Structure of rapidly solidified aluminium-silicon alloys

S. K. BOSE, R. KUMAR

National Metallurgical Laboratory, Jamshedpur 7, India

This paper present results obtained on rapid solidification of aluminium-silicon alloys from the liquid state. It shows that the limit of primary solid solubility is extended almost to the eutectic composition and that the large supersaturation is relieved on raising the annealing temperature to the range 110 to 450°C. This conclusion is based on measurements of lattice parameter and is also supported by corresponding changes in hardness and metallographic features.

1. Introduction

During the last decade, considerable experimental interest has been shown in the formation of metastable phases by rapid solidification from the liquid state [1]. From the structural standpoint, the results on various alloy systems [2-5] indicate the extension of the limits of primary solid solubility and/or formation of non-equilibrium intermetallic phases. Metallographic features of rapid solidification have also been studied [6]. The aluminium-silicon system has been chosen essentially because it forms the basis of a large number of industrial alloys and also because some information is available with regard to the distribution of silicon in the liquid state [7, 8].

2. Experimental techniques

Four aluminium-silicon alloys containing 5.8, 10.0, 12.6 and 18.4 at. % Si were prepared from 99.8% Al and Si of 99% purity. Rapidly solidified thin films of thickness varying between 0.1 and 0.15 mm were prepared by ejecting a fine stream of liquid alloy from an orifice in a graphite crucible axially centrifuging at 1400 r.p.m. and at 920°C onto a double-walled copper drum kept at 0°C. Experimental details of this technique have been described elsewhere [9]. The films thus obtained were metallographically examined in the etched and unetched conditions using Keller's reagent. X-ray and microhardness techniques were also used to study the nature of the alloys in the as-solidified condition, and after annealing in vacuum in the temperature range 110 to 450°C. Debye-© 1973 Chapman and Hall Ltd.

Scherrer photographs of selected cylindrical specimens were taken in a camera of 11.4 cm diameter using filtered CuKa radiation. These cylindrical specimens were extracted from selected regions of the rapidly solidified thin films and Nelson and Riley's extrapolation function was used to determine the lattice parameter. The microhardness of mounted specimens was also measured; for each alloy, an average of ten readings were taken.

3. Results and observations

Fig. 1 shows the variation of the lattice parameter and microhardness as a function of the silicon content of the alloys. The lattice parameter continuously decreases and the microhardness



Figure 1 Lattice parameter and microhardness values of rapidly solidified thin films of A1-Si alloys,



Figure 2 Cast structures of rapidly solidified alloys: (a) Al \times 270; (b) Al-5.8 at. % Si \times 350; (c) and (d) Al-10.0 at. % Si, \times 350; (e) Al-12.6 at. % Si, \times 350; (d) conventionally cast structure of Al-12.6 at. % Si alloy, \times 350; (g) cast structure of rapidly solidified Al-18.4 at. % Si alloy, \times 350; (h) Al-12.6 at. % Si Alloy showing pre-dendritic regions, \times 350.

correspondingly increases with increase in the silicon content. Recalling that the equilibrium phase diagram of the Al-Si system [10] shows the occurrence of an eutectic at 11.3 at. % Si and primary solubility decreasing from a maximum of 1.59 at. % at the eutectic temperature (557°C) to less than 0.1 at. % Si at room temperature, the most important structural change obtained by rapid solidification of the melt is an increase of the solid solubility of Si in Al to 10.0 ± 0.5 at. % Si, a six-fold increase over the maximum value of primary solid solubility under equilibrium condition.

Metallographic examination of as-solidified specimens revealed that the morphology of solidification showed considerable variety and a few typical microstructures are shown in Fig. 2. The microstructure of aluminium solidified under similar conditions is shown for comparison. The following observations can be made from the micrographs.

(a) The hypoeutectic alloy containing 5.8 at. % Si solidifies in an essentially dendritic manner (Fig. 2b), which is typical of that of aluminium (Fig. 2a).

(b) The alloy containing 10 at. % Si solidified in some regions as a single phase (Fig. 2c) and in others in a manner typical of an unmodified eutectic structure exhibiting the formation of proeutectic primary aluminium (Fig. 2d).

(c) The microstructure of the hypereutectic alloy containing 12.6 at. % Si (Fig. 2e) was similar to that of the conventionally cast unmodified Al-Si alloys of similar composition (Fig. 2f), but the phase distribution is refined as a result of rapid solidification. Such morphology is not observed in the case of alloys containing 18.4 at. % Si, in which the silicon is distributed in an extremely fine manner as shown in Fig. 2g. Idiomorphic crystals of Si are also present in the microstructure but with much reduced frequency of distribution as compared with that of the conventionally cast alloys.

(d) Pre-dendritic regions of solidification were observed in the alloys containing 10.0 and 12.6 at. % Si and are marked with arrows in Fig. 2c; a typical pre-dendritic region observed in the alloy containing 12.6 at. % Si is shown in Fig. 2h.

Biloni and Chalmers [11] and Kumar and Bose had earlier shown that solidification under rapid rates of heat extraction could lead to the formation of what were called "pre-dendritic regions" of characteristic morphology and these regions could nucleate dendritic solidification under favourable conditions.

The decomposition of the metastable supersaturated solid solution was studied in the temperature range 110 to 450° C by the use of (a) lattice parameter, (b) microhardness and (c) metallographic observations, and the results are summarized in Fig. 3a and b. Continuous increase of lattice parameter of the matrix as a result of holding in the temperature range 110 to 450° C for 30 min is shown in Fig. 3a. It can also be noted that almost identical values of the lattice parameter are established within 30 min of ageing at elevated temperatures as a result of



Figure 3 Variation in (a) lattice parameter, (b) microhardness, as a function of annealing temperature 30 min isochrones.



Figure 4 Metallographic structures showing the relief of supersaturation in thin films containing 12.6 at. % Si; annealed at (a) 250°C, 30 min, (b) 350°C, 30 min, (c) 450°C, 3 h, (d) 450°C, $7\frac{1}{2}$ h. All × 350.

relief of supersaturation through precipitation of silicon. These observations are also corroborated by corresponding changes in the microhardness values (Fig. 3b) which gradually decrease with increase in annealing temperatures. Metallographic observations shown in Fig. 4a and b support this contention of the relief of supersaturation through extensive precipitation in the matrix for the alloy containing 12.6 at. % Si when annealed at 250 and 350°C for 30 min. It is observed that the morphology of the silicon precipitates in the matrix is mostly spheroidal (Fig. 4c and d) and not acicular as in the conventionally cast alloys of similar composition.

4. Discussion

Extension of the limits of primary solid solubility as a result of rapid solidification is a fairly general metallurgical phenomenon and has been observed in a large number of systems [12-16]. Whilst the general pattern of decomposition observed in these alloys conforms to the general principles of precipitation, the extension of the limits of the primary solid solubility observed

in the Al-Si system deserves some special comments. Similar results in the Al-Si system were earlier reported by Itagaki et al [17]. Singh and Kumar [7] showed in a recent investigation that the distribution of Si in hypoeutectic alloys is different from that in the hypereutectic alloys in the liquid state. They showed that in the eutectic and hypoeutectic alloys, Si is present in association with Al in the form of (Al-Si) clusters, and in the hypereutectic alloys in the form of an (Si-Si) cluster. Preferential association of Si atoms with Al atoms in the eutectic and hypoeutectic alloys as (Al-Si) clusters may possibly be responsible for the presence of the primary phase in the cast structures, so typical of unmodified alloys. This is because monatomic Si atoms are not available in the liquid state to nucleate the solidification of silicon in the binary eutectic, since the diffusion of Si atom is hindered by their association with Al atoms, making nucleation of the Si phase difficult. Rapid cooling from the liquid state, therefore, extends the limit of solid solubility up to the eutectic composition for the same reason. Thus, in

hypoeutectic and eutectic alloys, the Al-Si association hinders Si diffusion sufficiently to quench in a solid solution, and in the hyper eutectic alloys the atoms are free enough to allow the nucleation of the two phases.

5. Conclusions

This investigation shows that the limit of primary solid solubility of silicon in aluminium can be extended almost to the eutectic composition by rapid cooling from the liquid state. This phenomenon can be attributed to the preferential association of the atoms of silicon with those of aluminium in the liquid state for hypoeutectic and eutectic aluminium-silicon alloys.

Acknowledgement

The authors thank Dr V. A. Altekar, Director, National Metallurgical Laboratory, Jamshedpur for permission to publish this paper.

References

- 1. T. R. ANANTHARAMAN and C. SURYANARAYANA, J. Mater. Sci. 6 (1971) 1111.
- 2. H. L. LUO, C. C. CHAO and P. DUWEZ, Trans. Met. Soc. AIME, 230 (1964) 1488.
- 3. A. TONEJC and A. BONAFAČIĆ, Fizika 2 (1970) 81.
- 4. C. JANSEN, B. C. GIESSEN and N. J. GRANT, Abstracts, Fall Meeting of the Met. Soc. AIME (Detroit, 1968).

- 5. R. KUMAR and S. K. BOSE, Scripta Met. 5 (1971) 515.
- 6. *Idem*, Metallographic studies of rapidly solidified Al.Cu alloys, presented in the third annual technical meeting of the International Metallographic Society Cleveland, Ohio, USA (Oct 1970) published in IMS proceedings (1970) 95.
- 7. M. SINGH and R. KUMAR, J. Mater. Sci. 8 (1972) 317.
- R. KUMAR, C. S. SIVARAMAKRISHNAN and R. MOHANTY, Structure of liquid Al-Si-Mg alloys. Presented at the Second International Conference on liquid Metals, Tokyo, Japan (September 1972).
- 9. R. KUMAR and A. N. SINHA, Trans. Indian Inst. Met. 22 (1969) 9.
- 10. M. HANSEN, "Constitution of Binary Alloys" (McGraw Hill, New York, 1958).
- 11. H. BILONI and B. CHALMERS, Trans. Met. Soc. AIME 233 (1965) 373.
- 12. H. L. LUO and P. DUWEZ, Canad. J. Phys. 41 (1963) 758.
- 13. B. C. GIESSEN, U. WOLFF and N. J. GRANT, J. *Appl. Cryst.* 1 (1968) 30.
- 14. I. V. SALLI and L. P. LIMINA, "Growth and Imperfections of Metallic Crystals" (Consultants Bureau, New York, 1968) p. 251.
- 15. R. C. RUHL, B. C. GIESSEN, M. COHEN and N. J. GRANT, J. Less Common Metals, 13 (1967) 611.
- 16. Idem, Mat. Sci. & Eng. 2 (1968) 314.
- 17. M. ITAGAKI, B. C. GIESSEN and N. J. GRANT, *Trans. Amer. Soc. Metals*, **61** (1968) 330.

Received 5 March and accepted 29 May 1973.