Journal of Mining and Metallurgy, 38 (3–4) B (2002) 153 - 162

MICROSTRUCTURE DEVELOPMENT DURING THERMOMECHANICAL TREATMENT OF Al-Mg-Si ALLOY

Z. Martinova* and G. Zlateva**

*University of Chemical Technology&Metallurgy,
1756 Sofia, Kl.Ohridski blvd. 8, Bulgaria
**Institute of Metal Science, Bulgarian Academy of Sciences,
1574 Sofia, Shipchenski Prohod str. 67, Bulgaria

(Received 20 August 2002; accepted 2 November 2002)

Abstract

The effect of natural aging and 95% cold deformation on the microstructure evolution and aging characteristics in commercial Al - 1 mass % Mg_2Si alloy subjected to thermomechanical treatment (TMT) was examined. Transmission electron microscopy observations, tensile tests and electrical conductivity measurements were carried out in order to correlate microstructural features to properties on each TMT step. It was established that pre-aging at room temperature affected the morphology of dislocation structure induced by next cold deformation. The observed transition from cellular to homogenous dislocation distribution was explained by the different stability of zones produced by pre-aging of different duration. Natural aging suppressed recovery processes during post-deformation artificial aging, especially after prolonged storage after quenching and at lower aging temperature. It influenced the morphology of precipitates produced by post deformation artificial aging also. The overall effect of TMT involving prior-deformation natural aging in the scheme, on hardness, tensile properties and electrical conductivity is discussed based on experimental microstruture observations.

Keywords: AlMgSi alloys, thermomechanical treatment, precipitation, microstructure, transmission electron microscopy

1. Introduction

A storage at ambient temperature often precedes in practice the standard procedures of plastic deformation and artificial aging of thermomechanically treated Al-Mg-Si alloys (6000 series). The schedule of thermomechanical treatment (TMT) in this case involves two-step aging and so the natural pre-aging duration becomes one of the manufacturing process variables. Microstructural changes in 6000 series alloys during Quenching-Natural Aging-Deformation-Artificial Aging (Q-NA-D-AA) scheme are more complicated and less investigated than the conventional T8 Quenching-Deformation-Artificial Aging (Q-D-AA) scheme. Most of investigations show that cold deformation after quenching suppresses GP-zones formation and accelerates the nucleation and growth of intermediate phases [1-5]. The properties of alloys after TMT including room-temperature pre-aging were studied for low to moderate (5-30%) [6-7] and severe (70-90%) degrees of deformation [6, 8-10].

The objective of this paper is to investigate the development of microstructure on each step of TMT including ambient temperature pre-aging and to correlate it to mechanical properties and electrical conductivity of Al-Mg-Si alloys.

2. Experimental procedure

Continuously cast and rolled 9,5 mm rods (Southwire production process) of two commercial Al-Mg-Si alloys (6201) were used for investigation. The chemical composition of alloys was like follows:

For Alloy 1: 0,62Mg, 0,56Si, 0,05Cu, 0,23Fe, bal.Al, in mass %;

For Alloy 2: 0,63Mg, 0,59Si, 0,12Cu, 0,13Fe, bal.Al, in mass %.

Homogenization of the parent material was carried out at 520°C for 4 hours, followed by cold water quenching. Quenched rods were naturally aged for different periods of times. Subsequently, Alloy 1 was 95% cold drawn to wire and Alloy 2 - 95% cold rolled to band without intermediate annealing. Two regimes of artificial aging were applied - 6h at 130°C and 6h at 170°C. They were chosen in order to clarify the different balance between electrical conductivity and tensile strength found in our previous work [10].

Microstructural investigations were carried out by means of transmission electron microscopy (TEM) on thin foils of Alloy 2. Details on tensile tests and measurements of electrical resistance performed on Alloy 1 are given in our earlier work [10]. Vickers Hardness was measured on Alloy 2 using Leitz Miniload 2 at 4,903N load.

3. Results and discussion

3.1. Structure and properties in Q and Q+NA temper

Process of recrystallization took place in the structure of severely deformed rod during 520^{o}C-solution treatment (Fig.1). Broken spheroidized AlFeSi intermetallides and rarely large non-dissolved Mg_2Si particles were randomly distributed in the quenched structure. Particles of this type were observed at the next TMT steps also, but they were fragmented by the cold deformation. The quenched α_{Al} solid solution demonstrated a faint contrast of spherical dark spots, especially sharp around extinction contours. This specific TEM contrast is usually due to strain fields around clusters of alloying atoms. It is known that solute clustering of Mg and Si atoms begins in AlMgSi alloys shortly after or even during quenching [11-12]. In our case it initiated obviously during thin foils preparation (up to 30 minutes after quenching). The contrast of Mg-Si clusters became stronger with NA duration. Clusters remained spherical for 94h NA (Fig.2) but after 500h NA some elongated zones (length up to 50nm, diameter up to several nm) could be detected (Fig.3).

The formation of Mg-Si clusters during NA resulted in higher hardness (Table 1) and strength (Table 2). The dependence of both characteristics on NA duration is typical for naturally aged AlMgSi alloys: rapid increase over the first 4 days followed by asymptotic slow down. The quenched specimen reached the strength of parent rod (Rm=230 Nmm^{-2}) after the first 5-6 days.

3.2. Structure and properties in Q+D and Q+NA+D temper

95% deformation of freshly quenched (Q) or shortly pre-aged (Q+NA^{4h}) specimens produced dislocation structure of cellular type: cells of diameter up to 1 μm with small number of dislocations in the interior and relatively wide dislocation walls (Fig.4). The contrast of cells interior was typical for homogenous solid solution. It indicated that zones, which had nucleated during Q or Q+NA^{short} tempers reversed under the severe deformation.

After longer NA the cellular structure became imperfect, the quantity of dislocations in cell interior increased and their density in cell walls decreased. This led to smaller mis-orientation of neighboring cells, to more homogenous distribution of dislocations and finally to complete disappearance of cells. In Q+NA^{500h}+D specimen only homogenous three-dimensional dislocations distribution was observed (Fig.5).

Table 1. Hardness HV in Q+NA temper, Q+NA+D temper, Q+NA+D+AA temper

NA duration [h]	HV^{Q+NA} [daN/mm^2]	ΔHV^{NA} [%]	HV^{Q+NA+D} [daN/mm^2]	ΔHV^{D} [%]	$HV^{Q+NA+D+AA130^\circ}$ [daN/mm^2]	$HV^{Q+NA+D+AA170^\circ}$ [daN/mm^2]
4	56		110	96	96	78
24	66	18	120	82	100	67
94	68	21	120	76	96	74
500	78	39	110	41	113	103

Table 2. Ultimate tensile strength Rm in Q+NA and Q+NA+D temper; elongation A in Q+NA+D temper and electrical conductivity γ in Q+NA+D temper.

Temper and NA duration [h]	Rm^{Q+NA} [MPa]	Rm^{Q+NA+D} [MPa]	ΔRm^{D} [%]	$À^{Q+NA+D}$ [%]	$ã^{Q+NA+D}$ [m.ohm^{-1}mm^{-2}]
Q	-	144	-	-	-
Q+D	-	354	146	4,0	28,30
Q+NA4+D	190	372	96	3,5	27,80
Q+NA24+D	217	396	82	3,0	27,30
Q+NA94+D	230	395	72	2,5	27,64
Q+NA500+D	231	390	69	1,5	27,44

The change of dislocation distribution with NA duration could be related to the development of clustering process, although no zones were distinguished in the highly deformed structure. Small sized clusters produced by short NA probably were easily destroyed by moving dislocations and could not affect the character of dislocation substructure. After long NA the number of Mg-Si clusters was high and they were stable enough to be destroyed. They pinned the dislocation lines at short distances thus preventing them from movement, interaction and annihilation, i.e. from formation of dislocation tangles (cell walls) and dislocation free volumes (cell interior). The process of deformation of $Q+NA^{long}$ specimens propagated by increase of density of three-dimensionally distributed dislocations and resulted in the observed homogenous dislocation substructure.

The effect of pre-aging on tensile strength of 6201 alloys deformed in Q or Q+NA tempers is shown in Table 2. It is seen that the absolute value of strength Rm^{Q+NA+D} increased, but the strength increment ΔRm^{D} decreased with NA duration. Similar dependence was observed for hardness increment ΔHV^{D} (Table 1). Both dependencies showed that the effect of clusters on work hardening in Q+NA+D specimens was smaller compared to the effect of dislocation substructure and that the cell substructure (after short NA) was more effective for strengthening compared to the three-dimensional substructure (after long NA).

Fig.1. Microstructure of parent rod of Alloy 2 after water quenching from 520°C (Q temper)

Fig.2. Microstructure of quenched rod of Alloy 2 after 94 hours aging at ambient temperature (Q+NA^{94} temper)

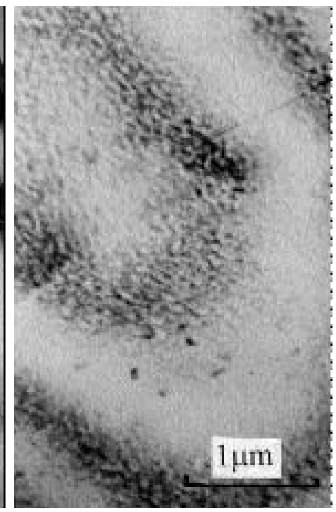

Fig.3. Microstructure of quenched rod of Alloy 2 after 500 hours aging at ambient temperature (Q+NA^{500} temper)

Work hardening resulted in some decrease of electrical conductivity and elongation in all investigated tempers (Table 2). The small variation of conductivity with NA duration probably was due to some balance between formation and growth of new clusters (zones) and reversion of a part of clusters already existing on this TMT step.

3.3. Structure and properties in Q+NA+D+AA temper

Artificial aging of 6201 alloys deformed in Q temper led to intensive precipitation in the whole structure (Fig.6, 7). Particles of two morphologies were distinguished in Q+D+AA temper: needle-like, with length-diameter ratio up to 10:1 and rod-like, with significantly smaller length-diameter ratio. At both AA temperatures the diameter of needle-like particles was under 15 nm, the length – 150 nm and of rod-like ones – 80 nm, resp.200 nm. Both types of particles showed preferred crystallographic orientation along $<001>_{Al}$ direction. On the base of morphological features we determined the needle-like phase as metastable coherent $\beta''(Mg_2Si)$ and the rod-like one – as semi-coherent β' (Mg_2Si).

Fig.4. Microstructure of Alloy 2 cold rolled to band immediately after quenching (Q+D temper)

Fig.5. Microstructure of Alloy 2 cold rolled to band after 500 hours aging at ambient temperature (Q+NA^{500}+D temper)

Fig.6. Microstructure of Alloy 2 cold rolled in quenched condition and subjected to artificial aging at 130 °C (Q+D+AA^{130} temper)

Pre-aging before deformation influenced dislocation structure as well as precipitation in Q+NA+D+AA scheme. The preliminary NA suppressed recovery processes, especially after prolonged NA and at lower AA temperature. The structure of Q+NA^{94h}+AA specimens showed signs of polygonization at both AA temperatures, but recrystallization was supressed. It missed at 130°C and the number of recrystallized grains at 170°C was strongly reduced (Fig.8). Almost no signs of recovery were observed in 500h pre-aged specimens, the dislocation density remained high, the polygonization was incomplete and no recrystallization developed even at 170°C (Fig.9).

Pre-aging up to 94h did not influence the type of phases precipitated in AA step, but affected their size and shape after NA^{long}, especially at $170^{o}C$. After 500h NA the length of β''-particles decreased up to 100 nm at both temperatures, the length of β'-particles – up to 150 nm. The diameter of β' increased up to 120 nm and their shape changed to round one at $170^{o}C$ (Fig.9). The diameter of β'' remained constant at both temperatures, but the number of particles decreased. This could be related to the stabilization of zones, which prevented transition to β -phase formation.

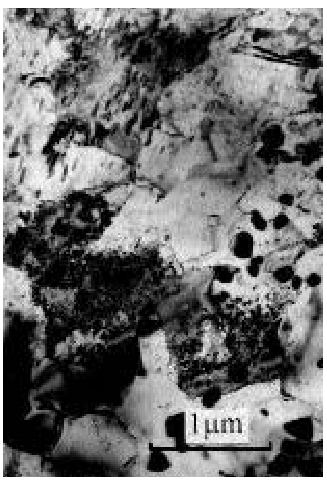

Fig.7. Microstructure of Alloy 2 cold rolled in quenched condition and subjected to artificial aging at $170^{o}C$ ($Q+D+AA^{170}$ temper)

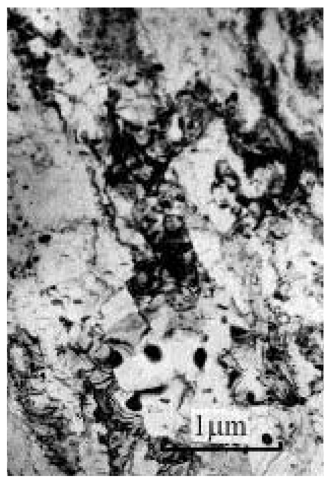

Fig.8. Microstructure of Alloy 2 cold rolled in naturally aged for 94 hours condition and subjected to artificial aging at $170^{o}C$ ($Q+NA^{94}+D+AA^{170}$ temper)

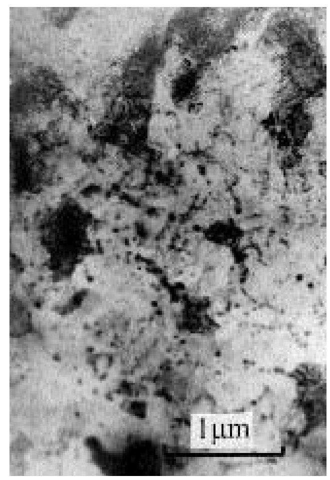

Fig.9. Microstructure of Alloy 2 cold rolled in naturally aged for 500 hours condition and subjected to artificial aging at $170^{o}C$ ($Q+NA^{500}+D+AA^{170}$ temper)

The influence of pre-aging on dislocation structure and precipitation after AA could be explained by the different level of solid solution decomposition and by the differences in dislocation substructure existing prior AA. The dislocation cells produced by deformation in Q+D and $Q+NA^{short}+D$ tempers supplied ready nuclei for polygonization and recrystallization. The accumulated energy of the three-dimensional dislocation substructure of $Q+NA^{long}+D$ temper was lower, the complexes of dislocations did not offer nuclei for polygonization and this prevented recovery processes.

The changes of structure caused by the two superimposed processes – precipitation and recovery - reflected on properties at the final TMT stage (Table 3). AA contribution to strengthening was negligible, especially at 170^{o}C (Table 1,2), when recovery processes probably eliminated the hardening effect of precipitation.

Table 3. Ultimate strength Rm, elongation A and electrical conductivity after TMT scheme Q+NA+D+AA with different NA duration

AA temperature	130^{o}C			170^{o}C		
NA [h]	Rm[MPa]	A [%]	ã [m.ohm^{-1}m^{-2}]	Rm [MPa]	A [%]	ã [m.ohm^{-1}mm^{-2}]
4	395	6,0	28,56	342	4,3	31,46
24	395	6,1	28,54	343	4,4	31,40
94	397	6,5	28,46	346	4,8	31,22
500	402	7,7	28,14	355	6,1	30,29

This result confirmed the available data on AA of other severely deformed alloys [6,13]. Electrical conductivity and plasticity increased after AA in all schemes. Electrical conductivity, which is an indicator for solid solution status, decreased with NA duration at both investigated AA temperatures, remaining higher at 170^{o}C. This fact confirmed our conclusions, based on microstructural observations, about the easier transition from zones formation to phase aging in Q+NA^{short}+D+AA and in Q+D+AA temper. On contrary, the stabilization of zones in Q+NA^{long}+D+AA temper reduced conductivity due to the hard phase precipitation.

4. Conclusion

Recent studies on precipitation sequence and kinetics in Al-Mg-Si system [12,13,14] confirmed experimentally that Si and Mg clusters and Mg-Si co-clusters developed during natural aging. From the chemical point of view co-clusters and GP-I zones are identical, (they differ in size and density of solute atoms only) though there is some ambiguity and disagreement in terminology. Kinetics parameters, derived by Gutta and Lloyd [14] from DSC curves for super-purity Al-0,8%Mg-0,9% Si showed that the natural aging continued for a long time at room-temperature and that the temperature of the peak, associated with co-clustering reaction, shifted to higher temperature range with aging duration. Our results permit conclusion to be made that co-clusters grew most probably to zones during room temperature storage.

The mechanism of preventing clusters/zones formation by cold deformation of quenched alloys has been established long ago [15]. The acceleration of aging process in cold deformed supersaturated solid solutions has been observed also. It has been explained by clustering of solutes around dislocations of increased density and shifting the aging reactions to lower temperatures [5], or by reducing the activation energy of β′ intermediate phases formation and preferential nucleation on dislocations [4]. These facts explain the precipitation of metastable phases during aging at 130°C - temperature, which lies in the range of GP-zone formation [14].

The mode of interaction of pre-existing clusters, co-clusters or zones with dislocations, introduced in Q-NA-D temper by severe deformation, depended on their stability, which should be higher the longer the pre-aging time was. Small unstable clusters (zones) of shortly pre-aged alloys dissolved and increased the solute concentration of the matrix. The homogenous dislocation distribution found in our work in 500h naturally aged and deformed specimens proved indirectly the gradually increasing stability of zones on this stage. Similarly, in an early work Ber [8] proposed that the zones formed during 24hrs natural aging of Al-0,73Mg-0,57Si alloy were successfully stabilized by 90% cold work**.** Our TEM investigations indicated that the duration of natural aging determined the substructure after deformation and controlled the subsequent processes of precipitation and formation of substructures at the final aging treatment.

The mutual influence of structural changes due to the two superimposed processes – precipitation and recovery - which took place in different extent during investigated schemes of TMT was decisive for mechanical and electrical properties. Higher hardness and strength, but lower conductivity obtained in alloys pre-aged for long time was supposed to be due to the stabilization of co-clusters (zones) and dislocation substructure. However, further study including quantitative estimation of volume fraction, number density, size and other parameters of precipitating phases should be carried out to clarify and determine more precisely the relation between precipitation sequence and strength.

References

1. H.L.Lee, Win-Hwa, S.L.Chan, *Scripta Metall*, 25 (1991) 2165
2. T. Christman, A.Needleman, S.Nutt, S,Suresh, *Mater. Sci Eng*. A, 107 (1989) 49
3. K. Matsuda, M.Terasaki, S.Tada, S.Ikeno, *J. Japan Inst. Light Metals*, 3 (1993) 127
4. T. Hirata, S.Matsuo, Trans. *JIM*, 15 (1973) 1
5. P.C.LoNg, Y.Ohmory, K.Nakai, Materals Trans. *JIM*, 4 (6) (2000) 690
6. M.H.Rabinovich, “Thermomechanical Treatment of Aluminium Alloys”,

Moskva,, Mashinostroenie, 82 (1972) 51 (in Russian)
7. L.S. Toropova, D. Uskin, M.Harakterova, *Non-ferrous Metals* 8 (1991) 93 (in Russian)
8. L.B. Ber, Y.Vainblat, V.Davidov, S. Hauyrov, N. Schtegoleva, *FMM*, 36 (3) (1973) 583
9. P. Ghosh, Z. Metallkunde, 82 (9) (1991) 727
10. Z. Martinova, D. Damgaliev, M.Hirsh, *JMM*, 38 (1-2)B (2002) 61.
11. I. Kovachi, J.Lendvai, E.Nagy, *Acta Met*., v.20, July, (1972)
12. M. Murayama, K. Hono, K.Kikuchi, *Mat. Sci. Eng*. A, 250 (1997) 127
13. G.A.Edwards, K.Stiller, G.l.Dunlop, M.G.Couper. *Acta Mater*., 46 (11) (1998) 3893
14. A.K.Gupta, D.L.Lioyd, *Met. Mat.Trans*. A,.30 (1999) 879
15. K.V.Chuistov, "Ageing of Metallic Alloys, Kiev", *Naukova dumka*, 1985 (in Russian)