Progress in Natural Science: Materials International 26 (2016) 391–397

HOSTED BY

Contents lists available at ScienceDirect

Progress in Natural Science: Materials International

journal homepage: www.elsevier.com/locate/pnsmi

Original Research

Effect of solidification rate on the coarsening behavior of precipitate in rapidly solidified Al-Si alloy

Zhiyong Cai [a,b], Chun Zhang [a,b,*], Richu Wang [b], Chaoqun Peng [b], Ke Qiu [b], Naiguang Wang [b]

[a] School of Materials Science and Engineering, Jiangxi University of Science and Technology, Ganzhou 341000, China
[b] School of Materials Science and Engineering, Central South University, Changsha 410083, China

ARTICLE INFO

Article history:
Received 8 August 2015
Received in revised form
27 May 2016
Accepted 4 August 2016
Available online 18 August 2016

Keywords:
Rapid solidification
Solidification rate
Super-saturated solutions
Grain growth
Aluminum Silicon alloy

ABSTRACT

The gas-atomized Al-Si alloy powder with different particle sizes was subjected to isothermal annealing for understanding the effect of solidification rate on precipitation and growth Si crystals. The results show that Si precipitates grew more quickly in the small samples with a large solidification rate due to high interfacial energy. Moreover, these Si crystals had a tendency to form a quasi-spherical shape after annealing at a low temperature or for a short holding time. Coarsening of the Si precipitates during annealing was examined using a LSW equation. Thermal stability of the rapidly solidified alloy was significantly influenced by its original microstructure as a result of high solidification rate. Furthermore, more serious clustering of Si-Si phase was also observed in the small samples, attributed to the rapid coarsening of the Si phases.

1. Introduction

Rapidly solidified materials exhibit superior mechanical properties because of the reduction of compositional segregation, refinement of secondary phases, and suppression of coarse primary phases [1–3]. Among various rapidly solidified alloys, hypereutectic Al-Si alloys have attracted increasing interesting due to their excellent properties, such as light-weight, good wear resistance, and low coefficient of thermal expansion (CTE) [4,5]. For these materials, effective modification of Si phases, e.g. size, morphology, and distribution, is of crucial importance for exerting the advantages of rapid solidification techniques [6]. However, the conventional rapid solidification routes, such as gas atomization for preparing powder and melt spinning for obtaining thin ribbon, suffer from the limitation on useful product size and hence can hardly be used directly. Consequently, such materials are almost unavoidably followed by a subsequent consolidation step including heat treatment at an elevated temperature, thus degrading the desirable aspects of microstructure obtained via rapid solidification [7,8]. The microstructure and mainly the local clustering of Si phase during heat treatment play an important role in determining the mechanical properties because fracture initiates at the clusters and grows rapidly through the matrix [9]. Hence, thermal stability is essential for rapidly solidified raw materials.

Matsuura et al [10]. studied the precipitation behavior of super-rapidly solidified hypereutectic Al-17 wt% Si alloy and found that Si phase (about 20 nm) dispersed in the Al matrix and grew gradually during sintering, while the size of Si phase remained at a submicro level. They also indicated that after a quick growth at the beginning of sintering, the growth rate decreased significantly with prolonging the holding time. In addition, the coarsening of Si phase in semi-solid state is observed, and the coarsening rate constants increased with either increasing or decreasing solid fraction, depending on the conditions of invariable temperature or Si content, respectively [11]. Furthermore, the growth of precipitated Si crystals is significantly influenced by the original microstructure of rapidly solidified alloy. Isothermal annealing of Al-12 wt% Si alloy ribbons showed that the microstructure features of rapid solidification disappeared when the annealing temperature was above 250 °C [7]. Their results indicated that the microstructure of rapidly solidified alloy was less stable. However, most of the rapidly solidified Al alloys are needed to be consolidated at the temperature above 250 °C to obtain near full-density.

Previous studies suggested that thermal stability is significantly influenced by the solidification rate. Graiss and Saad [12] studied the thermal stability of Sb-InSb eutectic alloys solidified unidirectionally in a vertical resistance furnace. They concluded that

* Corresponding author at: School of Materials Science and Engineering, Jiangxi University of Science and Technology, Ganzhou 341000, China.
E-mail addresses: caicsu@163.com (Z. Cai), zchun614@163.com (C. Zhang), rcwcsu@163.com (R. Wang), pcq2005@163.com (C. Peng), ranstrom@163.com (K. Qiu), wangnaiguang505@163.com (N. Wang).
Peer review under responsibility of Chinese Materials Research Society.

http://dx.doi.org/10.1016/j.pnsc.2016.08.002

Sb particles grew more rapidly in the fine particle (solidification rate of 30 mm h^{-1}) than the large sample (solidification rate of 15 mm h^{-1}). Vianco et al. [13] studied the coarsening behavior of Pb-rich phase particles in Sn-37 wt% Pb solders with solidification rate from 0.1 to 100 °C min^{-1} using a differential scanning calorimeter (DSC). Their results suggested that the Pb-rich phase particle grew more quickly in the sample solidified at large solidification rate than that at low solidification rate. Our previous work on gas-atomized Al-Si alloy showed that the microstructure characteristics were strongly influenced by the particle size [14,15]. The calculated relationship between solidification rate and particle size revealed that the deviation in solidification rate resulted in the difference of microstructure characteristics. Therefore, the solidification rate of rapidly solidified alloy should also affect the growth of precipitates during isothermal annealing. However, until very recently, few studies have been paid to the effect of solidification rate on crystal growth behavior in rapidly solidified alloys.

In this work, the growth of precipitated Si crystals in gas-atomized hypereutectic Al-27 wt% Si alloy with different particle sizes is investigated to clarify the effect of solidification rate on the coarsening behavior of precipitates. The growth of the Si crystals is studied as a function of solidification rate, annealing temperature, and holding time. The coarsening kinetics of different samples is calculated on the basis of a modified LSW equation.

2. Experimental procedure

In this study, an Al-27 wt% Si hypereutectic alloy with impurities less than 0.1 wt% was employed. The master alloy was prepared by induction melting at 950 °C using high purity Al (99.995 wt%) and single crystal Si. The molten metal was poured into a tundish, and then atomized through a graphite melt delivery nozzle with an inner diameter of 2.5 mm. The atomization was operated at a pressure of 0.9 MPa by an annular N_2 gas atomizer. The as-atomized powder was thereafter cooled down to room temperature under the protection of N_2.

According to the previous researches, powder particle size has a significant effect on the solidification rate of gas-atomized powder [14,16]. Hence, the as-atomized powder was mechanically sieved into five different particle size groups of 200–250, 90–125, 63–74, 38–50, and less than 25 μm, respectively, to investigate the effect of solidification rate on the thermal stability. Isothermal annealing of the samples with different particle sizes was carried out at 400, 430, and 450 °C for various time up to 10,240 min under the protection of argon. The heating rate from room temperature to the annealing temperatures was 10 °C min^{-1}. In each case, samples of 1 g were placed in a ceramic crucible and annealed since the fixed temperature was reached. Samples after isothermal treatment were water quenched and then dried for microstructure observations.

Samples used for microstructure observations were prepared by standard metallurgical methods, i.e., grinding on SiC abrasive papers and subsequently polishing with 1 μm diamond paste, followed by etching with Keller's reagent (1HF-1.5HCl-2.5HNO_3-95H_2O, by volume fraction). The powder microstructure was observed using a field-emission scanning electron microscope (FE-SEM, FEI QUANTA-200). X-ray diffraction (XRD) analysis was carried out with a Rigaku D/Max2500VB+ diffractometer using Cu Kα radiation at a scan step of 0.02 (°) s^{-1} from 20° to 120°. The size characteristics of primary Si phase and precipitated Si crystals were determined with a commercial image analysis software based on several hundreds of Si phase particles and precipitates under each condition on at least twenty single powder particles. The average size was denoted according to the diameter of an equivalent area, and the details of quantitative analysis were reported in Ref. [13].

3. Results and discussion

3.1. Microstructure characteristics

Fig. 1 shows the microstructure of samples with different particle sizes after isothermal annealing at 450 °C for 40–10,240 min, respectively, and the corresponding solidification rate is also presented [14]. It can be seen that the eutectic Si phase with bar-like or entangled structure disappears after annealing. At the same time, the primary Si phase with sharp corners becomes smooth, especially in the fine sample obtained with large solidification rate. However, the size of primary Si is stable after holding for 40 and 640 min. Generally, the precipitates grow and their number decreases gradually as the annealing time prolongs. Ullah et al [17]. suggested that the diffusion and interface kinetics control the formation of round and large crystals, i.e., the spheroidizing and Ostwald ripening occur simultaneously. Meanwhile, they also found that such a relatively fast process occurred during annealing at 600 °C for 60 min as a result of low solidification rate. After annealing at 450 °C for 10,240 min, the precipitated Si crystals grow to 1.82, 2.03, and 2.87 μm with the average solidification rate of 1.87×10^3, 2.02×10^4, and 6.28×10^5 °C s^{-1}, respectively [14].

Comparisons between the microstructure of samples with different solidification rates after annealing under the same condition, some obvious deviation is observed. Firstly, it is observed from Fig. 1a–c that, after isothermal annealing, the precipitated Si phase is more clearly observed from the Al matrix of the fine sample. Additionally, the eutectic Si is completely dismissed in the matrix of fine sample as compares with that of 63–74 and 200–250 μm. Such deviation is also found in the samples annealed at 400 and 430 °C, but less obvious owing to the lower precipitation kinetics. This result implies that the precipitation process is more rapid to perform in the sample with large solidification rate.

Secondly, it is observed that the morphology of the precipitates is also affected by the solidification rate under the same annealing condition. The precipitated Si with an aspect ratio of 2.64 exists in the samples of 200–250 μm after annealing at 450 °C for 10,240 min. However, in the samples of 63–74 μm and less than 25 μm, this value is near 1.0, indicating a quasi-spherical morphology. Moreover, it is more obvious that the sample possess precipitates with different sizes and morphologies after annealing at a lower temperature or for a shorter holding time, e.g. Fig. 1a–c. These results suggest that the alloy with large solidification rate is more favorable for the formation of quasi-spherical precipitates during annealing at a relatively low temperature or for a short holding time. The above results are contrary to the previous study, which reported that the formation of spherical precipitates requires elevated temperature annealing to increase the matrix diffusion rate [18].

Thirdly, it is observed that clustering of the precipitated Si occurs in the fine sample after annealing at 450 °C for 10,240 min, but less obvious in the large samples. Clustering of the precipitated Si results in difficulty in separation of the precipitates from the primary Si. Such Si-Si clustering contributes to the rapid coarsening of the precipitates after annealing at higher temperature or for a longer holding time. Therefore, the Si phase grow not only via attachment of Si atoms but also in the form of some Si-Si clusters to the surface of existing Si phase [19]. Such difference during annealing should come from the material with the deviation of original microstructure.

After isothermal annealing, the morphology of Si phase in powder over a special range of particle size is studied in more