore, comprehension of the underlying mechanisms of deformation induced grain refinement is essential both from theoretical and technological points of view, required for the development of advanced plastic deformation techniques to produce in large quantities for engineering applications.
1.2. THEORETICAL APPROACH

1.2.1. INTRODUCTION OF COPPER

1.2.1.1. CHARACTERISTIC AND CRISTALOGRAHY

Copper is a chemical element with the symbol Cu and atomic number 29. Copper is in group 11 of the periodic table, and it shares certain attributes with elements in that group: they have one s-orbital electron on top of a filled d-electron shell, and are characterized by having a high ductility and electrical conductivity. For its properties, copper is used as a conductor of heat and electricity, a building material, and a constituent of various metal alloys.

<table>
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<th>Table 1. Copper properties.</th>
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</tr>
<tr>
<td>Boiling Point</td>
</tr>
<tr>
<td>Melting Point</td>
</tr>
<tr>
<td><strong>ELECTRICAL PROPERTIES</strong></td>
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<tr>
<td>Electrical resistivity (20°C)</td>
</tr>
<tr>
<td>Temperature coefficient (0-100°C)</td>
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<td><strong>THERMAL PROPERTIES</strong></td>
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<td>Thermal conductivity</td>
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<td>Thermal expansion (25°C)</td>
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<td><strong>MECHANICALS PROPERTIES</strong></td>
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<tr>
<td>Young’s modulus</td>
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<tr>
<td>Tensile strength</td>
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<tr>
<td>Vickers Hardness</td>
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<td>Brinell Hardness</td>
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</table>
1.2.1.2. – STRUCTURE CRystalline

Copper doesn’t present allotropic variety. It has a stable structure at room temperature, high and low temperatures.

Copper has a structure formed by grains in a regular system known as face cubic centered (FCC). This means that the atoms are arranged like vertexes of a cube, and at the cube face centres. Other materials that tend to crystallize this into the structure are aluminium, silver or gold.

Materials with FCC structure tend to increase their sensitivity strain rate deformation due to a decrease in the activation volume in nanocrystal system.

The sliding’s direction of copper is <110> and their slip planes are {111} being that offer a minimum shear strength.

Fig. 1. Schematic illustration of structure FCC[2]

1.2.2.-INTRODUCTION OF HALL-PETCH RALTIONSHP

A general relationship between the yield stress and the grain size was proposed by Hall and extended by Petch. They address about how hardening is basically tied to grain size reduction. This method relies on the fact that the common crystallographic presentations thereof are different. Upon arriving at the grain boundary, dislocation stops and continues to deformation, which involves the application of more energy and therefore greater material strength.
The fact that curv dislocations cause to perform deformation is necessary to apply a high stress. Therefore, a fine-grained material is harder and tougher than a material with bigger grains. This behavior is defined by the Hall-Petch equation as below:

\[ \sigma_y = \sigma_0 + \frac{k_y}{\sqrt{d}} \]  

(1)

\( \sigma_y \) = Yield stress.

\( \sigma_0 \) = Friction stress, represents the total resistance to movement of network’s dislocations.

\( k \) = Constant of extent, measure of failure dislocations in front barriers.

\( d \) = Average grain diameter.

That is, the initial yield stress \( \sigma_0 \) is increased according to a constant \( k \) of the material average grain diameter \( d \), thus obtaining the new yield stress \( \sigma_y \).

The original dislocation model for the Hall-Petch equation was based on the concept that grain boundaries act as barriers to motion dislocation.

The factor \( k \) is the slope of the line obtained when \( \sigma_y \) is represent in front \( d^{-1/2} \). The term \( \sigma_0 \) is the intersection along the ordinate in a graph of \( \sigma_y \) against \( d^{-1/2} \). It’s interpreted as the shear stress required to move the dislocations unlocked along the sliding plane. This term depends largely on temperature, stress, and the content of impurities in the alloy.

The influence of grain boundaries on the properties may be important, but this depends on the exact conditions of deformation and the material. At low temperatures, the grain boundaries act as barriers to dislocation motion, thus strengthening the material. At elevated temperatures the grain can’t reduce the strength for both diffusion and dislocation motion.

The Hall-Petch relationship isn’t valid for materials with exceptionally large grains, or UFG, then one speaks of the inverse Hall-Petch.
In the reverse Hall-Petch the material softens when the grain size is reduced [4]. Many explanations have been proposed, which are summarized below. It seems reasonable to suppose that the mechanism of deformation is different in the samples according to the production of grain was essentially free of dislocation, and the severe plastic deformation, where the small grains are produced by breaking large grains through a dislocation intense activity. The most commonly proposed explanations are: increased creep diffusion, suppression of dislocation cells, different structures of grain boundaries, the poor quality of the sample (porosity and other defects), and the deformation in the grain boundaries.

Fig 2. Schematic Hall-Petch relation, showing the strength of materials as a function of grain size. First the grain size reduction leads to increase of the strength, the saturation of grain size between 20 and 30 nm, and then the rapid softening. Critical grain size $d_c$ where the transitions is strengthening (Hall-Petch) occurs to weaken (inverse Hall-Petch). [3]
1.2.3. - INTRODUCTION THERMAL OF CONDUCTIVITY

The thermal conductivity of a solid \( k \) is defined by relating the steady flow of thermal energy \( q \) along a long bar with a temperature gradient [5]:

\[
q = KA\frac{\Delta T}{\Delta x}
\]

(2)

Where \( q \) is the flow of thermal energy. The form of equation (2) defining the conductivity means that the thermal transfer energy is a process in which the energy not merely enters one end and goes directly straight to the other, but rather, diffuses through the sample undergoing multiple scattering events through collision scatterers, such as defects, grain boundaries, or very massive ion elements, among others. If energy is propagated directly undeflected through the sample, the expression of thermic flow wouldn’t depend on the temperature gradient \( \Delta T \), the ends sample and the length. The random nature of the process of conductivity introduces a temperature gradient and mean free path in the expression of the thermal flow. The average distance phonons traveling without being scattered or not interaction between them is called the mean free path.

From the kinetic theory of gases we find the following expression for the thermal conductivity:

\[
k = \frac{1}{3} C_V^{SP} \nu l
\]

(3)

Where \( C_V^{SP} \) is the specific heat at constant volume and \( \nu \) is the average rate of the particles. Debye considered the phonoms as particles and applied this result for the first time to describe the thermal conductivity in solid dielectrics. Thus, developing the elementary kinetic theory leads to the equation (3). The particle flowing the direction \( x \cos \frac{1}{2} n \langle |v_x| \rangle \), where \( n \) is the concentration of particles; on balance there is a flow of the same magnitude in the opposite direction. The sign \( \langle \cdots \rangle \) represents the mean value. \( C_V \) is the thermic capacity for unity mass, or heat specific from a particle. If the particle moving from a region with local temperature \( T + \Delta T \) to another temperature \( T \), the particle yields one \( C_V \Delta T \) of energy. The term \( \Delta T \) between the ends of a free path of the particle is given:
$$\Delta T = \frac{dT}{dx} l = \frac{dT}{dx} v_x \tau$$  \hspace{1cm} (4)

Where t is the time between two crashes. The net flow energy (due to particle flow in both directions) is:

$$Q = n \langle v_x^2 \rangle c_x \tau \frac{dT}{dx} = \frac{1}{3} n \langle v_x^2 \rangle c_x \tau \frac{dT}{dx}$$  \hspace{1cm} (5)

For the phonons v is constant and we can write (5) as:

$$Q = \frac{1}{3} C_v^{SP} v^4 \frac{dT}{dx}$$  \hspace{1cm} (6)

To develop a theory of thermal conductivity must know the rate at which phonons can transfer thermal energy within a crystal. Within the crystal the phonons are scattered continuously and can interact with them. We can tailor the kinetic theory (classical) driving thermal energy of the gases to the problem of the thermal energy transfer by phonons, which for all purposes is considered almost particles, and using equation (3) where v is the speed of sound. Thus, the main problem that arises when studying the thermal conductivity is to determine the behaviour of the average phonon free path due to different scattering mechanisms that can occur in the solid.

Several phonon scattering mechanisms that can limit the value of the average phonon free path:

1. Phonon interaction «Umklapp-processes».

2. Phonon scattering by point defects such as impurities, isotopes; crystal atoms with the same number of protons but different numbers of neutrons, etc.

3. Phonon scattering by the boundaries of the specimen or the crystallites.

2. – EXPERIMENTAL PROCEDURES

2.1. – PROCESS OF ACCUMULATIVE ROLL-BONDING.

The material used for this study is copper 99.95% purity arranged in sheets. The material has been processed previously annealing heat treatment at 600ºC during two hours in a protective argon’s atmosphere. Protective atmosphere is required over 400ºC, due to the hydrogen in the air reacts with impurities of oxygen that the sample contains. In the reaction are water particles that deposit on the grain’s edges generating a faulty material.

Annealing heat treatment is applied to eliminate the history of coming tensions from the manufacturing process of copper sheets. Thus we ensure that the samples doesn’t have any prior to the ARB process deformations.

In the Accumulative Roll Bonding process we begin from two sheets of copper with initial size 10mm width x 20 mm long x 1mm of thickness. Brushing the surface of the leaves to a polish is of prime importance before stacking them, in order to allow their adhesion. A strip is carefully placed on top of the other band, then inserted between the laminating rollers where the two layers of material are attached to each other due to the pressure exerted by the rollers during the laminate. Then, the material’s length is altered. New laminate is sectioned into two halves. The two band’s interfaces are treated on surface with a scraping with metallic fibers driven by an electric mill and cleaned with acetone to remove dirt and grease in order to improve the bond strength. Section strips are stacked and united again in the rollers. The entire process is repeated over and over again. The process can introduce ultra-high plastic strain without any geometric change if the reduction in thickness stays by 50% each lamination pass.
The ARB process has been carried out during seven cycles maximum at room temperature, a six cycles maximum for the cryogenic temperature and six cycles maximum for high temperature. All of them carried out without any lubrication and were brought to the air.

After seven cycles at room temperature, cracks begin to form due to a large amount of plastic strain in the material, making it impossible to continue with the subsequent lamination cycles.

Each cycle creates a 50% reduction, the initial thickness of the strip after \( n \) cycles is defined in equation (7) where \( t_0 \) is the initial thickness strip:

\[
t = \frac{t_0}{2^n}
\]

The total reduction \( r_t \) after \( n \) cycles is equation (8):

\[
r_t = 1 - \frac{t}{t_0} = 1 - \frac{1}{2^n}.
\]

Assuming the criterion of Von Mises’s efficiency and strain plain state, so to speak, without lateral displacement, equivalent plastic strain \( \varepsilon \) is expressed by equation (9)[8]

\[
\varepsilon = \left\{ \frac{2}{3} \ln \left( \frac{1}{2} \right) \right\} \times n = 0.80n.
\]
In case that the process is repeated over seven times, the thickness is reduced to 1/128. The initial thickness of 1.0 mm is reduced to 7.8 μm. The total reduction achieved is 99.2% and the total plastic strain equivalent is 5.6. It’s easy to introduce high stress inside the materials by the ARB process.

<table>
<thead>
<tr>
<th>No. of cycles</th>
<th>1</th>
<th>2</th>
<th>3</th>
<th>4</th>
<th>5</th>
<th>6</th>
<th>7</th>
<th>8</th>
<th>9</th>
<th>10</th>
<th>n</th>
</tr>
</thead>
<tbody>
<tr>
<td>No. of layers</td>
<td>2</td>
<td>4</td>
<td>8</td>
<td>16</td>
<td>32</td>
<td>64</td>
<td>128</td>
<td>256</td>
<td>512</td>
<td>1024</td>
<td>2^n</td>
</tr>
<tr>
<td>No of bonded boundaries</td>
<td>1</td>
<td>3</td>
<td>7</td>
<td>15</td>
<td>31</td>
<td>63</td>
<td>127</td>
<td>255</td>
<td>511</td>
<td>1023</td>
<td>2^n-1</td>
</tr>
<tr>
<td>Layer interval (μm)</td>
<td>500</td>
<td>250</td>
<td>125</td>
<td>62.5</td>
<td>31.2</td>
<td>15.6</td>
<td>7.8</td>
<td>3.9</td>
<td>1.9</td>
<td>0.96</td>
<td>1000/2^n</td>
</tr>
<tr>
<td>Total reduction (%)</td>
<td>50</td>
<td>75</td>
<td>87.5</td>
<td>93.8</td>
<td>96.9</td>
<td>98.4</td>
<td>99.2</td>
<td>99.6</td>
<td>99.8</td>
<td>99.9</td>
<td>(1 - 1/2^n) × 100</td>
</tr>
</tbody>
</table>
| Equivalent strain | 0.8| 1.6| 2.4 | 3.2 | 4.0 | 4.8 | 5.6 | 6.4 | 7.2 | 8.0 | \( \frac{1}{\sqrt{3}} \ln 2 \) n = 0.8 n

*Table 2. Geometrical changes of the materials during the ARB where two pieces of the sheets 1mm thick are roll-bond by 50% reduction cycle. [9]*

2.2. – ARB WITH TEMPERATURE VARIATION

2.2.1. – CRYOGENIC TEMPERATURE

Samples were immersed at cryogenic temperatures in liquid nitrogen for twenty minutes until reaching 77.35 Kelvin degrees, then the ARB was made quickly to avoid the rise of temperature. Immersion in liquid nitrogen was repeated before each rolling cycle.

2.2.2. – RECRYSTALLIZATION TEMPERATURE

For the specimens at high temperature, two rolling cycles at room temperature were performed, then the sample was deposited in the oven at 130ºC for 30 minutes. The third and fourth cycles were conducted at room temperature after that, and samples were heated again at 130ºC for
another 30 minutes. Finally a fifth and sixth cycles were done at room temperature. It’s possible to observe in the graph 1, that for 30 minutes at 130ºC, the recrystallization is 100%.

![Graph showing percent crystallization of pure copper as a function of time at constant temperature.](image)

*Fig. 4. Percent crystallization of pure copper as a function of time at constant temperature.* [10]

2.3. – MICROSTRUCTURE

The characterization technique of scanning electronic microscopy (SEM) and optical microscopy (OM) were used for the observation of the microstructure of samples.

Specimens with transverse and longitudinal sections to normal direction have been created.

The preparation of samples for the analysis consisted of sanding the surfaces with different thicknesses of sandpaper, increasingly finer from grade 200 up to 2000, followed by a polish with velvet and a suspension of 0.05 µm of silica particles to achieve a very smooth surface. The samples were corroded with a solution of CrO₃ 200 g, HCl (35%) 17 mL, N₂S₂O 20g, H₂O₃ 1000 mL.

The surface was rinsed with current water and soap to remove all traces of the attack and dried with air to try to avoid the formation of rust.

To observe the surface of the copper specimen an OM has been used, through a set of lenses that increases and reproduce the image up to a size 0.2 µm.
In order to observe more accurately the micro-structure of the copper, a SEM has been required. The SEM is based on the principle of optical microscopic, in which the light beam is replaced by an electrons beam. With this method we can up to the 100 nm with a resolution much higher than any other optical instrument.

Operation is to affect a sweeping beam of electrons onto the sample. The sample must be conductive, so in our case conductivity has been improved by driving with a Platinum sputtering.

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**Fig. 5. Schematic illustration of OM configuration.**

**Fig. 6. Schematic illustration of SEM configuration [11]**
2.4. – MECHANICAL PROPERTY. HARDNESS

A hardness test is simple and of high-performance, and is particularly useful for evaluating the different microstructural components of material properties.

Vickers procedure employs a diamond indenter in the shape of pyramid tetrahedral of 136°. Such indenter is applied perpendicularly to the surface of hardness to be measured, under the action of a load $F (N) - 50 g$ for this study-. This charge is maintained for some time $t = 5$ seconds for this study- after which it is removed, and measure the diagonal $d$ of the print that is on the surface of the sample (see figure 4). With this value and the appropriate tables it’s possible to get the Vickers hardness, which is characterized by $HV$ and defined as the ratio of the load applied and the area of the lateral surface of a print.

$$HV = \frac{F}{A} = \frac{F}{d^2/2 \cdot \sin 68} \Rightarrow HV = 0.1891 \cdot \frac{F}{d^2}$$

(10)

An AXS-1000A microdurometer has been used to measure hardness data, and the standard method has been followed as specified by ASTM 384-89 / UNE-EN 6507-2.

Fig. 7. Image of sample del microdurometer a) Geometry of the diamond’s pyramid b) Mark left on the material

Vickers hardness is considered a plastic strain because it leaves a permanent residual mark, left by the tip of the indenter geometry, and we can assume error tolerance neglectable since the force applied is large enough and the materials aren’t very elastic.
2.5. – THERMAL PROPERTIES

To take data for copper’s thermal properties, the sample has been arranged in a vacuum chamber to prevent propagation of thermal energy through the air. A heater has been attached to the chamber, plus two thermal sensors to adjust the heat flow accordingly.

Resistivity was measured with a digital Agilent multimeter (34401A 6½), while measuring current intensity required a Uni-T multimeter (UT805A). Data was taken in a range of different amperage values, from 10 mA up to 20 mA, for a greater accuracy in the calculus of electro-resistance.

The dimensions are: thickness 42 μm x width 1,5 mm x longitude 8 mm.

![Image of system used to measure the difference temperature.](image)

First, temperature data is taken with a sensor pointing up and a sensor pointing down. Temperature is applied through the main heater, so by finding out the inferred amperage we can deduct the electro-resistance that the sample provides. The experiment has been repeated both in upstream as downstream current. With the temperature and resistance data taken, following graphic has been created, where the relationship can be observed.
Sample with seven cycles ARB:

With the arithmetic mean of the two equations obtained in the graphs we get the following equation:

\[ R_1 = 0.00397 T + 1.0009 \] (11)

Where the temperature coefficient is 0.00397, \( R_1 \) is the electric resistance and \( T \) is the Up temperature sensor.

Following the same process for the Down sensor gives the equation:

\[ R_2 = 0.00387 T + 1.0069 \] (12)
Where $R_2$ equals electric resistance. The temperature coefficient is 0.0385 and $T$ is the Down temperature sensor.

Initial data for the electro-Resistance of sensor $R_1$ (Up) = 1,1016 KΩ and $R_2$ (Down) = 1,10336 KΩ.

Taking variations between 10 and 20 mA, we obtain different torque values $R_1$ (Up) and $R_2$ (Down).

To calculate the differential resistance:

$$\Delta R_1 = R_1 - R_1 \quad \Delta R_2 = R_2 - R_2 \quad (13)$$

For temperature differentials for sensor Up1 and the Down2 use differential resistance between temperature coefficients:

$$\Delta T_1 = \frac{\Delta R_1}{0.003977} \quad \Delta T_2 = \frac{\Delta R_2}{0.0038675} \quad (14)$$

The current flux provides it following equation:

$$q = KA \frac{\Delta T}{\Delta x} \quad (15)$$

Where $\Delta x$ = Length of sample and $\frac{\Delta T}{\Delta x} = \frac{\Delta T_1 - \Delta T_2}{0.008} \quad (16)$

Isolating the $K$ value of the thermal conductivity will be obtained.
3. RESULTS AND DISCUSSION

3.1. EVOLUTION OF MICROSTRUCTURE

3.1.1. MICROSTRUCTURE IN OM

To perform a comprehensible characterisation of a material, it’s necessary to learn about the structure presented, defined by its process of manufacture and composition. In our case of study, the material presented is in the state of reception from manufacture, without variation of composition. This is important because the manufacturing process affects the grain size and orientation.

A good preparation of the simples, accompanied by an accurate interpretation of the microstructures observed, can solve a large number of metallurgical problems during the development and production of copper. Given the large grain size of the studied alloys, it’s possible to see their morphology. Here are the cross sections of the samples taken with optical microscope to be examined:

![Fig. 9. Sample pure cooper without ARB.](image1)
![Fig. 10. Sample with 1 cycle ARB.](image2)

The main visible feature with OM, is that in pure copper, the structure is granular, with equiaxial grains in random orientations. The thickness of grains are around 25 μm. However in the first
cycle of ARB, there is a mixture of grains that have not deformed, without guidance with other grains that suffer a reduction in thickness down to the 15 μm, and an elongated morphology oriented in the direction of the effort. The twins within the grains can be identified, this tells us that there has change of stacking sequence in the grain. The twinning is a mechanism, supplementary to slip, by which the material is hardened. The twins have been formed as a result of the shear force applied parallel to the same plane, and in the same direction. The twin prevents the movement of dislocations, impeding the process of sliding, still a hardening mechanism and creating partition of grains on grain size.

In the second cycle, grains are visually well appreciated and the orientation of these is clearly seen. They are likely two different types of grains, the equiaxial and the morphologically elongated, both types coexisting.

In the third cycle we it is observed a severe reduction of thickness of grain size, from the 15 μm of the second cycle down to 3 μm. In the third cycle, grain boundaries become less visible since grains are much thinner and elongated in the direction of the laminate.

In this third cycle, the union during the laminate has been higher than in previous cycles. This indicates that the laminate from the third cycle improves enough union interfaces introduced in the previous ones.
In the fourth cycles, samples show a more uniform structure and beads with a fairly large strain. Average grain thickness is 1,2 μm, about half the thickness of a grain from the simple of three cycles. The density of dislocations is high due to the large deformation suffered by the material.

In the fifth cycles, it’s possible to observe in the sample small grains of recrystallization along with the UFS micro-structure. The beads’ average thickness has increased slightly due to the process of recrystallizing. It goes from 1,2 μm in 4th cycles up to 2,8 μm.
The suffering material is continuously recrystallizing, characterized by the subdivision of ultra-fine grain, recovered to form new ultra-fine grains, and the short range migration of grain boundaries. [13]

Fig. 16. Schematic diagram showing the continuous recrystallization of a highly deformed lamellar microstructure. (a) Initial structure, (b) Collapse of the lamellar boundaries, (c) Spheroidisation begins and (d) Further spheroidisation and growth. [14]

First, the material suffers a collapse of the laminar structure due to the surface stress that is generated during deformation. The critical condition of collapse occurs when the relationship between the length L in the direction of the grain and the length in the normal direction H (L/H) is $\sim 4$. [15]

It’s then when the grain size is relatively stable and the average movement of a free dislocation is shorter, that the formation of dislocation in the grain inside cells is extremely limited. Motion of dislocation slips easily without a blockage inside the limit of grain and creates subcells that eventually go away as a result of the dynamic recovery local. At the same time, dislocation (focus on Frank-Read) generators would be limited by the lack of dislocation cells inside the grains, which would further promote the dynamic recovery.

Then new grain growth happens by adiabatic warming.
The six cycles with liquid nitrogen created a grain size of 1.10 μm, smaller than the sample of six cycles at room temperature with a grain size of 1.30 μm, but notably the difference isn’t very large.

The rolling at low temperature eases the extraordinary accumulation of high densities of dislocations in copper. The temperature of strain has strong effects on the operating mechanisms and the grain size obtained in the ultrathin regimes and nanocristallins. Low temperatures of strain suppress dynamic recovery and, therefore, facilitate grain refining mechanisms.

Apparently, between the six cycles at room temperature with grain size of 1.30 μm and the seven cycles sample with grain size of 1.40 μm, there aren’t many differences.
3.1.2. – THICKNESS EVOLUTION OF GRAIN AT ROOM TEMPERATURE

A follow-up of the evolution of the thickness of grain with different cycles of ARB at ambient temperature has been performed with the optical microscope. The size of the grains in a metal affects its mechanical properties, so it’s important to know their nature.

Fig. 21. Evolution of the grain in RT image in OM.
It’s possible see that the average size of grain thickness decreases gradually as the number of ARB cycles increases, from 25 μm to approximately 1,42 μm. The following graph helps visualize the evolution of grain thickness according to the number of ARB cycles:

In the first cycle, it is observed a great decline in the thickness of the grain, as in a mash of cooper, there is no displacement of the anchored, these move freely by the slip plane without encountering any obstacles.

In the second cycle, the reduction of grain is minor, since the first cycle grains have different orientations, the dislocations that will pass the following grain will have to change the direction of their movement by breaking the barrier of the limit of grain. This becomes more difficult the reduction of grain.

*Graph 6. Thickness grain in μ m vs. Nº cycles ARB.*
In the third cycle, the difference in orientation has declined from the previous cycle. Dislocations have turned back to move more easily because they are more oriented to the bonded boundaries and the energy to cross grain boundaries is lower. [17]

The third cycle deformation is fairly high, that is to say, there has been a large number of moving dislocations that have formed new grains. These new grain boundaries act as barriers to the movement of dislocations, since they create an atomic disorder within the grain boundary that will produce a discontinuity of the planes of a grain sliding to another.

From fourth cycle, it starts to appear the effect of recrystallization which makes that in the fifth cycle, beads will have slightly increased in size since subgrains will have been formed.

The sixth cycle shows a further reduction of thickness of grain since dislocations have been able to move with more freedom through the interior of the new formed subgrains.

Finally, the seventh cycle remains with a constant grain size since dislocations are anchored and haven’t been able to move freely.
3.1.3. – EFFECT OF DIFFERENT TEMPERATURES

Samples have been selected and cross sectioned from the 6 cycle ARB process at different temperatures. They have been observed with optical microscope:

![Images of samples at different temperatures](image)

*Fig. 23. Effect of different temperatures in the cooper with 6 cycles ARB, images in OM.*

Between the grains of CT and RT there doesn’t seem to be much difference, either in its thickness or appearance. They both have elongated their morphology due to deformation.

In the sample with CT, due to the low temperatures, dynamic recovery has been supressed by dislocations, creating grains with more distortion and high elongated morphology. The grains are finer and less defined, its grain boundaries are diffuse and possible entanglements can be seen.

In the processed specimen at RT the observable grain limits are diffuse and not uniform due to dynamic recovery, since the mixing of the two structures can be found. The grains remain elongated but less pronounced than the CT morphology.

In the sample that has been annealed, intermediates have reduced tensions caused by the ARB process, and grains can be seen best defined, with less strain and less elongated morphology.
These grains have lower density of dislocations and subgrains better defined. Angles of dislocations are not observed.

3.1.4. – SEM MICROGRAPH. SURFACE MORPHOLOGIES

With the objective to obtain visual samples of grain deformation caused by different cycles in the process and different temperatures, the same samples used in the OM analysis were monitored by means of microscopy electron scanning (SEM).

Thanks to the micrographs we can observe clearly in the six ARB cycle simple at high temperature that the grain has completely recrystallized, with a grain area size of 11.4 μm X 3.8 μm and more homogeneous than the grains treated at room and cryogenic temperatures.

It is evident the high plastic deformation and the high level of orientation of grains in all the observed samples.
3.2. - MECHANICAL PROPERTY. HARDNESS

The hardness is related to the ease in which plastic strain can occur. The mechanical strength can be increased by reducing the mobility of dislocations. Higher mechanical forces will be required to initiate plastic strain. Restriction, and the impediment of the dislocations movement translates to a harder and more resistant material. [18]
The strain hardening values are increasing progressively as the cycles advance in the ARB process. Mostly this fact is predominant in the first stage, which increases hardness drastically. In the third cycle, a maximum is reached, so we can say that from this point forward, hardening mechanisms have less effect, ceasing to be an important mechanism in the structure as not only fails to increase, but decreases it slightly, to keep constant. It suffers from a saturation. The saturation is known as inverse Hall-Petch.

The saturation behavior of hardening showing more cycles are commonly played on UFG materials created by severe plastic strain [19]. The faster increase of hardness at low grain levels can be attributed to the tightening as a result of the subgrain formation. This behaviour is described by Hall-Petch.

The loss of hardness stress in the sample of 6 cycles is associated with a decrease in the intensity of dislocations due to recrystallisation initiated in the fifth cycle. The hardness values obtained in the sample sixth are similar to the initial values of pure cooper.

Contrary in the sixth cycle with cryogenic temperature, the specimen doesn’t suffer this loss of hardness stress since the deformation at low temperatures suppresses dynamic recovery and, therefore, facilitates mechanisms of grain refinement.
3.3. – THERMAL PROPERTIES

With the data obtained experimentally, the following graphic of flow of thermal current vs $\Delta T/\Delta X$ is represented:

Graph 8. Thermal current vs $\Delta T/\Delta X$.

On the basis of the Law of Fourier $q = KA \frac{\Delta T}{\Delta x}$ the thermal conductivity value $K$ is obtained.

Then the thermal conductivity of the sample treated with 7 cycles of ARB is 414.22 W/(mK).

While the thermal conductivity of pure copper is 389.6 W/(mK), thermally speaking it isn’t a very significant increase for applications where a high conductivity is required. But structurally speaking refers to which there has been a change in the microstructure. This increase in thermal conductivity is due to the sample with 7 ARB cycles having small grain size oriented, that is to say, it has a more ordered crystalline structure so that movement of the conducting electrons and phonons lattice is favoured. With the following equation the thermal conductivity is expressed in terms of grains size.
The simplified theory of phonon thermal conductivity as a function of grain boundary can be expressed as:

\[ K_L = \left( \frac{2k_S}{3} \right) + \left[ \left( \frac{3d_S}{l} \right) \left( \frac{k_S}{2k_0} \right) \right]^{1/4} \] (18)

where \( K_L \) is the phonon thermal conductivity, \( k_S \) is the phonon thermal conductivity of crystals having negligible phonon boundary scattering in large grain size, \( k_0 \) is the phonon thermal conductivity in the absence of alloy scattering, and \( d_s \) is the grain size. [20]

This result demonstrates that the quality of the crystallization can affect the value of the thermal conductivity.
CONCLUDING REMARKS

With the interpretation and analysis of the experimental results, the following conclusions can be made:

1. The proposed Accumulative Roll Process (ARB) promotes grain refinement in Cu, both at RT, CT and HT.
2. The procedure produces large deformation in grains and generates strain from the third cycle. The grain size can be controlled by heat treatment.
3. To obtain the highest hardness in the material it’s required not to exceed three ARB cycles, since after then the reverse Hall-Petch is generated and the material loses strength. The fact it’s only necessary to perform three cycles reduces time and energy in production. As the added advantage that this process also has high productivity and viability of production for large material and wouldn’t be difficult to work with wide bands in coils. The process of bonding roll is widely adopted in the production of metal.
4. If the purpose is to get a material increase in thermal conductivity, although it is experimentally true, it has been favourable in actual practice that in thermal applications the increase is negligible.
REFERENCES


ADDITIONAL BIBLIOGRAFY


