Original Article

Numerical simulation of cryogenic cyclic closed-die forging of Cu: hardness distribution, strain maps and microstructural stability

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\begin{abstract}
Cyclic closed-die forging (CCDF) appears to be an easy to operate deformation process, which imposes high levels of strain, even on difficult-to-deform materials. However, despite said potential advantages, the CCDF at cryogenic temperatures has not yet been investigated. Copper samples with dimensions of 10 mm × 10 mm × 20 mm were processed in up to six passes with interpass rotation, enabling the samples to return approximately to their initial dimensions after each pass. The intensity and homogeneity of plastic deformation was evaluated by mapping the Vickers hardness over the entire surface of the sample, and the resulting maps were compared with the strain and stress distribution estimated by FEM numerical simulation. The deformed microstructures were examined by optical and transmission electron microscopy. Cryogenic CCDF has proved to be effective in suppressing the recovery mechanisms of Cu samples, resulting in finer and more heterogeneous strains distribution than those deformed at room temperature. However, long-term observations by TEM have shown that these microstructures are inherently unstable, so that hardness decreases 50% after two years.

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\end{abstract}

1. Introduction

Conventional methods of plastic deformation, such as rolling, extrusion, forging, etc. are known to increase the mechanical strength of metals and alloys [1]. However, this improvement is usually accompanied by a decrease in deformation capacity [2–4]. Several unconventional processing routes have been developed in an attempt to achieve a balance between strength and ductility. These routes involve the production of ultrafine-grained (UFG) materials through various severe plastic deformation (SPD) techniques, including equal-channel

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angular pressing (ECAP), high-pressure torsion (HPT), accumulative roll-bonding (ARB) and cyclic closed-die forging (CCDF) [5–10].

In contrast to other SPD routes, CCDF imposes high strains on the billet during each pass, enabling a wide variety of materials to be processed, including difficult-to-deform alloys [11–14]. CCDF, which was developed by Gosh et al. [15], consists of the cyclic compression of a billet with dimensions of \( H \times W \) (height \( \times \) width) in the vertical direction inside a rectangular channel, normally resulting in a reduction of up to 50% in height. In the subsequent pass, the billet is rotated, and the sample maintains its initial dimensions. The equivalent deformation \( \varepsilon_{eq} \) achieved in this process can be estimated using Eq. (1), as follows:

\[
\varepsilon_{eq} = N \cdot \frac{2}{\sqrt{3}} \ln \left( \frac{H}{W} \right)
\]  

(1)

where \( N \) is the number of cycles, \( H \) is the height and \( W \) is the width of the specimen.

The literature describes the application of CCDF to difficult-to-deform materials such as magnesium and its alloys and titanium alloys [16–22]. Guo and coauthors [17] observed the simultaneous increase in strength and ductility of an AZ31 magnesium alloy processed by five CCDF passes at 350 °C. This behavior was attributed to grain refinement and to the deformation of the basal plane. Another investigation [19] concluded that CCDF is a very suitable route for the production of UFG composites, having a uniform distribution of nanoparticles in the matrix. However, although CCDF appears to be an easy to operate deformation process that can impose high levels of strain in metals and alloys, it is still little studied.

Many studies focus on the deformation behavior of copper and its alloys at low temperatures, but they employ rolling [23–27] and other SPD techniques, such as dynamic plastic deformation at cryogenic temperatures [28]. As a rule, mechanical behavior is strongly affected by deformation temperature, and in the case of copper, strength and ductility definitely improve when processed at cryogenic temperatures. However, the microstructural evolution and mechanical behavior of copper processed by CCDF at cryogenic temperatures has not yet been investigated.

This study evaluated the strength and ductility of a commercially pure copper processed by CCDF at room and cryogenic temperatures. In addition, the strain and stress distributions were estimated by FEM numerical simulations and compared to an experimental hardness map. Lastly, the long-term microstructural stability of the material processed at room and cryogenic temperatures was compared based on microstructural changes.

2. Materials and methods

Billets of commercially pure copper were machined into 10 mm \( \times \) 10 mm \( \times \) 20 mm samples for CCDF, annealed at 500 °C for 1h, and air-cooled. The schematic in Fig. 1 illustrates the CCDF die and the processing route; finishing and machining tolerances were specified to ensure a sliding die-plunger adjustment. The sample was placed at the center of the lower die, which has a cross section of 10 mm \( \times \) 20 mm and 10 mm height. A plunger with identical cross section forged the sample down, from the height of 20 mm to a thickness of 10 mm, with a constant anvil speed equal to 1.5 mm/min, which correspond to an initial strain rate equal to \( 1.25 \times 10^{-3} \) s^{-1}. After each pass, the sample was rotated simultaneously 90° around the z- and y-axes, so that after three passes it returned to its original orientation. Overall, the deformation process consisted of six passes (6×) performed at room temperature (RT) and four passes (4×) at cryogenic temperature (CT), each pass corresponding to an average equivalent strain equal to 0.8. The set die-plunger and sample were kept fully immersed in liquid nitrogen as a cooling medium during the CT passes.

The microstructures of samples in the annealed condition and after CCDF processing were examined on the XY plane. In preparation for optical microscopy (OM), samples were conventionally polished and etched with 90 ml H2O + 30 ml HNO3. For transmission electron microscopy (TEM), specimens were prepared from the compressed materials using a twin-jet electropolisher containing a solution of 33% HNO3 and 67% methanol at −30 °C, operating at 5 V. TEM observations were carried out in a Philips TECNAI G2 F20 TEM microscope operating at 200 kV, with the foil surface kept parallel to the XY plane. To evaluate the uniformity of deformation, Vickers microhardness tests were performed, which involved two-dimensional (2D) hardness mapping of about 200 measured points in 1 mm steps on the XY plane under a load of 300 gf.

RT and CT deformed samples were placed in long-term storage (2 years) in air (at room temperature). Microstructural changes in these samples were evaluated from microhardness measurements and examined by OM and TEM, as described above.

The CCDF process was numerically simulated using the finite element software package DEFORM 3D™, which deals with large strain problems inherent to most industrial processes. The program employed an implicit algorithm to solve residual equilibrium equations by a direct iteration method, considering a thermo-viscoplastic model, with a heat transfer coefficient equal to 5 N/s/mm²/°C and constant elastic properties. The Zerilli-Armstrong flow stress model [29] was applied, according to the general expression shown in Eq. (2).
\[ \sigma_f(\varepsilon_p, \dot{\varepsilon}_p, T) = \sigma_a + B \exp(-\beta(\dot{\varepsilon}_p)T) + B_0 \sqrt{\varepsilon_p} \exp(-\alpha(\dot{\varepsilon}_p)T) \]  

where \( \varepsilon_p \) represents the equivalent plastic strain, \( \dot{\varepsilon}_p \) the plastic strain rate, \( T \) the temperature, and \( B \) and \( B_0 \) are material constants. \( \alpha \) and \( \beta \) are the free temperature part of the model, given by the following expressions:

\[ \sigma_a = \sigma_0 + \frac{k_0}{\sqrt{l}} + K \rho \]  
\[ \alpha = \alpha_0 - \alpha_1 \ln(\varepsilon_p) \]  
\[ \beta = \beta_0 - \beta_1 \ln(\dot{\varepsilon}_p) \]

where \( \sigma_a \) is the contribution of solutes and initial dislocation density, \( k_0 \) represents the intensity of microstructural stress, \( l \) the average grain diameter, and \( K \) is zero for a FCC material. \( \alpha_0, \alpha_1, \beta_0, \beta_1 \) are material parameters dependent on the crystal structure. The numerical values of these constants, taken from Zerilli and Armstrong [29] are shown in the Appendix, while the effective values of \( \varepsilon_p \) and \( \dot{\varepsilon}_p \) are continuously calculated by the software. The contact forces were determined by the penalty method and a friction coefficient equal to 0.12 was adopted for all the numerical simulations. The tools were considered as rigid surfaces and the billet was discretized by fully integration elements and automatic remeshing for high strain levels.

3. Results and discussion
3.1. Microstructural evolution

Fig. 2 shows OM micrographs of microstructures of commercially pure copper after CCDF performed at room (1x-RT, 6x-RT) and cryogenic (1x-CT, 4x-CT) temperatures. These microstructures were observed immediately after CCDF processing and are hereafter referred to as the “as-deformed” condition.

The microstructures suggest that CCDF processing produced very intense grain refinement, regardless of the temperature. The annealed sample (Fig. 2a) shows equiaxed grains with an average size of 65 \( \mu \)m \pm 3 and some annealing twins inside the grains. After 1x at RT (Fig. 2b), the grains were smaller (42 \( \mu \)m \pm 5) and the shape followed the direction of compression. Several deformation twins were visible inside the grains and the microstructure was heterogeneous, with some undeformed regions around heavily deformed regions. After 6x-RT (\( \varepsilon_{eq} = 4.8 \)), the microstructure was more refined and more homogeneous, as indicated in Fig. 2c. The samples processed at CT showed a similar behavior (Fig. 2d and e), but the average grain size was smaller than in the samples processed at RT. At very low temperatures, the partial suppression of dynamic recovery may be attributed to increased dislocation accumulation, which leads to intense grain refinement. According to Wang and Ma [30], cryogenic deformation can be a strategy to produce refined microstructures with high dislocation density, increasing the material’s strength and ductility. The microstructural features will be discussed in greater detail later.

3.2. Hardness distribution after CCDF deformation

To gain a better understanding of the strain distribution after CCDF, 2D microhardness mapping was done, as shown in Fig. 3. Vickers microhardness was measured in the central region of an XY plane (see Fig. 1), avoiding the irregular ends of the samples.

In both RT and CT processing, hardness was distributed very heterogeneously after the first pass (Fig. 3a and d, respectively), but became more homogeneous at higher equivalent strains (Fig. 3c and e). This homogeneity may be attributed to the interpass rotation of the sample, which allows for the activation of different slip planes during deformation. Another important feature observed here is the increase in hardness promoted by the higher dislocation density in the CT-deformed samples (Fig. 3d and e).

The frequency distribution of the microhardness measurements shown in Fig. 4 offers a general overview of how this property varies along the XY plane of the processed samples. The 1x-RT sample depicted in Fig. 4a shows a quasi-Gaussian distribution, with most of the values lying between 110 HV and 130 HV and a total range of about 40 HV. In contrast, the 4x-RT sample shows a narrower hardness distribution, with the highest frequencies between 120 HV and 130 HV. As for the samples processed at CT, Fig. 4b clearly shows a larger range distribution, i.e., hardness is distributed more heterogeneously throughout the specimen in comparison to the same equivalent strain at RT. In addition, the 4x-CT sample shows higher hardness values than samples in any other analyzed condition. In short, two general tendencies can be inferred from Fig. 4: hardness values are higher after CCDF at CT, while the homogeneity is higher after processing at RT.

3.3. FEM simulation

The CCDF process was simulated numerically using the finite element software package DEFORM 3D\textsuperscript{TM}. The experimental inputs were the initial strain rate equal to \( 1.25 \times 10^{-3} \text{s}^{-1} \), and temperatures of 77 K (CT) and 298 K (RT). The constitutive material model for copper, as well as the input data employed in the software, were taken from Zerilli and Armstrong [29], since some results obtained using this model have been presented and evaluated by Banerjee [31], for a similar range of strain, strain rate and temperature. In our study, we compared simulated stress and strain distribution after CCDF with the experimental hardness mapping.

Fig. 5 compares the average values of the hardness maps after each CCDF pass with the average values of effective stress, estimated by numerical simulation at the correspondent level of equivalent strain. Note that, after a given number of passes, the equivalent strain for processing at RT is slightly higher than at CT. Clearly, CT processing leads to higher levels of hardness and flow stress than RT. Another evidence is that for RT processing hardness and flow stress tend to saturate after the third-fourth pass, which does not seem to be happening at CT.

After four passes (4x) at CT the hardness reaches \( \sim 147 \) HV, i.e., almost \( \sim 230\% \) higher than in the annealed condition (\( \sim 45 \) HV), while at RT a maximum of \( \sim 130 \) HV is reached near the fourth pass and does not increase with the subsequent
Fig. 2 – Microstructures of commercially pure Cu: (a) annealed; (b) 1x-RT (as-deformed); (c) 6x-RT (as-deformed); (d) 1x-CT (as-deformed); (e) 4x-CT (as-deformed).
Fig. 3 – Experimental Vickers microhardness maps of commercially pure Cu after CCDF processing (as-deformed): (a) 1x-RT; (b) 4x-RT; (c) 6x-RT; (d) 1x-CT; (e) 4x-CT.

Fig. 4 – Frequency distribution of hardness measured on the XY plane of Cu samples after CCDF at RT (a) and CT (b).
that is consistent with the present work. It is well known that hardness may be related to the energy stored during plastic deformation, such as compression in CCDF, and that an increase in hardness reflects an increase in stored dislocation density. Processing temperature is a fundamental parameter that determines the tendency for dynamic recovery \[22,32,33\]. At very low temperatures, the thermally activated mechanisms of dislocation climb and cross slip are partially suppressed; dislocation mobility is reduced and the annihilation of dislocations at grain boundaries decreases. This combination leads to an increase in dislocation density along with an increase in equivalent strain in CCDF at CT.

**Fig. 6** shows numerical maps of the effective strains along the surface of the specimens. A comparison with the hardness maps in **Fig. 3** indicates that the homogeneity of strain follows a similar qualitative evolution, i.e., the higher the number of passes the more homogeneous the deformation distribution. In **Fig. 7**, the average values of strain obtained by numerical simulation are compared with the theoretical values determined by Eq. (1). In addition to good agreement between numerical and theoretical values, the graph in this figure shows that the effect of processing temperature on strain level and distribution is negligible.
On the other hand, Fig. 8 shows that the corresponding levels of stress increased significantly when processing was performed at CT, which confirms that dynamic recovery mechanisms were hindered by the low temperature. It has been demonstrated [34] that low stacking-fault energy and low deformation temperatures favor the accumulation of both dislocations and twins, which may lead to increased strength, together with a high capacity for plastic deformation.

The work hardening behavior of some FCC alloys at CT was analyzed in a previous study [33], using the so-called Kocks–Mecking (K-M) plots, which are derived from experimental true stress–true strain curves in uniaxial tensile tests. The purpose of said tensile tests is to evaluate the deformation behavior of the material at RT and CT. A simultaneous increase in strength and elongation was observed (Fig. 9a) in annealed samples of commercially pure Cu when tested at CT in comparison to those tested at RT. The corresponding K-M plots (Fig. 9b) relate the work hardening rate $\theta = \varepsilon_0/d\varepsilon$ to the instantaneous stress $(\sigma - \sigma_0)$, where the slope $[-d\varepsilon/d(\sigma - \sigma_0)]$ of the linear segment is proportional to the dynamic recovery rate [34]. In the present case, this rate is clearly reduced by the low temperature, as shown by the slopes of the dashed lines in Fig. 9, together with maps of the simulated stress distribution and the experimental hardness distribution after CCDF at CT.

3.4. Microstructural stability – differences between RT and CT processing

Fig. 10 shows the measured hardness of samples in the as-deformed condition (i.e., immediately after CCDF) and after 2
years of storage in air at room temperature. The hardness of Cu in the annealed state is also shown.

The microstructures of pure copper developed during RT and CT processing showed thermal instabilities, i.e., static recovery and recrystallization may occur at room temperature after long-term storage, as indicated in Fig. 10. These differences can be attributed to self-annealing after severe deformation at cryogenic temperatures. Self-annealing is abnormal softening and grain coarsening after severe deformation, mainly cryogenic deformation, and this effect has been observed in different metals and alloys [35–39]. As can be seen in Fig. 10, the self-annealing effect of 6 × RT samples is almost negligible even after 2 years of storage, since the vacancy and dislocation densities are smaller during CCDF at RT. The 6 × RT samples show a smaller difference in measured hardness than the 4 × CT samples, i.e., after long-term storage, the hardness of the latter decreased by about 50%. According to Edalati and co-authors [39], samples of pure copper subjected to HPT at 100 K exhibit self-annealing within a few hours after processing, according to hardness measurements. In this study, this effect has been linked to a high density of lattice defects at large strains in Cu samples processed at cryogenic temperature, which is the driving force for the development of self-annealing.

Fig. 10 shows TEM micrographs of copper subjected to CCDF processing at room and cryogenic temperatures. The 6 × RT as-deformed sample (Fig. 11a) shows some recovered grains surrounded by regions with high dislocation density, but no twinning. This microstructure contains many deformation bands of about 200 nm in width. After two years of storage, the Cu sample subjected to 6 × RT (Fig. 11b) showed slight differences and a fairly homogeneous microstructure. However, several deformation bands were visible in the 4 × CT samples (Fig. 11c). In general, the dislocation density was high, but some grains contained a few defects. In addition, some grains showed deformation twinning. After long-term storage at room temperature, this sample underwent self-annealing as indicated in Fig. 11d. This microstructure is typical of partially annealed copper, with large grains with low dislocation density and some annealing twins, and some regions with high dislocation density (dark regions). This recovery behavior after severe cryogenic deformation has been observed in other studies [37–39], suggesting a limited microstructural stability, especially for low stacking-fault energy materials. It must be recalled that the possibility of producing fine dispersion of precipitates acting as substructure pinning sites, can limit self-annealing processes, which are the main causes of microstructural instability after cryo-deformation, for example, Zr additions in copper [40–42].

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Fig. 9 – (a) Tensile tests performed at RT and CT on annealed commercially pure Cu; (b) Kocks–Mecking plots derived from the true stress–true strain tensile curves [33].

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Fig. 10 – Copper samples processed by CCDF at RT and CT: Vickers hardness measured in the as-deformed condition and after long-term storage at room temperature.
4. Conclusions

The microstructural evolution and mechanical properties of commercially pure copper processed by CCMF were systematically investigated in terms of the effect of temperature, grain refinement, hardness mapping and FEM simulations. Our main findings are summarized below.

(1) Strain and hardness homogeneity increased as a function of the number of CCDF passes. Also, hardness increases in response to high levels of deformation at cryogenic temperatures.

(2) Cryogenic CCDF of copper produces more refined microstructures and more heterogenous strain distributions in commercially pure copper, compared to RT processing;

(3) In annealed samples, ultimate tensile strength and elongation to failure increased simultaneously when tested at CT, in comparison to tests performed at RT; the corresponding Kocks–Mecking plots shows that dynamic recovery rate is clearly reduced by the low temperature.

(4) The long-term storage showed that CCDF of commercially pure copper at CT promoted intense recovery and recrystallization of the microstructure, so that hardness measurements indicated a 50% decrease in hardness after two years.

Conflicts of interest

The authors declare no conflicts of interest.

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Appendix A.

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<th>- Input data applied to DEFORM 3D™ for numerical simulation of CCDF processing.</th>
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<td>$\sigma_0$ (MPa)</td>
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REFERENCES


