

FORMATION OF MICROSTRUCTURE IN AL-SI ALLOYS UNDER ULTRASONIC MELT TREATMENT

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Abstract

It is well known that ultrasonic melt treatment (UST) provides many benefits to casting processing, especially for the refinement or modification of as-cast structure. There is a lack for systematic studies on Al-Si alloys, although a number of reports are available on hypo-eutectic A356-type and hyper-eutectic (18-24 wt% Si) alloys, showing primary Al or Si refinement. In this paper, the effect of UST on the formation of microstructure was systematically analyzed in hypo-eutectic, near-eutectic and hyper-eutectic Al-Si alloys, including commercial piston alloys. The results show that UST usually results in the refinement of grains and primary Si particles when it is applied in a proper temperature range, while ultrasonic treatment during the whole solidification processing leads to coarsening effect on eutectic Si phase or primary Si particles.

Introduction

Cast Al-Si alloys have been widely used in many industrial areas due to their excellent castability, welding ability and corrosion resistance [1]. The presence of primary Si particles in near-eutectic or hyper-eutectic Al-Si alloys provides also an outstanding wear resistance [2].

Usually, fine grain structure and eutectic Si phase/primary Si particles with uniform distribution are desired in real casting practice. In order to modify the microstructure of Al-Si alloy, many techniques are available: (1) deliberate addition of several elements, the most common of which are sodium and strontium [3]; (2) high cooling rate [4]; and (3) mechanical modification, including high intensity shear [5], mechanical or electromagnetic stirring, and ultrasonic vibrations [6].

Ultrasonic melt treatment (UST) is known to induce refining effect in aluminum alloys [6–10]. The basic principle is introduction of acoustic waves with a frequency higher than 17 kHz into liquid metal. High frequency and high amplitude oscillations result in cavitation of the melt and also promote intense mixing through agitation. The reported mechanisms of refinement by UST range from affecting nucleation through local undercooling and wetting of substrates to fragmentation of forming crystals [6, 10].

There is a large number of publications on refined microstructures (grain structure, eutectic Si phase and primary Si particles) caused by UST, which show a promising application prospect for UST in Al-Si alloys. The refinement of both grain structure and primary Si particles has been reported by several researchers [11–14]. However, there have been several conflicting reports on refinement of eutectic Si phase in both hypo- and hyper-eutectic Al-Si alloys. Significant refinement of eutectic Si phases caused by ultrasonic treatment has been observed in Refs [15–17], while Refs [11–12, 18–21] reported the coarsening of eutectic silicon phase when UST was applied to various Al-Si alloy systems.

Although it seems obvious that UST affects the microstructures of Al-Si alloys during solidification processing, the ultrasonic treatment time, temperature range and chosen alloy systems differ from researcher to researcher, which makes it difficult to compare the reported results.

The aim of this paper is to present a systematic study of the effect of the treatment temperature range on the formation of microstructures in Al-Si alloys corresponding to different positions on the phase diagram. Several treatment temperature ranges were chosen for hypo-eutectic, near-eutectic and hyper-eutectic Al-Si alloys. A commercial Al-Si piston alloy was also analyzed after being treated at different temperatures.

Experimental procedure

In this study, hypo-eutectic Al-Si alloy (Al-5wt% Si), near-eutectic Al-Si alloy (Al-11wt% Si) and hyper-eutectic Al-Si alloy (Al-17wt% Si) were cast while applying UST at different temperature ranges, and subsequently analyzed. A commercial Al-Si piston alloy was also studied.

Experimental alloys were prepared using pure Si and 99.97 wt% pure aluminium. The commercial Al-Si piston alloy was obtained from industry (Kolbenschmidt, Germany). The main alloying elements in this commercial alloy are 10 wt% Si, 3wt% Cu, 1wt% Mg and 2wt% Ni.

The experimental setup for UST comprised a 5-kW ultrasonic generator, a 5-kW magnetostrictive transducer with water-cooling system and a niobium ultrasonic horn (sonotrode). Experiments were performed at the 4-kW generator power. The corresponding amplitude of vibrations was 40 μm , as measured by a contactless vibrometer.

In each experiment, the amount of melt was 0.35 kg. The alloys were first molten in an electric furnace in graphite crucibles and then treated by ultrasound in different temperature ranges. In the binary Al-Si alloys, the starting temperature of UST is 40–70 °C above the alloy liquidus. During the ultrasonic treatment, the crucible was exposed to air, and the melt temperature decreased continuously as monitored by a K-thermocouple. The UST was performed in the liquid phase for 10 s, or in the temperature range from the starting melt temperature to a temperature within the solidification range, or during the solidification until the alloy was getting mushy (about 0.9 solid fraction). After ultrasonic processing, the melt was poured either into a copper mold or directly solidified in the graphite crucible. Without ultrasonic treatment the pouring melt temperature was 40–70 °C above the liquidus. In the commercial alloy, the starting temperatures of UST were 650 °C, 670 °C, 700 °C and 720 °C. The treatment time was about 10 s. The reduction in melt temperature during UST for each experiment was 30 °C. Without ultrasonic treatment, the pouring melt temperatures were 650 °C, 670 °C, 700 °C and 720 °C, respectively.

Table 1. Casting conditions and the characteristics of experimental alloys

Alloy	Starting temperature for UST (°C)	Liquidus temperature (°C)	Temperature range of UST	Casting condition	
				mold	Cooling rate (K/s)
Al-5% Si	690	630	No-UST, cast at 690 °C	Copper mold	2
			690–660 °C (liquid state)	Copper mold	2
			690–610 °C (solidification range)	Copper mold	2
			Solidification range until the alloy is mushy	Graphite crucible	0.8
Al-11% Si	630	589	No-UST, cast at 630 °C	Copper mold	2
			630–600 °C (liquid state)	Copper mold	2
			Solidification range until the alloy is mushy	Graphite crucible	0.8
Al-17% Si	720	647	No-UST, cast at 720 °C	Copper mold	2
			720–690 °C (liquid state)	Copper mold	2
			720–620 °C (solidification range)	Copper mold	2
			Solidification range until the alloy is mushy	Graphite crucible	0.8
Commercial Al-10% Si-3% Cu-2% Ni-1% Mg alloy	650, 680, 700, 720	590	UST was applied for 10 s, the reduction of temperature for each starting temperature was 30 °C	Copper mold	2

The casting conditions and their characteristics are summarized in Table 1.

The size of the samples solidified in copper mold is Ø40 mm × 30 mm. The diameter of the samples solidified in graphite crucible is 50 mm, and the height is 45 mm. The microstructures and composition analysis were made on cross section taken from the middle of the samples. The microstructures were examined in a

Neophot-30 optical microscope after being ground, polished. The grain structure was analyzed after electrolytic oxidation at 20 VDC in a 3% HBF₄ water solution. Grain size and primary Si phase were measured using the linear intercept method and the statistical analysis of the results was performed. The chemical composition of all tested Al alloys was measured using a spark spectrum analyzer (Spectro, Kleve, Germany).

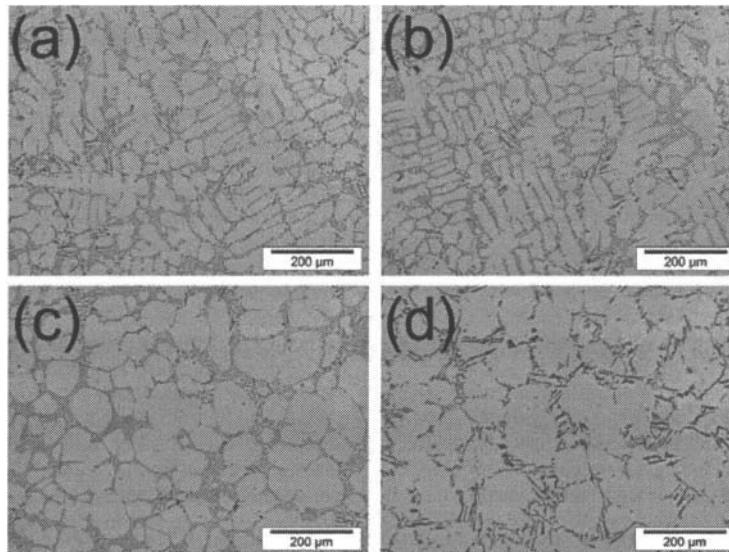


Fig. 1 As-cast microstructure of an Al-5 wt% Si alloy. (a) No-UST; (b) UST in the temperature range 690 to 660 °C; (c) UST in the temperature range 690 to 610 °C; (d) UST until the alloy becomes mushy (about 0.9 solid fraction).

Results and Discussion

1. Effect of UST on microstructure of a hypo-eutectic Al-Si alloy

An Al-5 wt% Si alloy was chosen in this study. The liquidus temperature of this alloy is about 630 °C, as determined by thermodynamic simulations.

Figure 1 presents the as-cast microstructure of this alloy with and without ultrasonic treatment. Acicular eutectic silicon was observed in non-treated alloy, as shown in Fig. 1(a). When UST was applied in liquid stage from 690 °C to 660 °C (Fig. 1(b)) and in the solidification range of primary aluminum from 690 °C to 610 °C (Fig. 1(c)) no obvious change in the morphology of eutectic silicon can be found. However, when the melt was treated until later solidification stages, eutectic silicon phase becomes coarser as illustrated in Fig. 1(d).

Generally, the eutectic silicon forms at 577 °C at normal casting condition. Therefore, it is understandable that no obvious effect of UST on morphology of eutectic silicon can be found when it is applied at a temperature above the eutectic temperature in an Al-Si alloy. Although lots of researches have been done when UST was applied near eutectic temperature or during the whole solidification processing, the reported results are contradictory. Jian et al. [15] used ultrasonic treatment on an A356 alloy and found that, when UST was applied, the average length and width of eutectic silicon decreased from 26 μm to 2 μm and 2.7 μm to 0.6 μm, respectively. And also the eutectic phase spacing was much smaller in the treated alloy. Puga et al. [16] also found the size, thickness and spacing between eutectic silicon lamellae decreased when UST was used on an AISi9Cu3 alloy. Similar results can be found elsewhere [17]. However, the opposite results were reported by Freedman and Wallace [18], Burbure et al. [19], Kocatepe and Burdett [20], Abu-Dheir et al. [21], Eskin and Eskin [11], and Feng et al. [12]. They found the coarsening of eutectic phase when UST was applied in different Al-based alloys. Our results on the formation of eutectic silicon phase are in agreement with the latter, “coarsening” results. Looking through the reports on the refined eutectic, we can find that this refinement is usually observed when UST is applied isothermally in the temperature range close to the eutectic reaction. The introduction of extra ultrasonic energy into the melt does not need to be considered. The formation of eutectic phase depends largely on the fragmentation effect caused by ultrasonic cavitation.

In our case, the temperature of the molten alloy decreased during UST. At a lower temperature close to the eutectic reaction, the

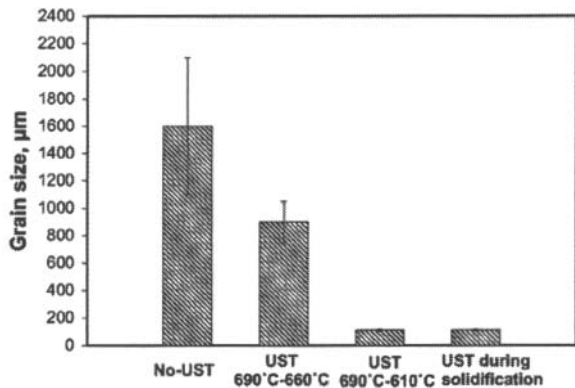


Fig. 2 Effect of ultrasonic treatment temperature range on grain size in an Al-5 wt% Si alloy.

mushy melt (the melt with high percentage of solid fraction) obstructed the further propagation of cavitation and acoustic flow in the melt, preventing the fragmentation. However, the continuous introduction of ultrasonic energy into the mush resulted in the heat input that slowed the cooling rate and changed the growth conditions of the eutectic, reducing the growth rate without causing the fragmentation of the crystals. Therefore, the eutectic structure coarsened when UST was applied at this temperature range. The relatively lower cooling rate in this experiment further increased the size of eutectic phases. This is in line with the experimental results which show that ultrasonic treatment additionally superheats the melt [11, 13].

As for grain structure, UST clearly led to the refinement of Al grains, as summarized in Fig. 2 and Fig. 3. In the non-treated Al-5 wt% Si alloy, fully grown primary Al dendrites can be observed. The average grain size was 1600 μm (Fig. 3(a)). Some dendrites even grown to more than 4 mm. The UST at the liquid state (from 690 to 660 °C) reduced grain size to some extent. This can be explained by transforming non-wetting oxide particles, which are always present in Al melt, into solidification sites [6, 7]. When UST was applied in the solidification range of primary aluminum, the morphology of primary grains changed to globular and equiaxial. The average grain size was about 100 μm (Fig. 3(c)). This result is consistent with our previous results [7–9] and reported results [6, 22–24]. The cavitation-assisted fragmentation is considered as the main contributor to the refinement of grain size in the hypo-eutectic Al-Si alloy in this study.

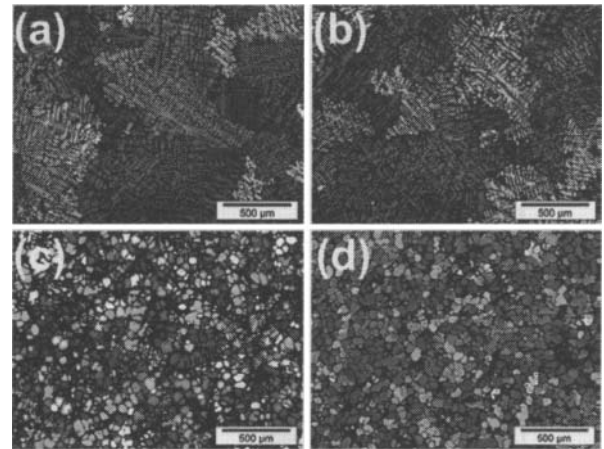


Fig. 3 Grain structure of an Al-5 wt% Si alloy. (a) No-UST; (b) UST in the temperature range 690 to 660 °C; (c) UST in the temperature range 690 to 610 °C; (d) UST until the alloy becomes mushy (about 0.9 solid fraction).

2. Effect of UST on microstructure in near-eutectic Al-Si alloy

In this Al-11%Si alloy, two treatment temperature ranges were used. As shown in Fig. 4, primary Si particles can be observed due to non-equilibrium solidification in the coupled zone [25]. When the melt was treated for about 10 s in the liquid state, from 630 to 600 °C, the size and morphology of Si particles and eutectic Si phases were similar to those in the non-treated alloy. This means that the treatment temperature in this case did not reach the non-equilibrium solidification range of primary Si. The average Si particle size was about 25 μm in both cases. When UST was applied until lower temperatures, the eutectic Si phase coarsened and became more discontinuous. The probable reason was

suggested above. It should be noted that although the average size of primary Si particle did not change a lot as compared with the non-treated alloy and the alloy treated from 630 °C to 600 °C, the morphology of part of Si particles was starting to change from faceted to more rounded, as shown in the central part of Fig. 4(c). Although not all particles changed in our experiment, the trend was clear and similar result had been reported before [13]. It was suggested that the fragmentation of large primary particles followed by aggregation of the fragmented Si is responsible for the spheroidization of the primary Si particles [13].

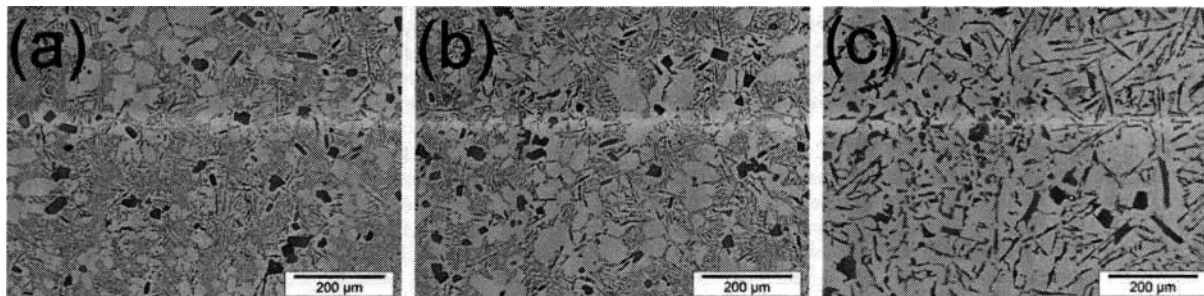


Fig. 4 As-cast microstructure of an Al-11 wt% Si alloy. (a) No-UST; (b) UST in the temperature range 630 to 600 °C; (c) UST until the alloy becomes mushy (about 0.9 solid fraction).

Due to a relatively narrow range of solidification of primary aluminum in this alloy as compared to the Al-5 wt% Si alloy, the UST did not result in grain refinement as significant as in the Al-5 wt% Si alloy, as shown in Fig. 5. The grain size decreased from 650 μm to 500 μm and further to 250 μm when UST was applied for 10 s in the liquid state and during solidification, respectively.

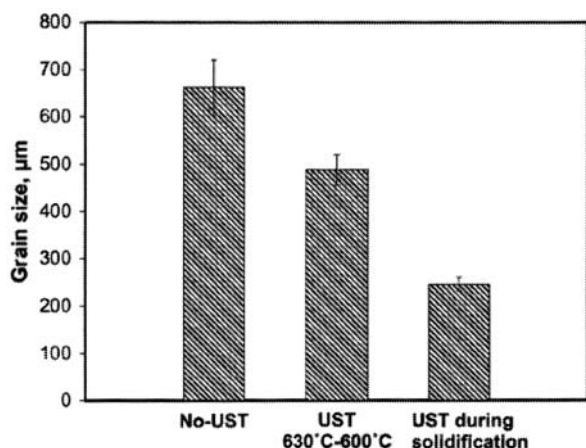


Fig. 5 Effect of ultrasonic treated temperature range on grain size in Al-11 wt% Si alloy.

3. Effect of UST on microstructure in hyper-eutectic Al-Si alloy

Good results have been reported for refining of primary Si in hypereutectic Al-Si alloys [11–14, 26]. However, the effect of temperature range of treatment on the formation of primary Si phase during UST remains unclear. In our study, an Al-17 wt% Si alloy was treated in three different temperature ranges in order to understand the mechanism of Si refinement.

Figure 6 shows the microstructures of the Al-17 wt% Si alloy without and after UST. When UST was applied for 10 s in the liquid state, from 720 to 690 °C, a dramatic refinement of primary

Si particles can be achieved. The average particle size decreased from 45 μm to 18 μm, as illustrated in Fig. 6(b). However, continuation of the ultrasonic treatment to lower temperatures, during the primary Si solidification, resulted in local coarsening, as illustrated in Fig. 6(c) and 6(d). The average size of primary Si particles is summarized in Fig. 7.

There are two factors that may influence the refinement of primary Si particles. On one hand, UST multiplies the nucleation sites for the formation of primary Si phases, either through transforming non-wettable particles present in the melt into solidification sites,

or through increased undercooling related to cavitation expansion and collapse [6]. On the other hand, the cavitation-assisted fragmentation may further reduce the size of primary Si phase [10]. As the primary Si refinement was achieved in our case when UST was performed above the liquidus of the Si primary formation, the former mechanism is more likely to act.

As we have discussed above, when the melt with high percentage of solid fraction is treated by ultrasound, the propagation of ultrasonic effect (i.e. cavitation) is limited except for the heat input and transfer. This heat input increases the alloy temperature or reduces the cooling rate during solidification. As a result, the primary Si particles coarsen and some previously refined Si particles may coalesce and agglomerate as can be observed in Fig. 6(c) and Fig. 6(d). The main difference between treatment temperature ranges from 720 to 620 °C and to the mushy zone is that in the latter case the UST caused the coarsening of eutectic Si phase as well. In addition, the relatively lower cooling rate in the graphite crucible also contributes to the formation of coarse eutectic phases.

4. Application of UST to a commercial Al-Si piston alloy

In order to check the application prospect of UST in Al-Si alloys, a commercial Al-Si piston alloy was used. A range of casting temperatures was chosen to resemble the real casting practice. The melt was treated for about 10 s in order to avoid any coarsening effect which has been shown previously in binary Al-Si alloys.

The results are illustrated by the plot in Fig. 8 and the microstructure in Fig. 9. Without UST, the size of primary Si particles decreased with increasing casting temperature. Ultrasonic treatment resulted in a consistent refinement of primary Si particles, but did not lead to any coarsening effect on eutectic Si. The results show that the closer the UST temperature to the liquidus, the greater the refinement effect is. This is in good agreement with cavitation-assisted nucleation mechanism which shows that the significant refinement usually appear when UST activates the nucleation sites and they have more chance to survive at a lower melt temperature.

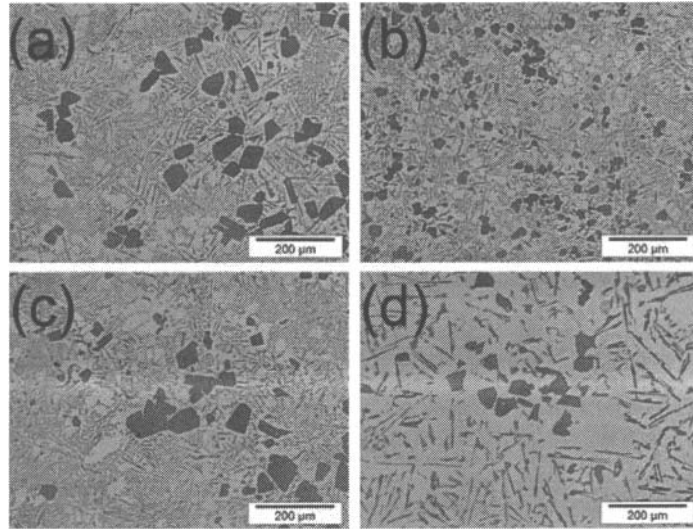


Fig. 6 As-cast microstructure of an Al-17 wt% Si alloy. (a) No-UST; (b) UST in the temperature range 720 to 690 °C; (c) UST in the temperature range 720 to 620 °C; (d) UST until the alloy becomes mushy (about 0.9 solid fraction).

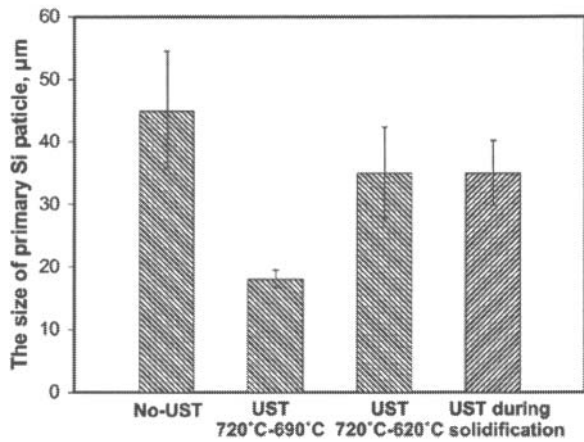


Fig. 7 Effect of ultrasonic treatment temperature range on the size of primary Si particles in an Al-17 wt% Si alloy.

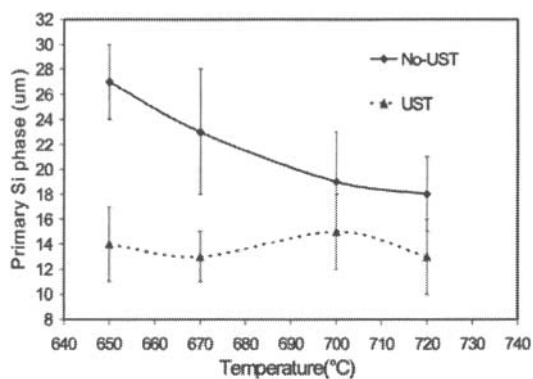


Fig. 8 Effect of ultrasonic treatment temperature and casting temperature on the size of primary Si crystals in the commercial piston alloy.

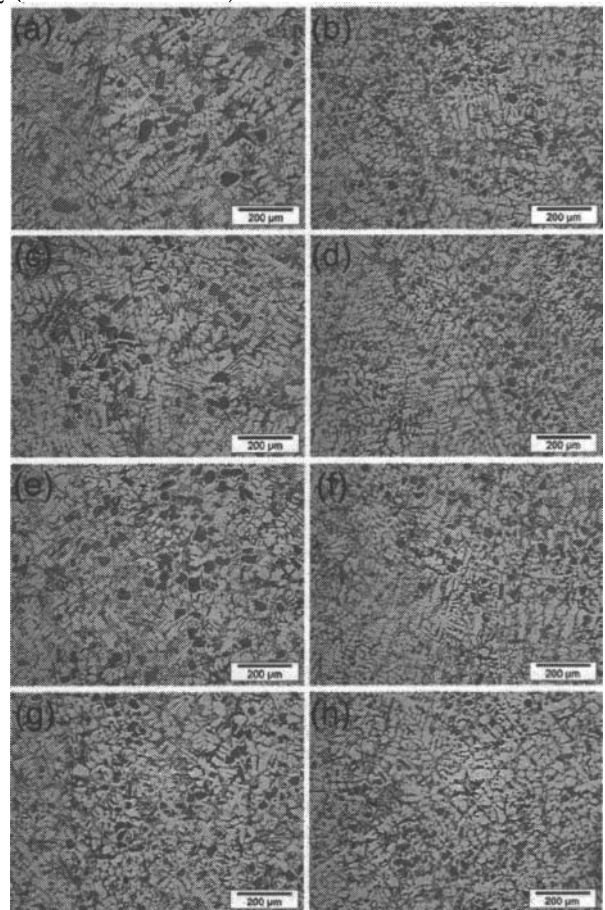


Fig. 9 As-cast microstructure of a commercial Al-Si piston alloy. (a) No-UST, cast at 650 °C; (b) UST, cast at 650 °C; (c) No-UST, cast at 670 °C; (d) UST, cast at 670 °C; (e) No-UST, cast at 700 °C; (f) UST, cast at 700 °C; (g) No-UST, cast at 720 °C; (h) UST, cast at 720 °C;

Conclusions

Experimental results show that ultrasonic treatment affects the formation of microstructures in hypo-eutectic, near-eutectic and hyper-eutectic Al-Si alloys when UST is applied in different temperature ranges with respect to the forming phases.

(1) A refined grain structure can be achieved when the melt is continuously treated during solidification of the primary Al phase. The cavitation-assisted fragmentation is considered as the main contributor to the refinement of grain size in a hypo-eutectic Al-Si alloy.

(2) When UST is applied during solidification, the Al-Si eutectic tends to coarsen. This might be due to the heat input from the ultrasonic treatment while the fragmentation mechanism cannot work as a result of limited propagation of acoustic cavitation and flows in the mushy material. As a result the multiplication of eutectic particles does not occur while their coarsening is facilitated in the hotter environment. In addition, the use of graphite crucible in this series of experiments might also reduce the cooling rate and promote the formation of coarse eutectic phases.

(3) A significant refinement of primary Si crystals is observed in hypereutectic alloys when UST was applied to the liquid phase close to the liquidus of the primary Si phase. However, UST performed during further solidification might result in coarsening and agglomeration of primary Si.

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