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Zhenjie Cui

Institute of Metal Research Chinese Academy of Sciences

Haichang Jiang (✉ hcjiang@imr.ac.cn)

Institute of Metal Research, Chinese Academy of Science

Duo Zhang

Institute of Metal Research Chinese Academy of Sciences

Yuanyuan Song

Institute of Metal Research Chinese Academy of Sciences

Desheng Yan

Institute of Metal Research Chinese Academy of Sciences

Lijian Rong

Institute of Metal Research Chinese Academy of Sciences

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Study of natural aging on nanoparticles and their strengthening effect in Al-Mg-Si alloy

Zhenjie Cui ^{1,2,3}, Haichang Jiang ^{1,3*}, Duo Zhang ^{1,2,3}, Yuanyuan Song ^{1,3}, Desheng Yan ^{1,3}, Lijian Rong ^{1,3*}

Abstract

Atom probe tomography (APT) and transmission electron microscopy (TEM) were employed simultaneously to study nanoparticles and their strengthening effect in artificial aging (AA) Al-Mg-Si alloy with different natural aging (NA). We discovered that NA leads to formation of different type of nanoparticles meanwhile affect their fraction and dimension. Separation of the contribution from different types of nanoparticles to the alloy strength is conducted. In LNA-AA, β' with a volume fraction of 0.16% contributes 54MPa to the yield strength while 0.14% β'' contributes 101MPa, which fully demonstrates that β'' is the main strengthening phase. The thermodynamic analysis was conducted for further research and the differential scanning calorimeter (DSC) results show that LNA forms more numerous clusters I than SNA and consumes more vacancies and solute atoms then does not have enough driving force to precipitate clusters II which are the nucleation sites of nanoparticles. When there is a certain distance between the nucleation sites, preferential growth of few β'' occur then converted to β' . The solute atoms in the region nearly which there is no nucleation site do not have enough driving

force to diffuse and precipitate but stay in the matrix, reducing the volume fraction of the nanoparticles.

Keywords: Al-Mg-Si alloy; natural aging; nanoparticles

1. Background

The precipitation sequence of the nanoparticles in the heat-treatable reinforced Al-Mg-Si alloys is generally considered as a supersaturated solid solution (SSSS) → Solute clusters → GP zones → β'' → $\beta'/\text{Type A/Type B/Type C}$ → β [1-4]. Previous studies have found that natural aging (NA) strongly affects the nanoparticles after artificial aging (AA) and negatively affects the alloy's properties. Røyset, Jostein et al. have revealed that NA 3600 hours leads to a lowering in the number density of AA nanoparticles and a decrease in the yield strength by 30MPa in an Al-Mg-Si alloy [5]. Tao, G. H. et al. proposed that NA 30 days reduces the AA peak hardness by 13HV in an Al-1.0wt% Mg-0.5wt% Si alloy which is related to the broad precipitate length distribution and appearance of large-sized β'' [6]. Wenbin Tu et al. reported that the AA hardening rate was dramatically slowed down by NA 14 days because NA clusters delayed the formation of β'' , leading to a drop of the peak hardness by 30HV in an Al-0.5at.% Mg-1.0at.% Si alloy [7]. Recently, we discovered that the NA also leads to the appearance of different types of nanoparticles so that the fractional and dimensional variation of each kind of nanoparticle can further affect the magnitude of precipitation strengthening. Transmission electron microscopy

(TEM) is effective in determining the structure of nanoparticles [8, 9]. However, the distinguishing of different nanoparticles is difficult only by TEM due to the size limitation of clusters and GP zones as well as the similar needle-like morphology of β'' and β' . The three-dimensional atom probe (3DAP) provides a near-atomic resolution and a capacity to record statistically meaningful data on the compositions, size, and volume fraction of nanoparticles [10, 11]. In this paper, TEM and 3DAP were employed simultaneously to reveal the fractional and dimensional variation of different nanoparticles in an artificial aging Al-Mg-Si alloy with different NA. We calculated the strengthening effect of the contribution from different AA nanoparticles and consequently reveal the essence of decreased strength.

2. Methods

Material

An Al-Mg-Si alloy with a nominal composition of Al-0.95wt% Mg-0.8wt% Si was prepared to study. The cast ingots were homogenized at 550°C/10h and forged into a bar with a diameter of 40 mm. The aging treatments started with a solid solution treatment of 4h at 525°C, water-quenched to room temperature. After the solution, samples were stored at room temperature for a different time before experiencing a 175°C/8h AA.

Characterization

The tensile tests were performed on Shimadzu Autograph Dcs-10T Electronic universal testing machines. The TEM sample was prepared by a Struers Tenupol-5 type

twin-jet electropolishing with a 30% methanol solution of nitric acid, and the microstructure was observed using a JEM 2100F transmission electron microscope. The APT samples of NA were prepared by electrolytic polishing and were completed within 10-15min. NA will not occur again in the APT sample after the preparation is completed which was proved by [12]. The APT experiments were carried out on LEAP 5000 XR with laser mode at a temperature of 25 K and a laser pulse frequency of 200 kHz. The 3D reconstruction and analysis are performed using IVAS 3.8.2 software. The thermal analysis was carried out on Q1000 DSC and the heating rate is 10 °C/min.

3. Results and discussion

The NA hardness curve of the Al-Mg-Si alloy is shown in Fig.1 (a), which is drawn by averaging seven hardness indentation data. The NA hardness firstly increases fast and then tends to be stable as the NA time reaches 24h. Samples of NA10min, 2h, 24h indicated by the black arrows on the NA hardness curve are selected for investigation, named SNA, MNA, and LNA, respectively. After experiencing a 175°C/8h AA, the engineering stress-strain curves of SNA-AA, MNA-AA, and LNA-AA are presented in Fig.1(b). The yield strengths of SNA-AA, MNA-AA, and LNA-AA are 280 MPa, 253MPa, and 230 MPa, respectively. According to the variation of strength, LNA-AA loses about 18% strength compared to the SNA-AA. It can be seen that NA makes the alloy strength obviously decrease.

Fig.1 (a) NA hardness curves of the Al-Mg-Si alloy. (b) Engineering stress-strain curves of SNA-AA, MNA-AA, and LNA-AA.

The TEM bright-field images of SNA-AA, MNA-AA, and LNA-AA with the electron beam parallel along the $\langle 001 \rangle_{\text{Al}}$ are shown in Fig.2 (a)~(c). The HRTEM images and FFTs viewed along with the $\langle 001 \rangle_{\text{Al}}$ are shown in Fig.2 (d)~(f). The SNA-AA has a fine and uniform distribution of the nanoparticles as shown in Fig.2 (a). The nanoparticles become coarser in MNA-AA and nanoparticles over 100nm appeared. The length of nanoparticles of LNA-AA becomes longer and a few considerably elongated nanoparticles exceed 200nm and the number of nanoparticles is significantly reduced. The FFT in Fig.2 (d) agrees well with the monoclinic β'' [8]. Fig.2 (e), (f) show the FFTs of the considerably elongated nanoparticles which is consistent with hexagonal β' [9]. β'' and β' coexist in MNA-AA and LNA-AA. The TEM images realize the statistics of average radius (R) of β'' and β' and the detailed data are shown in Table 1. The R of β'' is 1.93nm in SNA-AA and 2.15nm in LNA-AA, while for β' is 2.39 nm in MNA-AA and 3.23nm in LNA-AA. With the extension of NA, β'' gets coarser and some β' appear.

Fig.2 TEM bright-field images of (a) SNA-AA, (b) MNA-AA and (c) LNA-AA. HRTEM images and FFTs of the β'' and β' of (d) SNA-AA, (e) MNA-AA, and (f) LNA-AA, and the images were taken with the electron beam along the $\langle 001 \rangle_{\text{Al}}$ orientation.

Fig.3(a), (b) show the 3NN distribution of the experimental and randomized data of Mg and Si in SNA and Mg + Si in LNA. Comparing (a) and (b), Mg and Si solute atoms hardly aggregated in SNA while aggregated significantly in LNA. Fig.3 (c) and (d) show the 3D atom maps of evenly distributed Mg (red), Si (purple) of SNA, and Mg+Si clusters of LNA. Obviously, the formation of NA clusters in LNA is an important reason for the degradation of AA strength. It is commonly believed that the formation of NA clusters depletes the super-saturated solutes in the matrix as well as not being favorable nucleation sites for further nanoparticles [13, 14]. Instead, clusters that formed at early AA were confirmed to play the role of critical nuclei for β'' [6].

Fig.3 3D atom maps of Mg (red) and Si (purple) and the 3NN distribution of experimental and randomized data in the Al-Mg-Si alloy: (a), (c) SNA, (b), (d) LNA.

The 3D atom maps of Mg(Orange), Si(purple), and a 10.0 at% Mg+Si iso-concentration surface of SNA-AA, MNA-AA, and LNA-AA are shown in Fig.4. The nanoparticles in the MNA-AA and LNA-AA are coarsened and reduced in number compared with SNA-AA which is consistent with the TEM results in Fig.2. The inter-particle spacing in SNA-AA is small, that is, the nucleation sites of β'' are close to each other. The inter-particle spacing in MNA-AA becomes bigger and nanoparticles in LNA-AA are sparsely distributed. As reported in Ref. [6], long NA leads to the preferential growth and the formation of large-sized β'' which created a large inter-particle spacing.

Fig.4 The 3D atom maps of Mg, Si and a 10.0 at% Mg+Si iso-concentration surface of (a) SNA-AA, (b) MNA-AA (c) LNA-AA, Al(Green), Mg(Orange), Si(purple). The nanoparticles marked in yellow in (a), (c) were carried out 1D concentration analysis in the following part.

The maximum separation method is applied to distinguish the different nanoparticles of SNA-AA, MNA-AA, and LNA-AA according to previous studies with $d_{\max}=0.7\text{nm}$ and $N_{\min}=10$ [10, 15]. The statistical data of different nanoparticles have been listed in Table 1. The Clusters and GP zones at AA state are collectively referred to as spherical nanoparticles, abbreviated as GC. It is considered that the supersaturation of solute and

presence of vacancies is necessary for nucleation of GC during AA in small local regions of the Al lattice between the large nanoparticles [16]. The volume fraction of GC is increasing with the extending of NA time. A low fraction of GC in SNA-AA can be attributed to greater consumption of the solute by densely distributed β'' with a volume fraction of 0.74%, thus reduced solute supersaturation of the matrix between the nanoparticles. The falling of the volume fraction of β'' from 0.74% in SNA-AA to 0.21% in MNA-AA is dramatic and 0.22% β' appear in MNA-AA. Obviously, β' precipitates at the expense of β'' . The β'' and β' become coarser in LNA-AA and the overall volume fraction drop to 0.37% including 0.14% β'' and 0.16% β' . Additionally, it can be obtained by the ‘Solute in matrix’ that more solute atoms remain in the matrix at samples with longer NA. This is probably because those numerous NA clusters deplete the supersaturated solutes leading to the driving force of precipitation of solute atoms being reduced so that more of them remain in the matrix.

Table 1. The relative data of nanoparticles in the different states

Samples	Nanoparticles	R/nm	V_f^a	Solute in
SNA-AA	GC	0.82 ± 0.13^a	0.02%	0.45 wt% Mg 0.26 wt% Si
	β''	1.93 ± 0.16^b	0.74%	
	Sum	—	0.76%	
MNA-AA	GC	0.77 ± 0.12^a	0.05%	0.52 wt% Mg 0.31 wt% Si
	β''	1.95 ± 0.06^b	0.21%	
	β'	2.39 ± 0.1^b	0.22%	
	Sum	—	0.48%	
	GC	0.43 ± 0.08^a	0.07%	

LNA-AA	β''	2.15 ± 0.11^b	0.14%	0.61 wt% Mg
	β'	3.23 ± 0.27^b	0.16%	0.35 wt% Si
	Sum	-	0.37%	

^a Parameters determined from the APT data.

^b Average radius R of β'' and β' were measured in the bright-field images in Fig. 2.

Fig.5 (a), (b) demonstrate the 1D concentration profiles of representative β'' and β' marked in yellow in Fig.4 (a) and (c), respectively. The 1D concentration profiles which were measured by a selected cylinder with a moving step of 0.3nm distinguished β'' and β' in terms of their compositions. The Mg/Si ratio of β'' and β' are 0.83 and 1.84, and the content of Al is 32.13 at.% and 19.22 at.%, respectively. It indicates that the transformation process from β'' to β' is dominated by Mg, and Al is continuously discharged which is consistent with Refs [10].

Fig.5 The representative 1D concentration distribution of (a) β'' and (b) β' marked in yellow in Fig.4 (a), (c), respectively. The 1D concentration profiles were measured by a selected cylinder with a moving step of 0.3nm.

Separation of the contribution from different types of nanoparticles to the alloy strength is conducted. The yield strength σ_y of the Al-Mg-Si alloys consists of precipitation hardening σ_p , solid solution strengthening σ_s and lattice resistance σ_i . The overall yield strength calculation can be achieved by adding the various contributions as equation (1) [17-20]. The σ_p can be explained mainly from the modulus strengthening σ_{mod} , chemical strengthening σ_{chem} , coherency strengthening σ_{coh} and σ_{GC} which is the contribution from GC, as shown in equation (2) [17, 20-22]. Detailed information of equations and parameters for calculation is provided by the supplementary information. Input the data of Table 1 and Table S1, the calculation results of the various contributions are shown in Table 2. At the SNA-AA state, the yield strength is mainly derived from $\sigma_{\beta''}$ which is 221 MPa. At the MNA-AA state, β' with a volume fraction of 0.22% contributions 56MPa to the yield strength while 0.21% β'' contributes 119MPa. At the LNA-AA state, β' with a volume fraction of 0.16% contributions 54MPa to the yield strength while 0.14% β'' contributes 101MPa. It fully demonstrates that β'' is the main strengthening phase in Al-Mg-Si alloy and the appearance of large-sized β' is a major cause of the negative NA effect. σ'_y is the yield strength obtained by the tensile test in Fig.1(b). σ'_y is similar to the calculation results which proves that the selection of the calculation formula is reasonable. Importantly, the relationship between strength and the size and volume fraction of nanoparticles is not linear which means that the comparison of strength increments per unit volume fraction at different states is not meaningful.

$$\sigma_y = \sigma_p + \sigma_s + \sigma_i \quad (1)$$

$$\sigma_p = \sigma_{cut} = \sigma_{mod} + \sigma_{chem} + \sigma_{coh} + \sigma_{GC} \quad (2)$$

Table 2. The calculation results of the various contributions

Strength (MPa)		SNA-AA	MNA-AA	LNA-AA
$\sigma_{\beta''}$	σ_{mod}	14.6	7.8	6.5
	σ_{chem}	0.7	0.4	0.3
	σ_{coh}	206.1	110.4	94.6
	SUM	221	119	101
$\sigma_{\beta'}$	σ_{mod}	—	14.4	13.3
	σ_{chem}		1.1	0.7
	σ_{coh}		40.5	40.1
	SUM		56	54
σ_{GC}		2.3	3.6	4.2
σ_s		44.0	49.1	53.8
σ_i		10	10	10
σ_y		277	238	223
σ'_y		280	253	230

The thermodynamic analysis was conducted for further research and the DSC flow curves of SNA and LNA are shown in Fig.6. The exothermic peak at 50-100°C should be the formation of the clusters II which can be the nucleation sites of β'' [23, 24]. The trough at 200-250°C can be related to the dissolution of unstable clusters I formed during NA [25]. The peak around 250°C and 300°C are normally associated with the formation of β'' and β' , respectively [23, 25]. LNA has no exothermic peak of clusters II. Combining the APT results in Fig.3, it can be concluded that the LNA forms a lot of clusters I and

consumes more solute atoms then does not have enough driving force to precipitate clusters II. The trough of LNA at 200-250°C caused by the dissolution of clusters I delayed the precipitation process of nanoparticles, leading to the peak of β'' move toward a higher temperature.

Fig.6 DSC flow curves of the Al-Mg-Si alloy at SNA and LNA.

Based on the above discussion, the AA process of SNA and LNA have been summarized through the schematic diagram in Fig.7. Solute atoms of SNA hardly aggregated and are evenly distributed as shown in Fig.3 (a), (c). During initial AA, sufficient solute atoms quickly precipitate a large amount of cluster II which acts as nucleation sites to form fine and dense β'' during the peaking AA. LNA formed numerous clusters I which will dissolve into the matrix during initial AA. Few clusters II formed during initial AA as shown in Fig.6. It takes time to dissolve cluster I and only when the solute supersaturation in the matrix reaches the nucleation condition, the formation of cluster II can occur. Under this condition, the distance between clusters II is usually very large. With the extending AA time, preferential growth of β'' on the few clusters II occur

then converted to β' . This preferential growth process is easier to capture the surrounding solute atoms, which directly restricts the growth of the GC. Additionally, the solute atoms in the region nearly which there is no nucleation site do not have enough driving force to diffuse and precipitate but stay in the matrix. This is the fundamental reason for the significant decrease in the volume fraction of the nanoparticles in LNA-AA.

Fig.7 The schematic diagram of the AA process of SNA and LNA.

4. Conclusion

- (1) We discovered that NA leads to the formation of different types of nanoparticles in AA Al-Mg-Si alloy meanwhile affect their fraction and dimension. The volume fraction of GC and β' is increasing with the extending of NA time, while the volume fraction of β'' is continuously declining from SNA-AA to LNA-AA.

The β'' and β' become coarser in LNA-AA and the overall volume fraction drop from 0.74% to 0.37%.

(2) Separation of the contribution from different types of nanoparticles to the alloy strength is conducted. At the SNA-AA state, the yield strength is mainly derived from $\sigma_{\beta''}$ which is 221 MPa. At the LNA-AA state, β' with a volume fraction of 0.16% contributes 54MPa to the yield strength while 0.14% β'' contributes 101MPa. It fully demonstrates that β'' is the main strengthening phase in Al-Mg-Si alloy and the appearance of large-sized β' is a major cause of the negative NA effect.

(3) LNA formed numerous clusters I which will dissolve into the matrix during initial AA. Few clusters II formed during initial AA. With the extending AA time, preferential growth of β'' on the few clusters II occurred then converted to β' . This preferential growth process is easier to capture the surrounding solute atoms, which directly restricts the growth of the GC. Additionally, the solute atoms in the region nearly which there is no nucleation site do not have enough driving force to diffuse and precipitate but stay in the matrix. This is the fundamental reason for the significant decrease in the volume fraction of the nanoparticles in LNA-AA.

Abbreviations

3D: Three-dimensional; APT: Atom probe tomography; HRTEM: High-resolution transmission electron microscopy; TEM: Transmission electron microscopy; UA: Under aging; DSC: differential scanning calorimeter.

Declarations

Availability of data and materials

All datasets are presented in the main paper or in additional supporting files.

Competing interests

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Authors' contributions

Zhenjie Cui: Conceptualization, Methodology, Investigation, Formal analysis, Validation, Writing - Original Draft.

Haichang Jiang: Conceptualization, Writing - review & editing.

Duo Zhang: Methodology, Validation.

Yuanyuan Song: Resources, Methodology.

Desheng Yan: Resources.

Lijian Rong: Supervision, Conceptualization, Writing - review & editing.

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Authors' information

¹ *CAS Key Laboratory of Nuclear Materials and Safety Assessment, Institute of Metal Research, Chinese Academy of Science, Shenyang 110016, China*

² *School of Materials Science and Engineering, University of Science and Technology of China, Shenyang 110016, China*

³ *Shi-changxu Innovation Center for Advanced Materials, Institute of Metal Research, Chinese Academy of Sciences, Shenyang 110016, China*

Zhenjie Cui: Email address: zjcui17s@imr.ac.cn

Duo Zhang: Email address: dzhang15s@imr.ac.cn

Yuanyuan Song: Email address: songyuanyuan@imr.ac.cn

Desheng Yan: Email address: dsyan@imr.ac.cn

* Corresponding author:

Prof. Haichang Jiang, Tel: +86 024 2397 1976; Email address: hcjiang@imr.ac.cn

Prof. Lijian Rong, Tel: +86 024 2397 1979; Email address: ljrong@imr.ac.cn

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