Light Metals 2015
Edited by: Margaret Hyland
TMS (The Minerals, Metals & Materials Society), 2015

The Influence of Microchemistry and Processing Conditions on the Softening Behavior of Cold-Rolled Al-Mn-Fe-Si Alloys

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Keywords: Recrystallization, Microchemistry, Second-phase particles, Precipitation

Abstract

Motivated by an increased focus on recycling of aluminium alloys, for which elements like Mn, Fe and Si tend to increase, a comprehensive investigation of the softening behavior during annealing after cold rolling of Al-Mn-Fe-Si alloys has been carried out. It is clearly demonstrated that the kinetics and final microstructure are the result of a delicate balance between processing conditions and microchemistry. In general, at the same processing condition, more and finer dispersoids, whether pre-existing or formed during annealing (concurrent precipitation) strongly affect the kinetics and either mainly suppress recrystallization or give coarse non-equiaxed recrystallized grains. Faster softening kinetics is observed at large deformations and conditions less affected by dispersoids, together with equiaxed fine grains. The precipitation behavior before the completion of recrystallization is accelerated by high solid solution levels of Mn, large deformations and high temperature. However, even with a strong supersaturation of Mn in solid solution, annealing at high temperature accelerates recrystallization so that it is mainly completed before substantial concurrent precipitation take place, giving a fine equiaxed grain structure. Moreover, annealing treatments at low heating rates produce more inhomogeneous microstructures than isothermal annealing.

1. Introduction

AA3xxx series alloys belong to the group of non-heat treatable aluminum alloys, with Mn as the main alloying element, but commonly also with significant amounts of Fe and Si. The additions of Mn influence the material properties in various ways. In addition to going into constitute particles formed during solidification, supersaturated Mn in solid solution after solidification will precipitate as Mn-bearing dispersoids during subsequent thermomechanical processing, changing the microchemistry of the alloy. It is well established that particles are of the outmost importance in recrystallization of alloys containing second-phase particles [1-5]. Fine and numerous dispersoids, typically formed during homogenization and/or subsequent heat treatments, tend to hinder boundary motion and slow down and even hinder recrystallization and grain growth through a Zener drag effect [6]. The size distribution of these dispersoids also plays a role [7]. On the other hand, coarse constituent particles larger than ~1μm, can accelerate recrystallization by particle stimulated nucleation (PSN) [8]. It is well documented that different homogenization procedures may provide different microchemistry states (solid solution levels, constituent particles and dispersoids) [9, 10]. Under certain circumstances, back-annealing of cold deformed conditions may give a recrystallized microstructure of coarse highly elongated grains [11-13]. The effect of microchemistry, introduced by different homogenization procedures, on the microstructural evolution during *isothermal* annealing of a cold-rolled Al-Mn-Fe-Si alloy at one specific processing condition was investigated in [14]. In the present study, in addition to the influence of microchemistry, the effects of processing conditions including different cold deformations, annealing temperatures and heating rates on the softening behavior in terms of softening kinetics, precipitation behavior and grain structure are also investigated. Significant variations of these parameters are frequently encountered during industrial processing conditions. Thus a systematic investigation of their effect on the softening behavior of these materials is not only of academic interest, but also of outmost industrial importance. Their effect on the recrystallization texture is, however, not included in the present work as they are presented elsewhere [13, 15, 16].

The present work is part of a comprehensive investigation on particle-annealing interactions in AA3xxx type alloys. The results presented in the current paper is not only intended to give a better understanding and quantitative description of the different phenomena occurring during the annealing after cold deformation of these alloys, but can also serve as basis of validation and further development of relevant existing physically based numerical models [17,18].

2. Experimental

2.1 Alloy conditions and annealing treatment

The investigated alloy was supplied by Hydro Aluminium in the form of DC-cast AA3xxx extrusion billets. The as-received material was in the as-cast state, with the chemical composition given in Table 1. More details about the received material can be found in [13].

Table 1 Chemical composition of the studied alloy, in wt. %.

Alloy	Si	Fe	Mn	Others
C1	0.152	0.530	0.390	<0.01

The as-cast condition was labelled C1-0 to be consistent with previous investigations on the same alloy [13-16]. To provide material conditions of different microchemistry, *i.e.* solute level and second-phase particle, the as-cast material was subsequently homogenized with two different procedures. The homogenization treatments were performed in an air circulation furnace with a temperature accuracy of ± 2 K, starting from room temperature (about 20 °C). One set of the samples were heated at 50°C/h to 450 °C and kept for 4 hours, referred to as the C1-2 condition. The other set of the samples were subjected to a two-stage homogenization treatment. The samples were first heated at 50°C/h to 600 °C for 4 hours, and then cooled at 25°C/h to 500°C where they were kept for another 4 hours, to give the C1-3 condition. Materials were water quenched to room temperature at the end of the homogenization procedure to freeze the state of supersaturation/precipitation.

The three variants were subsequently cold rolled to different strains of ε = 0.7, 1.6, 3.0. The cold rolling was performed at room temperature in multiple passes with heavy lubricated rolls. The rolled sheets were either subsequently isothermally back-annealed in a pre-heated salt bath or non-isothermally heated at different heating rate to desired target temperatures in the range 300-500 °C and with different holding time in the range of 5-10^5 s, followed by quick water quenching. Unless specifically mentioned (section 3.5), the annealing experiments were all conducted in salt bath, i.e. isothermal annealing.

2.2 Microstructure characterization

The softening and precipitation behavior during annealing was followed by Vickers hardness (VHN) and electrical conductivity (EC) measurements performed on the RD-TD plane of sheets. Each reported value was obtained by averaging eight measurements. EC was measured by a Sigmascope EX 8 instrument at room temperature of about 293 K (20°C). Metallographic examinations of constituent particles and dispersoids were done by backscattered electron channeling contrast imaging in a Zeiss Ultra 55 field emission gun scanning electron microscope (FEG-SEM). Images were captured and then analyzed using standard image analysis software Image-J, by which the characteristic size parameters of constituent particles, equivalent diameter *d* and number density were measured. For the hardness measurements, a load of 1 Kg, a loading time of 15 s and a loading speed of 100µms^{-1} were used.

The microstructures of the annealed sheets were characterized by means of EBSD (Electron backscattered diffraction) in a Zeiss Supra/Ultra 55 SEM equipped with TSL software for selected conditions. For all of the micrographs presented in this paper, the horizontal direction corresponds to the rolling direction (RD) while the vertical direction is the normal direction (ND). The grain size was measured as the equivalent circular diameter in the RD-ND cross section. The typical step size for the EBSD maps is 1-2 µm.

3. Results and discussion

3.1 The microstructure and microchemistry before annealing

The micrographs of the particle distribution for the three variants (before deformation) are presented elsewhere [13]. From this work it is clear that the variant homogenized at the lower temperature, i.e. C1-2, obviously has more and finer dispersoids than its counterpart C1-3, as shown in Table 2. It is important to note that the dispersoids were shown to have a more or less even spatial distribution [13]. The constituent particles increase slightly in size for C1-2 as compared to C1-0. When homogenized at a higher temperature (C1-3), the constituent particles were observed to grow to 1.10 µm. The Mn level in solid solution decreases from C1-0 to C1-3.

Table 2 Electrical conductivity, concentrations of solutes, diameter and number density of particles in the alloys studied

	Concentration of Mn (wt%)	Constituent particles		Dispersoids	
		Diameter (µm)	Number density (mm^{-2})	Diameter (µm)	Number density (mm^{-2})
C1-0	0.35	0.88	2.8e4	-	-
C1-2	0.16	0.96	2.9e4	0.054	1.3e6
C1-3	0.11	1.10	2.1e4	0.127	5.5e4

3.2 Effect of microchemistry

The effect of microchemistry, in terms of solute level and second phase particle structures on the softening behavior of cold-rolled Al-Mn-Fe-Si alloy was firstly investigated. The samples of the three variants were cold rolled to a deformation strain of 3.0, and then were subjected to isothermal annealing at 300°C, the softening and precipitation behavior are shown in Fig.1. Hardening upon annealing was observed for all the three variants for the first 5s, which was also observed by Govindaraj et al [19] using the same material, and suggested by them to be due to some sort of clustering or precipitation mechanism. After this 5s, it is clearly shown that the softening becomes less pronounced and much slower with increasing level of Mn in solid solution, and as seen from Fig. 1b, significant concurrent precipitation does take place for C1-0. The C1-3 variant, on the other hand, obviously experiences the weakest concurrent precipitation, which at least partly explains why a fully soft (recrystallized) state was reached after annealing for 10^5s for this variant (Fig. 2). C1-2, for which concurrent precipitation is only slightly stronger than for C1-3, the softening behavior resembles that of the C1-0 condition, which signifies that a large number of pre-existing dispersoids may also suppress and/or strongly retard recrystallization.

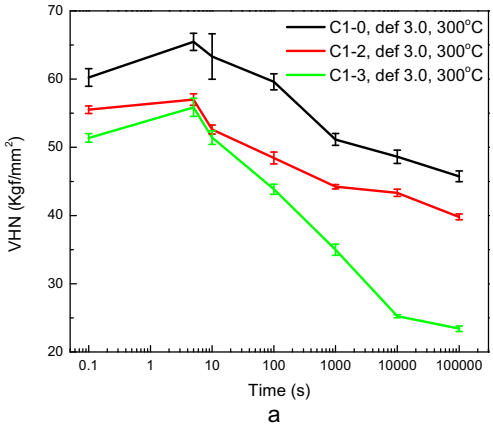

Fig.1 The effect of microchemistry on the a) softening kinetics; and b) precipitation behavior of samples cold rolled to a strain of ε = 3.0, and annealing at 300°C

The effect of microchemistry on the annealed microstructures is illustrated in Fig.2. The significant concurrent precipitation for C1-0 (Fig.1b) has totally suppressed the nucleation of

recrystallization. C1-2, which has a large number of pre-existing fine dispersoids, only a few recrystallized grains are visible. Most of these grains are coarse and elongated in the RD direction, which may indicate that some additional concurrent precipitation, which preferentially take place along the original high angle grain boundaries and thus provide a stronger Zener drag in the ND direction than in the RD direction, has taken place. A fully recrystallized state was only obtained for C1-3 where the effect of concurrent precipitation and dispersoids in general is limited. The slightly larger constituent particles might also have played a role through particle stimulated nucleation (PSN).

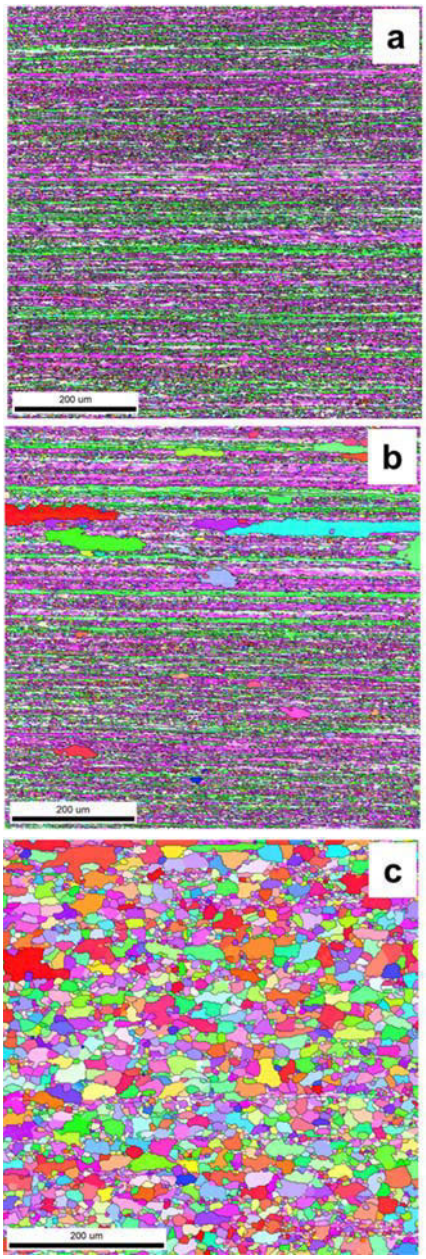

Fig.2 The effect of microchemistry on the annealed microstructure on samples cold rolled to a strain of ε = 3.0 before they were annealed in salt bath at 400°C and kept for 10^5s
a) C1-0; b) C1-2; c) C1-3

3.3 Effect of cold deformation

The as-cast variant C1-0 was used to illustrate the effect of cold deformation on the softening and precipitation behaviors..In terms of kinetics, it is nicely demonstrated that large deformations promote faster recrystallization (see Fig.3a). This is first and foremost due to the larger stored energy, i.e. a higher driving force for recrystallization. However, for the present alloy and conditions, an additional (retarding) effect from significant concurrent precipitation is experienced for the condition(s) for which recrystallization is already slow (lower strains), as can be seen in Fig.3b. Fully recrystallized state was reached after annealing of 10^3s and 10^4s for the sample deformed at ε=3.0 and ε=1.6 respectively, while recrystallization was still not completed even after annealing for 10^5s for the sample with the smallest deformation of ε=0.7.

The precipitation behavior, on the other hand, shows a somewhat different cold deformation dependcy with annealing time, as shown in Fig.3b. The fact that the sample deformed to 0.7 has the largest EC value after annealing for 10^5s may seem counter intuitive, since it is well documented that heavy deformation will accelerate precipitation [20], and actually this is the case for the current study before recrystallization is completed (~3000s). For the sample deformed to ε = 0.7, on the other hand recrystallization has not completed yet after annealing for 3000s, i.e. heterogeneous nucleation sites for (concurrent) precipitation are available throughout the complete annealing period and explains why this condition ends up with the largest EC value.

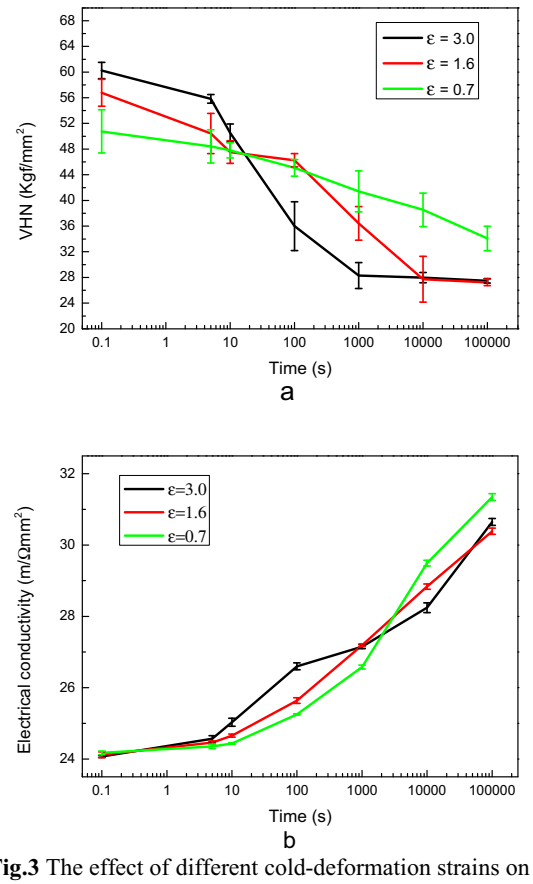

Fig.3 The effect of different cold-deformation strains on the a) softening kinetics; and b) precipitation behavior of the C1-0 condition isothermally annealed at 400°C

The different cold deformations are also strongly reflected in the annealed microstructures, Fig.4. Recrystallization is not yet completed for the sample deformed to ε = 0.7, while the recrystallized grain size clearly decreases with increasing rolling strain from ε = 1.6 to 3.0. It is also interesting to note the decreasing grain size in the ND direction with increasing cold deformation. This is mainly because after deformation the original grain boundaries tend to be aligned with the RD direction and as concurrent precipitation preferably takes place at these boundaries, increasing deformation leads to smaller distance between these old grain boundaries in the ND direction.

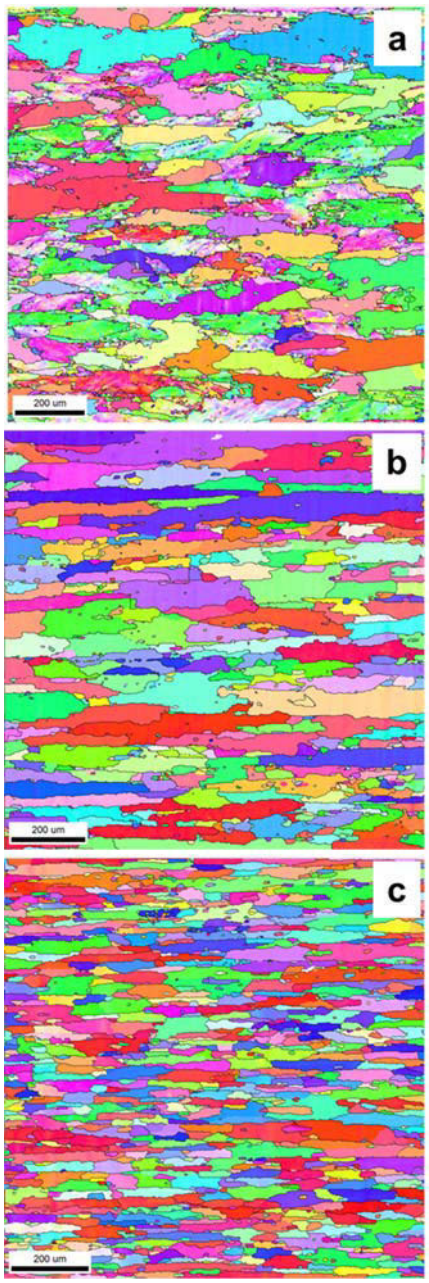

Fig.4 The effect of deformation on the annealed microstructure. of C1-0 samples deformed to different rolling strains and annealed in salt bath at 400°C and kept for 10^5s. a) ε=0.7; b) ε=1.6; c) ε=3.0

3.4 Effect of annealing temperature

. For the present case, the material condition was fixed, i.e., the C1-0 variant deformed to ε = 3.0, while the effect of different annealing temperatures was investigated. At the lowest annealing temperature of 300°C, the kinetics is quite slow and the actual softening limited (i.e. no or limited recrystallization), while fully while recrystallization completed within 5s at 500°C. As for the degree of cold deformation, the actual kinetics is a combined effect, in this case of temperature alone and the (concurrent) precipitation behavior which also changes with annealing temperature, as shown in Fig.5b. For the two highest annealing temperatures recrystallization is completed before any significant precipitation has taken place. The fact that the sample annealed at the intermediate temperature (400°C) end up at the highest EC value (although only slightly lower than at 500 °C) do indicate that precipitation is close to optimum at this temperature and decreases again at higher temperatures. At the lowest temperature of 300°C, the EC curve steadily increases during annealing but its absolute value is consistently lower than for the other two cases, indicating the lowest amount of precipitation in this case. Still the softening kinetics is slowest for this case clearly illustrating that the concurrent precipitation effect is a secondary effect, and the softening behavior is mainly due the fairly low annealing temperature with decreased nucleation of recrystallization as well as boundary migration rate (both thermally activated processes).

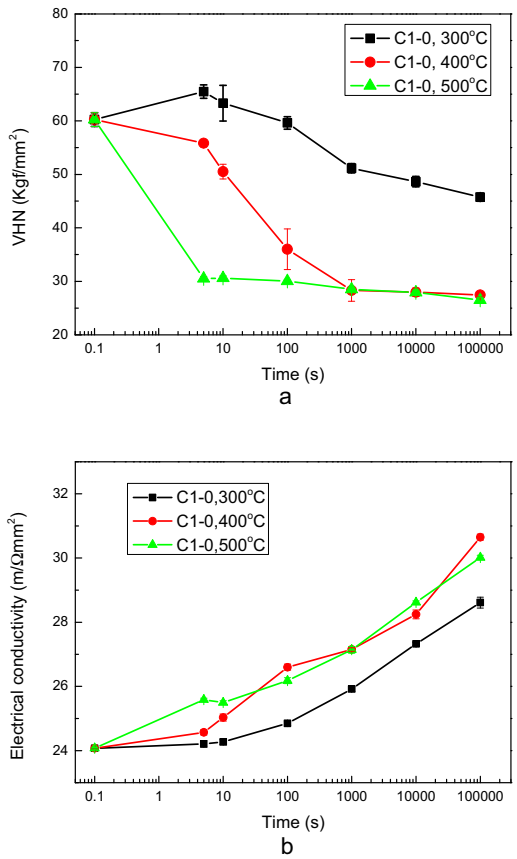

Fig.5 The effect of annealing temperature on the a) softening kinetics; and b) precipitation behavior of C1-0 samples cold deformed to a strain of ε = 3.0

The resulting microstructures following annealing at the different annealing temperature are shown in Fig.6. As indicated above recrystallization was totally suppressed for C1-0 when annealing at 300°C for 10^5s, as also illustrated by the Fig. 6a. At 500°C, recrystallization completed within 5s (see Fig.5a), and an equiaxed fine grain structure is obtained (Fig. 6c). Also for annealing at 400°C, a fully recrystallized grain structure is obtained. The grain structure of clearly larger and elongated grains, as compared to the 500°C condition (Fig. 6b), is explained by the stronger Zener drag effect in the ND direction caused by concurrent precipitation, as discussed previously.

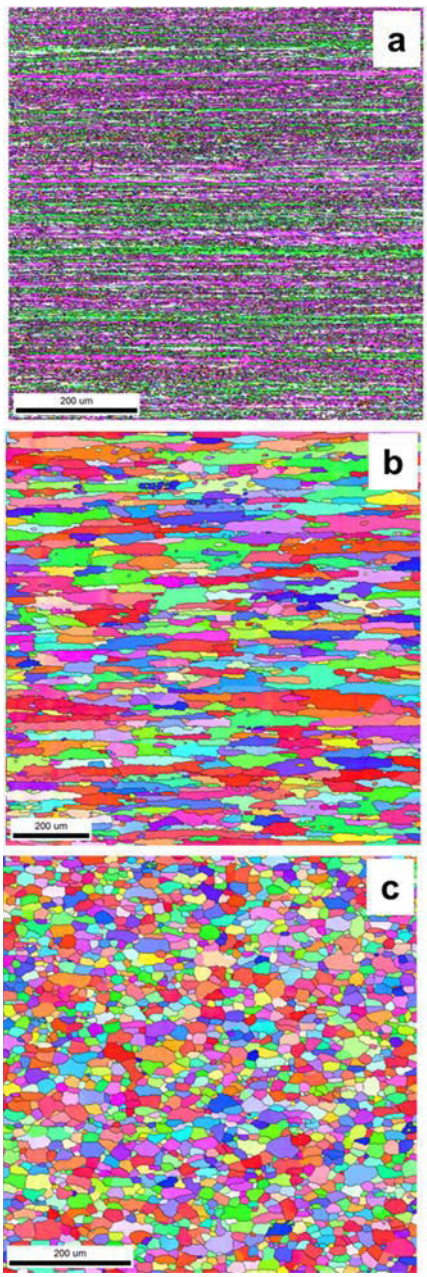

Fig.6 The effect of annealing temperature on the annealed microstructure of C1-0 samples that were first deformed to a strain of ε = 3.0 before annealing in salt bath at different target temperatures and kept for 10^5s. a) 300°C; b) 400°C; c) 500°C

3.5 Effect of heating rate

It has been previously shown that low heating rates give slower softening kinetics using the same material [21]. The effect of heating rate on the corresponding annealed microstructures was analyzed using the C1-0 variant deformed to ε = 3.0 and then subsequently heated at different heating rates to 400°C and kept for 10^5s. Four different heating rates were tested, namely 50, 100, 200 and 7.6×10^6 °C/h where the latter one corresponds to isothermal annealing. As shown in Fig.7, faster heating rate in general produces finer grain sizes, the average grain size decreased from 149μm to 56μm when the heating rate was increased from 50°C/h to 7.6×10^6 °C/h. The fact that the average grain size in the ND direction for the pan-cake shaped grains decreases with increasing heating rate is mainly due to the increased nucleation activity, since the same starting material was used and less interaction with concurrent precipitation is expected with faster heating rate, but still, a clear elongated grain structure was observed even with the fastest heating rate (see Fig.7d). As we learn from Fig.5a, recrystallization completed at ~1000s for C1-0 sample deformed to ε=3.0, and significant concurrent precipitation has taken place within this time (Fig.5b). These precipitates are preferentially located along grain/subgrain boundaries [22], and give larger Zener drag effect along the ND direction, leading to the elongated grain shape.

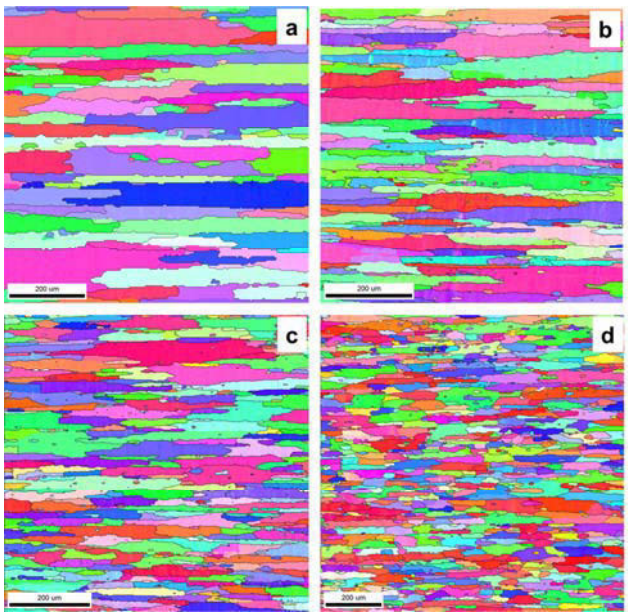

Fig.7 The effect of heating rate on the annealed microstructure of samples that were first deformed to 3.0 before being annealed at different heating rates to 400°C and kept for 10^5s
a) 50°C/h; b) 100 °C/h; c) 200°C/h; d) 7.6×10^6 °C/h

In terms of numerical modelling, the results shown in this paper indicate that the microchemistry state, i.e., solute level of Mn, size and spatial distribution of the second-phase particles, should be considered in order to get adequate predictions of kinetics and final microstructures. A physically based semi-analytical softening model [17] has previously been tested against selected conditions of similar Al-Mn-Fe-Si alloys at different processing conditions. Even though the solute drag and Zener drag effects are both incorporated (by classical equations), it is extremely challenging to get reasonable results for the cases

strongly affected by dispersoids and concurrent precipitation. More experimental investigation on the interaction between microchemistry and recrystallization are needed, where the individual effects of solute drag, pre-exiting dispersoids and concurrent precipitation should be quantified. Coupled models [e.g. [18]] which take into account of the interaction between recrystallization and microchemistry should be helpful to obtain a better understanding of this complicated phenomenen, since this interaction is evolving with time and it is very often difficult to quantify experimentally.

4. Conclusions

In this paper, a comprehensive investigation of the softening behavior during annealing after cold rolling of Al-Mn-Fe-Si alloys has been carried out. The softening and concurrent precipitation behaviors of the samples have been monitored by hardness and electrical conductivity measurements, respectively, and the final microstructure has been characterized by EBSD. In general, at the same processing condition, more and finer dispersoids, whether pre-existing or formed during annealing (concurrent precipitation) strongly affect the kinetics and give coarse and highly elongated grains. Faster recrystallization kinetics is observed at large deformations and conditions experiencing limited effects of dispersoids, together with equiaxed fine grains. The precipitation before the completion of recrystallization is accelerated by a high supersaturation level of Mn, large deformations and medium to high annealing temperature. However, even with a high supersaturation of Mn, annealing at high temperature accelerates recrystallization kinetics and recrystallization is often completed before substantial concurrent precipitation take place, in which case an equiaxed fine grained structure also is obtained. Moreover, annealing treatments at low heating rates produce more inhomogeneous microstructures than isothermal annealing.

Acknowledgements

This research work has been supported by the KMB project (193179/I40) in Norway. The financial support by the Research Council of Norway and the industrial partners, Hydro Aluminium and Sapa Technology is gratefully acknowledged. KH acknowledges the financial support from NTNU, through the Strategic Area Materials.

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