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Study of Cryogenic Rolling of FCC Metals with Different Stacking Fault Energies

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Aluminum, copper and silver samples, all of them face-centered cubic (FCC) metals, were rolled at room and cryogenic temperatures until equivalent strains (ε) were between 3.23 and 4.13. The cryogenic temperature (CT) and room temperature (RT) rolled samples were evaluated by hardness tests and X-ray diffraction (XRD), which indicate influence of stacking fault energy (SFE) on process. Lower SFE metals tend to exhibit dislocation densities significantly increased and as consequence, hardness too. It was also noted that after sometime exposed to RT, the materials rolled at CT present hardness decrease.

Keywords: *Cryogenic Deformation, Cryogenic Rolling, Stacking Fault Energy, Recovery.*

1. Introduction

Deformation at cryogenic temperatures (CT), or at temperatures close to liquid nitrogen (LN_2) temperature partly suppress dynamic recovery during deformation, allowing a higher density of defects to be produced in the material compared in a deformation at room temperature (RT). Annealing uses part of the stored energy in the material for the formation of new grains with low density of defects. If done after cryogenic deformation, the grains could achieve submicrometric or even nanometric scales.[1-4]

Stacking fault energy (SFE) has marked influence in the work hardening process, as well as in the evolution of deformation substructures. In addition, the deformation substructures affect the microstructural changes during subsequent annealing of work hardened metals. On one hand, planar slip is the predominant mode of deformation in metals at room temperature, leading to the development of dislocation cells arrangement for high SFE materials (aluminum), as well as planar arrays of dislocations for materials with lower SFE (like copper and silver). On the other hand, as stated by Gapontseva et al.[5], deformations at low temperatures, even for intermediate and high SFE materials, results in higher dislocation density, promote twinning deformation and as consequence, the materials present higher strength combined with good ductility and tend to have smaller grain size.[4-8]

In the present work, commercially pure samples of aluminum (Al), copper (Cu) and silver (Ag) were rolled at room and cryogenic temperatures until approximately 99% of overall thickness reduction, or equivalent strains (ε) between 3.23 and 4.13, in order to detect structural changes. These materials were chosen because of their well-known SFE levels: aluminum (166 $mJ.m^{-2}$, or high SFE), copper (78 $mJ.m^{-2}$, or medium SFE) and silver (22 $mJ.m^{-2}$, or low SFE).[4]

2. Experimental

Al Alfa Aesar® (99.95% purity), Cu Alfa Aesar® (99.9% purity) and Ag Alfa Aesar® (99.9% purity) rods of approximate length of 3.5 mm and ϕ 6.35 mm were rolled for initial thickness adjustment ≈ 5 mm, from round sections to flat bars. The materials were then recrystallized at 345 ºC (Al) and 400 ºC (Cu and Ag) for 15 min in argon atmosphere (heating rate of 30 $ºC.min^{-1}$ and cooling at furnace).

The samples were rolled at room temperature (RT) and at liquid nitrogen (LN_2) temperature in a laboratory rolling mill with increasing thickness reduction per pass (initial reduction ~10%) until the total reduction was ~99% overall the thickness (final thickness between 0.05 and 0.10 mm) for all the materials.

During the cryogenic rolling, the samples were cooled by dipping into the LN_2 prior and thereupon immediately of each pass. The temperature was monitored during rolling using a thermocouple and datalogger system until the thermocouples rupture. For each material, two samples were rolled with the thermocouple attached in the middle of the sample, one for the cryogenic rolling and one for the room temperature rolling.

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Although the recovery seems to be the most reasonable process at room temperature, some authors such as Dong-Hyun et al.[9] and Konkova et al.[10] pointed out the recrystallization at RT with these metals. According to that, after completion of the rolling process, specimens were stored at LN_2 until testing, to avoid extensive recovery or even recrystallization at RT, and removed from LN_2 right before each test and the tests were performed immediately after the rollings.

The microhardness (HV) of the sheets were measured at room temperature on a Shimadzu HMV-2T tester at a load of 98.07 mN (for Al and Ag) and 245.2 mN (for Cu) for 12s, and all samples were selected at least 6 points to test.

X-ray diffractions (XRD) were performed with a MRD Panalytical X-ray diffractometer™, at LNNANO (Brazilian Nanotechnology National Laboratory) at CNPEM (National Center for Research in Energy and Materials), Campinas, Brazil. X-ray diffraction measurements were performed on samples of copper, silver and aluminum at room and cryogenic temperatures, using CuKα radiation (λ = 1.542 x 10^{-10} m) and scanning over a 2θ range of 35° to 100°. The low temperatures were reached using LN_2. The diffraction data collected was analyzed by the Panalytical software High- ScorePlus™, using a pseudo-Voigt function and the background was subtracted. In the case of aluminum samples, only one diffraction peak was observed due to the high degree of texture and thus, there is not enough information for the analysis. XRD was performed on samples of aluminum, copper and silver rolled at room temperature and also at cryogenic temperatures using a developed apparatus for these purpose.

Microstrain and crystallite size were calculated from method shown in Gong et al.[11] Dislocation density was calculated as follows:[12]

$$\rho = \frac{(2\sqrt{3})(\varepsilon^2)^{1/2}}{d_{XRD}\,b}$$

Where d is the grain (crystallite) size, b is the absolute value of the Burgers vector, and $(\varepsilon^2)^{1/2}$ is microstrain. For FCC metals, $b = ((\sqrt{2})/2)a$, and a is the lattice parameter.

3. Results and Discussion

Temperature was controlled during rolling to assure that it was low enough to keep the major defects quantity, due to minimize recovery, annihilation and rearrangement of dislocations. Figure 1 shows the temperature variation for rolling process at room temperature for Al, Cu and Ag. Figure 2 shows the temperature variation for rolling process at cryogenic temperatures. It can be seen that the temperature did not exceed 26.5 ºC for RT rollings. It was expected that CT rollings presented lower temperatures than detected when compared to RT rollings, but according to Huang, et al.[13] at very low temperatures (approaching absolute zero), heat capacity is quite small and the energy converted into heat in deformation leads to a very large temperature rise. If the heat can be rapidly dissipated by conduction along the length of the specimen or through the surface to the surrounding cooling medium, the temperature enlargement is slighter. It can also be observed that the harder it is to deform the material, more the temperature rises during its rolling. On the other hand, the majority of steps of rolling CT process could keep the samples on lower than 0 °C and the temperature rise lasted less than 0.5s (which is a very short time interval).

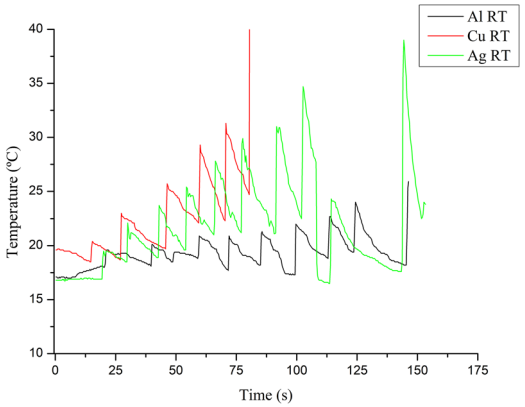

Figure 1. Temperature variation during RT rolling processes for Al, Cu and Ag.

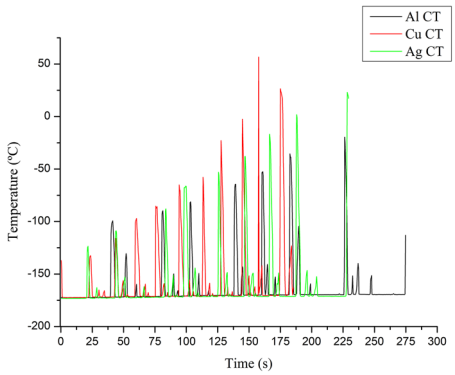

Figure 2. Temperature variation during CT rolling processes for Al, Cu and Ag.

At each rolling step, the energy stored in the materials in form of dislocations increases and more energy is converted into heat, as can be seen in Figures 1 and 2 that show that the temperature rises a little more each pass.

Figure 3 shows a crescent tendency of the force registered by the rolling mill, because of the increasing density of defects along the rolling process. It can also be seen that all materials rolled at CT present higher force values. It could be explained by the change in deformation mechanism from dislocation glide to twining and by the combination of higher dislocation density and a relatively large homogeneous deformation-length scale in the microstructure.[13,14] According

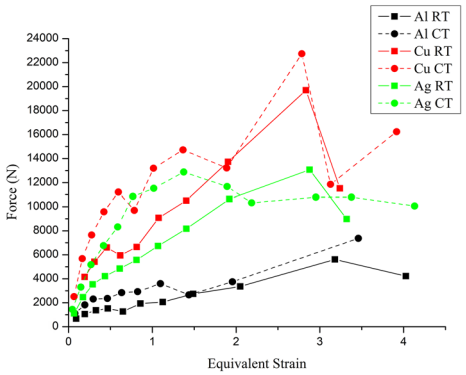

Figure 3. Force variation over rolling steps.

to that, hardness tests were executed for the materials rolled at RT, in the longitudinal section.

Figure 4 shows the hardness of RT rolled materials in function of equivalent strain. It can be observed that the hardness tends to increase slightly for all the materials, but after certain roll reduction percentage, the aluminum's hardness tends to drop to a constant value and has no significant change with deformation, as was also observed by Edalati and Horita[15] and Kawasaki, Ahn and Langdon[16]. As stated by Xu, Horita and Langdon[17], it happens due to cross slip and easy dynamic recovery because of aluminum's high SFE, but as stated by Khatibi et al.[18], it happens because of the relatively high homologous temperature, which in this case is 0.32.

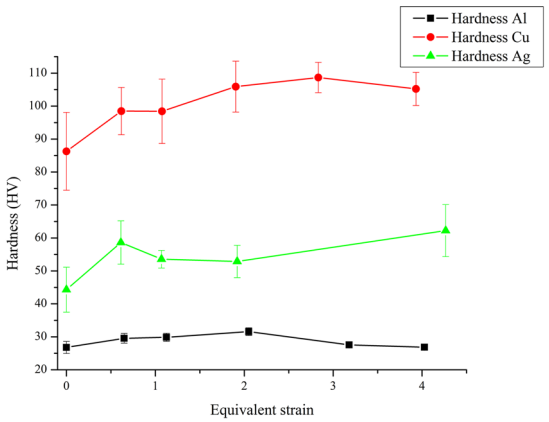

Figure 4. Hardness in function equivalent strain for RT rolled materials.

Table 1 compares the hardness of the samples after rolling to ~99% of total deformation at RT and CT. Data shows that cryogenic rolling is more efficient in keeping the defects, since their hardness are larger if compared to the RT rolled materials. It can be observed that hardness increase percentage is directly proportional to SFE in this case, that is not what was expected. Maybe it is due to dislocation rearrangement and annihilation during RT rolling after certain

Table 1. Hardness of materials rolled up to approx. 99%.

Material	Hardness (HV)	Relative hardness increase (%)
Al CT	35.8	34.6
Al RT	26.6	
Cu CT	137.5	30.7
Cu RT	105.2	
Ag CT	71.9	15.6
Ag RT	62.2	

deformation degree, leading to softening of the materials and causing larger difference between hardness and, therefore, larger values of hardening increase percentage for materials with higher SFE.

In samples deformed at CT, aging at RT decreased hardness, as can be seen in Figure 5. The percentage of hardness decrease is higher for Ag (45.8%), which has low SFE and lower for Al (26%), which has high SFE and that could be associated with static recovery. Copper presented a hardness decay of 30.2%, that is between Al and Ag. Al has high SFE, so even at low temperatures, the dynamic recovery suppression is not as effective as for low SFE as Ag and the dislocation density tends to be smaller inside materials with higher SFE. Ag and Cu have greater defects densities (especially vacancies) and the heating from CT to RT can provide enough energy to enable grain boundary movement, which could explain the hardness decay, that is associated with the static recovery.[14,19-21]

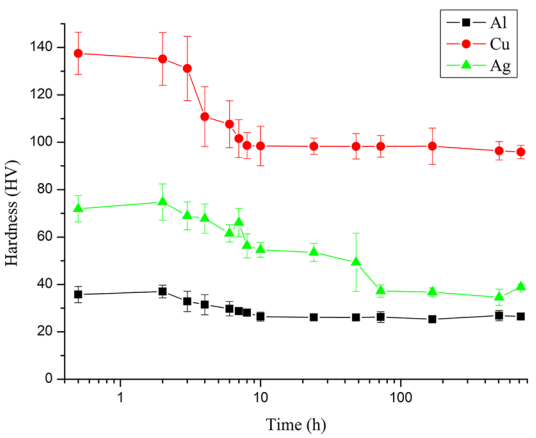

Figure 5. Hardness decay after CT rolled materials removal from LN_2.

Figures 6, 7 and 8 show the diffractograms obtained from the aluminum, copper and silver samples, respectively, deformed at room and cryogenic temperatures.

Tables 2 and 3 show the X-ray diffraction results of deformed copper and silver samples at room and cryogenic temperatures. The values of interplanar distance and lattice parameter were calculated from the obtained diffraction peaks. The contraction values for the interplanar distance

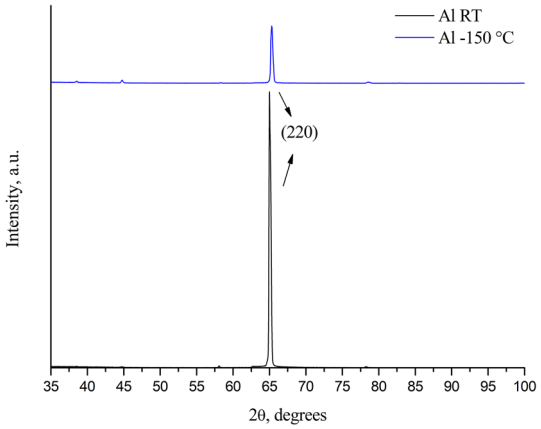

Figure 6. XRD aluminum rolled at RT and CT.

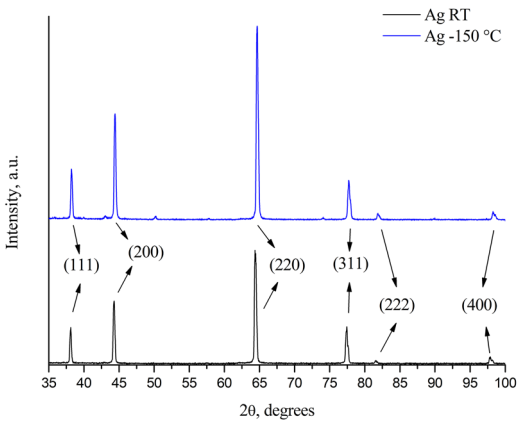

Figure 8. XRD silver rolled at RT and CT.

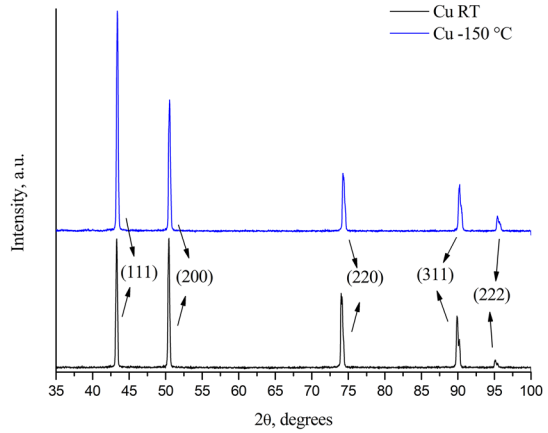

Figure 7. XRD copper rolled at RT and CT.

and the volume of the unit cell were estimated for the identified peaks. The volumetric contraction in silver was higher than in copper.

Table 4 displays the values of cumulative roll reduction, microstrain, crystallite size and dislocation density for rolled samples at RT and CT. It is shown that the crystallite size parameter does not change for this high deformation degree, so a steady state grain size is reached for copper and silver.

Also in Table 4, it can be observed a significant increase in dislocation density and microstrain, at cryogenic temperatures compared to room temperature, especially for silver, of lower SFE. This phenomenon can be explained by lower dynamic recovering level, for lower SFE materials.

Table 2. Calculated volumetric contraction based on lattice parameter for X-ray diffraction peaks of copper.

λ =1.541874 Å								
h k l	Experimental 2θ at 25ºC	d (Å)	a (Å)	Experimental 2θ at -150ºC	d (Å)	a (Å)	d contraction (%)	volume contraction (%)
1 1 1	43.268	2.091	3.622	43.352	2.087	3.615	0.18%	0.55%
2 0 0	50.401	1.811	3.621	50.491	1.808	3.615	0.17%	0.50%
2 2 0	74.069	1.280	3.620	74.293	1.277	3.611	0.26%	0.78%
3 1 1	89.908	1.091	3.619	90.190	1.088	3.610	0.25%	0.74%
2 2 2	95.120	1.045	3.619	95.442	1.042	3.610	0.26%	0.77%

Table 3. Calculated volumetric contraction based on lattice parameter for X-ray diffraction peaks of silver.

λ=1.541874 Å								
h k l	Experimental 2θ at 25ºC	d (Å)	a (Å)	Experimental 2θ at -150ºC	d (Å)	a (Å)	d contraction (%)	volume contraction (%)
1 1 1	38.085	2.363	4.093	38.227	2.354	4.078	0.36%	1.08%
2 0 0	44.262	2.046	4.093	44.404	2.040	4.080	0.30%	0.92%
2 2 0	64.391	1.447	4.093	64.647	1.442	4.078	0.35%	1.07%
3 1 1	77.382	1.233	4.090	77.697	1.229	4.076	0.34%	1.03%
2 2 2	81.546	1.180	4.089	81.867	1.177	4.076	0.32%	0.98%
4 0 0	97.855	1.023	4.091	98.290	1.019	4.077	0.33%	1.00%

Table 4. Parameters obtained from X-ray diffraction of samples of copper and silver rolled at room and cryogenic temperatures.

	Cumulative roll reduction	Microstrain <ε>	Cristallite size (nm)	Dislocation Density – ρ (m^{-2})
Copper RT	0.98	$6.01x10^{-4}$	6.8	$1.20x10^{15}$
Copper CT	0.98	$1.88x10^{-3}$	7.2	$3.57x10^{15}$
Silver RT	0.99	$8.08x10^{-5}$	6.0	$1.61\ x10^{14}$
Silver CT	0.98	$1.74x10^{-4}$	5.9	$8.88\ x10^{15}$

4. Conclusions

Force and hardness data indicate that CT rollings are efficient in partial suppression of dynamic recovery. Nevertheless, the microstructure is not stable and after aging at RT, hardness decrease is observed. This provides an indirect evidence of microstructural changes in metals, which could be related to the static recovery/recrystallization.

Sometime after the removal of the samples from LN_2, they presented hardness decay, larger for silver, which has the lowest SFE and lower for aluminum, that has the highest SFE. We used the values of SFE reported by Humphreys and Hatherly.[4] According with them, aluminum, copper and silver have different values of SFE.

It can be observed that silver presented substantial volumetric contraction (0.92 to 1.08%) and, in comparison, copper displays less volumetric contraction (0.5 to 0.78%).

It is shown with the parameters calculated from the X-ray diffraction that for the degree of reduction of the rolled samples (~ 0.99), it is reached a steady state grain size. Also, a significant increase in dislocation density, especially for silver, of lower stacking fault energy with processing at cryogenic temperature due to dynamic recovery suppression.

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