Different aging behaviors at surface layer and central region of a die-casting A380 alloy during heat treatment

Zi-hao Yuan¹, Zhi-peng Guo¹, *Shou-mei Xiong^{1,2}

1. School of Materials Science and Engineering, Tsinghua University, Beijing 100084, China

2. State Key Laboratory of Automobile Safety and Energy, Tsinghua University, Beijing 100084, China

Abstract: Microstructural and hardness evolutions of a vacuum-assistant die-cast A380 (Al-8.67wt.%Si-3.27wt.%Cu) alloy during heat treatment were investigated. Isothermal DSC test at 200 °C revealed that the precipitation reaction in the surface layer was faster than that in the central region. This corresponded with the hardness evolution that the surface layer hardened faster. The hardness increment in the surface layer was higher than that in the central region. Further experimental evidences indicated that the differences were due to the different amounts of heterogeneous nucleation sites for precipitation in the two parts. The influence of the characteristic as-cast microstructure on the artificial aging process is analyzed and discussed in detail.

Key words: high pressure die casting (HPDC); heat treatment; macro-segregation; kinetics; precipitation; externally solidified crystals (ESCs)

CLC numbers: TG146.21/249.2

Document code: A

Article ID: 1672-6421(2017)06-506-07

High pressure die casting (HPDC) is one of the most efficient casting processes for mass production of precisely shaped components^[1-2]. However, the gas entrapment during the turbulent filling leads to the formation of gas pores in die castings, which could diminish the mechanical properties. Besides, these gas pores expand during the subsequent solution treatment (1–8 h at 510–525 °C^[3-5]), leading to the formation of unacceptable surface blisters^[6]. From a traditional viewpoint, the die castings are not suitable for heat treatment.

However, in the past few decades, new technology advancement, such as vacuum-assisted die casting and pore-free die casting, could reduce the porosity significantly. Besides, recent studies^[7-10] demonstrated that a low temperature solution treatment was sufficient to enable the die castings to gain a great improvement in strength during the artificial aging process without surface blister.

Die castings have a special microstructure distribution across the section. The surface layer contains more solute, thus more eutectic phases, compared with that

of the central region^[11-13]. In addition, due to rapid cooling^[14-18], much finer microstructure would form at the casting-die interface, whereas the central region exhibits much coarser grains and the ESCs (Externally Solidified Crystals) which originally nucleate inside the shot sleeve^[19, 20].

Characteristic microstructure distribution has a great influence on the mechanical performance of the die-cast components^[11, 21, 22]. The surface layer plays an important role in material failure. Thus the strength of the surface layer is more important, especially when the load contains components of bending and/or tension in service. Besides, the featured macro-segregation could affect the microstructural evolution of die-cast components during heat treatment, which has been reported by Song et al.^[23] in high-vacuum die-cast (HVDC) specimens of AZ91D magnesium alloys.

However, very limited studies were performed on die-cast aluminum alloys, to investigate the impact of the characteristic heterogeneity on the evolution of the microstructure and mechanical properties during heat treatment. A further understanding of this impact would help optimizing heat treatment practice based on particular requirements in service. In this work, HVDC specimens of A380 alloy were prepared to investigate the microstructural evolution during heat treatment by performing ICP, SEM, DSC and TEM analysis. The microstructural evolution at both the surface layer and

*Shou-mei Xiong

Male, Ph.D., Professor. Research interests: Al and Mg alloy high pressure die casting and microstructure simulation during solidification.

E-mail: smxiong@tsinghua.edu.cn

Received: 2017-08-11; Accepted: 2017-09-15

the central region were studied, respectively and in contrast. Results showed that the surface layer has different kinetic features of precipitation from that in the central region. Further detailed analysis and discussion are also performed on the effect of the macro-segregation, microstructure morphology and grain size on the precipitation behavior.

1 Experimental methods

The composition of A380 alloy used in this study is listed in Table 1. The HVDC components were produced on a TOYO BD-350V5 cold chamber die casting machine. With optimized operation parameters listed in Table 2, the die castings with excellent and stable mechanical properties were produced in large numbers without interruption. Figure 1 shows the configuration of the component, which comprises three bars and one 170 mm × 30 mm × 2.5 mm plate. The sample, as marked with the square in Fig. 1, was chosen for the subsequent analysis.

Powder samples were carefully prepared at the surface layer and central region for ICP (inductively coupled plasma) test to identify the contents of Si and Cu. The specimens covered with MgO powder (for homogeneous heating and less oxidation)

Table 1: Composition of A380 alloy used in this study (wt.%)

Si	Cu	Fe	Mn	Zn	Ni	Mg	Al
8.67	3.27	0.82	0.42	2.57	0.03	0.08	bal.

Table 2: Key operation parameters of die casting process

Parameters	Value
Melt temperature	690 °C
Slow shot speed	0.05 m·s ⁻¹
Gate speed	57.9 m·s ⁻¹
Vacuum degree	~90 kPa
Filling rate	16%
Intensification pressure	79 MPa

Fig. 1: Specific die casting with 3 bars and one 2.5 mm thick plate

were solutionized at 490 °C for 30 min, followed by a water quenching. To investigate the microstructural evolution during the solution treatment, the die-cast and the as-quenched samples at both the surface and the center region were ground, polished, etched with keller's etchant, and observed using JEOL 7001F SEM.

Parts of the water-quenched specimens were aged immediately after quenching for different times (0–5 h) at 200 °C. To study the aging response, the Vicker's hardness of the surface layer and the central region of the specimens were determined separately under a small load of 1 kg holding for 30 seconds.

Samples for DSC of about 25 mg were prepared at both the central region and the surface layer (thinner than 0.1 mm). Within 5 min after quenching, the isothermal DSC tests were performed using a TA Instrument DSC Q2000 at a temperature of 200 °C for 5 hours to investigate the precipitation process.

Samples of 90-minute artificial age were chosen for TEM observation. The surface layer and the central region were ground to a thickness of ~30 μm. Sample discs with 3 mm in diameter were then prepared and ion milling thinned. Liquid nitrogen was used as the cooling agent to protect the meta-stable precipitates from the high temperature due to the ion flux. The specimens were examined by JEOL 2010 TEM operated at 200 kV to observe the precipitates in α-Al grains.

2 Results and discussion

2.1 Typical cross-section of thin-walled A380 die-cast specimen

The metallographical images of the thin-walled die-cast specimen are shown in Fig. 2a, b and c. The microstructure at the center was much coarser (Fig. 2a and Fig. 2b). The grains at the surface layer are eutectic microstructure, which are too small to see clearly under optical microscope, while the central region contains large dendritic α-Al grains besides those tiny eutectic grains. The ICP result (Fig. 2d) shows the content of Si and Cu at the surface layer and the central region of the die casting. Both Si and Cu concentrations exhibit a larger content at the surface layer. The weight fraction of Si at the surface layer is 10.24wt.%, which is very close to the composition of eutectic point according to the Al-Si phase diagram. The Si concentration at the central region is about 8.6wt.% while for Cu, it is 3.59wt.% and 3.24wt.% at the surface layer and the central region, respectively.

2.2 Microstructural evolution during solution treatment

Figure 3 shows a typical microstructure of the surface layer and the central region before and after the solution treatment. According to the previous study^[13], the average cooling rate of the surface layer until the end of solidification of die-cast

Fig. 2: Typical die-cast microstructure of (a) surface layer and (b) central region, (c) Typical microstructure of thin-walled die-cast specimen on cross-section, (d) ICP result of samples at surface layer and central region

aluminum alloy could be as high as $\sim 200 \text{ K} \cdot \text{s}^{-1}$, while that of the central region was only $\sim 50 \text{ K} \cdot \text{s}^{-1}$. The eutectic microstructure exhibits a fully divorced morphology at both the surface and the central region (Fig. 3a and Fig. 3b) due to the high cooling rate. Both Si and Al_2Cu exhibit as semi-continuous networks along the α -Al grain boundaries before the solution treatment. The surface layer (Fig. 3a) contains α -Al grains with a size of 3–5 μm , which is much finer than those at the central region (Fig. 3b). The large dendrite-like α -Al grains with size of $\sim 20 \mu\text{m}$ in the central region (Fig. 3b) are known as ESCs (Externally Solidified Crystals), which refers to the primary α -Al grains that nucleated originally in the slot sleeve and then grew in the mold cavity.

After a 30-minute solution treatment at 490°C , all the Al_2Cu is dissolved into the α -Al matrix (Fig. 3c and Fig. 3d). The original large dendrite-like α -Al grains can still be recognized, as marked with “ESC” in Fig. 3d. All the eutectic Si is fragmented into pieces. In particular, the eutectic Si particles in the surface layer coarsen significantly to a size of $\sim 2 \mu\text{m}$ (Fig. 3c), which is of the same size as those at the central region (Fig. 3d). In the central region, the Si particles distribute in the interdendritic regions between large α -Al grains. On the contrary, the absence of large ESCs at the surface layer make the Si particles distribute more uniformly (Fig. 3c). A few Al_7FeCu_2 are generated during the solution treatment^[24, 25], which exhibits as bright particles with a size of less than $1 \mu\text{m}$.

According to the metallographical image in Fig. 2a and the ICP result in Fig. 2b, the thickness of the surface layer in the present study is about 0.3 mm. The evaluated diffusion distance of the solute, i.e. $\sqrt{Dt} = 8 \mu\text{m}$, where $D = 3.035 \times 10^{-14} \text{ m}^2 \cdot \text{s}^{-1}$ is the diffusion coefficient of Cu in α -Al matrix at the solutionizing temperature (490°C)^[26], and $t = 1,800 \text{ s}$ is the aging time. Thus, the solution treatment under such a low temperature and short time is enough to homogenize the solute distribution in the microstructure. After solution treatment, the distribution of Cu atoms is uniform in the α -Al grains. The macro-segregation of the die-cast specimens, on the contrary, is not affected during the heat treatment. The concentrations of solute elements in the surface layer and the central region remain unchanged after solution treatment and aging, which is very similar with the results in Ref. [8].

2.3 Heat release during isothermal aging

The isothermal DSC results of the as-quenched samples obtained at both the surface layer and the central region are shown in Fig. 4. The rate of heat release per unit mass increases (heat flow $\dot{Q}$) initially, reaches a maximum and then decreases to a very low level after a characteristic aging time t_f . The t_f obtained for the surface layer is about 100 minutes, while that of the central region is about 140 minutes. The total amounts of heat release (the area under the curve in Fig. 4) per unit mass at

Fig. 3: A typical A380 die casting microstructure of (a) and (c) the surface layer, and (b) and (d) of the central region. (a) and (b) show as-cast microstructure, (c) and (d) for microstructure after solution treatment

two locations are almost the same, implying that the amounts of precipitate are very close.

The precipitation process during artificial aging is considered to follow the Johnson-Mehl, Avrami, Kolmogorov (JMAK) kinetics^[27]:

$$f_r = 1 - \exp(-kt^n) \quad (1)$$

where f_r is the relative volume fraction of precipitates, which refers to the ratio of current precipitate volume to the final

precipitate volume after t_{test} , $t_{\text{test}}=5$ h. f_r is related to the fraction of the positive heat effect due to the precipitation process during isothermal calorimetry^[27]:

$$f_r = \frac{\int_0^{t_{\text{test}}} \dot{Q} dt}{\int_0^{t_{\text{test}}} \dot{Q} dt} \quad (2)$$

The evolution of f_r during artificial aging that was calculated with the DSC traces exhibits as scatters (Fig. 5). According to

Fig. 4: Isothermal DSC results of surface layer and central region at 200 °C

Fig. 5: Evolution of relative volume fraction during artificial age at 200 °C

Eq. (1), the fitting curves of the surface layer and the central region have the same k value. But the n values are 1.1467 and 0.9996, respectively, for the surface layer and the central region. The k value is thought to be an Arrhenius-type quantity that is affected by temperature, while a bigger n value of the surface layer means a faster precipitation. The surface layer almost

finishes precipitation at about 100 minutes, earlier than the central region does.

The obtained total heat release at both the surface layer and the central region are almost the same. It revealed the fact that solution treatment under 490 °C could not dissolve all the Cu atoms into the α -Al matrix. According to thermodynamic

Fig. 6: Bright-field TEM images and diameter distribution of precipitates, including (a) and (b) for α -Al grain interior of surface layer, (c) and (d) for α -Al grain interior of central region, (e) and (f) for an α -Al grain near to a Si particle in central region

calculation, the Cu dissolved in the α -Al matrix at the surface layer is about 3.64wt.% at 490 °C, while that of the central region is about 3.37wt.%.

2.4 Precipitates during isothermal aging

Bright-field TEM images of samples of 90-minute artificial aging are shown in Fig. 6. The zone axes are near to $[001]\alpha$. Disc-like precipitates, which are commonly believed to be the main reason of hardening in Al-Cu alloys during aging, are identified as θ' according to the diffraction pattern (see insets of Fig. 6). They precipitate with the orientation of $\{001\}/\{001\}$ α and $[001]/[001]\alpha$. The diameter distribution of precipitates is shown in Fig. 6. The measured average diameter of precipitates at the surface layer is about 47 nm (Fig. 6a and Fig. 6b). The precipitates have different average diameters in different zones of the central region. The average diameter of precipitates in the interior of large α -Al grains (ESCs) is about 57 nm (Fig. 6c and Fig. 6d), while that of the zones near the Si particles is about 40 nm (Fig. 6e and Fig. 6f).

As mentioned in section 2.3, due to the presence of ESCs in the central region (Fig. 3d), the Si particle distribution is less uniform than that of the surface layer. At the surface layer, the densely-distributed Si particles induce a relatively higher and more uniform distribution of plastic strain, as well as the dislocation network. Almost all the α -Al matrix at the surface layer belongs to “fast zone”. On the contrary, α -Al matrix of the central region is divided into two kinds, “fast zone” and “slow zone”. That is why the sample from the surface layer stops heat release at an earlier time during isothermal DSC test (Fig. 4 and Fig. 5).

2.5 Hardness response to isothermal aging

The hardness evolution during artificial aging is shown in Fig. 7. The surface layer hardens much faster than the central region. After solution treatment, the hardness of both the surface layer and the central region is very low and close to 70–75 HV1. The surface layer is only slightly harder than the central region. As aging proceeds, the difference of hardness between the surface

layer and the central region increases. After 90 minutes, the hardness of the surface layer approaches its peak 140 HV1, while that of the central region is only 113 HV1. The central region achieves its peak hardness after 120 minutes. The times to reach the peak hardness are very close to the results obtained from isothermal DSC test. Further aging leads to a slow decrease of the hardness.

According to the observation of microstructure, the only distinct microstructural evolution happening in the aging process is precipitation. The Al-Si binary alloys normally have no obvious age-hardening effects^[28]. Thus, the hardening effects during artificial aging are mainly due to the θ' precipitates. The small difference between the surface and the center at the beginning of artificial aging is the result of the mixed effect of different solute supersaturation, Si particle density, grain size of α -Al and dislocation density. During the artificial age process, the temperature is so low^[29] that the atom mobility is insufficient to trigger distinct dislocation annihilation and grain coarsening. Thus the effects of the change of grain size and dislocation density on hardness increment can be neglected. Even if the recovery and grain coarsening happen during aging, the surface layer would soften more due to denser dislocation and smaller grain size. That means the hardness increment of the surface layer due to precipitates could be even greater.

What else affects the precipitation except the dislocation density mentioned above? One possible answer is grain boundary. In theory, the grain boundary could act as the site for heterogeneous nucleation of precipitates. However, that might be a secondary factor in the present study. Few precipitates at the grain boundaries were found, neither was the concomitant PFZs (Fig. 8). That may be the result of the rapid quench after solution treatment and the relative low aging temperature.

Cu segregation was another factor affecting precipitation. A higher supersaturation of Cu in α -Al matrix would provide a larger driving force for nucleation and growth for the hardening phase. However, it might be not important in this study. As mentioned before, the actual supersaturation is very close according to both DSC the test and thermodynamic calculation.

Fig. 7: Evolution of hardness of surface layer and central region versus aging time at 200 °C

Fig. 8: Grain boundary in surface layer sample after 90 min artificial aging at 200 °C

The difference in Cu content between the surface layer and the central region is not the main factor affecting precipitation.

3 Conclusions

The hardness evolution of a vacuum-assistant die-cast A380 alloy during heat treatment was investigated in this work with particular attention focused on precipitation hardening at different parts. Accordingly, the following conclusions can be drawn:

(1) Low temperature and short time heat treatment could improve significantly the hardness of the vacuum-assistant die castings.

(2) During artificial aging at 200 °C, the surface layer of the die castings hardens more and faster than the central region, which is the result of faster precipitation reaction, as well as more and finer hardening phases in α -Al grains.

(3) Because of the shrinkage mismatch with Si, the α -Al matrix could be divided into two zones according to the magnitude of the dislocation density. The surface layer contains a larger proportion of regions with high dislocation density due to the absence of the ESCs, thus has lower activation energy of precipitation, and precipitates easier than the central region.

References

- [1] Zhang W S, Xiong S M, Liu B C. Study on a CAD/CAE system of die casting. *Journal of Materials Processing Technology*, 1997, 63: 707–711.
- [2] Mohamed A, Samuel F H. A Review on the Heat Treatment of Al-Si-Cu/Mg Casting Alloys. In: *Heat Treatment - Conventional and Novel Applications*, edited by Czerwinski F, InTech, 2012: 55–72.
- [3] Hirose S, Oguri Y, Sato T. Experimental and Computational Investigation of Formation of Precipitate Free Zones in an Al-Cu Alloy. *Materials Transactions*, 2005, 46: 1230–1234.
- [4] Boyd J D, Nicholson R B. The coarsening behaviour of θ'' and θ' precipitates in two Al-Cu alloys. *Acta Metallurgica*, 1971, 19: 1379–1391.
- [5] Ozhoga-Maslovskaja O, Gariboldi E, Lemke J N. Conditions for blister formation during thermal cycles of Al-Si-Cu-Fe alloys for high pressure die-casting. *Materials & Design*, 2016, 92: 151–159.
- [6] Lumley R N, O'Donnell R G, Gunasegaram D R, et al. Heat treatment of high-pressure die castings. *Metallurgical and Materials Transactions A*, 2007, 38A: 2564–2574.
- [7] Timelli G, Lohne O, Arnberg L, et al. Effect of Solution Heat Treatments on the Microstructure and Mechanical Properties of a Die-Cast AlSi7MgMn Alloy. *Metallurgical and Materials Transactions A*, 2008, 39: 1747–1758.
- [8] Lumley R N, Polmear I J, Curtis P R. Rapid Heat Treatment of Aluminum High-Pressure Diecastings. *Metallurgical and Materials Transactions A*, 2009, 40A: 1716–1726.
- [9] Lumley R N, O'Donnell R G, Gunasegaram D R, et al. The role of alloy composition in the heat treatment of aluminium high pressure die castings. *Metallurgical Science and Technology*, 2013, 26: 2–11.
- [10] Lee S G, Patel G R, Gokhale A M. Inverse surface macro-segregation in high-pressure die-cast AM60 magnesium alloy and its effects on fatigue behavior. *Scripta Materialia*, 2005, 52: 1063–1068.
- [11] Otarawanna S, Gourlay C M, Laukli H I, et al. Microstructure Formation in AlSi4MgMn and AlMg5Si2Mn High-Pressure Die Castings. *Metallurgical and Materials Transactions A*, 2009, 40: 1645–1659.
- [12] Timelli G, Fabrizi A. The Effects of Microstructure Heterogeneities and Casting Defects on the Mechanical Properties of High-Pressure Die-Cast AlSi9Cu3(Fe) Alloys. *Metallurgical and Materials Transactions A*, 2014, 45: 5486–5498.
- [13] Guo Z, Xiong S, Cho S, et al. Development of an inverse heat transfer model and its application in the prediction of the interfacial heat flux. *Acta Metallurgica Sinica*, 2007, 43: 607–611.
- [14] Guo Z, Xiong S, Cho S, et al. Study on heat transfer behavior at metal/die interface in aluminum alloy die casting process - I. Experimental study and determination of the interfacial heat transfer coefficient. *Acta Metallurgica Sinica*, 2007, 43: 1149–1154.
- [15] Guo Z, Xiong S, Cho S, et al. Study on heat transfer behavior at metal/die interface in aluminum alloy die casting process - II. Effect of process parameters on the interfacial heat transfer. *Acta Metallurgica Sinica*, 2007, 43: 1155–1160.
- [16] Guo Z, Xiong S, Cho S, et al. Study of interfacial heat transfer coefficient between metal and die during high pressure die casting process of aluminum alloy ADC12Z. *Acta Metallurgica Sinica*, 2007, 43: 103–106.
- [17] Guo Z, Xiong S, Cho S, et al. Effects of alloy materials and process parameters on the heat transfer coefficient at metal/die interface in high pressure die casting. *Acta Metallurgica Sinica*, 2008, 44: 433–439.
- [18] Laukli H I, Gourlay C M, Dahle A K. Migration of crystals during the filling of semi-solid castings. *Metallurgical and Materials Transactions A*, 2005, 36A: 805–818.
- [19] Bi C, Xiong S, Li X, et al. Development of a Fluid-Particle Model in Simulating the Motion of External Solidified Crystals and the Evolution of Defect Bands in High-Pressure Die Casting. *Metallurgical and Materials Transactions B*, 2016, 47: 939–947.
- [20] Wen W, Luo A A, Zhai T, et al. Improved bending fatigue and corrosion properties of a Mg-Al-Mn alloy by super vacuum die casting. *Scripta Materialia*, 2012, 67: 879–882.
- [21] Song J, Xiong S, Li M, et al. The correlation between microstructure and mechanical properties of high-pressure die-cast AM50 alloy. *Journal of Alloys and Compounds*, 2009, 477: 863–869.
- [22] Song J, Xiong S. The correlation between as-cast and aged microstructures of high-vacuum die-cast Mg-9Al-1Zn magnesium alloy. *Journal of Alloys and Compounds*, 2011, 509: 1866–1869.
- [23] Lise Dons A. The Alstruc homogenization model for industrial aluminum alloys. *Journal of Light Metals*, 2001, 1: 133–149.
- [24] Cerri E, Evangelista E, Spigarelli S, et al. Effects of thermal treatments on microstructure and mechanical properties in a thixocast 319 aluminum alloy. *Materials Science and Engineering A*, 2000, 284: 254–260.
- [25] Du Y, Chang Y A, Huang B, et al. Diffusion coefficients of some solutes in fcc and liquid Al: critical evaluation and correlation. *Materials Science and Engineering A*, 2003, 363: 140–151.
- [26] Esmaeili S, Lloyd D J, Poole W J. Modeling of precipitation hardening for the naturally aged Al-Mg-Si-Cu alloy AA6111. *Acta Materialia*, 2003, 51: 3467–3481.
- [27] Chen R, Xu Q, Guo H, et al. Modeling the precipitation kinetics and tensile properties in Al-7Si-Mg cast aluminum alloys. *Materials Science and Engineering A*, 2017, 685: 403–416.
- [28] Volin T E, Balluffi R W. Annealing kinetics of voids and the Self-diffusion coefficient in aluminum. *Physica Status Solidi*, 1968, 25: 163–173.

The work was financially supported by the National Natural Science Foundation of China – China Aerospace Science and Technology Corporation Aerospace Advanced Manufacturing Technology Research Foundation (NO. U1537202), and the Special Funding Program on Transformation of Scientific and Technological Achievements at Jiangsu Province (No. BA2015041).