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Optimization of a technological process for heat treatment of Cu99.9 with different degrees of plastic deformation

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Abstract. ***Different degrees of plastic deformation of Cu99.9 have been achieved by using different technological forming processes. Recrystallization annealing was carried out at different time-temperature regimes. The influence of the degree of plastic deformation and the recrystallization annealing conditions on the structure of the studied material (Cu99.9) is discussed. The technological process of heat treatment is optimized.***

Keywords: recrystallization annealing, plastic deformation, microstructure, Cu99.9

I. INTRODUCTION

Recrystallization and grain growth are of crucial importance in regulating the mechanical properties and performance characteristics of thermomechanically treated metals and alloys [1, 2]. This is especially true for strong plastic deformation (SPD)-deposited ultrafine-grained (UFG) and nano-grained (NG) structures, which typically have degraded thermal stability and low ductility due to the high stored excess energy and dense dislocation arrangement resulting from plastic deformation (PD) [3,4]. More well-known methods for the intensive plastic deformation of copper are: Equal Angle Channel Drawdown (ECAP) [5,6], Hot Pressure Torsion (HPT) [7,8] and Dynamic Plastic Deformation (DPD) [9,10].

Recrystallization consumes the excess energy introduced by plastic deformation and restores the properties of the metal by reducing the dislocation density and by generating new, essentially defect-free, recrystallized grains [11, 12]. The deposited energy in the plastically deformed metal depends on the magnitude of the cold plastic deformation undergone (the degree of plastic deformation), and the larger it is, the larger areas of the polycrystallite are covered by the recrystallization process and the more numerous are the newly formed recrystallization nuclei. From these very numerous recrystallization centres, a large number of grains are nucleated, which remain small in size due to limited growth opportunities [13]. Initially, the new, though equiaxed grains still have a certain "hereditary orientation" which gradually disappears. The second stage of the process, which takes place with prolonged heating or heating to higher temperatures, consists in the nucleation of the primary recrystallized equiaxed grains, at the cost of the fusion of adjacent grains [14, 15]

At a high degree of plastic deformation, the recrystallization annealing processes have been studied, and optimal parameters of the recrystallization annealing regime have been indicated in the literature studied. Of interest are products with a lower degree of PD and the possibility of structure refinement by heat treatment. The parameters of the recrystallization annealing process depend on: the nature of the metal (alloy), the degree of plastic deformation, the heating temperature and the holding time at this temperature. The recrystallization temperature threshold depends on the purity of the metal, the homogeneity of the structure and the degree of deformation. There are no unambiguous relationships between these parameters [16, 17, 18, 19].

In order to establish the optimum mode of the recrystallization process after cold PD, it is necessary to conduct experimental studies. This will establish the optimum heat treatment regime that secures the desired

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set of physical and mechanical performance at the most energy efficient parameters.

The purpose of the present research is to ensure a structure of the material after plastic deformation and recrystallization annealing minimum 10 points, with a hardness not less than 55 HB.

II. MATERIALS AND METHODS

For the planned experiments, segments of workpieces manufactured from Cu 99.9 obtained by two different plastic deformation (PD) processes were isolated. The first workpiece was produced by spin-forming with a relatively low PD rate of 60%, and the second by stamping with an even lower PD rate of 40%. The workpieces produced by both processes were cut into segments and subjected to different recrystallization annealing regimes shown in Table 1.

TABLE 1. HEAT TREATMENT MODES

№	Heating temperature, C^o	Retention time, min	Cooling
1	I^{-st}200 II^{-nd}280	I^{-st} 120 II^{-nd}180	under running water for 2-3 min
2	I^{-st} 200 II^{-nd} 280	I^{-st} 180 II^{-nd}180	
3	I^{-st} 200 II $^{-nd}$ 350	I^{-st} 120 II^{-nd} 10	
4	I $^{-st}$ 200 II $^{-nd}$280	I^{-st}120 II^{-nd} 40	
5	350	20	

The recrystallization temperature threshold of pure copper is 133°C but the process of new grain formation at this temperature is very slow. For the present experiments, five regimes of recrystallization annealing were selected.

Modes 1÷4 represent two-stage recrystallization annealing with mode parameters: heating temperature 200°C and holding time 120min. The purpose of the first heating is to pre-exist the recovery (the processes of relaxation and polygonization) in the cured metal. Then, in the second stage, recrystallization of the annealed sample bodies takes place.

The fifth heat treatment mode is the recommended mode for recrystallization annealing of Cu 99.9 in which the pre-annealing process is absent. The mode parameters are: heating temperature 350ºC; holding time at the specified temperature - 20 min.

To investigate the microstructure of Cu99.9 samples, metallographic sections were prepared for microstructural analysis. The samples were wet-ground on grinding wheels numbered 240, 320, 400 and 600, 800 and 1000. After grinding the same were mechanically polished with diamond paste and lubricant. The microstructure of copper was developed with hydrochloric acid reagent (HCl- 200 ml) and triferric chloride (FeCl3- 20 g). The structure was qualitatively characterized using a Leika FlexaCamC1 metallographic microscope with 20X magnification. And the determination of the grain size of the structure with a module to the microscope software LAS X Grain Expert..

The macrohardness of the investigated Cu99,9 segments was determined by the Brinell method using a FOUNDRAX BRIN400D hardness tester.

III. RESULTS AND DISCUSSION

The results of all heat treatment modes are presented in Tables 2 (for the workpiece produced by spinning) and 3 (for the workpiece produced by stamping).

TABLE 2. RESULTS OF THE STUDY OF THE SPINNING

Annealing mode	Grain size microstructure number	Hardness HB $_{2.5/62.5/30.}$
1	11-12	62,9
2	9	49,3
3	9	51,2
4	9	51,2
5	9	50,8

TABLE 3. RESULTS OF THE STUDY OF THE STAMPING

Annealing mode	Grain size microstructure number	Hardness HB $_{2.5/62.5/30.}$
1	6	62,6
2	5	61,7
3	6	59,9
4	7	62,6
5	6	57,4

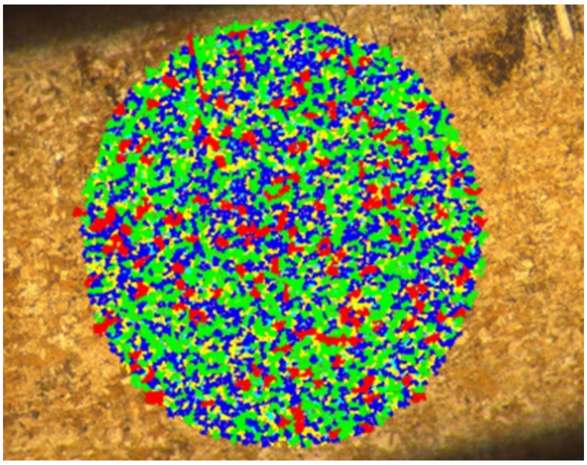

Fig. 1. Software grain measuremen after mode №1 after spinning

A fine structure with dispersively distributed crystals that are rounded was observed on the examined metallographic slit field of Cu99.9 after spinning and recrystallization annealing by mode 1 (Fig. 1). The measured and calculated conditional grain diameter was 7.75 μm, which bal. Software measurements of the microstructure show that the percentage content of different grain sizes varies. The predominant grain sizes were 11 bal - 25.97%, 12 bal - 31.58% and 13 bal - 21.82%. As not a few crystals were crystallized with size 14 Ball - 8.78%. The measured hardness of the copper so annealed was 54.6 Hb $_{2.5/62.5/30.}$

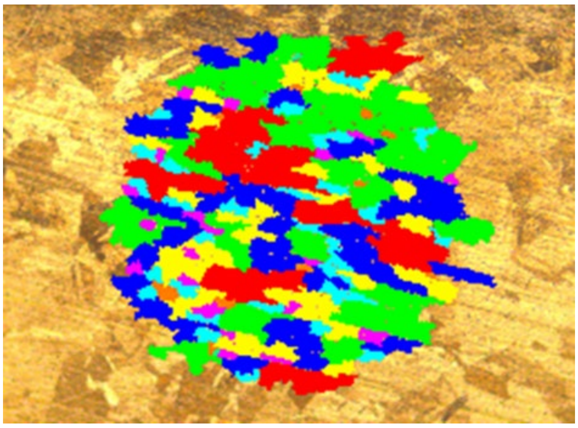

Fig. 2. Software grain measuremen after mode №1 after stamping

After mode 1 of the sample body made by stamping with a PD of 40%, we observe much larger grains (Fig. 2). Software measurements of the microstructure show that the percentage of different grain sizes varies, but the predominant amount is 6 Ball The measured hardness of the copper annealed in this way is 62.6 Hb $_{2.5/62.5/30}$. At practically the same macrohardness we observe a considerable difference in grain sizes. The probable reason for this is that, at this low degree of PD, deformation took place unevenly and some of the crystals remained undeformed. The energy required to nucleate new grains in the recrystallization annealing process was probably insufficient.

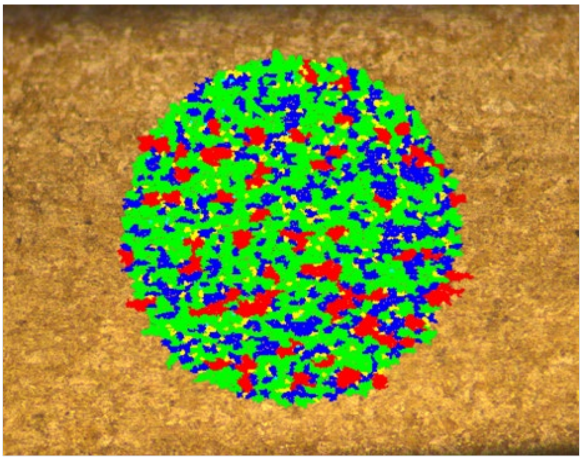

Fig. 3. Software grain measuremen after mode №1 after spinning

In Fig. 3, we observe the microstructure of a sample body fabricated by spinning and subjected to heat treatment mode No. 5 without pre-treatment. In the literature studied [17,18], the recommended temperature for recrystallization to take place is 350ºC and the retention time is 10 to 20 min, according to the degree of plastic deformation, for this reason the same parameters of the recrystallization annealing regime were used. The results after annealing in mode No. 5 of the spun workpiece were satisfactory but much lower than those in mode No. 1. The measured and calculated conditional average grain diameter was 13.9 µm, which corresponds to the 9th ball. Software measurements of the microstructure show that the percentage content of the different grain sizes is different. The predominant grain sizes were 9 ball - 27.18%, 10 ball - 31.28% and 11 ball - 21.47%. The measured hardness of the copper so annealed was 50.8 Hb 2.5/62.5/30.

The microstructure of the segment of the workpiece produced by stamping is shown in Fig. 4. On the investigated metallographic slit field of Cu99.9, after plastic deformation conducted in the cold state of the sample body with subsequent recrystallization at 350℃, holding for 20 min and subsequent cooling in running water for 1-2 min, multiple ucropped grains surrounded by grains of very small size are observed, from which we judge that secondary recrystallization has taken place due to the small PD degrees. The measured and calculated conditional mean diameter of the grains is 44.26 µm, which corresponds to the 6-7th ball. Software measurements of the microstructure show that the percentage content of the different grain sizes is different. The predominant grain sizes were 5 score - 17.86%, 6 score - 12.50%, 7 score - 21.43% and 8 score - 19.64%. The results of all the measured crystals are summarized in Figure 5. The measured hardness of the as-annealed copper was 57.4 Hb $^{2.5/62.5/30.}$.

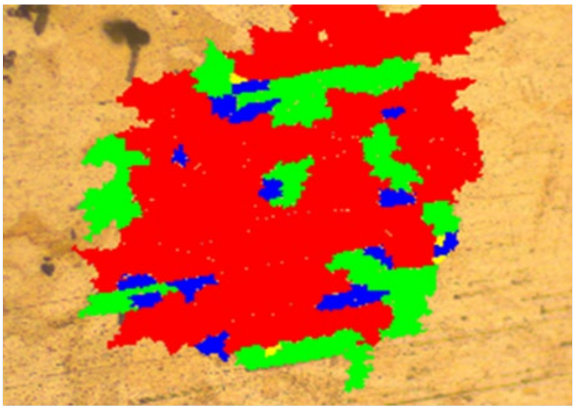

Fig. 4. Software grain measuremen after mode №1 after stamping

Tables 2 and 3 report the results of the grain size studies conducted after recrystallization as well as the measured macrohardness. The results show that for the workpiece produced by spinning, the results are very good, the microstructure is fine and relatively uniform. In the mode of recrystallization annealing with pre-recovery (Mode No. 1), the microstructure corresponds to 11-12 points, which is difficult to achieve even in articles with a high degree of plastic deformation (at Cu99.9, such a degree of PD is assumed to be more than 70%). The macrohardness of the sample body so heat-treated is 62.9 Hb $_{2.5/62.5/30}$, which, combined with the high microstructure score, provide a very positive set of physical and mechanical indicators. In recrystallization annealing modes 2, 3, 4 and 5, the size of the grain corresponds to score 9, and the macrohardness is commensurate and lower than that in mode 1.

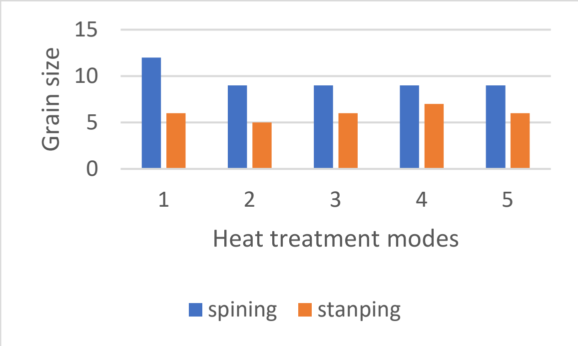

Fig. 5. Results of grain size determination after annealing

In the recovery process, different physical properties are recovered in different temperature range. The recovery mechanism is explained by the lattice resistance of the pre-deformed polycrystallite decreasing with increasing temperature in parallel with increasing atom mobility. In grains where the internal stresses were close to the yield stress Re, submicroscopic slips occurred which damped the internal stresses, thus removing the genus II stresses and part of those of genus III. By removing these stresses artificially after the first heating, the aim is to secure a microstructure free of stresses with ordered defects, which is a prerequisite for a finer dispersed structure after recrystallization annealing (the second stage of the heat treatment process). To a large extent, this explains the high score of the microstructure after the first heat treatment mode of the spin-formed workpiece. The low degree of plastic deformation of the workpiece produced by shimming is the reason for the deteriorated microstructure after heat treatment.

In mode 5 of the workpiece produced by stamping, we observe the mechanism of grain fusion, which consists in the gradual "dissolution" of the grain boundaries and the merging of many small grains into one large grain. The migration of grain boundaries is a diffusion process whose rate is determined by the rate of self-diffusion and therefore this process is characteristic of heating at higher temperatures, and at the low PD (40%) a temperature of 350ºC appears to be high enough for the 20 min retention period for the process to proceed to secondary recrystallisation.

IV. CONCLUSIONS

The most refined and homogeneous microstructure is possessed by the sample body produced by spinning, which was subjected to a quenching (at 200°C for 120 min) before the primary recrystallization process (280°C for 180 min). With the investigations carried out, it is shown that the time provided for the ordering of the defects in the crystal lattice during the polygonization process prepares the microstructure for primary recrystallization and this favours the nucleation of a larger number of centers on which to grow a greater number of crystals.

From the investigations carried out, it can be seen that the specimens subjected to the five different recrystallization annealing regimes of the spin-coated workpiece are in the primary recrystallization zone. This is evidenced by both the microstructure and the measured macrohardness.

The experiments performed show that at very low PD levels (40%), the different annealing regimes do not improve the microstructure. At all heat treatment regimes, the microstructure is coarse and heterogeneous.

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