Effect of Hot Rolling on the Thermomechanical Properties of a Superelastic Cu-Al-Be-Cr Alloy

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Shape memory alloys are generally produced by casting processes and are subsequently homogenized. However, to obtain semifinished products on an industrial scale, the ingots from the casting process must be hot worked. In particular, final bar and sheet products can be obtained by hot rolling process. During intense hot work, surface oxidation of the material and microstructural changes may cause modifications to its original thermomechanical properties. In this sense, the present work aimed to study the correlation of the superelastic behavior in a Cu-Al-Be-Cr alloy before and after subjecting it to the hot rolling thermomechanical process. Abnormal grain growth was observed for a hot rolled sample with 30% reduction in initial alloy thickness. This abnormal growth in relation to non-rolled alloy caused an increase in phase transformation temperatures, a reduction in residual strain, a reduction in induction stress and an increase in alloy superelasticity.

Keywords: *Abnormal grain growth, superelasticity, hot rolling.*

1. Introduction

Cu-Al-Be system polycrystalline shape memory alloys (SMAs), modified with inoculant addition, have good shape memory and superelasticity properties, with a lower processing cost than conventional NiTi system SMAs, making them, attractive for technological applications[1-5]. In addition, Cu-Al-Be SMAs can be modified for use at low temperatures. For example, the addition of only 0.1% beryllium (Be) by weight is required for a phase transformation temperature reduction of these alloys by approximately 100 °C[6]. They have stable β or austenitic phases and, when subjected to high temperatures, tend to stabilize the β phase at room temperature through rapid cooling or quenching[7].

Araya et al.[8] studied the properties of the superelastic Cu-11.8% Al- 0.5% Be SMA alloy at room temperature to evaluate its application in seismic resistance projects. For this work, wires previously heated for different periods of time were produced to evaluate the effect of grain size. According to the authors, the increase in grain size led to a gain in equivalent damping, a reduction in direct transformation and final stresses.

The hot working process of SMA alloy ingots previously obtained by casting and further homogenization may be detrimental to the compositional stability and thermo-mechanical properties of the alloys. This is due to the intense oxidation and microstructural changes due to the high temperatures required for the process, especially in hot rolling[9]. Several studies seek to remedy or minimize this inconvenience.

Narendranath et al.[10] studied the effects of different aging temperature treatments after hot rolling on martensitic phase transformation behavior and mechanical behavior of NiTi shape memory alloys. In their results they concluded that the stress induced martensite is more stable in relation to the thermal martensite. According to them, this is due to the change in the morphology of variant accommodation, from the self-accommodation of thermal martensite to the oriented state of tension-induced martensite. Superelasticity, associated with stress-induced martensitic transformation, increases with aging temperature gain (350 °C - 550 °C). This is associated with increased Ti_3Ni_4 and consequently network distortions.

Liu et al.[11] investigated the effects of lamination and heat treatment on the CuAlMn shape memory alloy microstructure and superelasticity. These alloys were processed via unidirectional solidification, followed by rolling and heat treatment. The alloy showed high workability and good superelasticity through hot rolling control. The alloy reached a reduction of 80% in the first pass, maintaining columnar grain structure. After two passes, followed by annealing at 800 °C, the superelastic deformation of the alloy reached 5.9%, although it recrystallized. This same alloy, cold rolled at room temperature, showed a reduction rate of around 50 ~ 70%, maintaining a two-phase columnar grain microstructure ($\beta_1 + \alpha$). Due to the precipitation of the α phase and the high annealing temperature, the grains may undergo abnormal growth, with diameters ranging from hundreds μm to more than 1 cm in diameter. The abnormal rate of grain growth is higher than that of the common polycrystalline alloy, and its superelastic deformation can reach about 7%.

In this sense, the main objective of this study is to determine the influence of hot rolling (HR) on the mechanical properties, on transformation temperatures and on superelasticity of a Cu-Al-Be-Cr alloy.

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2. Materials and Methods

The alloy with a nominal composition of Cu-11.8Al-0.6Be-0.3Cr (% by weight) was melted in an uncontrolled JUNG muffle furnace in a graphite crucible in an approximate amount of 700 g and cast in a rectangular section mold with 120 mm long x 22 mm wide and 40 mm high. The obtained ingot was homogenized at 850 °C for 12 hours to improve dissolution of the alloying elements.

After homogenization the ingot was sectioned into three samples: two with 10 mm and one with 15 mm thick by wire EDM. The sample measuring 15 mm thickness was then subjected to the hot rolling (HR) process to reduce its thickness to 1.5 mm (this sample was designated 100% HR). The with 10 mm of thickness sample was hot rolled to a final thickness of 9.7 mm (this sample was designated 30% HR). The other 10 mm thickness sample was non-hot rolled and was designated to 0% HR.

Figure 1 shows the specimens for mechanical tests, for the samples with and without laminate, that were made by wire erosion. After preparation, the specimens were heated at 850 ° C for 30 min and then quenched with water to room temperature to obtain the shape memory effect.

To reveal the phases between which the alloy is studied, the specimens were prepared for metallographic analysis using a Carl Zeiss microscope, model Axiotech 30 at room temperature for structural characterization, allowing the visualization of the size of the alloy grain in each sample, as well as the recording of their images through the software Shortcut to Analysis.

Superelasticity tests were performed on a Shimadzu 50 kN-EHF servomotor test machine equipped with a heating and cooling chamber. The test jaws are designed to accommodate the sample heads and thus avoid any possibility of slipping during their execution.

Transformation temperatures were investigated by Differential Scanning Calorimetry using a Shimadzu DSC-60. The samples were heated and cooled at a rate of 10 °C/min under a nitrogen atmosphere with constant flow with a rate of 50 ml/min.

3. Results and Discussion

The grain size in Cu-based shape memory alloys significantly influences their mechanical properties. It means that the alloy with larger grain size can achieve higher superelastic
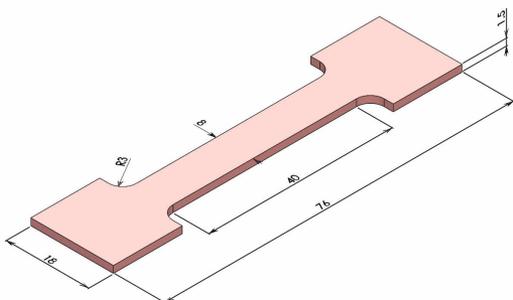

Figure 1 - Schematic drawing of the specimen via electroerosion. Dimensions in (mm).

stress[12-14]. Figure 2 shows the macrostructures of the rolled and non-rolled Cu-11.8Al-0.6Be-0.3Cr specimens. It was found that the non-hot rolled samples Figure 2a have very similar grains in relation to the samples that went through the hot rolling process Figure 2b, especially the sample of Figure 2c which showed an abnormal grain growth. This abnormal grain growth has been observed in SMAs undergoing cyclic heat treatments[15,16].

According to Kusama et al.[17] The mechanism responsible for abnormal grain growth is the migration of grain boundaries that consume the subgrains formed during cyclic heat treatments, and that the growth rate increases with increasing disorientation between the subgrains.

There is a degree of critical deformation for the dynamic recrystallization process to occur and when the degree of deformation exceeds the critical value, the recrystallized grains are gradually refined with increasing deformation[18]. This fact explains why the microstructure developed by the 100% HR sample has lower average grain sizes than the 30% HR sample. This indicates that for the 100% HR sample the dynamic recrystallization process occurred during hot rolling.

In Figure 3 it was observed that the predominant phase in all micrographs, at room temperature, is the austenite phase. It was also verified that the non-hot rolled sample, Figure 3a, presented an average grain size around 284.15 μm, while the samples 100% HR (Figure 3b) and 30% HR (Figure 3c) presented average grains around 403.4 μm and 734.17 μm, respectively. Thus, the lamination with a 30% reduction in the sample thickness provided a coarser microstructure with an average grain size of approximately 158% larger than the non-hot rolled samples.

Figure 4 shows the DSC curves for the 0% HR, 30% HR, and 100% HR samples. It was possible to identify the respective start and end points of the phase transformations: A_s and A_f in heating, M_s and M_f in the cooling of specimens with and without hot rolling. Peaks corresponding to the direct martensitic (austenite → martensite) and reverse (martensite → austenite) transformations were found to shift to the left as the grain size of the tested sample decreases.

Table 1 summarizes the results of the DSC analysis, where it was observed that after lamination the samples had increased martensitic transformation temperatures. Martensitic transformation temperatures are influenced by the grain size of the austenite phase[19,20]. Montecinos and Cuniberti[19] would evaluate the influence of austenite grain size on the Cu-Al-Be alloy M_s, and reported that M_s decreases with grain size reduction.

In order to quantify superelasticity, induction stress and residual strain cyclic tensile tests were performed. Figure 5 shows the load-unloading assay at different temperatures for 0% HR (Figure 5a), 100% HR (Figure 5b) and 30% HR (Figure 5c) samples. It is noteworthy that the maximum strain imposed for the 0% HR sample was only 3% due to the fragility presented by the alloy in this condition. Already the samples 100% HR and 30% HR were submitted to a strain amplitude of 5%.
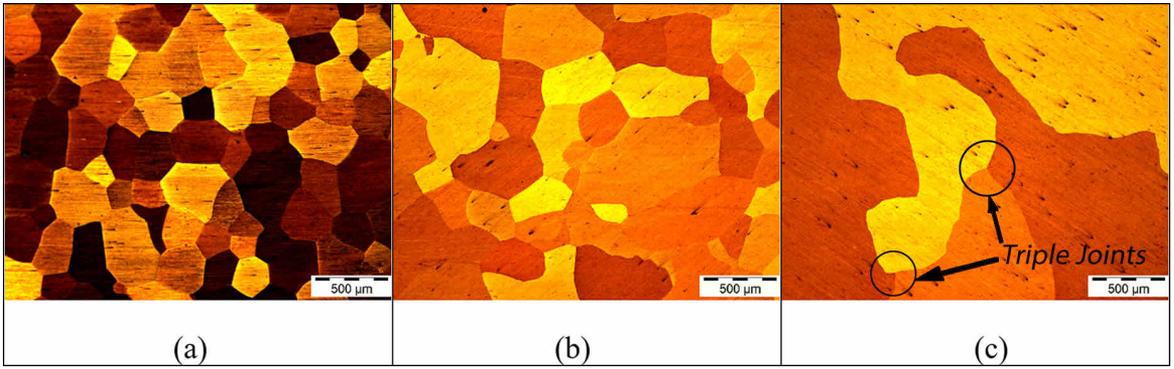
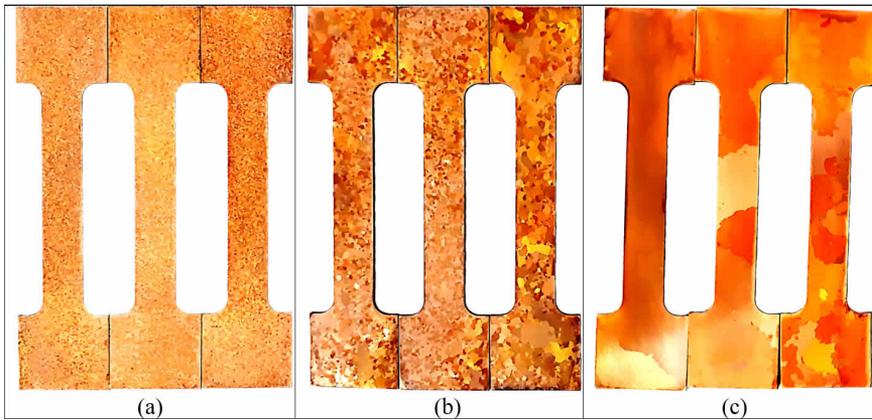**Figure 2 -** Macrography of the specimens (a) 0% HR (b) 100% HR and (c) 30% HR.

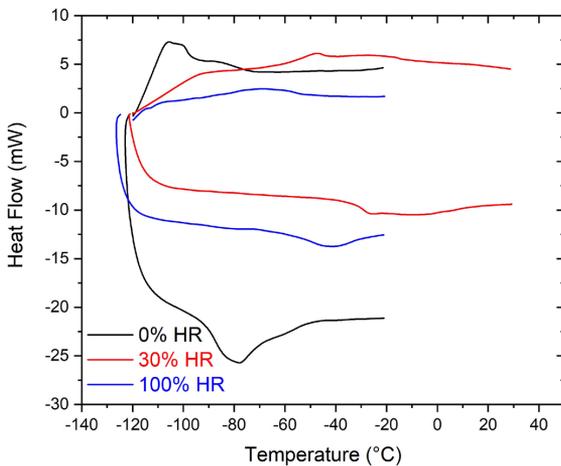**Figure 3 -** Micrograph of specimens (a) non-hot rolled (b) with 100% hot rolling and (c) with 30% hot rolling.

Figure 4 - DSC of Cu-Al-Be-Cr alloy alloy.

Table 1 - Cu-Al-Be-Cr alloy phase transformation temperatures.

Transformation Temperature (° C)			
	0% HR	**30% HR**	**100% HR**
A_s	-91.87	-34.37	-63.44
A_f	-61.87	10.62	-21.87
M_s	-88.44	-41.25	-49.06
M_f	-116.87	-62.5	-92.50

It was observed in Figure 5 that increasing the test temperature requires a higher stress to impose the same strain for both conditions (0% HR, 30% HR and 100% HR). For 100% HR condition (Figure 5b), for example, for a maximum deformation of 4% the corresponding stress was approximately 266.1 MPa at a temperature of 25 °C, while at a temperature of 100 °C a stress of around 301.4 MPa was required to impose the same deformation of 4%.

In addition, it was found that samples 0% HR and 100% HR, without abnormal grain growth, had a higher transformation slope ($A \rightarrow M$) than the sample with abnormal grain growth (30% HR). The higher this transformation slope, the greater the stress applied to the evolution of the martensitic transformation by stress. The lower transformation slope presented by the 30% HR sample can be explained by the smaller number of triple joints presented by the sample compared to the others. According to Liu et al.[21] the low energy straight grain boundary and the absence of triple grain boundary junctions in polycrystals with continuous columnar grain structure can significantly reduce the blockage of martensitic transformation at the grain boundaries.

Figure 6 shows the evolution of the induction stress of the martensitic transformation with temperature for the 0% HR samples that were subjected to a maximum deformation of 3% and for the 30% HR and 100% HR samples both subjected
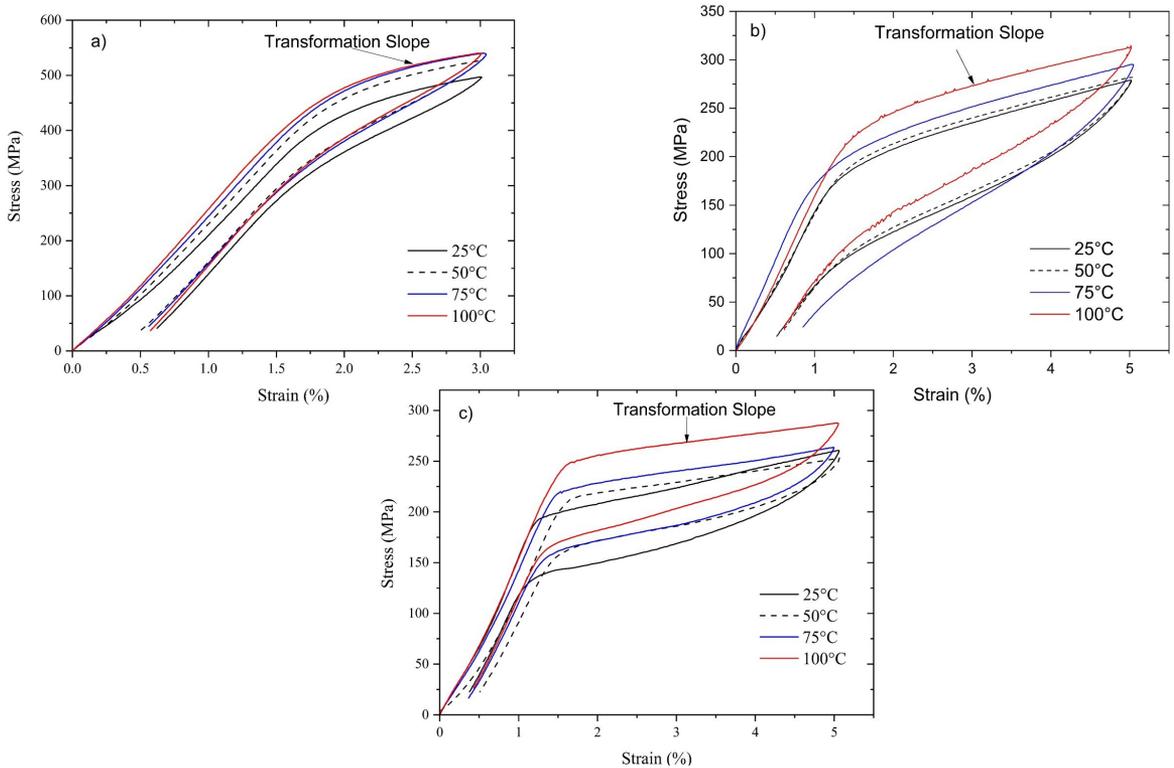
Figure 5 - Superelastic behavior at different Cu-Al-Be-Cr alloy temperatures: (a) 0% HR (a) 100% HR and (b) 30% HR.
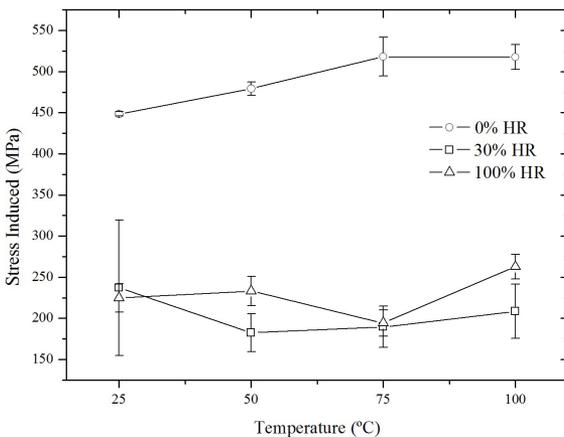

Figure 6 - Stress induced as a function of lamination degree and temperature.

to 5% deformations. The results show that the martensite induction stress for the 0% HR samples was higher than those reached for the 30% HR and 100% HR samples for the same test temperature. This difference in induction stress is associated with the grain size of the samples. Montecinos and Cuniberti[22] verified an induction stress dependence on the grain size for Cu-Al-Be alloy and reported an increase in induction stress with decreasing grain size.

Figure 7 shows the evolution of residual strain (Figure 7a) and superelasticity (Figure 7b) as a function of temperature

for the 30% HR and 100% HR samples, both subjected to a maximum deformation of 5%. As shown in Figure 7a, the residual strain initially increases for the 100% HR sample when the temperature increases from 25 °C to 75 °C and decreases with increasing temperature from 75 °C to 100 °C. For the 30% HR sample, there is a reduction in residual strain by increasing the test temperature from 25 °C to 50 °C, followed by a slight increase when the test temperature increases from 50 °C to 100 °C. The behavior of superelasticity as a function of temperature for the 30% HR and 100% HR samples was contrary to the residual strain, as observed in Figure 7b.

The effect of the degree of lamination on residual strain and superelasticity can also be observed in Figures 7a and 7b, respectively. At all test temperatures, the residual strain for the 30% HR sample was lower than the 100% HR sample, while the superelasticity was higher. This behavior is associated with the abnormal grain size presented by the 30% HR sample, as well as a smaller number of triple joints.

Yamagishi et al.[23] reported that the superelasticity of Mg-Sc alloy strongly depends on the grain size and that the maximum superelasticity at room temperature was obtained on the alloy with the largest grain size. Increasing grain size decreases the number of triple joints in the alloy microstructure, and consequently improves its superelasticity due to increased deformation compatibility at grain boundaries[24].
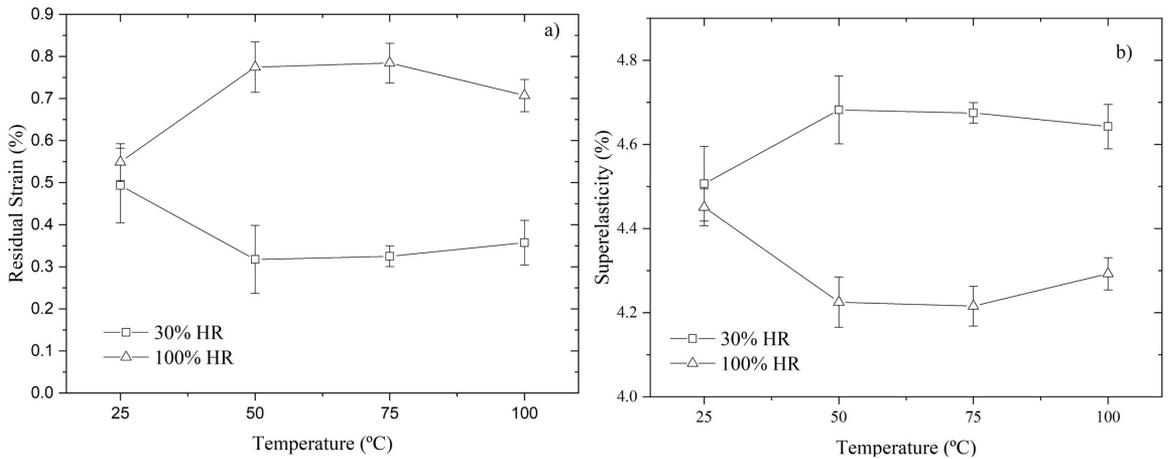
Figure 7 - a) Residual strain as a function of lamination degree and temperature. (b) Superelasticity as a function of lamination degree and temperature.

4. Conclusion

The influence of hot rolling on the thermomechanical and microstructural properties of a Cu-Al-Be-Cr alloy was studied using DSC, optical microscopy and cyclic tensile tests at different temperatures, therefore the following conclusions were obtained.

- Hot rolling caused a rise in phase transformation temperatures. This change in temperatures is associated with increased grain size of hot rolled samples.
- The 30% HR sample presented a lower transformation slope (induction of martensite by tension) than the other samples. This fact can be explained by the smaller number of triple joints presented by the sample.
- The induction stress of martensite at the same test temperature for the hot rolled samples (30% HR and 100% HR) was lower than the non-hot rolled sample (0% HR). This difference in induction stress is associated with the grain size of the samples.
- The 30% HR sample presented, for the same test temperature, a smaller residual strain than the one presented by the 100% HR sample, consequently a higher superelasticity. The difference in superelasticity and residual strain presented by the samples can be attributed to the abnormal grain growth of the 30% HR sample.

5. References

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