Study on texture and dynamic recrystallization behavior of high purity copper during reverse extrusion

Chunyu Li, Qichi Le, Ke Hu, Lei Bao, Bowen Ma, Yonghui Jia, Xuan Wang, Weiyang Zhou and Guojun Xu

Key Lab of Electromagnetic Processing of Materials, Ministry of Education, Northeastern University, 314 Mailbox, Shenyang 110819, People’s Republic of China

1 Author to whom any correspondence should be addressed.

E-mail: qichil@mail.neu.edu.cn

Keywords: dynamic recrystallization, reverse extrusion, microstructure evolution, grain size, high-purity copper

Abstract

The dynamic recrystallization (DRX) behavior of high purity copper was studied by the combination of numerical simulation and experiment. Specifically, extrusion experiments were performed in the temperature range of 200 °C~800 °C. The effects of temperature and deformation on the microstructure of the deformation zone were studied. It was found that the grain size decreased with the decrease of temperature and the increase of deformation. The optimized extrusion temperature of 500 °C~550 °C was determined based on grain size and uniformity of microstructure. The special grain boundaries, grain orientation and recrystallization texture during extrusion were studied by electron backscattered diffraction (EBSD) technique. It was found that the increase of the special grain boundary Σ3 and the texture of (111) parallel to the ED direction have a positive effect on grain refinement and microstructure uniformity. As the temperature increases, the proportion of recrystallized grains increases initially, and finally decreases. A cellular automaton (CA) method that couples the developed DRX model was implemented to investigate the evolution of microstructure during high-purity copper extrusion using DEFORM-3D software, and the results were verified by experiments. The microstructure evolution and average grain size distribution predicted by numerical simulation agreed well with the experimental results.

1. Introduction

Copper has good electrical conductivity, thermal conductivity, high chemical stability, corrosion resistance, low electrical resistivity and high migration resistance. It can be combined with low dielectric constant materials to reduce capacitance, thereby reducing resistance and increasing the speed of operation of integrated circuits [1]. The replacement of aluminum alloys with high-purity copper materials as a target is one of the important trends in the future. Up to now, high purity metal targets sputtered are mainly produced by casting and powder metallurgy. Since the manufactured high-purity copper ingot has the defects of coarse grains, inhomogeneity of structure, and less density, it cannot be directly used as a raw material of the target, so it is necessary to refine the grains and make the structure uniform by means of plastic manufacture. High-purity copper materials subjected to plastic deformation have been used to study the refinement of grain leading to better mechanical properties [2, 3].

In recent years, the market demand for high-purity copper targets has been increasing, which means that the requirements of products are also increasing. In order to reduce the cost and improve the quality of products, many companies have adopted reverse extrusion technology. Compared with other plastic deformation modes, the extruded product is in a three-dimensional compressive stress state, which greatly improves the plasticity and strength of the metal. The product has the advantages of good surface quality and high dimensional accuracy. A large number of experimental results show that the purity, microstructure and texture of high-purity copper have a great influence on the utilization rate of sputtering targets [4]. Mark Coleman et al [5].
found that the increase of grain size was not obvious with the increase of special grain boundaries $\Sigma 3$. Therefore, the increase of the special grain boundary can effectively inhibit the growth of the grains. It was found that grain orientation affects the sputtering rate of the target. In a certain range, the rate sputtered of target materials on different crystallographic planes is also different, and the rate sputtered of (111), (100), (110) crystallographic planes gradually decreases. In addition, sputtering rate of target materials with different grain orientation is also different, and the sputtering rates of [110], [100], [111] grain orientations gradually decrease [6]. Grain size is one of many factors. The smaller the grain size, the higher the sputtering speed of the target. In addition, the grain size also affects the uniformity of the target sputtered thin film. The smaller the grain size, the more uniform the thin film is deposited in the thickness of the target. Therefore, it is important to refine the grains. Grains will be refined by DRX during extrusion. The temperature and deformation level can greatly affect the generation of recrystallization. If the evolution of microstructure and DRX behavior can be predicted, it is of great significance for application of targets in the future. As a mathematical calculation method, finite element analysis can predict the flow of materials during the forming process, and the stress field, strain field and microstructure properties after deformation, which improves production efficiency, saves resources and reduces costs.

According to C-K Hu et al [7], since copper plating has a fine grain size than vapor deposition copper (CVD), the copper plating has a longer failure time than CVD copper. Pardis et al [8]. conducted a multi-pass extrusion study on pure copper by cyclic expansion-extrusion (CEE) process and found that compared with other large plastic deformation modes, finer grain structure can be obtained, and different passes will affect the final texture of pure copper. K A et al [9], optimized the extrusion process parameters based on DEFORM-3D software and inputs effective process parameters including friction coefficient, ram velocity, and die length to reduce extrusion force. Huang et al [10], studied the thermal deformation behavior of pure copper and established a constitutive equation to study the flow stress curve of copper during dynamic recrystallization. Many empirical models have been proposed to describe recrystallization behavior [11–13]. Among these methods, the cellular automaton (CA) method is widely used, and it is a dynamic system based on time and space discretization. Each unit is also discretized, following deterministic or probabilistic transition rules. Due to the high efficiency and flexibility of the time scale, the CA model can well simulate the evolution of microstructure during DRX [14]. Up to now, the recrystallization process in the fields of copper [15, 16], aluminum alloy [17, 18], steel [19–22], magnesium alloy [23, 24], titanium alloy [25] nickel alloy [26], etc have been studied using the CA method.

Therefore, in this study, the CA method was adopted to predict the evolutions of microstructure and grain size under reverse extrusion by DEFORM-3D software. Texture and microstructure evolutions of the high purity copper were studied by EBSD. It has been found that proportion increase of the special grain boundaries and the texture with a specific orientation have a positive influence on grain refinement and microstructure uniformity.

2. Experimental materials and procedures

2.1. Experimental materials

High pure copper was selected as the experimental material for the research. The purity of high pure copper is 4 N, which means that the mass fraction of Cu is over 99.99%. Due to the high purity, it can be used as an excellent raw material for sputtering targets. Compared to other materials, it possessed higher resistance to migration and lower resistivity.

2.2. Experimental rote

300 ton hydraulic equipment was used for reverse extrusion test. Pure copper ingot (47(diameter) × 50(height) mm) was used in this experiment. The initial grain size of the billet material was 4 mm. The initial temperature of extrusion was set as 300 °C–800 °C and the initial temperature of the sample was 350 °C–850 °C. The extrusion speed was set as 2 mm s−1. After the extrusion test, the extrusion bar was cooled in the air. A cross-sectional view of the assembly drawing was shown in figure 1.

In this study, the OLYMPUS DSX500 metallographic microscope was used to observe the microstructure. The microstructures were observed by scanning electron microscopy (SEM) and a ULTRA PLUS field-emission scanning electron microscope. Electron back-scattered diffraction (EBSD) analysis was performed on the selected regions using an Oxford HKL Channel 5 software. Because the microstructure of the extruded rod and the part of remaining should be carefully observed, the sample should be ground with water on 240#, 400#, 800#, 1500#, 3000#, 5000# sandpapers until there is no obvious scratch. And then, the sample was electro-polished, etched and observed using a microscope.
Figure 2 shows the cross section and longitudinal section macrostructures of high-purity copper ingot. The sampling position of the extruded billet was shown in figure 2. From the edge to center areas, the fine-grained, columnar grain and equiaxed grain zones in as-cast high purity copper were observed. The coarse columnar grains occupy a higher proportion, and grow along the direction of heat transfer in semi-continuous casting.

2.3. Finite element analysis model

2.3.1. Material constitutive equation

According to the material deformation resistance model provided in DEFORM, the constitutive equation model is [27]:

$$\dot{\varepsilon} = A [\sin h(\alpha \sigma)]^n \exp \left( -\frac{Q}{RT} \right)$$

Where $\dot{\varepsilon}$ is the material strain rate, $\sigma$ is the flow stress, and $Q$ is the deformation activation energy of the material, $R$ is the gas constant, $T$ is the thermodynamic temperature, $A$, $\alpha$, $n$ are several material-dependent constants. The specific parameter values are listed in table 1 [28].

Thermal activation energy was a very important material parameter based on the thermal deformation conditions. It not only determines the critical conditions of dynamic recrystallization, but also the dynamic relationship between softening mechanism and hardening mechanism in thermal processing. The Arrhenius model describing the deformation behavior of the material consists of the material deformation activation
energy and an extensive empirical model. This model was suitable for the case where the sample processing temperature was above the material recrystallization temperature and the stress-strain curve has a significant peak value, which was the main reason why the model was used.

2.3.2. Simulation parameters

In order to effectively analyze the reverse extrusion process, it was necessary to obtain the changes of material temperature field, stress field, strain field and during extrusion by means of finite element analysis. During the simulation, the sample was viewed as an elastoplastic body; other components were regarded as rigid bodies in the finite element calculation process, regardless of the deformation.

Pure copper ingot (47(diameter) × 50 (height) mm) was used in this simulation. The initial grain size of the billet material was 4 mm. The initial temperature of extrusion was set as 550 °C—750 °C and the initial temperature of the sample was 600 °C—800 °C. The extrusion speed was set as 2 mm s⁻¹. The friction coefficient between the workpiece and the extrusion die was set to 0.3, and the friction coefficient between the billet and the extrusion barrel was defined as 0.2. The friction type was defined as frequent friction factor method, and the heat transfer coefficient was 30 N s⁻¹ mm⁻¹ °C⁻¹. The process parameters of high purity copper and other components used in the finite element model were listed in table 2.

2.4. Cellular automaton model

In this research, the nucleation and growth of grains were established using the CA method. Firstly, the shape of the cell was selected as a quadrilateral, which means that there were four cells adjacent to the cell. Then, the size of the selected area was set up. Thirdly, the boundary conditions was determined between the cells. The neighbor of the Moore type was adopted in this work, and the radius length of the neighbor was set to 1 μm. In addition, it was necessary to select the type of dynamic recrystallization during the deformation process. The DRX type was a discontinuous dynamic recrystallization process in this study. This recrystallization has a high dislocation density during deformation. It makes the local grain boundary have higher energy, so that the energy in this region cannot be in disorder and produce recrystallization. Since the nucleation and growth of recrystallized grains are spontaneous, this approach is considered to be discontinuous. The critical dislocation density value was selected based on the dislocation density model. It determined the critical dislocation density of DRX nucleation, and its size needed to be constantly adjusted to make the analog value closer to the experimental result. Then the initial grain size was set to 4000 um, the distribution of grain boundary orientation was randomly selected by default, and the initial dislocation density was set to 0.01. The following discussion will focus on the dislocation density model and the grain nucleation and growth model.

2.4.1. Critical dislocation density model

It is generally believed that the metal is plastically deformed due to the movement of dislocations. Two models are mainly used to calculate the dislocation density under different deformation mechanisms in the simulation. The Laasraoui-Jonas model is mainly used for the dynamic recovery mechanism. In terms of work hardening, the Kocks-Mecking model is set up. Dislocation density is considered as a function of the deformation process in the Laasraoui-Jonas model (equation (2)) [29]:

\[ d\rho_i = (h - r\rho_i) d\varepsilon \]  

(2)

Where \( \rho_i \) is the initial dislocation density, where the value is set to 0.01. The work hardening coefficient \( h \) and the dynamic recovery coefficient \( r \) are regarded as coefficients related to temperature and strain rate, and the functions are as follows (equations (3) and (4)) [30]:

| Parameters | Values |
|------------|--------|
| Heat capacity (N/(mm²·C)) | 3.272 |
| Thermal conductivity (W/(m·K)) | 370 |
| Heat transfer coefficient between tooling and workpiece (N/(°C·mm²)) | 30 |
| Heat transfer coefficient between tooling / workpiece and air (N/(°C·mm²)) | 0.02 |
| Poisson’s ratio | 0.33 |
| Emissivity | 0.15 |
| Young’s modulus (N/m²) | 110000 |
is related to both the strain rate $\dot{\varepsilon}$, the dislocation density of the parent phase grains, and $\delta_0$ is the strain rate calibration constant, here is set to 1.

Dynamic recovery is actually a softening effect. As the metal is plastically deformed, the dislocation density of the metal is reduced due to the existence of dynamic recovery. In the CA model, a number of cells $N$ are randomly selected at different time steps, reducing the dislocation density by half. The number of cells $N$ can be expressed as (equation (5)) [31]:

$$N = \left(\frac{\sqrt{2} RC}{K}\right)^2 h(d_0)^{1-2m}$$

Where $R$ and $C$ are the number of rows and columns in lattices, $h$ is the work hardening coefficient. $K$ is a constant, and it is generally set to 6030.

2.4.2. Nucleation and growth model
The dynamic recrystallization nucleation rate $\dot{n}$ is related to both the strain rate $\dot{\varepsilon}$ and the temperature $T$ [32].

$$\dot{n} = C \dot{\varepsilon}^m \exp\left(-\frac{Q_a}{RT}\right)$$

Where $C$ and $m$ are constants, $Q_a$ is the activation energy, and $R$ is the gas constant of 8.314 J mol$^{-1}$ K$^{-1}$. After dynamic recrystallization stage, there is a large difference of dislocation density between the newly formed grains and the parent phase grains, which provides a driving force for the growth of recrystallized grains. Due to the recrystallization driving force, the newly formed crystal grains can be continuously grown until the dislocation density was completely eliminated. However, as the deformation continues, the strain gradually increases, reaching the critical dislocation density, and the recrystallized grains are formed at the grain boundaries, thereby repeating the process of nucleation and growth. The dynamic recrystallization critical dislocation density model can be expressed as (equation (7)) [33]:

$$\rho_c = \left(\frac{20\gamma_i \dot{\varepsilon}}{3Mh^2\tau^2}\right)^{\frac{1}{3}}$$

Where $\gamma_i$ is the grain boundary energy, $M$ is the mobility of grain boundary movement, $\tau$ is the dislocation line energy, $l$ is the free path of dislocation. The relationship between the recrystallized grain growth rate $v_i$ and the driving force $f_i$ satisfies the following conditions (equation (8)) [34]:

$$v_i = mf_i$$

Where $m$ is the grain boundary migration rate [32], it can be obtained by the following formula (equation (9)):

$$m = \frac{b\delta D_{ob}}{KT} \exp\left(-\frac{Q_h}{RT}\right)$$

Where $b$ is the Burger’s vector, $\delta$ is the grain boundary thickness, $D_{ob}$ is the grain boundary self-diffusion coefficient when the temperature is absolute zero, $Q_h$ is the grain boundary diffusion activation energy, and $K$ is the Boltzmann constant. Table 3 shows the values of them:

| $Q_b$/Jmol$^{-1}$ | $b$/m | $\mu$/Pa | $\delta D_{ob}$/m$^2$s$^{-1}$ | $K$/Jk$^{-1}$ |
|------------------|-------|----------|-------------------|-------------|
| 104000           | $2.56 \times 10^{-10}$ | $4.21 \times 10^{10}$ | $3.5 \times 10^{-15}$ | $1.38 \times 10^{-23}$ |

Where $m$ is the strain hardening sensitivity coefficient, $Q_b$ is the activation energy, $h_0$ is the hardening constant, $r_0$ is the recovery constant, and $\delta_0$ is the strain rate calibration constant, here is set to 1. After dynamic recrystallization stage, there is a large difference of dislocation density between the newly formed grains and the parent phase grains, which provides a driving force for the growth of recrystallized grains. Due to the recrystallization driving force, the newly formed crystal grains can be continuously grown until the dislocation density was completely eliminated. However, as the deformation continues, the strain gradually increases, reaching the critical dislocation density, and the recrystallized grains are formed at the grain boundaries, thereby repeating the process of nucleation and growth. The dynamic recrystallization critical dislocation density model can be expressed as (equation (7)) [33]:

$$\rho_c = \left(\frac{20\gamma_i \dot{\varepsilon}}{3Mh^2\tau^2}\right)^{\frac{1}{3}}$$

Where $\gamma_i$ is the grain boundary energy, $M$ is the mobility of grain boundary movement, $\tau$ is the dislocation line energy, $l$ is the free path of dislocation. The relationship between the recrystallized grain growth rate $v_i$ and the driving force $f_i$ satisfies the following conditions (equation (8)) [34]:

$$v_i = mf_i$$

Where $m$ is the grain boundary migration rate [32], it can be obtained by the following formula (equation (9)):

$$m = \frac{b\delta D_{ob}}{KT} \exp\left(-\frac{Q_h}{RT}\right)$$

Where $b$ is the Burger’s vector, $\delta$ is the grain boundary thickness, $D_{ob}$ is the grain boundary self-diffusion coefficient when the temperature is absolute zero, $Q_h$ is the grain boundary diffusion activation energy, and $K$ is the Boltzmann constant. Table 3 shows the values of them:

Where $f_i$ can be obtained by the following equation (equation (10)). Where $\gamma_i$ is the interfacial energy, $r$ is the grain radius, $\rho_{m}$ is the dislocation density of the parent phase grains, and $\dot{\rho}_i$ is the dislocation density of the ith recrystallized grains [32]. Where $\tau$ is the energy of the dislocation line per unit length, it is acquired by the following formula (equation (11)).

$$f_i = 4\pi r^2 \tau (\rho_m - \rho_i) - 8\pi \eta \gamma_i$$

$$\tau = 0.5\mu b^2$$
Table 4. Material parameter values involved in the calculation.

| $\alpha$ | $n_i$ | $m_i$ | $d_0$ | $Q_s$ | $R$ |
|----------|-------|-------|-------|-------|-----|
| 0.0039   | 0     | 0.11  | 4     | 28658.19 | 8.314 |

Table 5. Values of material parameter used in JMAK.

| $K_d$ | $a_s$ | $m_s$ | $\beta_0$ | $Q_s$ | $R$ |
|-------|-------|-------|-----------|-------|-----|
| 1.457 | 0.0039 | 0.11  | 1.388     | 28658.19 | |

Where $\gamma_i$ is gained by the following formula (equation (12)) [35].

$$\gamma_i = \gamma_{\text{imm}} \left( 1 - \ln \frac{\theta_i}{\theta_{\text{mm}}} \right)$$  \hspace{2cm} (12)

Where $\theta_i$ is the difference between the recrystallized grains and the adjacent crystal grains, $\theta_{\text{mm}}$ is the difference in the grain boundary of the large angle, and $\gamma_{\text{imm}}$ is the grain boundary energy.

2.4.3. Dynamic recrystallization volume fraction and grain growth model

According to Devadas, C [36] et al the starting of recrystallization is related to peak strain $\varepsilon_p$. The peak strain $\varepsilon_p$ can be expressed by the following formula (equation (13)). The same results have been also demonstrated in other literatures [37].

$$\varepsilon_p = \alpha_1 d_0^{m_1} \dot{\varepsilon}^{m_1} \exp \left( \frac{Q_s}{RT} \right)$$  \hspace{2cm} (13)

Where $d_0$ is the initial grain size, $\dot{\varepsilon}$ is the deformation rate, $Q_s$ is the deformation activation energy, and $T$ is the deformation temperature. $\alpha_1$, $n_1$, $m_1$ are constants. Table 4 shows the values in the CA model [28]:

The relationship between critical strain and peak strain can be expressed as (equation (14)) [28, 37]:

$$\varepsilon_c = 0.82 \varepsilon_p$$  \hspace{2cm} (14)

It is desirable to refine grains by recrystallization during thermal deformation. In this study, the JMAK equation was chosen to describe the dynamic recrystallization kinetic relationship. The calculation method is as follows (equations (15) and (16)) [38]:

$$X_{\text{drex}} = 1 - \exp \left[ -\beta_0 \left( \frac{\varepsilon - \varepsilon_{0.5}}{\varepsilon_{0.5}} \right)^{K_d} \right]$$  \hspace{2cm} (15)

$$\varepsilon_{0.5} = a_s d_0^{h_0} \dot{\varepsilon}^{m_0} \exp \left( \frac{Q_s}{RT} \right)$$  \hspace{2cm} (16)

Where $X_{\text{drex}}$ is the dynamic recrystallization volume fraction, $\varepsilon_{0.5}$ is the strain at the time of 50% dynamic recrystallization, $Q_s$ is the activation energy when dynamic recrystallization occurs. Regardless of the influence of the initial grain size, the influences of deformation rate and temperature change are mainly studied. $K_d$, $\beta_0$, $a_s$, $h_0$, $n_0$, $m_0$ are constants. Table 5 shows the used material parameters in JMAK simulation [28]:

The steady state recrystallized grain size is related to the strain rate and temperature. Their effects on the final grain size can be expressed as a function of the Zener–Hollomon parameter ($Z$) (equations (17) and (18)) [28, 39, 40]:

$$Z = \dot{\varepsilon} \exp \left( \frac{Q}{RT} \right)$$  \hspace{2cm} (17)

$$d_{\text{drex}} = 12636 Z^{-0.176}$$  \hspace{2cm} (18)

3. Results and discussion

3.1. Microstructure analysis in deformation zone

Figure 3 shows a schematic diagram of macrostructure in the deformation zone parallel to the extrusion direction. In figure 3, the microstructures of (a), (b), (c), (d), (e) are corresponding to the coordinates (0, 0), (0, 3), (0, 6), (0, 9), (0, 11), and the deformation level corresponding to these five positions are about 9%, 33%, 61%, 78%, 94%. Figure 3 shows that when the extrusion temperature was 450 °C, DRX was observed in the
deformation zone, and the annealing twins were produced in the partially recrystallized grains. As can be seen from figures 3(a) to (e), as the deformation level increases, the grain are further refined. Moreover, figure 4 shows the microstructures of different temperature at parallel extrusion direction of extrusion zone at deformation of 9%. As shown in figure 4, when the extrusion temperature raised from 450 °C to 500 °C, the grain size at the same deformation level (9%) decreases obviously because DRX is incomplete at less deformation level in deformation zone at 450 °C. When the extrusion temperature raised to 500 °C~550 °C, the grains in the deformation zone have gradually translated fully recrystallized grains, and the raw grains have been replaced by fine recrystallized grains. As the extrusion temperature continued to rise to 600 °C~650 °C, the recrystallized
grains begin to grow. When the extrusion temperature exceeded 750 °C, grains in the deformation zone grow abnormally due to excessive deformation temperature.

3.2. Extrusion rod grain size analysis

Figure 5 shows the relation between grain size, deformation and temperature at the central axis of the deformation zone. As shown in figure 5(a), as increasing of the deformation level, the grain size gradually decreases at a fixed extrusion temperature. The magnitude of grain size reduction is extremely obvious at an extrusion temperature of 450 °C. When the extrusion temperature raises to 500 °C~550 °C, as the deformation level increases, the magnitude of grain size reduction is not significant. However, the grain size is significantly reduced compared to the grain size at 450 °C. It is observed from figure 5(b) that as the extrusion temperature rises from 450 °C to 800 °C, the average grain sizes at different deformation level at the central axis of the deformation zone (figure (3)) decrease at first and then increase, and finally become stable. The grain size reduces to a minimum at temperature of 500 °C~550 °C, and substantially stabilized after continuously increasing to 750 °C. The reasons for this phenomenon are as follows: when the extrusion temperature raises from 450 °C to 500 °C, the material is more susceptible to deformation and produces DRX, resulting in grain refinement. When the extrusion temperature raises to 500 °C~550 °C, all the coarse grains in the deformation zone are completely refined into fine recrystallized grains, thus the grain sizes in the deformation zone decrease. When the temperature rises to 600 °C~650 °C, the fine recrystallized grains in the deformation zone grow up due to higher temperature, resulting in the increasing of grain size in the deformation zone. When the extrusion temperature raises to 750 °C, the partial recrystallized grains abnormally grow due to the excessive extrusion temperature, which causes that the average grain sizes obviously increase. The above results show that the extrusion temperature of 500 °C~550 °C is the optimum processing temperature considering the grain size and the uniformity of the microstructure.

Figure 6 shows the microstructure maps of core of extrusion bar with low extrusion temperature. When the extrusion temperature was 300 °C~350 °C, there are fine equiaxed grains in the gap of the deformation band, and high-purity copper partially generates DRX at the position where the distortion energy of the deformation band is high. The metal fluidity increases due to plastic deformation and increasing temperature, the original long strip-shaped deformation band is broken into irregular blocks, and the nucleation rate of grain is increased, which accelerates the generation of DRX. When the extrusion temperature raises to 400 °C, there is substantially no deformation band in the extruded rod, and the microstructure becomes more uniform than 350 °C.

Figure 7 shows the relationship between the grain size of extrusion rod core and temperature. As shown in figure 7, stage 1 represents incomplete recrystallization and stage 2 represents complete recrystallization. The recrystallized grains tend to grow gradually as increasing of the extrusion temperature, because of the higher the deformation temperature, the faster the grain boundary migration rate. Therefore, high-purity copper begins to generate DRX at 300 °C. Deformed grains begin to translate into the recrystallized grains and sub-grains. When the extrusion temperature raises from 400 °C to 450 °C, the change of grain size is more obvious. DRX produces sufficiently at 400 °C~450 °C, and most of the grains are refined. When the extrusion temperature exceeds 450 °C, DRX has generated fully. As elevating of extrusion temperature, the fully dynamic recrystallized grains gradually grow.
3.3. Texture and special boundary $\Sigma 3$ analysis during deformation

Figure 8 shows EBSD map and scatter plot of a high-purity copper ingot. It is obvious that the raw grains are coarse and the distribution of grain is relatively scattered. Therefore, there isn’t obvious texture in the as-cast high-purity copper.

In order to investigate the microstructure evolution of high-purity copper after deformation, the EBSD maps of high-purity copper at different extrusion temperatures and the misorientation distribution were obtained as shown in figure 9. As shown in figures 9(a) and (b), the microstructure of elongated deformed grains and substructure in extrusion rod was observed, which includes a large number of low angle grain
boundaries (LAGBs) and high density of dislocations. The deformed grains exhibit the \( \langle 111 \rangle \) and \( \langle 001 \rangle \) parallel to ND, respectively. Figure 9(c) shows that when the extrusion temperature is raised to 450 °C, the microstructure of the sample is refined, and the distribution of grain size is more homogeneous, and about 17.6 \( \mu m \). Besides, the texture is subsequently weakened, and the grain boundaries are mainly composed by high angle grain boundaries (HAGBs). There are a large number of annealing twin boundaries \( \Sigma 3 \) in the microstructure, and the misorientation reaches the peak around 60° (figure 9(d)), indicating that when the extrusion temperature rises to 450 °C, sufficiently recrystallized was formed in high purity copper during the extrusion. It can be seen from figures 9(e) and (f) that when the extrusion temperature raises to 800 °C, the grain boundaries are composed of HAGBs, meanwhile the misorientation distributes at 60°, and the grain size increases significantly than that of the extrusion sample at 450 °C. That also matches the results in figure 5(b).

Figure 10 shows that special boundary maps of high purity copper with different extrusion temperatures. As shown in figure 10(a), there are very few special grain boundaries at low temperature, and the \( \Sigma 3 \) grain boundaries are scattered. As increasing of the extrusion temperature, the proportion of special grain boundaries increases quickly at 450 °C, and special grain boundaries are mainly \( \Sigma 3 \) and \( \Sigma 9 \). Most of the annealed twin boundaries are distributed inside the grains, and a small portion of the annealed twin boundaries are flat. These coarse flat twin boundaries are mostly distributed at the bifurcated grain boundaries or the trigeminal grain.

**Figure 9.** EBSD maps of extruded rods with different extrusion temperatures: (a) 200 °C; (c) 450 °C; (e) 800 °C; The misorientation distribution of extrusion rod at different extrusion temperatures: (b) 200 °C; (d) 450 °C; (f) 800 °C.
boundaries, that is, the twin and grain boundaries intersect. As shown in figure 10(c), when extrusion temperature increases to 800 °C, the proportion of the Σ9 grain boundaries decreases. According to Randel et al [5], the Σ3 grain boundary can hinder plastic flow and effectively inhibit grain growth. As shown in figures 10(b) and (c), it can be observed that the grain size of the extrusion rod at 450 °C is lower than the grain size of 800 °C. Therefore, the fine grains the extrusion rod can be acquired at medium extrusion temperature.

Figure 11 shows the recrystallized pole figures of high purity copper with different extrusion temperatures, as can be seen from figure 11(a), the recrystallized texture of the extruded high purity copper at 200 °C exhibits ⟨100⟩ parallel to the ED direction, and the texture intensity value is 21.47. When the extrusion temperature rises to 450 °C, the recrystallization texture presents ⟨111⟩ of the grains parallel to the ED direction. It can be seen from figure 11(b) that the distribution of recrystallization texture is relatively randomized in the extruded high purity copper at 450 °C, and the texture intensity is lower than that in the extruded high purity copper at 200 °C, and the texture intensity value is 3.41. This is because there are a large amount of twins in the extruded high purity copper at 450 °C, grains exhibit anisotropy, thus texture strength is weakened. Furthermore, according to related research [41], the ⟨111⟩ boundary plane has a low surface energy, which can hinder the migration of grain boundaries and inhibit grain growth. As shown in figure 11(c), the ⟨120⟩ and ⟨100⟩ textures were observed in the extruded high purity copper at 800 °C, and the texture intensity value is 5.14. The reason is that the recrystallized grains generate secondary recrystallization, and partial grains dominate the grain growth direction, thus the texture intensity value increases.

3.4. Effect of temperature on DRX behavior
Figure 12 shows a microstructure of the relationship between temperature and DRX. The red areas in the picture represent deformed grains, the blue areas represent recrystallized grains, and the yellow areas represent sub-grains. It can be observed from figure 12 that the DRX generated is inadequate at the time of low temperature extrusion, and almost all of the microstructure in the extrusion rod is composed of deformed grains. When the extrusion temperature raises to 450 °C, the deformed grain is almost fully translated into sub-grain and recrystallized grain. The reason is that elevating the temperature leads to increasing in the distortion energy required for DRX, which is favorable for the nucleation of DRX and increasing in the proportion of recrystallized grains. When the extrusion temperature raises from 450 °C to 800 °C, the proportion of sub-grain increases and the proportion of recrystallized grain decreases. The reason is that the recrystallized grains translate into sub-grain. In addition, figures 12(b) and (c) exhibit that the dynamic recrystallized grains grow obviously as increasing of the temperature. Because elevated temperature favors grain boundary migration, and large grains absorb small grains, which promotes grain growth.
Figure 13 shows the statistic map of recrystallization extent in extrusion rods with different extrusion temperatures. As shown in figure 13, when the extrusion temperature is 200 °C, the proportion of deformed grains in the extrusion rod is 86.6%, and the sum of the sub-grains and recrystallized grains is 13.4%. This indicates that the extent of DRX is very low at low extrusion temperature and the microstructure of the extrusion rod is mainly composed of deformed grains. As the extrusion temperature increases to 450 °C, the proportion of deformed grains decreases quickly to 0.3%, the proportion of sub-grain increases to 66.8%, and the proportion of recrystallized grains increases to 32.9%, which indicates that the deformed grains in the extruded rod has sufficiently generated DRX. Furthermore, there are a large number of sub-grains in the extrusion rod. These sub-grains are formed by the initial deformed grains or the dynamic recrystallized grains subjected the deformation. When the extrusion temperature increases to 800 °C, the proportion of deformed grains is less than 0.1%, the proportion of sub-grain increases to 71.5%, and the proportion of recrystallized grains decreases to 28.4%. The reason is that the partial dynamic recrystallized grains are translated into sub-grains. The transformation of this part of the recrystallized grain is due to the fact that the grains which have fully generated DRX are in the recovery stage.

3.5. Extrusion rod microstructure analysis using CA model

Figure 14 shows microstructures with a strain rate of 0.1 s⁻¹ under 550 °C, 600 °C, 650 °C, and 700 °C. It can be seen that DRX grains has been produced at 550 °C. When the temperature is above 550 °C, the recrystallized grains in the extruded rod are almost in the form of relatively straight equiaxed grains, with the presence of annealed twins. Most of the grains are refined. It is observed that the recrystallized grain size is related to the thermal deformation conditions. As increasing of the temperature, the recrystallized grains grow and grain
boundaries become more straight. Figure 15 shows the recrystallized grain microstructures at a strain rate of 0.1 s$^{-1}$ under different temperatures predicted by the CA model. Regions of different colors represent recrystallized grains of different orientations that are produced in newly formed regions. Compared with the microstructures shown in figure 15, the simulation results are well matched to the experiment.

The grain sizes predicted by the CA model are 17.6 μm, 22.76 μm, 26.13 μm, 28.2 μm, 35.53 μm, 39.59 μm, 47.27 μm, 52.52 μm, at temperatures of 450 °C, 500 °C, 550 °C, 600 °C, 650 °C, 700 °C, 750 °C, 800 °C, respectively. The simulation results are in good agreement with the experiment. As shown in figure 16, the relative error is −5%~5%, and the calculated values are very close to the experimental data, which proves that the model has a good prediction accuracy. After statistical calculation, the minimum error between the actual and numerical is 0.34%, the maximum error is 3.84%, and the average value is 1.53%. The simulation results are well matched with the experimental data. It shows the reliability and practical applicability of the theoretical

![Figure 12. Relationship between extrusion temperature and DRX: (a) 200 °C; (b) 450 °C; (c) 800 °C.](image)

![Figure 13. Statistic map of recrystallization extent in extrusion rods with different extrusion temperatures.](image)
model. In summary, the CA model in this study can accurately predict the evolution of microstructure in high-purity copper during extrusion.

4. Conclusions

In this research, the DRX behavior of high-purity copper under different temperatures (200 °C–800 °C) were investigated by numerical simulations and experiments. The simulation results were verified by experiment. By analyzing the microstructure in the deformation zone, the optimum extrusion temperature range is determined. The DRX texture and special grain boundary $\Sigma 3$ of high purity copper were studied by EBSD. Conclusions can be drawn as follows:

(1) As the extrusion temperature rises from 450 °C to 800 °C, the average grain sizes at different deformation level at the central axis of the deformation zone decrease at first and then increase, and finally become stable. As increasing of the extrusion temperature, the microstructure uniformity increases and the grain size increases. When the temperature is 500 °C–550 °C and the deformation level is 78%, the recrystallized grains are fine and the uniformity of the microstructure is high.

(2) When the extrusion temperature is 200 °C, there is not almost special grain boundary in the extruded rod. As the extrusion temperature increases, the special grain boundary $\Sigma 3$ in the extrusion rod increases rapidly and eventually stabilizes.

(3) At low extrusion temperature, the grains in the extrusion rod are mainly deformed grains. When the temperature raises to 450 °C, the deformed grains in the extruded rod are translated into dynamic recrystallized grains and sub-grains. When the temperature elevates to 800 °C, the proportion of sub-grains

![Figure 14. Microstructures of extrusion temperature with a strain rate of 0.1 s$^{-1}$ at: (a) $T = 550$ °C, (b) $T = 600$ °C, (c) $T = 650$ °C, and (d) $T = 700$ °C.](image)
Figure 15. The CA model simulated microstructures with a strain rate of 0.1 s\(^{-1}\): (a) \(T = 550\, ^\circ\text{C}\), (b) \(T = 600\, ^\circ\text{C}\), (c) \(T = 650\, ^\circ\text{C}\), and (d) \(T = 700\, ^\circ\text{C}\).

Figure 16. Comparison of calculated grain size and experimental grain size.
increases, and the proportion of recrystallized grains decreases. There are abnormally grown grains in the extrusion rod at 800 °C.

(4) The experimental results show that the CA model can well predict the DRX behavior and microstructure evolution during hot deformation for high-purity copper with a high prediction accuracy.

Acknowledgments

The authors express their appreciation for the financial support of the National Key Research and Development Program of China (2017YFB0305504) and National Natural Science Foundation of China under (51771043).

ORCID iDs

Chunyu Li https://orcid.org/0000-0001-5591-8554
Ke Hu https://orcid.org/0000-0002-5029-4444
Yonghui Jia https://orcid.org/0000-0002-7557-1794
Weiyang Zhou https://orcid.org/0000-0002-7216-7155

References

[1] Hu C-K and Harper J M E 1998 Copper interconnections and reliability Mater. Chem. Phys. 52 5–16
[2] Yu H et al 2019 High thermal stability and excellent mechanical properties of ultrafine-grained high-purity copper sheets subjected to asymmetric cryorolling Mater. Charact. 153 34–45
[3] Horky J et al 2013 Effect of microstructural stability on fatigue crack growth behaviour of nanostructured Cu Mech. Mater. 67 38–45
[4] Pavate V et al 1997 Correlation between aluminum alloy sputtering target metallurgical characteristics, arc initiation, and in-film defect intensity Microelectron Manuf. 42–7
[5] Randle Y and Coleman M 2009 A study of low-strain and medium-strain grain boundary engineering Acta Mater. 57 3410–21
[6] Wang H, Zaluzec M J and Rigsbee J M Microstructure and mechanical properties of sputter deposited Cu/CuTax Alloys Metall Mater. Trans. A 9 917–25
[7] Hu C-K et al 1995 Copper interconnection integration and reliability Thin Solid Films 262 84–92
[8] Pardis N et al 2015 Microstructure, texture and mechanical properties of cyclic expansion–extrusion deformed pure copper Mater. Sci. Eng. A 628 423–32
[9] Francy K A, Rao C S and Gopalakrishnaiyah P 2019 Optimization of direct extrusion process parameter on 16MnCr5 and AISI1010 using DEFORM-3D Procedia Manuf. 30 498–505
[10] Huang S, Shu D, Hu C and Zhu S 2016 Effect of strain rate and deformation temperature on strain hardening and softening behavior of pure copper Trans. Nonferrous Met. Soc. China 26 1044–54
[11] Cao Z et al 2019 Cellular automaton simulation of dynamic recrystallization behavior in V–10Cr–5Ti alloy under hot deformation conditions Trans. Nonferrous Met. Soc. China 29 98–111
[12] Xiao N, Hodgson P, Rolfe B and Li D 2018 Modelling discontinuous dynamic recrystallization using a quantitative multi-order-parameter phase-field method Comput. Mater. Sci. 155 298–311
[13] Steiner M A, McCabe R J, Garlea E and Agnew S R 2017 Monte Carlo modeling of recrystallization processes in α-uranium J. Nucl. Mater. 492 74–87
[14] Yazdipour N, Davies C H J and Hodgson P D 2008 Microstructural modeling of dynamic recrystallization using irregular cellular automata Comput. Mater. Sci. 44 566–76
[15] Zhang H et al 2019 Study of dynamic recrystallization behavior of T2 copper in hot working conditions by experiments and cellular automation method J. Alloys Compd. 784 1071–83
[16] Hallberg H, Wallin M and Ristimaa M 2010 Simulation of discontinuous dynamic recrystallization in pure Cu using a probabilistic cellular automaton Comput. Mater. Sci. 49 25–34
[17] Hu Y et al 2018 CA method with machine learning for simulating the grain and pore growth of aluminum alloys Comput. Mater. Sci. 142 244–54
[18] Zhang T, Lu S, Wu Y and Gong H 2017 Optimization of deformation parameters for dynamic recrystallization of 7055 aluminum alloy by cellular automaton Trans. Nonferrous Met. Soc. China 27 1327–37
[19] Chen M-S et al 2017 Modeling and simulation of dynamic recrystallization behavior for 42CrMo steel by an extended cellular automaton method Vacuum 146 132–51
[20] Zhi Y, Liu X and Yu H 2014 Cellular automaton simulation of hot deformation of TRIP steel Comput. Mater. Sci. 81 104–12
[21] Qian M and Guo Z X 2004 Cellular automata simulation of microstructural evolution during dynamic recrystallization of an HY-100 steel Mater. Sci. Eng. A 365 180–5
[22] Chen M-S, Lin Y C and Ma X-S 2012 The kinetics of dynamic recrystallization of 42CrMo steel Mater. Sci. Eng. A 556 260–6
[23] Li X et al 2017 Simulation of dynamic recrystallization in AZ80 magnesium alloy using cellular automaton Comput. Mater. Sci. 140 95–104
[24] Chen M-S, Yuan W-Q, Li H-B and Zou Z-H 2017 Modeling and simulation of dynamic recrystallization behaviors of magnesium alloy AZ31B using cellular automata method Comput. Mater. Sci. 136 163–72
[25] Xu W et al 2017 Study on the dynamic recrystallization behavior of Ti-55 titanium alloy during hot compression based on Cellular Automaton model method Procedia Eng 207 2119–24
[26] Liu Y-X et al 2015 Study of dynamic recrystallization in a Ni-based superalloy by experiments and cellular automaton model Mater. Sci. Eng. A 626 432–40
[27] Cai J et al 2011 Constitutive equations for elevated temperature flow stress of Ti–6Al–4V alloy considering the effect of strain Mater. Des. 32 1144–51
[28] Jun W 2009 The study of copper microstructure during continuous extrusion *Non-ferrous metals* 5 41–44
[29] Quelennec X, Martin E, Jiang L and Jonas J J 2010 Work hardening and kinetics of dynamic recrystallization in hot deformed austenite *J. Phys. Conf. Ser.* 240 012082
[30] Gourdet S and Montheillet F 2003 A model of continuous dynamic recrystallization *Acta Mater.* 51 2685–99
[31] Goetz R L and Seetharaman V 1998 Modeling dynamic recrystallization using cellular automata *Scr. Mater.* 38 405–13
[32] Ding R and Guo Z X 2001 Coupled quantitative simulation of microstructural evolution and plastic flow during dynamic recrystallization *Acta Mater.* 49 3163–75
[33] Roberts W and Ahlblom B 1978 A nucleation criterion for dynamic recrystallization during hot working *Acta Metall.* 26 801–13
[34] Huang Y and Humphreys F J 1999 Measurements of grain boundary mobility during recrystallization of a single-phase aluminium alloy *Acta Mater.* 47 2259–68
[35] Read W T and Shockley W 1950 Dislocation models of crystal grain boundaries *Phys. Rev.* 78 275–89
[36] Devadas C, Samarasekera I V and Hawbolt E B 1991 The thermal and metallurgical state of steel strip during hot rolling: III. Microstructural evolution *Metall. Trans. A* 22 335–49
[37] Mirzadeh H and Najafiizadeh A 2010 Prediction of the critical conditions for initiation of dynamic recrystallization *Mater. Des.* 31 1174–9
[38] Quan G-Z et al 2011 Constitutive modeling for the dynamic recrystallization evolution of AZ80 magnesium alloy based on stress–strain data *Mater. Sci. Eng. A* 528 8051–9
[39] Derby B 1991 The dependence of grain size on stress during dynamic recrystallisation *Acta Metall. Mater.* 39 955–62
[40] Watanabe H et al 2001 Grain size control of commercial wrought Mg-Al-Zn alloys utilizing dynamic recrystallization *Mater Trans* 42 1200–5
[41] Randle V et al 2008 Five-parameter grain boundary distribution of commercially grain boundary engineered nickel and copper *Acta Mater.* 56 2363–73