

## REFINEMENT OF PRIMARY AND EUTECTIC SILICON PHASES IN SHAPE CASTING OF HYPER-EUTECTIC Al-Si ALLOYS

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### Abstract

Hyper-eutectic Al-Si alloys composed of low-density materials with excellent castability are good candidates for aerospace and automobile applications. Our efforts over the last few years have demonstrated that such hyper-eutectic Al-Si alloys with no primary silicon phase but with a eutectic microstructure that assumes nano-fibrous morphology for the eutectic silicon phase can be grown by directional solidification. In pursuit of this effort, more hyper-eutectic Al-Si alloys containing ultra-refined primary and eutectic silicon phases have been grown by an alternate casting method known as shape casting. The process of this fabrication and the microstructure of the resulting alloys are presented here.

### Introduction

The ever-increasing desire in the automotive and aerospace industries to reduce the weight of various mechanical parts such as engine blocks (for fuel efficiency purposes) has inspired a myriad of investigations into the development of newer lightweight materials. Among the new lightweight materials being sought are hyper-eutectic Al-Si alloys that possess excellent wear and corrosion resistance, low density, low coefficient of thermal expansion and good strength [1-3]. Attainment of such properties in the alloys requires a high volume fraction of a very fine morphology silicon phase uniformly distributed in the matrix of the aluminum. Al-Si alloys with such a microstructure can be economically successful if the components made of the alloys are produced by shape casting processes.

The phase diagram of Al-Si binary system [4] is shown in Figure 1. It reveals that a hyper-eutectic alloy is formed with a silicon composition that exceeds 12.2 at% (11.7 wt%) silicon. The microstructure of hyper-eutectic alloys contains two major components, the primary phase and the eutectic phase. Conventional casting resulting in hyper-eutectic Al-Si alloys in which the primary silicon phase appears as large particles exhibiting a variety of morphologies such as coarser platelets—star-like and blocky—while the eutectic structure consists of an aluminum-rich solid solution of silicon and virtually pure silicon, forming the matrix of the microstructure. The large particles of primary silicon can cause considerable tool wear and machining difficulties [5] and greatly aggravate tensile strength, ductility and impact strength of the alloys. Refining and controlling primary silicon particles in such alloys are an effective way to overcome those disadvantages. Recently, based upon the concept of the solubility of barium (Ba) metal in the silicon phase Shamsuzzoha and his coauthors [6-8] have demonstrated that hyper-eutectic alloys containing up to 17-wt% Si can be produced by directional solidifications without a primary silicon phase being present in the microstructure. The silicon in these alloys appears nano and close to nano-size fibers

at higher growth rate (2500 $\mu\text{m}/\text{sec}$ ) and moderate growth rate (250 $\mu\text{m}/\text{sec}$ ) respectively. The dual refinement of primary and

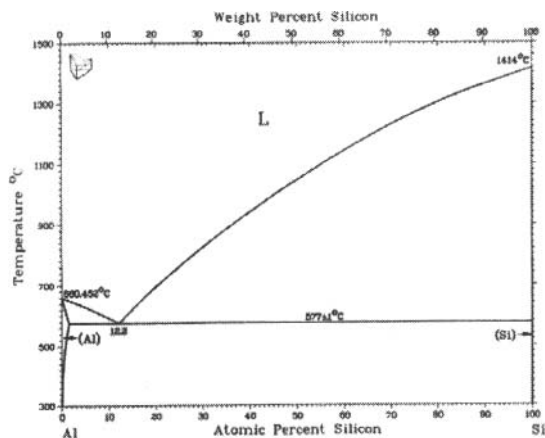


Figure 1. The Al-Si Eutectic Phase Diagram

eutectic silicon has been attributed to the solidification morphology of Si that has barium in solution [8]. Since directional solidification at lower growth of around 200-250 $\mu\text{m}/\text{sec}$  and of a thermal gradient of 50-70 K/cm approaches conditions typically present in sand casting, a simple shape casting of Al-17% Si-3% Ba alloy in a graphite mold was considered to produce alloys that can also possess very refined primary and eutectic silicon.

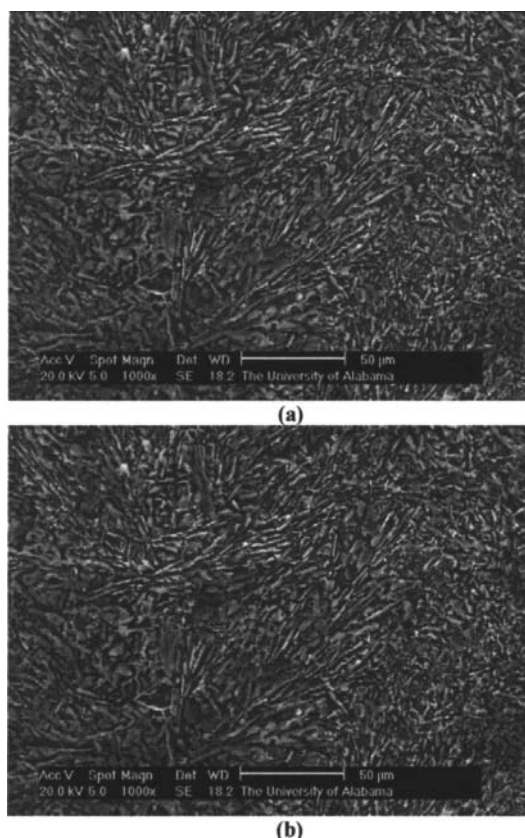
### Experimental Procedures

The process involves an initial casting of master alloy, which has a composition of Al-17% Si-3% Ba. The master alloy was produced from high purity (99.999%) aluminum and silicon. A resistance furnace was used for melting of the alloy constituents and the barium. The melt of the mixture was kept at 900°C in an argon gas atmosphere for some time, and then cast in the form of billets. The billets were mechanically reduced into pieces, and the reduced samples were then melted in an argon gas environment. The melt was then well stirred and poured in a graphite mold of 10 mm diameter and 150 mm height. Longitudinal and transverse section specimens taken from near the center of the grown samples were used to determine the microstructure and thin foil preparation. A few flat samples taken out from the solidified alloy using a diamond saw were applied to determine average hardness of a thus grown alloy. The morphology of the silicon phase of the alloy was investigated using a JOEL JSM-7000F SEM. Crystallography of the microstructure was studied by a FEI-Tecna F-20 TEM operating at 200 keV. For thin foil preparation, cylindrical specimens of 3mm diameter were cut by an ultrasonic disc cutter manufactured by GATAN Inc. These cylinders were then

ground gently with a 600 grade silicon carbide paper of 200  $\mu\text{m}$  grade and then dimpled by mechanical polishing. These discs were then ion-milled to a thickness of less than 100 nm to allow electron beam transmission. For the determination of chemical composition the EDX facility attached with the JSM-7000F SEM was used. The hardness of the alloy was measured using a Wilson Wolpert 401 MVD micro hardness tester with a load of 50 gm for 10 seconds.

### Result

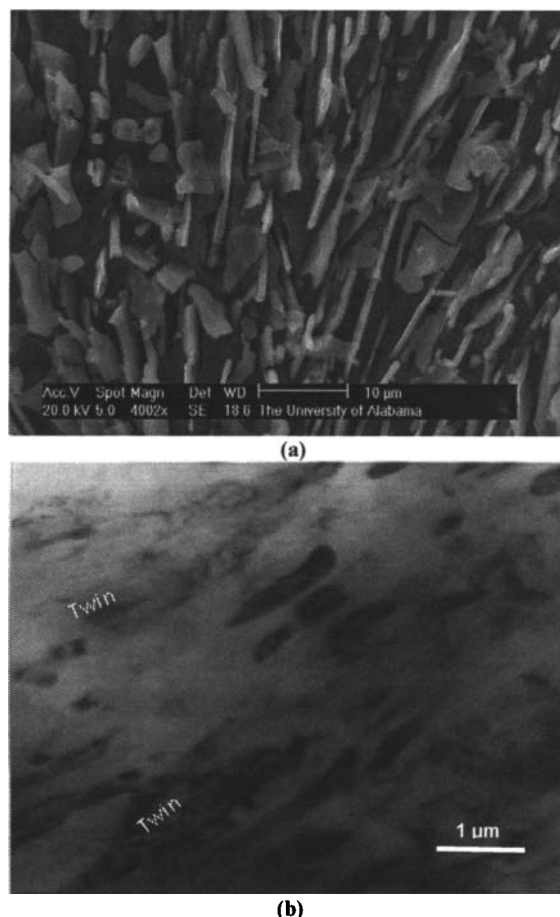
The microstructure of none of the cast alloys could be well-resolved by optical microscopy. Hence, samples taken of the alloys were deep etched and then investigated by scanning electron microscopy to study the morphology of silicon phase only. Such investigations on the cross sectional samples revealed that the microstructure covering a diameter of about one millimeter at the center is slightly refined. Figure 1a and 1b SEM micrographs of longitudinal and cross sectional samples of a thus grown shape cast alloy that has a composition of Al-17% 3% Ba. Energy dispersive x-ray spectrometry equipped with



**Figure 2. SEM micrographs of deep etched sample of a shape cast Al-17%/Si-3% Ba alloy showing very fine flake morphology of silicon in the microstructure (a) as viewed along the longitudinal section of the shape cast alloy (b) as viewed along the transverse section of the alloy.**

the SEM for the alloy showed an overall composition that is close that of Al-17 wt% Si. The microstructure in both longitudinal and cross-sectional micrographs appears similar in appearance and exhibits very fine eutectic as matrix. The

microstructure in these micrographs is also free of any large idiomorphic primary silicon. The silicon crystals in these micrographs are of eutectic silicon phase and assume a very fine flake-like morphology. The size of flake width in the alloy was found to be between 750 to 2000 nm, which is at least 20 times finer than those found in sand cast unmodified alloys [9], and about 3 times finer than those found in impurity or chill- modified alloys [10-12].



**Figure 2. SEM Micrograph (a) and TEM micrograph (b) of shape cast Al-17%/Si-3% Ba alloy, showing eutectic silicon flakes present in the microstructure to assume divergence from a point of origin.**

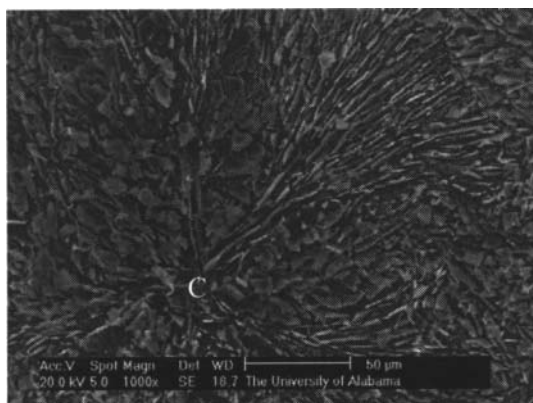
This silicon crystals found in the microstructure follow a wheat sheaf growth pattern originating from a point of origin. The diverging tendency from an origin for the growth pattern of flake-type silicon crystals is also evident in high-magnification SEM and TEM micrographs shown in Figures 3a and 3b. TEM studies revealed that the silicon phase in the sample is comprised of parallel layers of co-zonal twins. The TEM micrographs of Figure 4b show one of such individual silicon particles having their external surface parallel closely to internal {111} twins, which is the characteristic of the crystal that is grown by the twin-plane re-entrant (TPRE) mechanism [13, 14]

Even though the microstructure of the alloy is free of large particles of primary silicon, it contains many locations in which a star-like assembly of silicon crystal is present. Figure 4



**Figure 4. Longitudinal section SEM micrographs of deep etched shape cast Al-17%/Si- 3% Ba alloy, showing star-like (Points A and B) and fish bone (Point B) morphologies of silicon crystal present in the microstructure of the alloy.**

shows locations of two such star-like assemblies of silicon crystal, marked as A and B. In location B one of the arms of the star has its own side arms in the form of a fish bone structure. The size of an individual member of such a fish bone assembly is close to that of a typical eutectic silicon flake present in the microstructure. A similar type of fish bone assembly of crystals was also found in Al-Ge eutectic system, and is termed as the assembly of eutectic Ge phase [15]. This seems to suggest that these silicon crystals of the fish bone assembly found in the presently shape cast Al-Si alloys are also eutectic crystal assembly. TEM studies on the crystallography of this star assembly for silicon crystals are currently under progress in order to identify whether these crystals belong to eutectic or primary phase.



**Figure 5. Transverse section SEM micrographs of deep etched Al-17% Si- 3% Ba alloy, showing divergence of silicon flakes from a point of origin located at the center of a star like assembly of silicon crystal.**

However, the individual arm of such a star always assumes a size that is comparable to that of an individual eutectic silicon indicating the star to be an assembly of eutectic silicon crystals.

It was also found that most of the silicon flakes in the vicinity of the star assembly just discussed appears to diverge from the center of the assembly of silicon star. Figure 5 shows another SEM micrograph showing the point of origin for a group of diverging eutectic silicon at a position marked by C. This feature of the microstructure allows an easy identification of many nucleation centers in the sample.

The average width of a Si particle in these primary Si free alloys suggests that the cast alloy is likely to exhibit marked improvements in physical and mechanical properties over existing similar hyper-eutectic Al-Si alloys grown by other methods and contains microstructures of eutectic and primary silicon.

**Table I. Mechanical properties of hyper-eutectic alloys grown by different methods**

Hyper- eutectic Al-Si alloy	Micro-hardness (kg/mm <sup>2</sup> )
Vacuum spray deposited	151±15
Conventional cast	~ 100
Presently cast	135± 10

Table I summarizes the hardness of this sample and compares properties of this data with that of similar alloys prepared by vacuum plasma spray techniques (Al-21wt% Si) [16] and by conventional casting (Al-17 wt% Si) [17].

### Discussion

The relatively finer microstructure found at the center of the presently cast cylindrical samples support an important assumption that the last liquid was solidified at the mold center, and the initiation of solidification was started either at the mold wall or in the melt that exists between mold wall and the mold center. Present observation of star-like assembly of silicon crystals acting as the point of origin for the neighboring eutectic silicon also suggests that some nucleation of solids do occurs in the melt existing between from the mold wall and the mold center. The understanding of impurity-free unmodified modified Al-Si eutectic solidification establishes that flake type silicon in these alloys is grown by non-co-zonal twinning mechanism [18], but the size of the flake depends upon the solidification rate. In the solidification of present hyper-eutectic Al-Si alloys, the crystal growth of Si by the mechanism of co-zonal twinning is likely to yield flake-type Si phase in the microstructure. However, the low solidification rate that typically occurs in a graphite mold is expected to produce the silicon flakes with size that is typically found in a sand cast Al-Si eutectic alloy [19]. Whereas, silicon flakes found in the present alloy are comparatively refined. This difference possibly arises from the presence of barium in solution in the silicon. However, further work will be required to confirm this.

### Conclusion

The microstructure of the presently developed shape-cast high-strength hyper-eutectic Al-Si alloy discussed in this paper reveals that the entire silicon content of the alloy assumes very refined flake-like morphology. Appearances of such morphology of silicon in the alloy can be attributed to the crystallization behavior of silicon that has Ba in the form of solid solution. It appears that the solid solution of Ba in silicon effects the crystallization of both the primary and eutectic silicon in the solidification of shape cast hypereutectic Al-Si alloy. The effect allows the hyper-eutectic melt either not to nucleate any primary silicon crystal or to nucleate those primary silicon crystals that after growth assume small in size and remain indistinguishable from the eutectic silicon phase.

### References

1. I.J. Polmear, *Light Alloy*, 2nd edition (Edward Arnold, London 1989).
2. P. David, *Modern Casting* 82(2), (1990), 55.
3. M.D. Hanna, S.Z. Lu and A. Hellawell, *Met Trans A* 15, (1984), 459.
4. J. L. Murray, and A. J. McAlister, *Bulletin of Alloy Phase Diagram*, 5( 1), (1984)74-84.
5. J. C. Miller, "Machining High Silicon Aluminum," 11th International Die Casting Congress and Exposition, Cleveland, OH, paper No. G-T, 81-035. (June 1-4, 1981).
6. M. Shamsuzzoha, and F. R. Juretzco, "Dual Refinement of Primary and Eutectic Silicon in Hypereutectic Al-Si Alloys," *Aluminium Alloys for Transportation, Aerospace, and Other Applications*, Edited by: Subodh K. Das, Welmin Yin (The Minerals, Metals, and Materials Society) (2007),153
7. M. Shamsuzzoha, and F. R. Juretzco, Amwarul Haque "Development of High strength Hypereutectic Al-Si Alloys by Refining the Constituent Silicon Phase," *Aluminium Fabrication, Characterization and Applications*, Edited by: Welmin Yin Subodh K. Das, (The Minerals, Metals, and Materials Society), (2008),207.
8. M. Shamsuzzoha, L. Nastac, and J. T. Berry, "Nano-refinement of Eutectic and Primary Silicon Phases in Al-Si Cast Alloys for High Strength Structural Applications", Submitted for publication, *International Journal for Metal Casting*, 2011).
9. L. F. Mondolfo, "Aluminium Alloys", *Structure and Properties*, Butterworth and Co. Ltd., London, U. K. , (1976), 759.
10. M. Shamsuzzoha, and L. M. Hogan, *Phil. Mag. A* 54(4), (1986), 459.
11. S. Lu, and A. Hellawell, *J. Crystal Growth*, 73, (1985), 318.
12. M. Shamsuzzoha, and L. M. Hogan, *Journal of Crystal Growth*, 82 (4), (1987), 598.
13. R. S. Wagner, *Acta Met.*, 8, (1960), 57.
14. D. R. Hamilton, and R. G. Seidensticker, *J. Appl. Phys.* 31, (1960), 1165
15. A. Hellawell, "The Microstructure and Crystallography of Al-Ge Eutectic", *Trans. Met. Society, AIME*, 239, (1967), 1049.
16. T. Laha, A. Agarwal, T. McKechnie, T. Rea, and S. Seal, *Acta Mat.*, 53, 5429, (2005), 5429.
17. B. J. McDonald, and M.S. J. Hashmi, *J. Mater. Process. Technology*, 120, (2002), 341.
18. M. Shamsuzzoha, L. M. Hogan, and J. T. Berry, "Growth Crystallography of Eutectic Phases in Unmodified Al-Si Casting Alloys," *AFS Trans.*, 69, (1992), 619.