Accelerated recrystallization by electric current flash heating in cold-rolled Al-5Cu alloy under the influence of concurrent precipitation

Shubo Wang[a], Hailong Jia[a], Yanjun Li[a,*]

[a] *Department of Materials Science and Engineering, Norwegian University of Science and Technology (NTNU), N-7491 Trondheim, Norway*

* *Corresponding author. E-mail address: yanjun.li@ntnu.no.*

ABSTRACT

The influence of flash heating by using continuous direct current on the recrystallization behavior of an Al-5Cu alloy under the influence of concurrent precipitation of Al_2Cu precipitates has been investigated. In comparison to isothermal annealing in salt bath, recrystallization kinetics of the Al-5Cu alloy is greatly accelerated by direct current flash heating. Full recrystallization can be achieved within a much shorter time during direct current flash heating than that by salt bath isothermal annealing. As a result, the grain structure is much finer. By excluding the heating rate effect, it is shown that the accelerated nucleation kinetics during recrystallization is due to the athermal effect of the applied current.

Keywords:

Aluminum alloys; Electric current; Recrystallization; Microstructures; Precipitation

1. Introduction

Heat treatment is a crucial process to optimize the performance and mechanical properties of metals and their final products. Traditionally, the most used equipment for heat treatments of metals is radiative furnace or gas burning furnace. In some cases, pre-heated salt bathes are also used to take advantage of the uniform temperature and relatively high heating rate (typically in the order of magnitude of 10^2 °C /s) [1]. Alternatively, heating can also be generated when an electric current passes through a metal part, which is known as Joule heating or resistivity heating.

It has been known for decades that the application of external electric current or electric field can enhance dynamic recovery leading to improved plasticity of metals during forming [2, 3], and affect the solid state phase transformations in metals during heat treatment, such as formation of intermetallic compounds, precipitation, crystallization and recrystallizations [4-6]. Specifically, Conrad et al. [7-9] showed that high density (800 A/mm^2, 90 μs duration) direct current pulses with a frequency of 2 Hz could enhance the rates of recovery and recrystallization of cold-drown Cu. Xu et al. showed that both low density direct current and alternative current (≤ 10 A/mm^2) could promote the nucleation kinetics of recrystallization of cold worked Ti [10]. A grain refinement effect was seen at lower annealing temperatures [7], whilst enhanced grain growth was observed at higher annealing temperatures [9, 10]. In these cases, the applied electric current was coupled with conventional furnace heating, where the Joule heating effect could be neglected. It was suggested that the interaction between dislocations and drifting electrons played an important role during the process.

Electric current has also been applied to heat treat samples by exploiting the thermal Joule heating effect. Accelerated recrystallization has been observed in as deformed Cu samples that annealed by applied currents, for example by high density electric current pulses (140-280 A/mm^2) for ~ 10^{-4} s [11], by high frequency (200-600 Hz) electropulsing (74-100 A/mm^2) for 9 s [12] and low density direct current (5.3-6.2 A/mm^2) at 300 °C for minutes [13]. Recent studies showed that similar acceleration effect on recrystallization was found in hot rolled AA2024 Al alloy using alternating current (50 Hz, 200 A/mm^2) for 240 ms [14], cold rolled AA6061 alloy using high frequency (600-800 Hz) electropulsing (32-36 A/mm^2) for 15 s [15] and cold rolled AA3xxx alloy using alternative current (50 Hz, 33 A/mm^2) for 1,500 s [16]. Some researchers have proposed that the acceleration effect of current on recrystallization was mainly due to the thermal Joule effect, *i.e.* local heating effect and high heating rate [11]. However, it has been accepted by more researchers that the effect of electric current on promoting dislocation motion plays a more critical role[12-16].

So far, most investigations on the effects of electric current have been focused on the recrystallization of single phase aluminum alloys. However, most commercial Al alloys contain a large amount of alloying elements and impurities in supersaturated solid solution. During the recrystallization process of cold deformed aluminum alloys, precipitation might occur (so called concurrent precipitation) and interact with recrystallization. Precipitation of fine dispersed precipitates occurring prior to or concurrently with recrystallization can induce a strong Zener drag effect, retarding the nucleation of recrystallization [17-19]. Therefore, it is of interest to investigate the influence of electric current heating on the recrystallization behavior of cold rolled aluminium alloys under the effect of concurrent precipitation of precipitates.

It is well known that high density of θ' and θ-Al_2Cu precipitates will precipitate in deformed Al-Cu alloys during heat treatments [20]. In this study, a flash heating of a cold rolled binary Al-5Cu alloy by using a moderate density direct current has been done. The recrystallization behavior under the influence of electric current and concurrent precipitation have been studied.

2. Experimental

The experimental material used in this study is a binary Al-5 wt.% Cu alloy prepared by melting and casting. The as-cast Al-5Cu alloy slabs were homogenized at 540 °C for 20 hours to dissolve most of the eutectic Al_2Cu particles into the solid solution. Afterwards, the as-homogenized slabs with an initial thickness of 20 mm were cold rolled at room temperature to a final thickness of 1 mm (95% thickness reduction), which corresponds to a logarithmic true strain of $\varepsilon = 3.0$. The as-rolled sheet was machined into dog-bone shape samples with a cross section of 4.6 (± 0.1) × 1.0 (mm^2) , and then subjected to direct current flash heating with a constant current density of ~ 65 A/mm^2. Direct current was performed by a power supply with program controllable current/voltage output and duration. The grip sections of each sample were screwed to copper electrodes to assure a good electric contact, as schematically described in Fig. 1. It was not practical to attache a thermocouple directly to the tested sample because of the associated disturbance from the current [21]. Therefore, temperature evolution of the sample surface was measured by a non-contact infrared (IR) temperature sensor. Due to the low and complex emissivity of Al alloys, a thin layer of black high temperature paint was coated on the smooth surface of the samples to improve the accuracy of measurement by IR sensor. The measured temperature by the IR sensor was calibrated by using measurements recorded by a K type thermocouple attached to the samples that heated in an air circulation furnace. During the electric current flash heating experiments, the recorded temperature by IR sensor was regarded as bulk temperature because of the thin thickness of the sample. The samples were flash heated to different peak temperatures by applying different current duration, followed by air cooling. The electric current direction is parallel to the rolling direction (RD) of the samples. As a comparision, the as-rolled samples were also subjected to isothermal annealing in a salt bath for different durations at the same temperature as the peak temperature of electric current flash heating, then cooled in air.

Vickers hardness was measured using a 1kgf loading and 15 s dwelling time. The resulting hardness was based on an average of at least 5 readings on the polished RD-ND cross section (ND: normal direction). Metallographic examination of the RD-ND cross section was performed using optical microscopy (OM), scannning electron microscopy (SEM) in backscattered electron (BSE) mode and electron backscattered diffraction (EBSD). The samples for OM study were first mechanically polished down to 1 μm and then anodized using Barker's reagent. For BSE and EBSD study, the mechanically polished samples were further electropolished using a Struer A2 electrolyte at -25 °C. EBSD patterns were acquired by a Hitachi SU6600 FEGSEM equipped with a Nordif EBSD detector. The EBSD data were indexed and analyzed by the commercial OIM-TSL® software. All the samples for OM, SEM and EBSD characterization were examined in the center of the RD-ND cross section.

3. Results and discussion

The microstructure of the as-cast and as-homogenized Al-5Cu alloy has been studied in a previous work [22]. After homogenization treatment, most of the coarse skeletal eutectic Al_2Cu particles were dissolved into the Al matrix, while a small fraction of Al_2Cu particles remains in the bulk. After cold rolling to a thickness reduction of 95%, as can be seen from Fig. 2a, the coarse grains in the as-homogenized Al-5Cu alloy have been deformed into a fiber structure which is parallel to the RD. The Al_2Cu intermetallic particles have been broken into smaller pieces with a more globular shape and are aligned as strings parallel to the RD (Fig. 2b). It should be noted that no obvious precipitation can be found after several hours' storage at room temperature, as shown in Fig. 2c.

Fig. 3a shows the typical temperature evolution of the sample during electric current flash heating. After reaching the peak temperature, the sample cools down immediately in air. The instant heating rate (inset of Fig. 3a) was calculated by taking a derivative of the recorded temperature profile. As can be seen, the heating rate increases sharply in the first second, which is corresponding to the ramping of the input current (green dashed line). After the current reaches the desired value, the heating rate gradually increases with time until a maximum of 142 °C/s is achieved. In addition to the non-perfect square shaped current profiles, this heating rate variation during heating should also be attributed to the electric resistivity of the samples which is temperature and solid solution level dependent [23-25]. In the end of heating cycle, the heating rate sharply drops to a minus value before the current reduces to zero. The average heating rate during the whole electric current flash heating cycle is calculated as ~85 °C/s.

Fig. 3b shows the Vickers hardness of Al-5Cu samples after electric current flash heating to different peak temperatures. The hardness of the as-cast (55 Hv) and as-homogenized (77 Hv) sample are also shown as references. This increasing hardness (~ 22 Hv), after homogenization, is due to the solid solution strengthening of Cu solutes [26]. After a 95% thickness reduction, the sharp hardness increase to 174 Hv is mainly due to the storage of dislocations [22]. After electric current flash annealing, hardness of the samples decreases to 75 Hv with increasing peak temperature until 407 °C (corresponding to 4.5 s duration of current). With further increase of the peak temperature to 520 °C, the hardness does not decrease further implying a full recrystallization has already been achieved by flashing heating to a peak temperature of 407 °C.

Fig. 4a shows the EBSD grain structure of the sample after 4 s electric current flash heating to a peak temperature of 385 °C. Strain free grains surrounded by high angle grain boundaries (HABs) can be scarcely seen. Most of the grains still keep a fiber character. Within those long coarse grains, a large fraction of low angle boundaries (LABs) can be observed. It indicates that only recovery instead of recrystallization has occurred in the alloy, which is consistent with the hardness curve shown in Fig. 3. Fig. 4b presents the BSE-SEM image of the sample. A high density of Al_2Cu precipitates have formed in the alloy, most of which are aligned with the former deformation band boundaries, showing the occurrence of the concurrent precipitation during flash heating.

Fig. 4c shows the EBSD grain structure after 4.5 s electric current flash heating to a peak temperature of 407 °C. In agreement with the hardness measurement, a full recrystallization has happened. However, most of the recrystallized grains show elongated shape, which is a typical recrystallization structure under the influence of concurrent precipitation [17-19, 27]. The growth of the recrystallized grains into the ND direction are greatly constrained by the fine precipitates aligned along the deformation band boundaries. Fig. 4d shows the concurrent precipitation of Al_2Cu precipitates, which are slightly coarser than that shown in Fig. 4b. By the grain structures of Fig. 4a and Fig. 4c, it can be concluded that the recrystallization of the alloy happens in between 385 °C and 407 °C within 0.5 s.

To illustrate the effect of electrical current on the recrystallization process, comparison experiments of isothermal annealing in a salt bath at 407 °C were conducted. The average heating rate of Al sheets of 1 mm in thickness after immersing into a stirring salt bath of ~ 400 °C has ever been measured to be about 250 °C/s [28]. This heating rate is basically much higher than that by flash heating using a 65 A/mm^2 direct current in this study. As shown in Fig. 5, the hardness value decreases sharply after only 1 s annealing, and then gradually decreases with further increasing annealing time until 60 s, where a similar hardness value (58 Hv) as the as-cast one is achieved.

Fig. 6a shows the EBSD grain structures after 3.5 s isothermal annealing in a salt bath at 407 °C. As can be seen, only partial recrystallization has occurred. Most of the patterns of the retained deformed structures in the EBSD map cannot be properly indexed or show very low confidence index (CI) value which is less than 0.1. The recrystallization fraction is estimated to be ~ 39%. Fig. 6c shows the grain structure after 4.5 s isothermal annealing at 407 °C. Full recrystallization has not yet been realized, where the recrystallization fraction is measured to be ~ 72%. Fig. 6b shows that after 3.5 s annealing a high density of Al_2Cu particles have precipitated out in the alloy. Further coarsening of particles with longer annealing time for 4.5 s can be seen in Fig. 6d.

A comparison between the BSE-SEM images of Al_2Cu precipitates in the electric current flash heating treated samples and those salt bath isothermally annealed shows that the precipitates in the former sample are finer and with relatively higher number density, especially in the samples annealed for 4.5 s (Fig. 4d *vs.* Fig. 6d). Considering the faster heating rate of sample during up-quenching into the salt bath (250 °C/s) than the electric current flash heating (85 °C/s), this can be attributed to the longer soaking time at high temperature regime in the former heat treatment, which causes the coarsening of precipitates and reduces the number of precipitates.

The highly dispersed and finer precipitates in the electric current flash heating treated sample shown in Fig. 4d are supposed to have stronger retarding effect on the nucleation of recrystallization. However, a much finer recrystallized grain structure was achieved in the flash heating treated sample (Fig. 4c) than those salt bath isothermally annealed (Fig. 6a and c). Meanwhile, a complete recrystallization was achieved in the Al-5Cu alloy by 4.5 s electric current flash heating to 407 °C (Fig. 4c), while the sample up-quenched into 407 °C salt bath for 4.5 s only shows partial recrystallization (Fig. 6c). These results demonstrate that continuous electric current flash heating has strongly accelerated the nucleation kinetics of recrystallization and grain growth of recrystallized grains.

So far different mechanisms, *e.g.* Joule heating thermal effect, in terms of higher heating rate [11] and athermal effect by drifting electrons [7-9], have been proposed to explain the acceleration effect of electric current on recrystallization. In the present study, as the heating rate of salt bath isothermal annealing is faster than that of the direct current flash heating, the heating rate effect caused by electric current can be excluded. This result is consistent with the result of Huang et al., which shows that at exactly the same heating rate the recrystallization process of cold rolled aluminium alloys during electric current heating is faster than that with furnace heating [16]. Thus, the accelerated recrystallization in the present Al-5Cu alloy should also be due to the athermal effect. This athermal effect may be attributed to the enhanced vacancy diffusion flux, J, by the applied electric current [29]:

$$J = J_t + J_e = -D\frac{\partial c}{\partial x} + \frac{Dc}{kT}Z^* e\rho j_e \tag{1}$$

where c is the vacancy concentration, D is the diffusivities, k is the Boltzmann constant, T is the absolute temperature, e is the charge on an electron, ρ is the resistivity, j is the electrical current density, Z^* is the effective charge. The J_t, J_e terms represent the vacancy flux related to thermal effect and electric current, respectively. For J_e term, $Z^* e\rho j_e$ represents the magnitude of the electron wind force [4]. This force is mostly attributed to the momentum transfer of electrons in the defective areas containing dislocations [30, 31]. These areas also provide numerous diffusion paths to improve the diffusion rate. This implies that the vacancy diffusion flux in the as-deformed materials can be enhanced tremendously by the applied continuous current.

Dislocation gliding and climbing, which are proportional to the vacancy diffusion flux [32], play an indispensable role in the nucleation process of recrystallization [33]. Thus, the enhanced vacancy flux indicated by Eq. (1) facilitates dislocation climbing, which is essential for the formation of subgrains surrounded by LABs (Fig. 4a) and the subsequent migration of grain boundaries and formation of HABs. As a result, dislocation-free grains above critical size surrounded mainly by HABs, namely nuclei of recrystallized grains, can be easily formed. This research work shows that even under the strong pinning effect by the fine particles concurrently precipitated during annealing process, the athermal effect is strong enough to accelerate the recrystallization kinetics of the alloy.

4. Conclusions

In summary, we have shown that electric current flash heating can strongly enhance the recrystallization kinetics of cold-deformed aluminium alloys even under the strong retarding force of concurrent precipitation. A fully recrystallized grain structure of Al-5Cu alloy are obtained by electric current flash heating for only 4.5 s to a peak temperature of 407 °C. However, only partial recrystallization can be achieved by isothermal salt bath annealing at the same peak temperature for the same time. It is suggested that the accelerated recrystallization kinetics is due to the enhanced vacancy diffusion flux by the electric current in the deformed material, which stimulates the dislocation climbing and therefore the nucleation of recrystallization grains.

Acknowledgment

SW acknowledges financial support from NTNU for the PhD project. The authors would like to thank Dr. Yijiang Xu for providing the as-cast Al-5Cu alloys.

References

[1] ASM Handbook. Volume 4: Heat Treating, 2016.

[2] H. Conrad, Electroplasticity in metals and ceramics, Mater. Sci. Eng. A 287(2) (2000) p. 276-287.

[3] B.J. Ruszkiewicz, T. Grimm, I. Ragai, L. Mears, J.T. Roth, A Review of Electrically-Assisted Manufacturing With Emphasis on Modeling and Understanding of the Electroplastic Effect, J Manuf. Sci. Eng. 139(11) (2017) p. 110801-110801-110815.

[4] H. Conrad, Effects of electric current on solid state phase transformations in metals, Mater. Sci. Eng. A 287(2) (2000) p. 227-237.

[5] J.D. Guo, X.L. Wang, W.B. Dai, Microstructure evolution in metals induced by high density electric current pulses, Mater. Sci. Technol. 31(13) (2015) p. 1545-1554.

[6] C.-L. Liang, K.-L. Lin, The microstructure and property variations of metals induced by electric current treatment: A review, Mater. Charact. 145 (2018) p. 545-555.

[7] H. Conrad, N. Karam, S. Mannan, Effect of prior cold work on the influence of electric current pulses on the recrystallization of copper, Scripta Metall. 18(3) (1984) p. 275-280.

[8] H. Conrad, N. Karam, S. Mannan, A.F. Sprecher, Effect of electric current pulses on the recrystallization kinetics of copper, Scripta Metall. 22(2) (1988) p. 235-238.

[9] H. Conrad, N. Karam, S. Mannan, Effect of electric current pulses on the recrystallization of copper, Scripta Metall. 17(3) (1983) p. 411-416.

[10] Z.S. Xu, Y.X. Chen, Effect of electric current on the recrystallization behavior of cold worked α-Ti, Scripta Metall. 22(2) (1988) p. 187-190.

[11] T. Konkova, I. Valeev, S. Mironov, A. Korznikov, M. Myshlyaev, S.L. Semiatin, Effect of electric-current pulses on grain-structure evolution in cryogenically rolled copper, J. Mater. Res. 29(22) (2014) p. 2727-2737.

[12] R.F. Zhu, J.N. Liu, G.Y. Tang, S.Q. Shi, M.W. Fu, Properties, microstructure and texture evolution of cold rolled Cu strips under electropulsing treatment, J. Alloys Compd. 544(Supplement C) (2012) p. 203-208.

[13] D. Fabrègue, B. Mouawad, C.R. Hutchinson, Enhanced recovery and recrystallization of metals due to an applied current, Scr. Mater. 92 (2014) p. 3-6.

[14] X. Xu, Y. Zhao, B. Ma, J. Zhang, M. Zhang, Rapid grain refinement of 2024 Al alloy through recrystallization induced by electropulsing, Mater. Sci. Eng. A 612(Supplement C) (2014) p. 223-226.

[15] Y.S. Zheng, G.Y. Tang, J. Kuang, X.P. Zheng, Effect of electropulse on solid solution treatment of 6061 aluminum alloy, J. Alloys Compd. 615(Supplement C) (2014) p. 849-853.

[16] K. Huang, C. Cayron, R.E. Logé, The surprising influence of continuous alternating electric current on recrystallization behaviour of a cold-rolled Aluminium alloy, Mater. Charact. 129 (2017) p. 121-126.

[17] K. Huang, K. Zhang, K. Marthinsen, R.E. Logé, Controlling grain structure and texture in Al-Mn from the competition between precipitation and recrystallization, Acta Mater. 141(Supplement C) (2017) p. 360-373.

[18] Q. Zhao, K. Huang, Y. Li, K. Marthinsen, Orientation Preference of Recrystallization in Supersaturated Aluminum Alloys Influenced by Concurrent Precipitation, Metallurgical and Materials Transactions A 47(3) (2016) p. 1378-1388.

[19] J.G. Morris, W.C. Liu, Al alloys: The influence of concurrent precipitation on recrystallization behavior, kinetics, and texture, JOM 57(11) (2005) p. 44-47.

[20] H. Jia, R. Bjørge, L. Cao, H. Song, K. Marthinsen, Y. Li, Quantifying the grain boundary segregation strengthening induced by post-ECAP aging in an Al-5Cu alloy, Acta Mater. 155 (2018) p. 199-213.

[21] M.E. Davenport, P.M. Magee, G. Leppert, THERMOCOUPLE ATTACHMENT TO A DIRECT-CURRENT HEATER, J. Heat Transfer (1962) p. Medium: X; Size: Pages: 187-188.

[22] H. Jia, R. Bjørge, K. Marthinsen, Y. Li, The deformation and work hardening behaviour of a SPD processed Al-5Cu alloy, J. Alloys Compd. 697(Supplement C) (2017) p. 239-248.

[23] A. Matthiessen, C. Vogt, On the influence of temperature on the electric conducting-power of alloys, Philos. Trans. R. Soc. London 154 (1864) p. 167-200.

[24] M. Kaveh, N. Wiser, Impurity and temperature dependence of the electrical resistivity of dilute alloys of aluminum, Physical Review B 21(6) (1980) p. 2291-2308.

[25] T.S. Orlova, A.M. Mavlyutov, A.S. Bondarenko, I.A. Kasatkin, M.Y. Murashkin, R.Z. Valiev, Influence of grain boundary state on electrical resistivity of ultrafine grained aluminium, Philos. Mag. 96(23) (2016) p. 2429-2444.

[26] G.P.M. Leyson, W.A. Curtin, L.G. Hector Jr, C.F. Woodward, Quantitative prediction of solute strengthening in aluminium alloys, Nat. Mater. 9 (2010) p. 750.

[27] K. Huang, K. Marthinsen, The effect of heating rate on the softening behaviour of a deformed Al–Mn alloy with strong and weak concurrent precipitation, Mater. Charact. 110 (2015) p. 215-221.

[28] T. Furu, R. Ørsund, E. Nes, Subgrain growth in heavily deformed aluminium—experimental investigation and modelling treatment, Acta Metall. Mater. 43(6) (1995) p. 2209-2232.

[29] D.G. Pierce, P.G. Brusius, Electromigration: A review, Microelectron. Reliab. 37(7) (1997) p. 1053-1072.

[30] C.M. Tan, A. Roy, Electromigration in ULSI interconnects, Materials Science and Engineering: R: Reports 58(1) (2007) p. 1-75.

[31] C. Basaran, M. Lin, Damage mechanics of electromigration induced failure, Mechanics of Materials 40(1) (2008) p. 66-79.

[32] P.M. Anderson, J.P. Hirth, J. Lothe, Theory of Dislocations, Cambridge University Press 2017.

[33] J. Humphreys, G.S. Rohrer, A. Rollett, Recrystallization and Related Annealing Phenomena (Third Edition), Elsevier, Oxford, 2017.

Figure captions

Fig. 1

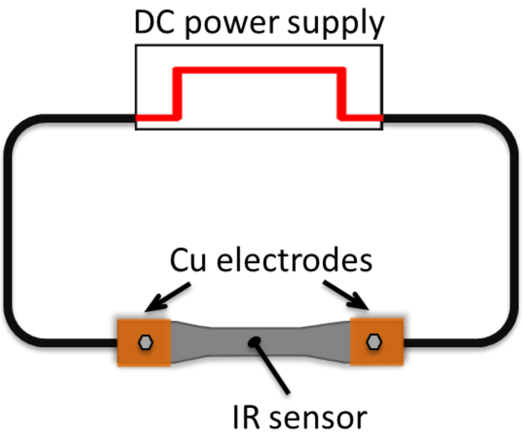

Fig. 1 Schematic description of the experimental set-up.

Fig. 2

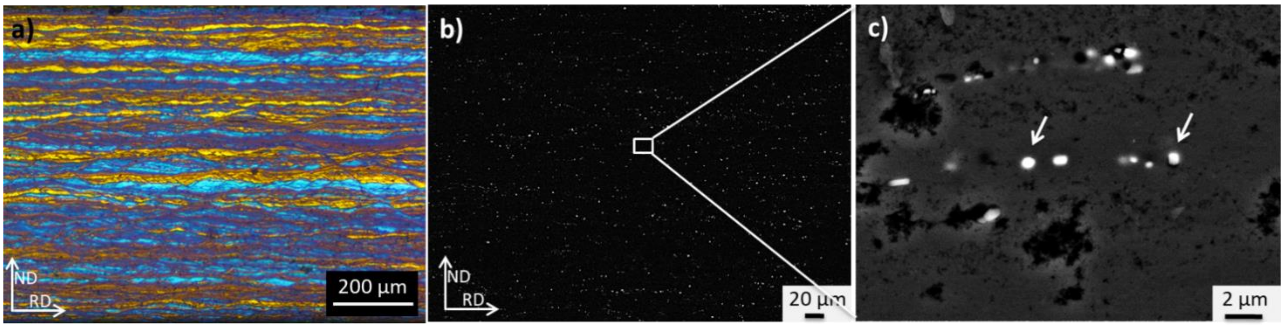

Fig. 2 (a) Optical micrograph of the as-rolled Al-5Cu alloy. (b) BSE-SEM image showing the distribution of primary Al_2Cu *particles after cold rolling. (c) Enlarged image of the microstructure within the white rectangle in (b) showing no formation of precipitates after cold rolling and room temperature storage.*

Fig. 3

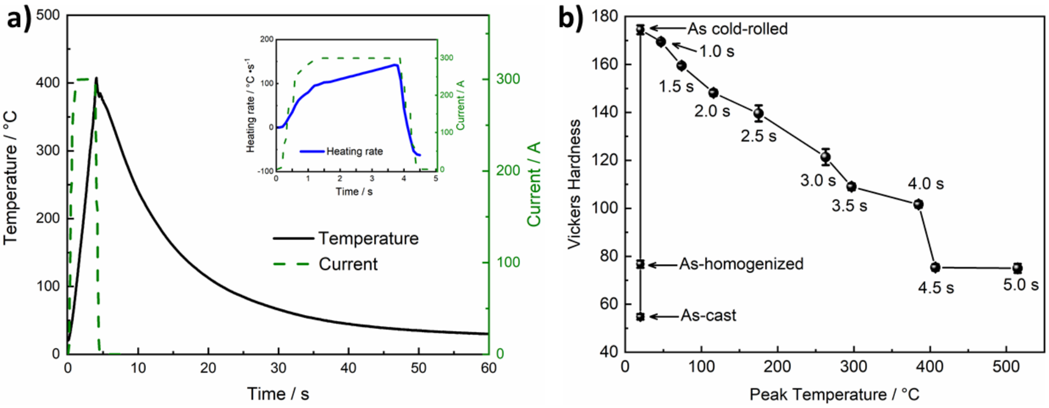

Fig. 3 (a) Typical temperature evolution and corresponding input electric current of the Al-5Cu alloy during 4.5 s electric current flash heating; the insert shows the heating rate (solid blue line) evolution during the heating cycle. (b) Vickers hardness evolution of the as-rolled Al-5Cu alloy after electric current flash heating to different peak temperatures; corresponding duration of current applied to reach the peak temperatures are also indicated in the figure.

Fig. 4

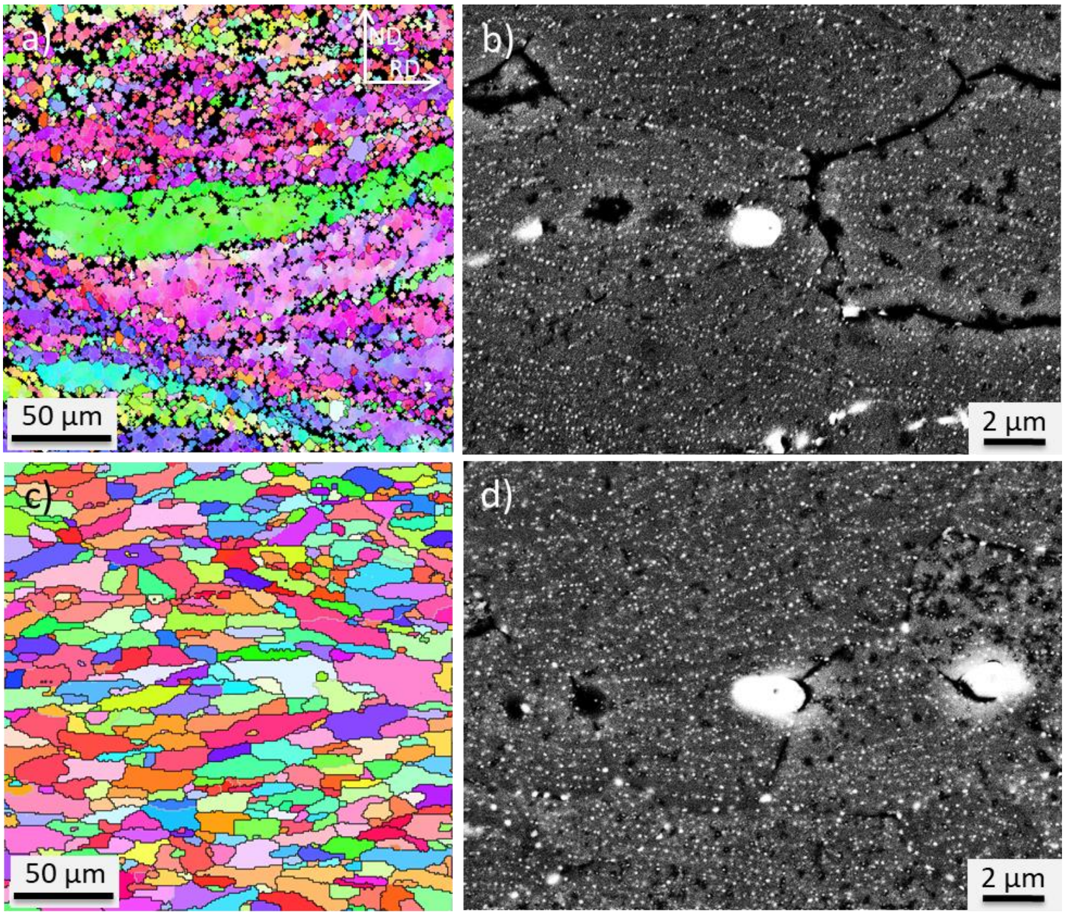

Fig. 4 EBSD orientation maps and corresponding BSE-SEM images of the Al-5Cu alloy after a 65 A/mm² direct current flash heating for 4 s to a peak temperature of 385 °C (a and b) and for 4.5 seconds to a peak temperature of 407 °C (c and d), respectively. In the EBSD orientation maps, orientation pattern of points has a CI (confidence index) value lower than 0.1 were shown in black; high angle boundaries (HABs) with misorientation angles >15° are shown as solid black lines, while low angle boundaries (LABs) with misorientation angles between 2° and 15° are shown as solid grey lines.

Fig. 5

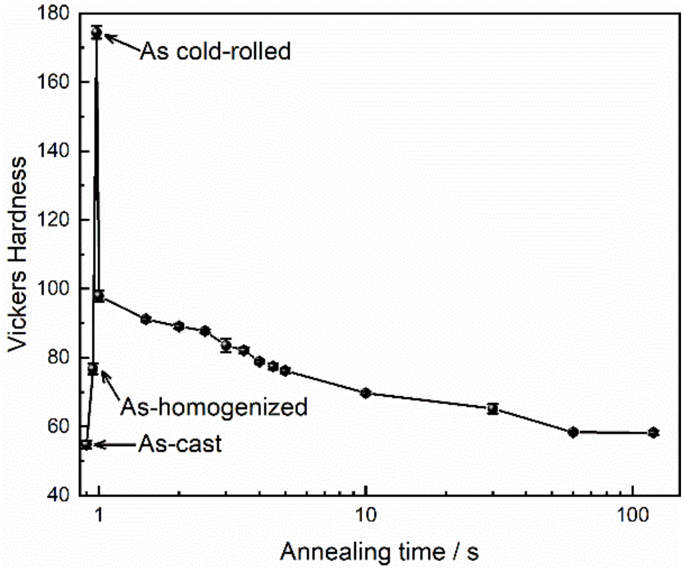

Fig. 5 Vickers hardness evolution of the Al-5Cu alloy isothermally annealed in a 407 °C salt bath.

Fig. 6

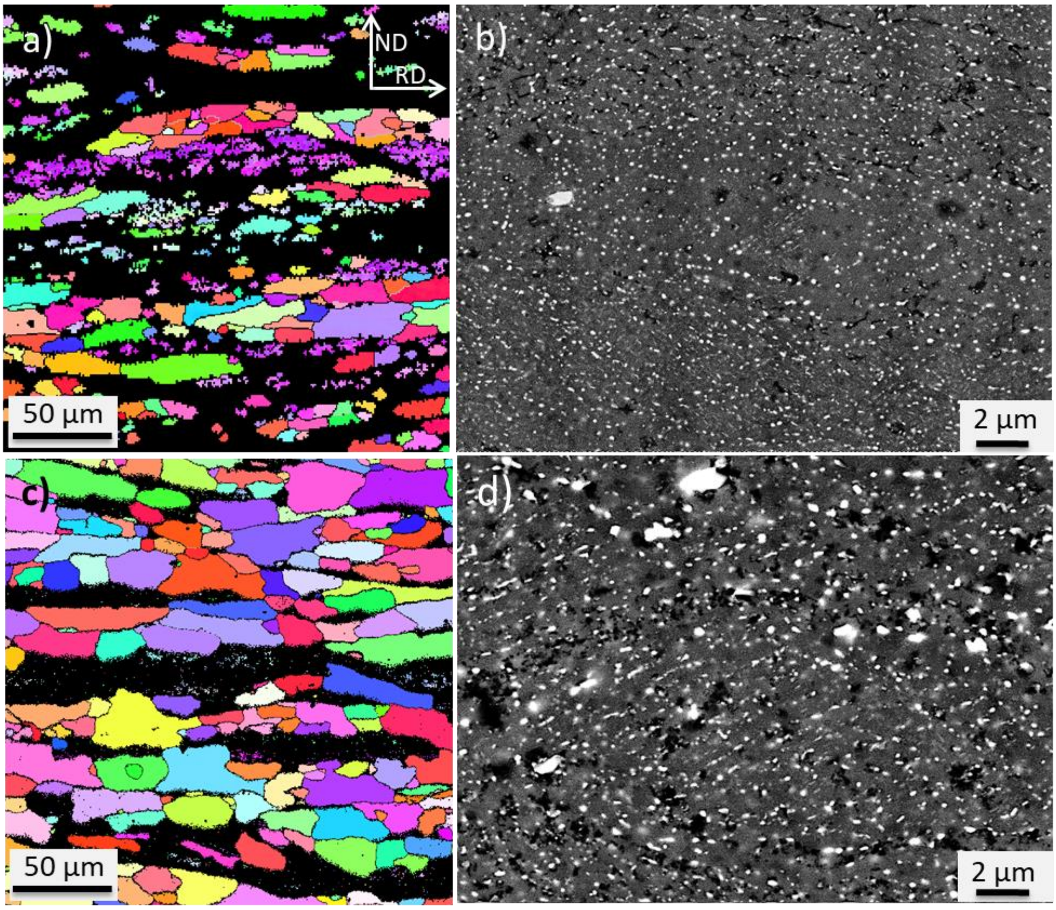

Fig. 6 EBSD orientation maps and BSE-SEM images of the Al-5Cu alloy isothermally annealed in a 407 °C salt bath for 3.5 s (a and b), and for 4.5 s (c and d), respectively. In the EBSD orientation maps, orientation pattern of points has a CI (confidence index) value lower than 0.1 were shown in black; high angle boundaries (HABs) with misorientation angles >15° are shown as solid black lines, while low angle boundaries (LABs) with misorientation angles between 2° and 15° are shown as solid grey lines.