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Microstructural Evolution and Thermal Stability of Pure Copper Processed by Severe Plastic Deformation

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Abstract: Annealed pure copper samples of 99.97 wt. % processed at room temperature by Equal Channel Angular Pressing (ECAP) technique with route B_c up to 8 passes followed by cold rolling up to 75% reduction in thickness. The sample was followed by isothermal annealing at different temperatures. The recrystallized microstructure was detected by using EBSD technique. Isochronal and isothermal annealing was performed to investigate the material's behavior by measuring the change of Vickers microhardness. With increasing annealing temperature, the microstructure becomes more homogeneous while the microhardness decreases dramatically. A modified standard JMAK- microhardness Model used to analyze the Kinetics of recrystallization of the annealed deformed samples, in this approach the JMAK model expressed in terms of microhardness measurements. JMAK exponent, n, the temperature dependent constant, k, and the activation energy, Q, for recrystallization are obtained.

Keywords: UFG, ECAP, EBSD, cold Rolling, recrystallization

1. Introduction

Processes with severe plastic deformation (SPD) may be defined as metal forming processes in which an ultra-large plastic strain is introduced into a bulk metal in order to create ultra-fine grained metals [1]. The main objective of a SPD process is to produce high strength and lightweight parts with environmental harmony.

In the conventional metal forming processes such as rolling, forging and extrusion, the imposed plastic strain is generally less than about 2.0. When multi-pass rolling, drawing and extrusion are carried out up to a plastic strain of greater than 2.0, the thickness and the diameter become very thin and are not suitable to be used for structural parts.

In order to impose an extremely large strain on the bulk metal without changing the shape, many SPD processes have been developed such as equal channel angular pressing (ECAP) [2,3], accumulative roll-bonding (ARB) [4,5], high pressure torsion (HPT) [6,7], repetitive corrugation and straightening (RCS) [8], cyclic extrusion compression (CEC) [9], torsion extrusion [10], severe torsion straining (STS) [11], cyclic closed-die forging (CCDF) [12], super short multi-pass rolling (SSMR) [13] have been developed.

Equal channel angular pressing (ECAP) is one of the important SPD methods for obtaining extremely fine grain structures in metals and alloys in bulk forms that may be used in a wide range of structural applications [14]. A large body of research has been published on the efficiency of this technique for grain refinement of a number of metallic materials and alloys [15–17].

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During ECAP process, a metal billet is pressed under a certain load to pass a die with two channels of equal cross section and induce a severe shear deformation to metal [18, 19]. The intersecting angle between channels is generally 90^o [20]. The unchanged cross-sectional area of the sample through the ECAP process makes possible to press the samples through the die a number of times in order to achieve a very high strain [21].
Metals that have been subjected to SPD usually exhibit improved mechanical properties because of high dislocation density and grain refinement but at the same time exhibit poor thermal stability [22]. The thermal stability of a material is a very important aspect that has to be considered in order to know the limitations arising from the changes in its microstructure and mechanical properties when it is subjected to heat treatments.

Recrystallization Kinetics

The kinetics of recrystallization is usually described by the JMAK (Johnson and Mehl-Avrami- Kolmogrov) model [23], when the stored energy is homogeneous and the nuclei are randomly distributed. The JMAK model can be expressed as:

$$X_v = 1 - \exp(-kt^n) \quad (1)$$

where Xv and t are the recrystallized fraction and time respectively, k is a kinetic parameter depending on the annealing temperature, nucleation rate and growth rate, it can be expressed as

$$k = k_o \exp\left(-\frac{Q}{RT}\right) \quad (2)$$

where k_o is a constant, Q activation energy, R the gas constant (R=8.314472 mol^{-1} K^{-1}) and T is the absolute annealing temperature. n is the Avrami or JMAK exponent, reflects the nucleation rate and/ or the growth morphology. Graphically this quantity is the slope in a plot of lnln (1 - X (t))-1 vs ln t; termed an Avrami plot.
The current method for analysis using a modified JMAK model for evaluating the kinetics of recrystallization is proposed. The model uses microhardness data instead of microstructure and provides a unified equation that can be used to determine the parameters of recrystallization.
The aim of the present work is to investigate the thermal stability of ultrafine grained copper processed by ECAP preceded Cold Rolling (ECAP CR).

2. Experimental Work

A high purity (99.97%) copper sample with size 32×32×160 mm was annealed at 600 °C for 1 h to achieve a homogenous microstructure prior to ECAP deformation Fig (1). The sample exhibits an equiaxed grain microstructure with an average grain size of about 100 μm with Vickers hardness HV= 64.
The procedure of ECAP consists in forcing a specimen to pass through two channels of equal cross-section interesting at an angle Φ. After passing through the channels, the sample, while retaining its original geometry, has undergone a shear deformation which under typical conditions (Φ= 90°, Ψ = 20°) i.e. perpendicular channels. The ECAP-die is shown in Fig. (2). in this work, the sample was deformed at room temperature with route B_C for 8 passes. In route B_C the sample is rotated by 90^o in the same direction between consecutive pressings. The total equivalent strain introduced in this ECAP process was ε = 8.45 The sample was then subjected to cold rolling at room temperature with 75% total reduction in thickness.

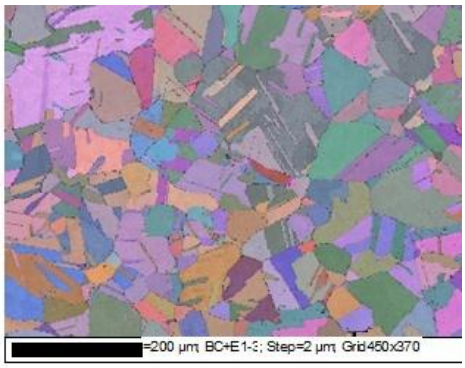


Fig. 1. Microstructure of the initially annealed sample before deformation

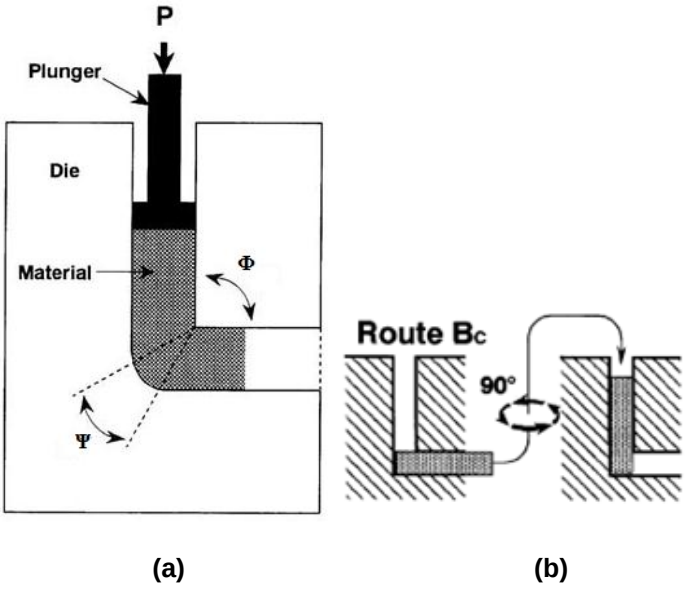


Fig. 2. (a) Illustration of ECAP die, (b) Route B_C

After (ECAP CR) deformation isochronal annealing was carried out on selected samples at various temperatures ranging from 100 to 350 °C in oil, in which the sample was held at each temperature for 50 second. In addition, samples were subjected to isothermal annealing at the same range of the temperature. The specimens were cooled in air after each heat treatment step.

The thermal stability of the mechanical properties was monitored by applying Vickers microhardness measurements using Wolpert Wilson hardness tester at room temperature. A load of 100 g force was applied for 10 s and more than 20 random indentations were made to obtain a representative bulk hardness value (HV0.1). To evaluate morphological evolution of samples after (ECAP CR) and after annealing, cross-sections of some of the Samples used in the present investigation were studied by using EBSD technique.

3. Results

3.1 Thermal Stability

The recrystallization temperature of a material is characterized by softening due to the decrease in dislocation density and the nucleation of new grains [24]. The response of hardness to isochronal heat treatment of ECAP processed specimen to 8 passes followed by cold Rolling is shown in Fig. 3. The thermal stability curve can be divided into three distinctive regions. For low temperature, starting from room temperature to 200 ^{o}C, the

microhardness values were almost stable, this is could be referred to that copper is a metal with low stacking fault energy (SFE). We can also see that the hardness in early stage is insensitive to the annealing while fall rapidly in a very short time in the second stage, The second region ranged from 200 to 280 ^{o}C. This region is characterized by a significant drop of hardness. The recrystallization of materials makes new grains without internal dislocation which causes this severe reduction. Upon further annealing, the third stage of thermal stability curve started at 280 ^{o}C and finished at about 350 ^{o}C. This region showed a slight reduction in microhardness with a gentle slope and is related to grain growth of materials.
From the numerically determined differential curve of the thermal stability curve the recrystallization temperature could be obtained. The differential curve was determined by differentiating the curve of the Vickers hardness values, which were normalized to the initial hardness HV_o [25]. Therefore the recrystallization temperature of the ECAP CR processed copper could be determined to be about 240 ^{o}C. An extrusion preceded ECAP specimen to a strain ε = 2.5 processed at 200 ^{o}C had a lower recrystallization temperature of 220 ^{o}C in comparison to 240 ^{o}C of (ECAP CR) specimen [25]. The different stored deformation energy for these different deformation procedures is the reason for the different recrystallization temperatures.

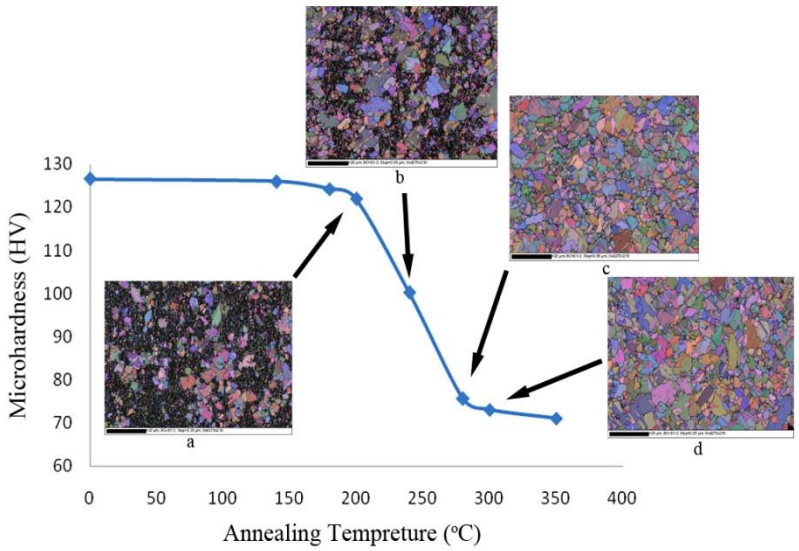


Fig. 3. Microstructure evolution during isochronal annealing for 50 seconds at different temperatures (a) 200°C, (b) 240°C, (c) 280°C, (d) 300°C

The effect of isothermal annealing on the microhardness can be seen in Fig .4, similar trend were observed where three clear stages can be seen. It can be observed that in all thermal stability curves no recovery takes place before recrystallization due to the low stacking fault energy of copper as mentioned before. With increasing the annealing temperature the acceleration of the softening of the material takes place. Fig 4 shows the fact that the higher the annealing temperature, the earlier the onset to recrystallization and grain growth where it is dropped from 30 minutes to 90 seconds as the temperature increasing from 140^{o}C to 200^{o}C respectively. We can notice that the time interval of the first stage is related to the annealing temperature. Annealing at low temperature resulted in increasing this period, while annealing at higher temperature makes it shorter.

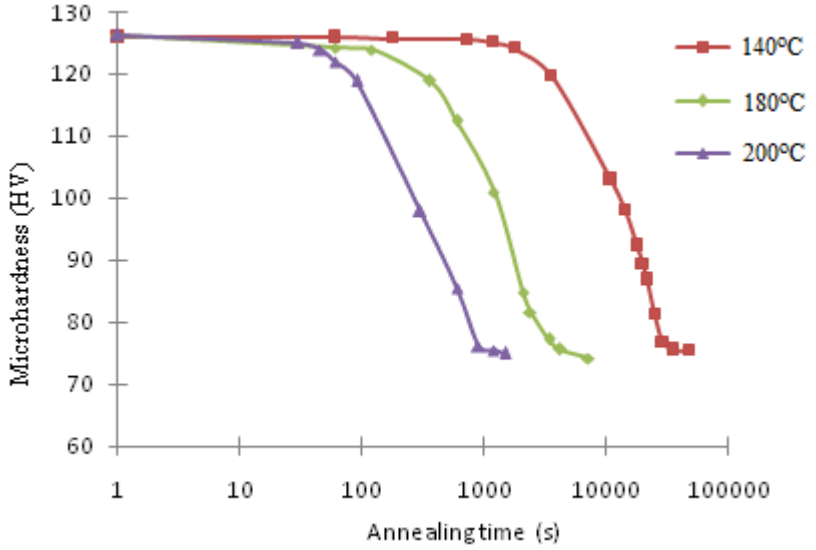


Fig.4. Variation of the microhardness vs. time at different annealing temperatures of (ECAP CR) sample.

3.2 Microstructure

Micrographs of annealed samples for various regions of thermal stability curve, which are presented in Fig. 3 shows that the nucleation of new grains in the boundaries of subgrains started at 200 ^{o}C (Fig.3a) and the recrystallization phenomena finished at 280 ^{o}C making a uniform and fine structure where the nucleated grains consumed the surrounding heavily worked grains and then new grains grew and formed equiaxed grains (Fig.3c). At 300 ^{o}C significant grain growth has occurred and the new microstructure consists of coarse grains with annealing twins.

Figure 5 shows an EBSD micrograph that represents the evolution of microstructure of a specimen during isothermal annealing at T = 140 ^{o}C. The microstructure of the materials annealed for 2 hours shows new grains partially evolved along some original grain boundaries with about 21 % recrystallized grains (Fig. 5a). Annealing for 5 hours produces a microstructure that was about 65 % recrystallized (Fig. 5b). The recrystallized microstructure reaches about 81% with further annealing to 6 hours (Fig. 5c); the microstructure was fully recrystallized when the sample was annealed at 8 hours (Fig. 5d).

Table (1) lists the changes in the recrystallized volume fraction, X_V in the annealed specimen to 140 ^{o}C as calculated by using EBSD technique.

3.3 The Modified JMAK Microhardness Model

To verify the recrystallized fraction achieved from the modeling, the microhardness test can be used, experimentally. The following relationship can be given for the recrystallized fraction [26]:

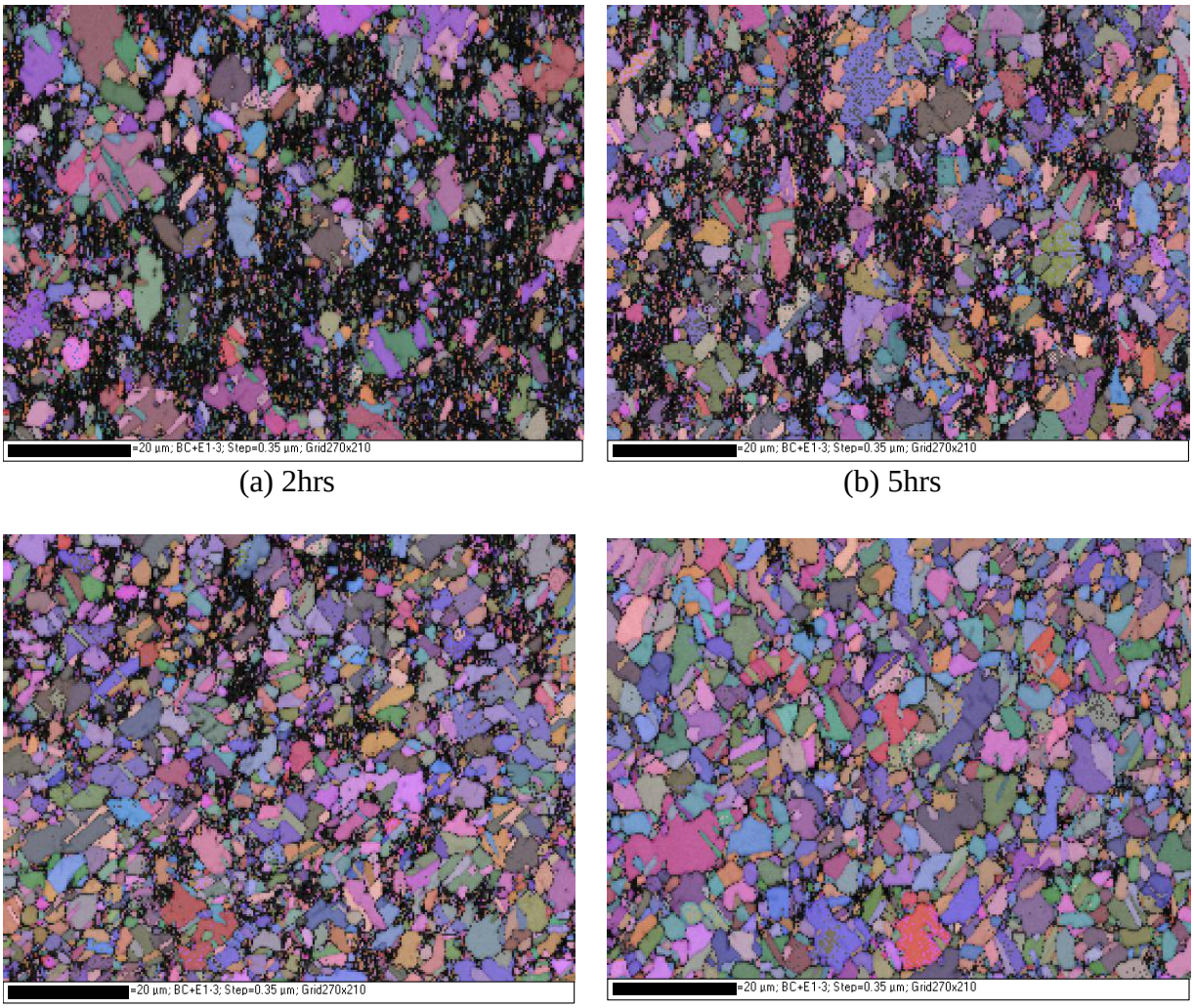
$$X_R = \frac{H_O - H_t}{H_O - H_{ann}} \tag{3}$$

where H_0 is the microhardness of the deformed material, H_t the microhardness of the annealed material at time t, and H_{ann} is the microhardness of the fully annealed material.

From (1), (3)

$$\frac{H_O - H_t}{H_O - H_{ann}} = 1 - \exp(-kt^n)$$

$$H_t = H_O - (H_O - H_{ann})[1 - \exp(-kt^n)] \tag{4}$$



(a) 2hrs (b) 5hrs

(c) 6hrs (d) 8hrs

Fig. 5. EBSD microstructure of the copper sample after 8 ECAP passes followed by cold rolling annealed at 140 ^{o}C at different times.

Table 1. Recrystallized volume fraction of the annealed specimen to 140 ^{o}C as a function of time

Time	30 mins	1 hr	2 hrs	3 hrs	4 hrs	5 hrs	6hrs	7 hrs	8hrs
X_V (%)	5	15	21	40	55	65	81	94	96

From JMAK equation

$$ln(1 - X_V) = (Kt^n)$$

$$ln\{\ln\left[\frac{1}{(1-X_V)}\right]\} = ln\,(K) + n\,ln(t)$$

In this technique the restoration kinetics during recovery, recrystallization and grain growth can be computed from JMAK equation using the microhardness data.

Plotting of the fraction recrystallization X_V (Fractional Transformed X_R) against annealing time (t) as shown in Fig.6 shows us the same results in Fig.4 where the sigmoid shape of three stages of recovery, recrystallization and grain growth are clearly evident.

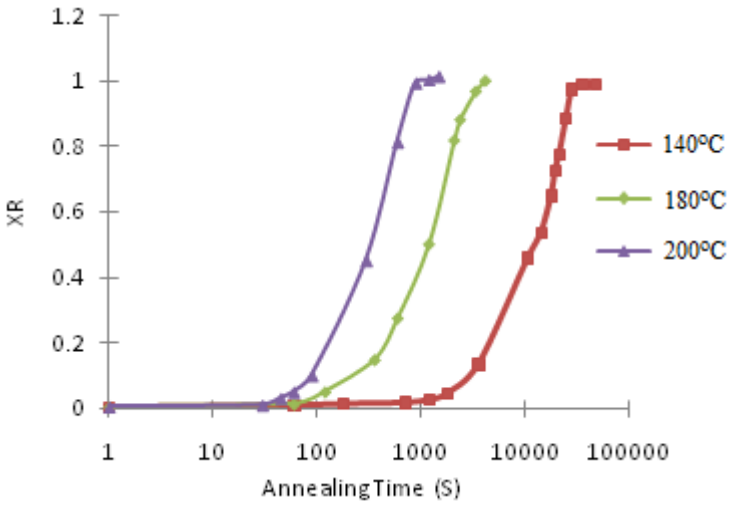


Fig. 6. (Fractional Transformed (X_R)) VS. Annealing Time (t).

Using the modified JMAK model to analyze our data and to plot the relation $\ln\left(\ln\left(\frac{1}{1-X_R}\right)\right)$ versus$(\ln t)$, the values of the JMAK exponent, n, and the temperature independent constant, k, were evaluated from the plot shown in Fig.7, the summary of the recrystallization kinetic parameters for the different specimen is presented in Table.2.

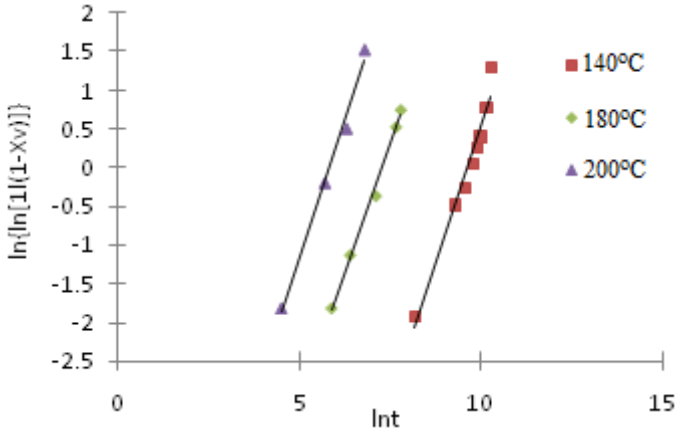


Fig. 7. JMAK plot for recrystallization kinetics for the annealed specimens

Table 2. Restoration kinetics parameters of recrystallization

	Recrystallization kinetic parameters	
	n	ln *k*
(T = 140 ^{o}C)	1.4	-13.77
(T = 180 ^{o}C)	1.3	-9.72
(T = 200 ^{o}C)	1.4	-8.24

The activation energy Q

The temperature dependent constant, k, can be described in the form of the equation (2) which also can be expressed as:

$$\ln k = \ln k_o - \frac{Q}{RT} \tag{5}$$

Plotting ln*k* versus 1/T will give a straight line whose slope equal to Q/R.

Figure (8) represents plots of lnk versus 1/T for the recrystallization (where T is the annealing temperature in Kelvin). The apparent activation energy, Q, computed from this plot is 149 kj/mol.

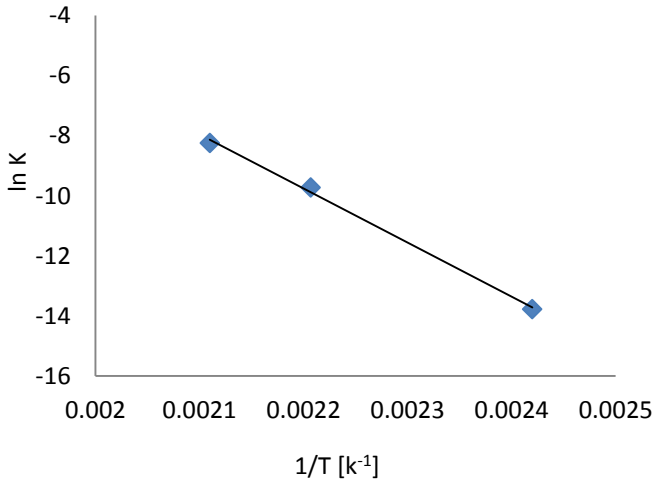


Fig. 8. A plot of lnk vs 1/T for the recrysallization

4. Discussions

ECAP processes generate large number of dislocations during deformation, which rearrange into a lower energy configuration upon annealing. Copper, as a low stacking fault energy material (SFE: 78 mJ/m^2) [27], shows an indistinctive recovery processes where climb and cross- slip of dislocations are difficult, as these are the mechanisms that usually control the rate of recovery. This explains the stable value of hardness without a remarkable change in the first stage in the thermal stability curve Fig 3.

During recovery there is no significant change in the microstructure, and the subtle changes cannot be accurately detected by the conventional stereology or OIM measurement [23]. Microhardness is inherently related to dislocation density, and can thus be used to measure the fractional transformed during recovery.

On the other hand, recrystallization is driven by the remaining part of the stored energy, although there may be competition between recovery and recrystallization for this energy, especially at the early stages of recrystallization. Recrystallization can take place by the nucleation and growth of new grains at the expense of the recovered matrix. These nuclei of newly formed grains are relatively strain-free, and on reaching the critical size, are surrounded or partially surrounded by high angle grain boundaries (HAGBs) with high mobility. They are, then energetically capable of growing into the matrix by the migration of their HAGBs. Recrystallization results in a drastic drop of the microhardness due to the fact that a large volume of dislocations are swept by the migration of HAGBs which have a much higher mobility than LAGBs.

As recrystallization proceeds, impingements among these new grains occur. Finally, the entire specimen is replaced by totally recrystallized grains. When primary recrystallization is complete, the structure is not yet stable, and further growth of the recrystallized grains may occur by the migration of grain boundaries with the grain-boundary free energy as the driving force.

The value of the JMAK exponent, n, for recrystallization kinetics in the three samples was about the same, and its average was 1.35 which means that at this range of temperature the recrystallization kinetics in this material is insensitive to the change in annealing temperature. The apparent activation energy for recrystallization in this study was estimated to be 149kJ/mol. Which is an intermediate value between the activation energy for self diffusion of

copper (Q_{sd}=200kj/mol [28]) and the activation energy for grain boundary diffusion (Q_{gb}=100 kj/mol [28]). However this value is close to the value reported by Takayama et al. [29].
The fraction recrystallized, X_V, measured in table 1, is fairly matched to the fraction transformed, X_R, measured by the microhardness data Fig .6, which is an evidence that the proposed model gives us a precise and quick analysis of the kinetics of recrystallization.

5. Conclusions

Severe plastic deformation under (ECAP CR), allowed pure copper to be highly deformed with a mainly granular ultrafine structure. In this deformed state, copper has high level of lattice strains and displayed high hardness values.
Isochronal annealing for 50 seconds up to temperature of 200°C shows that neither the microhardness nor the grain size is affected considerably, However the dislocation density decreased which referred to the low stacking fault energy of copper. While at higher temperatures up to 280°C, a full recrystallized structure is obtained. The recrystallization temperature of the ECAP CR processed copper could be determined from the thermal stability curve to be about 240 ^{o}C.
The microstructure evolution could be observed during annealing characterized by the appearance of new larger grains in the deformed structure. In the course of annealing the fraction of these larger grains grew at the expense of the deformed microstructure, and this proceeded faster with increasing the annealing temperature. The annealed microstructure becomes more homogeneous with increasing of annealing temperature
The modified JMAK-microhardness model is expressed in terms of microhardness data, and the results show that the model provides the time exponent (reaction order), n, and activation energy, Q, for recrystallization. For ECAP CR copper under study, the kinetics of recrystallization showed a temperature-independent time exponent, with average values of 1.35. The estimated activation energy was 149kJ/mol for recrystallization.

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