STRUCTURE TRANSFORMATIONS DURING ANNEALING OF TWIN-ROLL CAST Al-Fe-Mn-Si (AA 8006) ALLOY SHEETS II. EFFECT OF HOMOGENIZATION AND HEATING RATE

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The kinetics and temperature range of structure transformations during annealing of twin-roll cast AlFeMnSi (AA 8006) alloy sheets were studied using electrical resistometry, light metallography and transmission electron microscopy. The effects of homogenization and heating rate were investigated, as well. Recrystallization response curves were determined by Vickers microhardness measurements. It was found that recrystallization starts and is completed at much lower temperatures in the homogenized material than in the non-homogenized one. Coarse grained heterogeneous structure is formed in the non-homogenized material, while fine grain homogeneous structure is typical for the homogenized alloy. The heating rate of the final annealing affects the temperature range of structure transformations and the resulting strip grain structure. The grains are more homogeneous in quickly heated samples, particularly in the homogenized material.

Key words: Al-Fe-Mn-Si alloys, twin-roll casting, phase transformations, electrical resistometry, recrystallization

TRANSFORMACE MIKROSTRUKTURY PŘI ŽÍHÁNÍ PLYNULE LITÝCH PÁSŮ ZE SLITINY Al-Fe-Mn-Si (AA 8006) II. VLIV HOMOGENIZACE A RYCHLOSTI OHŘEVU

Vliv homogenizace a rychlosti ohřevu na kinetiku a teplotní interval fázových transformací ve slitině AlFeMnSi (AA 8006) byly studovány pomocí elektrické rezistometrie,

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světelné metalografie a transmisní elektronové mikroskopie. Rekrystalizační křivky byly stanoveny pomocí měření mikrotvrdosti HV0,1. U homogenizované slitiny je počátek i konec rekrystalizace posunut k podstatně nižším teplotám než u nehomogenizovaného materiálu. Zrna v nehomogenizované slitině jsou hrubá, zatímco pro homogenizovaný materiál je typická rovnoměrná jemnozrnná struktura. Rychlost ohřevu finálního žíhání ovlivňuje teplotní rozmezí vzniku precipitátu a výslednou strukturu zrn. V materiálech podrobených rychlému ohřevu jsou zrna rovnoměrnější, zvláště pak u homogenizovaného materiálu.

1. Introduction

Thin sheets of AlFeMnSi (AA 8006) alloy prepared by continuous twin-roll casting (TRC) offer several economical and metallurgical advantages over conventionally produced DC-cast materials [1, 2, 3]. The microstructure of TRC AlFeMnSi final gauge products involves finely dispersed intermetallic compounds and a very fine grain size, resulting in a good combination of strength and ductility. However, as TRC AlFeSiMn alloys were developed recently, their response to heat treatment was investigated only a little [4–6]. The downstream processing of AlFeMnSi (AA 8006) twin-roll cast (TRC) sheets involves cold rolling to a thickness from 0.10 to 0.30 mm and final annealing to obtain quarter-hard (H22) or soft (O) tempers. Our recent results [7] indicated that long-term homogenization treatment prior to cold rolling substantially contributes to the improvement of final gauge strip properties.

This paper reports the impact of the homogenization treatment on structure transformations during annealing of the final gauge material. The effects of cold rolling reduction and annealing time on the structure transformations of non--homogenized material were reported in the preceding paper [8].

2. Experimental details

The investigated material was cut from a commercial alloy AA 8006 twin-roll cast strip 8.5 mm thick. The composition of the alloy in wt.% was: 1.5 Fe, 0.4 Mn, 0.16 Si, other elements < 0.015, balance Al. The microstructure of the as-cast alloy was described in [8]. Two materials with the same final cold reduction were prepared and heat treated at final thickness in the same manner:

1. <u>Non-homogenized material</u>: an as-cast strip was cold rolled on a laboratory mill to final thickness of 0.3 mm (96.4% reduction, true strain $\varepsilon = 3.9$).

2. <u>Homogenized material</u>: the same as-cast strip was processed in industrial conditions by cold rolling to 5.6 mm thickness, then it underwent a homogenization annealing at 580 °C and cold rolling to thickness of 0.18 mm (96.8% reduction, true strain $\varepsilon = 4.0$).

Both samples were subjected to linear heating in the range from 20 to 605 °C at a rate of 1 °C/min. Phase transformations were detected by the evolution of electrical resistivity (for more details see [8]). Microstructure examinations of samples quenched from several temperatures were carried out. The recrystallization response curves of cold-rolled materials were obtained by the measurement of Vickers microhardness HV0.1 after 0.5 h annealing at temperatures in the range from 220 to 510 °C. The size and the number of particles were measured using light microscope linked with an image analysis system. Grain structure in the long transverse strip plane was examined in polarised light on samples anodised in Barker's solution. The precipitates, intermetallic phases and matrix substructure were observed at 200 kV using transmission electron microscope (TEM) JEOL JEM 2000 FX equipped with an X-ray energy dispersive spectrometer (XEDS) LINK AN 10 000. Thin foils were prepared parallel to the rolling plane of the sheet.

3. Results

3.1 Microstructure evolution during linear heating

The ratio of resistivity (RRR) values of the initial non-homogenized and homogenized material are 2.3 and 4.8, respectively. This indicates that the concentration of alloying elements in the solid solution of the homogenized sample is significantly lower than in the non-homogenized one. As cold rolling does not affect matrix solute concentration, this difference is preserved in the cold-rolled thin sheets and influences the annealing response.

The evolution of resistance with temperature $R = f(T)$ and the normalised differential curves $(\mathrm{d}R/\mathrm{d}T)/R = f(T)$ of both materials are plotted in Fig. 1. The

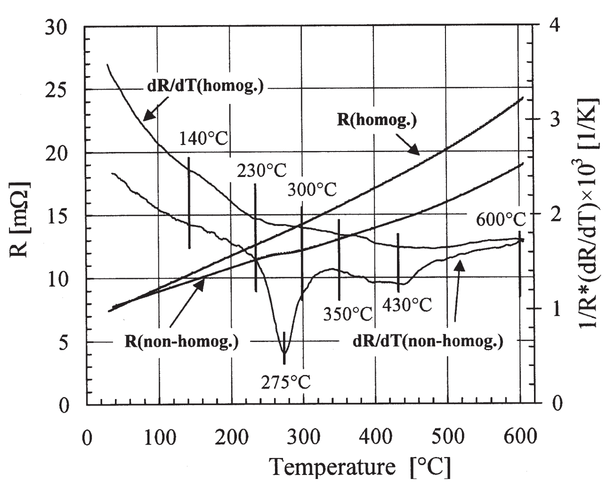

Fig. 1. Temperature dependence of resistivity R and the normalised differential resistivity $(\mathrm{d}R/\mathrm{d}T)R$ of strips cold rolled from as-cast and from homogenized materials during heating at the rate of 1 °C/min.

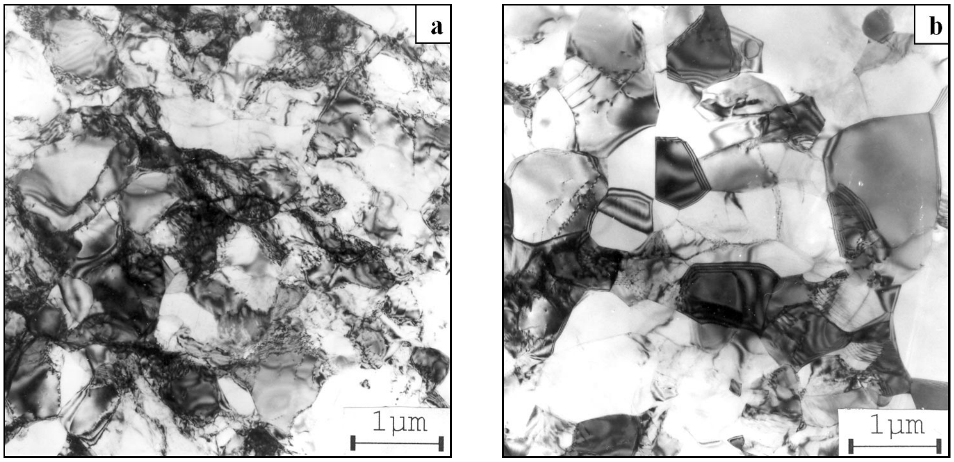

Fig. 2. TEM micrographs of the substructure of the homogenized material: a) as-rolled, b) after linear heating at 1 ℃/min and quenching from 230 ℃.

differential curve of the non-homogenized sample exhibits two pronounced features – local minima at 275 and 440 ℃. The first valley is due to the redistribution of solutes trapped by dislocations caused by the intensive process of dislocation recovery [8]. The resistivity evolution above 340 ℃ is due to second-phase precipitation and coarsening at lower temperatures and their partial dissolution at higher temperatures. For more details on the substructure and phase transformations of the non-homogenized sample, see [8].

TEM examination of the homogenized and cold rolled sample revealed the substructure which is typical for deformed low-alloyed Al materials. The dislocations were arranged in cells (~ 0.5 μm in size) and subgrain boundaries (Fig. 2a). The second phase composition, the density and the size of the particles were similar to those in the sample after homogenization-like annealing [8]. Coarse plate-like particles (1–4 μm in diameter) rich in Al and Fe were very frequent. Coarse ellipsoidal Al, Fe and Mn particles and smaller Al, Fe, Si and Mn particles were also observed.

The curve $(\mathrm{d}R/\mathrm{d}T)/R = f(T)$ of the homogenized sample is much smoother than that of the non-homogenized one. Between 230 and 330 ℃, no pronounced valley is observed. Nevertheless, above 230 ℃ there is a change in slope of the curve. TEM of a sample quenched from 230 ℃ indicated that the matrix was partially recrystallized (Fig. 2b). The recrystallization nuclei size was 1 to 2 μm. These nuclei were not observed by light microscopy, probably due to their small size.

A slow decrease of $(\mathrm{d}R/\mathrm{d}T)/R$ is observed between 250 and 390 ℃ (Fig. 1). The resistivity changes are supposed to be caused by precipitation of the complex

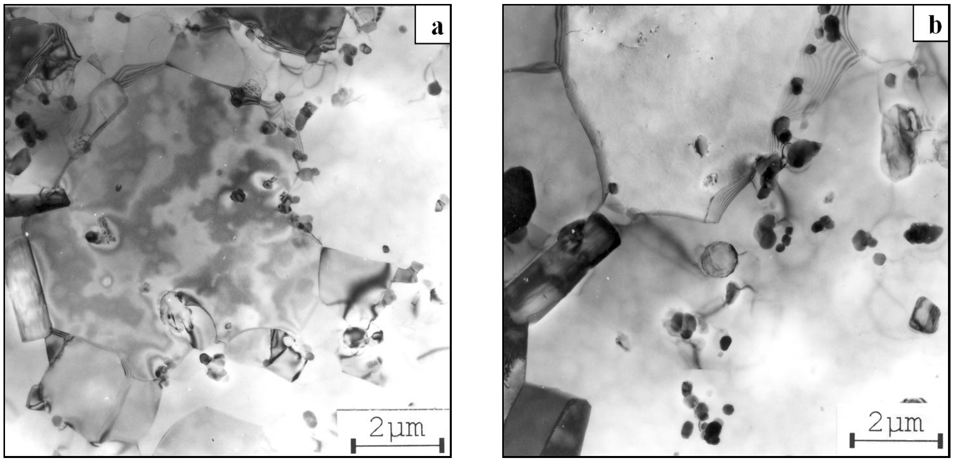

Fig. 3. TEM micrographs of the second phase in the homogenized material subjected to linear heating at 1 ℃/min: a) after quenching from 300 ℃, b) after quenching from 430 ℃.

α-Al(Fe,Mn,Si) phase. The size of precipitates in the sample quenched from 300 ℃ was about 300 nm (Fig. 3a), whereas their density in the sample quenched from 350 ℃ was higher and their mean size was estimated to be 500 nm. The size of recrystallized grains gradually increased from 2 to 6 μm, but grains of 10 μm in diameter were also observed. In areas with high particle density, the size of recrystallized grains was 2 to 3 μm, whereas in areas with low particle density the grain size was larger – up to 10 μm. This observation proves that particles exert a dragging force on the boundaries of growing grains. In samples quenched from 300 ℃ and higher temperatures, recrystallized grain nuclei were also observed in light micrographs (Fig. 4a). Recrystallized grain size increases with increasing temperature, grains of 20 μm were observed in samples quenched from 350 ℃.

Another change in the slope of the $(\mathrm{d}R/\mathrm{d}T)/R$ curve of the homogenized material was observed at 390 ℃ – Fig. 1. The $(\mathrm{d}R/\mathrm{d}T)/R$ continues to decrease and a plateau is observed between 430 and 480 ℃. TEM analysis of the sample quenched from 430 ℃ indicated that the resistivity evolution above 390 ℃ is due to the formation of the orthorhombic Al_6(Fe,Mn) phase (Fig. 3b). Coarsening of previously formed α-Al(Fe,Mn,Si) particles was also observed. The Al matrix in the sample quenched from 430 ℃ was almost fully recrystallized and the grain size was slightly uneven (Fig. 4b). The grain size (measured in the rolling direction) in the sample quenched from 430 ℃ was 10 μm, whereas at 600 ℃ almost 18 μm. The grain sizes in the short transverse direction were 8 μm and 15 μm, respectively. In contrast, the grains in non-homogenized samples were significantly coarser. The

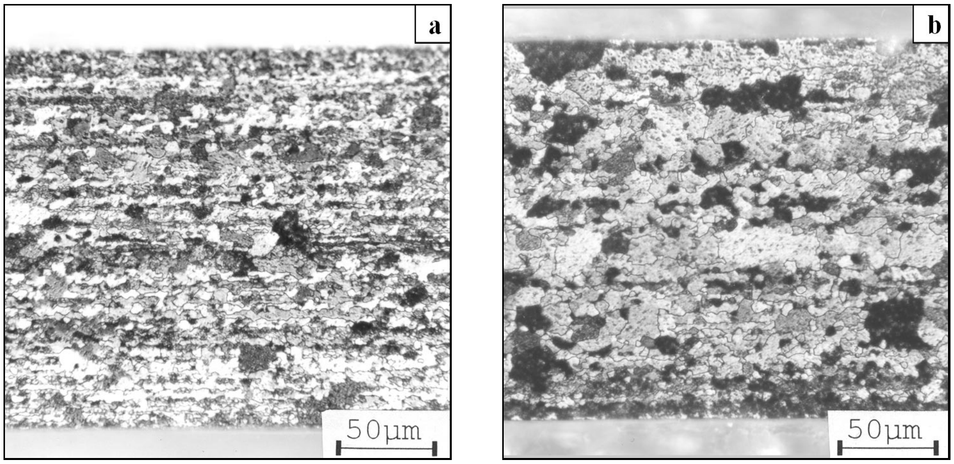

Fig. 4. Polarised light micrographs of the grain structure evolution in the homogenized material during linear heating at 1 ℃/min – sample quenched from: a) 300 ℃, b) 430 ℃.

corresponding grain sizes in the sample quenched from 600 ℃ were ~2900 μm (in the rolling direction) and ~240 μm (short transverse direction) [8].

Above 480 ℃, the curve of $(\mathrm{d}R/\mathrm{d}T)/R$ of the homogenized material gradually increases (Fig. 1). TEM and light microscopy examinations proved that small precipitates of both α-Al(Fe,Mn)Si and Al_6(Fe,Mn) are dissolved. The dissolution of the precipitates leads to solid solution enrichment in alloying elements resulting in a resistivity increase. In parallel, large primary particles of α-Al(Fe,Mn)Si and Al_6(Fe,Mn) coarsen. This process does not contribute to the resistivity increase. Table 1 shows particle size and shape characteristics of homogenized and non-homogenized samples heated to different temperatures (area A, length $D_{\max}$, number density N_{A}, volume fraction V_{v}, and fraction F_{r} of particles with length

Table 1. Particle characteristics of thin strips heated at 1 ℃/min and quenched from different temperatures

Annealing parameters		Particle characteristics			
Sample/ Temperature [℃]	Total time [hours]	A [μm^2]	$D_{\max}$ [μm]	$\bar{N}_{\mathrm{A}}$ [10^3 mm^{-2}]	F_{r} [%]
homog./as-rolled	0	0.51	0.93	102.9	6.0
homog./230	3.5	0.54	0.95	103.7	6.2
homog./430	7	0.46	0.88	119.4	5.2
homog./600	10	0.58	0.97	82.5	8.1
non-homog./600	10	0.28	0.74	321.3	0.8

> 2 μm). The character of particle populations in the as-rolled sample and in the sample quenched from 230 °C is almost the same. The particle mean size in the 430 °C sample is smaller than after quenching from 230 °C. The 430 °C sample contains significantly larger numbers of small particles resulting in the decrease of the mean value $D_{\max}$. TEM observations also showed the presence of a dense, new Al_6(Fe,Mn) precipitate.

As expected, the largest particle size and the lowest density were found in the samples quenched from 600 °C. A significant difference between the particle populations in the homogenized and the non-homogenized samples was observed. The non-homogenized sample contains approximately four times more particles than the homogenized one. The mean size and the fraction of coarse particles are significantly smaller than in the homogenized sample. No attempt to measure the particles in the non-homogenized samples quenched from lower temperatures was made. In these samples the size of the particles was close to the resolution limit of the light microscope.

3.2 Recrystallization response curves

The plots of Vickers microhardness HV0.1 as a function of temperature (quick heating) for both non-homogenized and homogenized materials annealed for 0.5 h are shown in Fig. 5. The microhardness evolution in the non-homogenized sample annealed at a slow heating rate (1 °C/min) is also plotted. In the whole temperature range the non-homogenized material exhibits higher hardness than the

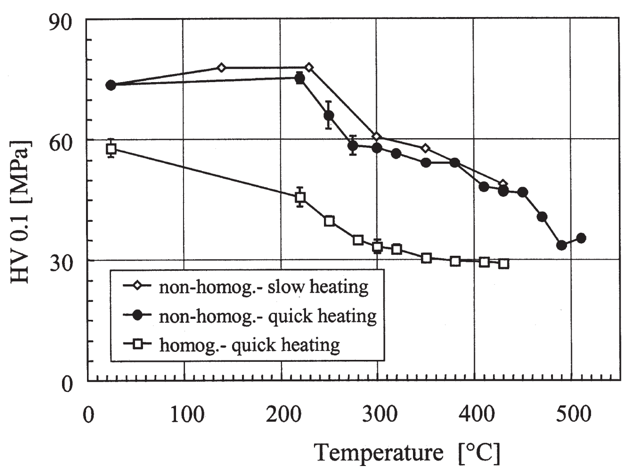

Fig. 5. Plot of hardness as a function of annealing temperature of non-homogenized and homogenized material – samples annealed for 0.5 h in a pre-heated furnace.

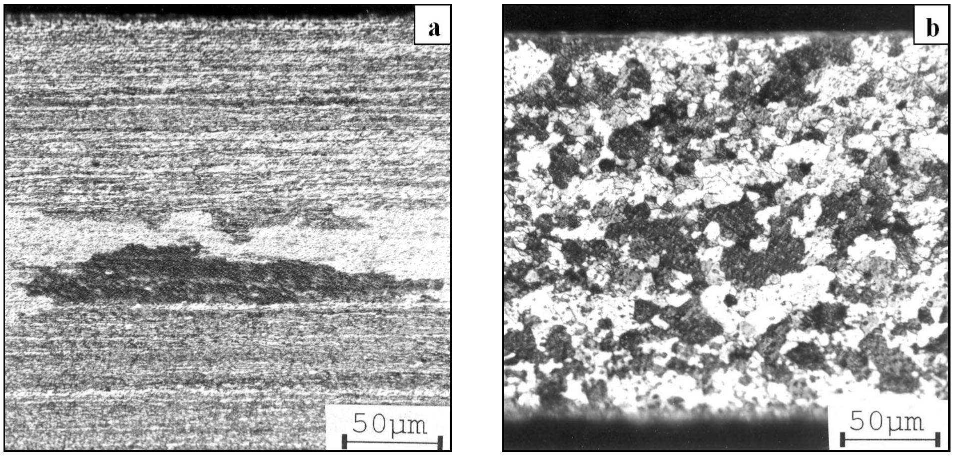

Fig. 6. Polarised light micrographs of the grain structure of samples after annealing for 30 min at 430 ℃ in a pre-heated furnace: a) non-homogenized, b) homogenized material.

homogenized one. The shapes of the curves of homogenized and non-homogenized material are different. The decrease in the non-homogenized sample shows two plateaux: from 270 to 380 ℃ and from 410 to 450 ℃. In the homogenized material hardness HV0.1 decreases with increasing temperature much more gradually. In the non-homogenized material the first recrystallization nuclei were observed at 410 ℃, whereas in the homogenized material at 220 ℃.

The heating regime does not significantly influence hardness change with temperature. The grain structure of both materials annealed at 430 ℃ for 0.5 h is shown in Fig. 6. In the non-homogenized material (Fig. 6a) only a few coarse grains are observed in the central part of the strip, while the homogenized material (Fig. 6b) is fully recrystallized. The homogenized material has fine grain structure with even equiaxed grains, whereas very coarse prolonged grains of uneven size are observed in the non-homogenized samples.

4. Discussion

4.1 Effect of homogenization on structure transformations

As mentioned in section 3.1, the concentration of alloying elements in the matrix of the non-homogenized sample is significantly higher than that of the homogenized one. On the other hand, the number of coarse particles is significantly higher in the homogenized sample. Both solid solution content and particle dispersion affect the deformation during cold rolling and also the kinetics of deformation recovery and phase transformations during the annealing of strips after cold rolling.

The interparticle spacing can be assessed using the data of particle measurements (Table 1). For randomly distributed particles the centre to centre nearest neighbour spacing Δ_2 in a plane is given by [9]:

$$\Delta_2 = 0.5 N_{\mathrm{A}}^{-1/2}. \tag{1}$$

The interparticle spacing Δ_2 in the homogenized as-rolled sample (prior to final annealing) estimated using Eq. (1) was 1.6 μm while interparticle spacing in the non-homogenized sample annealed up to 600 ℃ was 0.9 μm. TEM examinations of as-rolled samples showed that the dislocation cells and subgrain size in the non--homogenized and homogenized material was about 1 μm and 0.5 μm, respectively. A comparison of Δ_2 and the cell size proves that in the non-homogenized material the cell size is larger than the interparticle spacing. Therefore, the recovery process, consisting in rearrangement of dislocations, is hindered by the densely spaced particles. Moreover, higher concentration of solute atoms in non-homogenized samples exert a larger pinning effect on dislocations than in homogenized material. On the other hand, the cell size in the homogenized material is smaller than the interparticle spacing and thus the recovery is only a little affected by the presence of particles. This is one of the reasons why recrystallization starts at lower temperatures and is completed sooner in the homogenized material. Furthermore, the presence of coarse and strong (non-deformable) particles in the homogenized material contributes to the localization of deformation in the vicinity of particles. The presence of these deformation zones promotes particle stimulated nucleation (PSN) of recrystallization [9]. As both the magnitude of lattice rotation in the deformation zone and the number of zones formed per particle increase with particle size, larger particles support PSN more effectively than smaller particles. Such zones probably are not formed or are too small in the non-homogenized material due to small particle size. Therefore, the recrystallization in the non-homogenized material is very retarded both by the closely spaced particles and by the lack of deformation zones where PSN can occur.

The situation becomes more complicated if we consider the interaction between the processes of precipitation and recrystallization. A shift in the onset of precipitation (from 350 to 300 ℃) towards lower temperatures due to homogenization was observed. The non-homogenized material exhibits much higher solute saturation than the homogenized one. This difference in connection with the slope of the phase diagram of the system Al-Fe-Mn-Si in the temperature range between 300 and 480 ℃ provides the obvious evidence of this shift – the less concentrated solution begins its decomposition at a lower temperature. It was found that in the homogenized material heated at a low rate (1 ℃/min) recrystallization starts at about 230 ℃ (confirmed by TEM observations). TEM observations also showed that the new small particles forming above 300 ℃ have small effect on moving grain boundaries. Therefore, recrystallization in the homogenized material is affected by precipitation only in a small extent. On the other hand, the number of tiny particles observed in the samples of non-homogenized material annealed above 350 ℃ is very high and the particles are closely spaced. Therefore, the particles can effectively pin boundaries and retard recrystallization. At temperatures above 350 ℃ only a few recrystallization nuclei are formed in the central part of the samples, where larger particles were found. These nuclei thus grow to a very large size (Fig. 6a).

4.2 Effect of heating rate on grain structure

In the homogenized samples the first recrystallization nuclei were found in the sample annealed at 220 ℃ for the fast heating regime, but only at 300 ℃ in the slowly heated sample. In the non-homogenized material, the first nuclei were found at 410 and 430 ℃ for the rapidly and slowly heated samples, respectively. Therefore, the increase in the heating rate shifts the start of recrystallization towards lower temperatures.

The heating rate (and hold period) also affects the grain shape and size homogeneity. In homogenized material grains in slowly heated samples (Fig. 4b) are more flattened and their size is much more uneven than in the quickly heated samples (Fig. 6b). Furthermore, the recrystallization takes more time to be completed when using slow heating. For example, the sample quenched from 430 ℃ after slow heating is still only partially recrystallized, whereas the sample put in a furnace preheated to 430 ℃ is fully recrystallized within 2 minutes. The effect of heating rate on the recrystallization kinetics and grain size are both due to deformation recovery prior to recrystallization. When samples are heated at a high rate, the amount of recovery occurring before recrystallization is reduced, thus a larger driving force for recrystallization is preserved in comparison with the slowly heated samples. Furthermore, the low diffusivity of the solute Fe and Mn atoms [10] involved in the precipitation at high heating rate causes that the recrystallization of homogenized material is completed below 350 ℃, i.e. before precipitation can occur. Therefore, precipitates and solutes (low in quantity in the homogenized material) do not hinder subgrain and grain boundary movement. The grains can grow up to the size controlled by spacing of the particles that are present in the material before annealing.

When the homogenized samples are heated slowly, a larger extent of recovery occurs, and this process lowers the driving force for recrystallization. Moreover, in spite of the fact that recrystallization starts as first, when the temperature range of solid solution decomposition is reached, precipitation interferes with recrystallization. Therefore, precipitates formed above 300 ℃ can pin high angle grain boundaries and recrystallization is further slowed down. The resulting grain structure exhibits a typical heterogeneity. Prolonged grain shape is observed in slowly heated homogenized and all non-homogenized samples. The grain shape anisotropy is due to the pinning effect of particles, which are more closely spaced in the original dendrite boundaries parallel to the rolling plane. Such specific second phase distribution heterogeneity is observed in homogenized TRC strips and is very pronounced in non-homogenized strips. The effect of heating rate on the grain size and the shape in non-homogenized material is not so pronounced. In both fast and slowly heated materials, the motion of dislocations, subgrain and grain boundaries is strongly disturbed by solute atoms and precipitates, which act as obstacles effectively hindering recrystallization.

5. Conclusions

The study of structure transformations occurring during annealing of AA 8006 twin-roll cast cold rolled strips showed that:

1. Long-term high temperature homogenization significantly influences the kinetics of structure transformations occurring during annealing of thin cold rolled strips. A shift towards lower temperatures of the onset of precipitation due to the homogenization was observed.

2. The homogenization significantly facilitates the recrystallization of cold-rolled materials. Recrystallization starts and is completed at much lower temperatures in the homogenized material than in the non-homogenized one. This is caused by the difference both in second phase dispersion and in solid solution concentration induced by homogenization.

3. Coarse grain heterogeneous structure is formed during annealing of samples non-homogenized prior to cold rolling. On the other hand, fine grain homogeneous structure is typical for strips subjected to a suitable homogenization treatment.

4. The heating rate significantly influences recrystallization kinetics. The recrystallization begins at lower temperature in both homogenized and non-homogenized materials when the samples are subjected to fast heating. The resulting grain structure is more homogeneous in quickly heated samples, especially in the homogenized material.

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