Degassing effect of ultrasonic vibration in molten melt and semi-solid slurry of Al-Si alloys

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Abstract: In the process of semi-solid slurry preparation with direct ultrasonic vibration (UV) by dipping the horn into the melt, one of the questions is whether the gas content in the melt would be increased or not by the cavitation effect of ultrasonic vibration. By application of quantitative gas content measurement technique, this paper investigated the effect of the ultrasonic vibration on the gas content of both the melt and the semi-solid slurry of Al-Si alloys, and the variations of the gas contents in two kinds of aluminum alloys, i.e., A356 alloy and Al-20Si-2Cu-1Ni-0.6RE alloy (Al-20Si for short). The results show that ultrasonic vibration has an obvious degassing effect on the molten melt, especially on the semi-solid slurry of Al-Si alloy which is below the liquidus temperature by less than 20 ℃. The ultrasonic degassing efficiency of the A356 alloy decreases with the reduction of the initial gas content in the melt, and it is nearly unchanged for the Al-20Si alloy. The gas content of both alloys decreases when the ultrasonic vibration time is increased. The best vibration time for Al-20Si alloy at the liquid temperature of 710 ℃ and semi-solid temperature of 680 ℃ is 60 s and 90 s, respectively; and the degassing efficiency is 48% and 35%, respectively. The mechanism of ultrasonic degassing effect is discussed.

Key words: ultrasonic vibration; semi-solid slurry; aluminum alloy; gas content; degassing
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During the melting of aluminum alloy, it is easy for the melt to absorb gases, and the research results indicated that the hydrogen content in the gases reaches 80% or more [1,2]. Therefore degassing is essentially hydrogen gas eliminating. Ultrasonic vibration, as an environmentally green way of degassing, has attracted a lot of research internationally. It was verified that ultrasonic vibration has the effect of degassing on high-temperature molten aluminum and its alloys. Xu et al [3-5] compared the degassing effects between ultrasonic vibration and argon degassing for A356 alloy, and pointed out that ultrasonically assisted argon degassing is a more efficient method than argon degassing alone. These studies were almost all carried out in the temperature range of 620 to 740 ℃, and the degassing effects were evaluated through measuring the specimen's density which solidified under reduced pressure, or the pinhole distribution on the sectional plane of specimens [3-7]. H. Puga [6] studied the degassing efficiency of AlSi9Cu3 alloy with ultrasonic vibration and combined with melt stirring, in which the ultrasonic degassing temperature was 700 ℃ and the degassing effect was evaluated through measuring the specimen's density solidified under reduced pressure, and he found that melt stirring significantly improves degassing efficiency, and such improvement depends on the melt temperature. Li [7] studied the influences of ultrasonic time, power, frequency and other parameters on the gas content of pure aluminum and its alloy, in which the treatment temperature was 40 ℃ above the alloy's liquidus temperature, and the gas content was also evaluated through the ingot density. The ingot density was increased after ultrasonic vibration.

Most of the above studies were carried out under a temperature about 40 to 100 ℃ higher than the liquidus line, and show a lack of quantitative measurement of the gas content. Almost all studies used the density measurement method to indirectly estimate the gas content and then evaluated the degassing effects.

Recently, the authors [8-10] developed a preparation process of semi-solid slurry with direct ultrasonic vibration (DUV), i.e., the slurry was made by dipping the horn directly into the melt and non-dendritic structure was obtained through ultrasonic vibration. This process has important application prospect. Because the temperature of semi-solid slurry preparation is relatively low, and the slurry is in solid/liquid two-phase co-existent state, the apparent viscosity of the slurry is higher than the melt. With ultrasonic vibration at the semi-

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solid temperature, would the solid particles hinder ultrasonic cavitation bubbles floating and escaping from the surface, or lead to the increase of gas content and pinholes? This is one of the important issues which affect the development of ultrasonic semi-solid process. There are no related studies reported in the literature.

This paper utilizes a quantitative gas content measurement technology for aluminum alloys to directly measure the variation of gas content. The effects of ultrasonic vibration on degassing efficiency at molten melt and semi-solid temperatures have been studied. The main contents of the study include the effects of slurry temperature, initial gas content, and time and power of ultrasonic vibration on the degassing efficiency.

1 Experimental procedure

Two kinds of Al alloys were used in the experiments: the hypoeutectic Al-Si alloy A356 and a new high silicon alloy Al-20Si-2Cu-1Ni-0.6RE (Al-20Si for short in the following). The semi-solid temperature range of A356 alloy is 555 to 615 ℃[8], and for Al-20Si alloy is 533 to 694 ℃ [11]. The chemical compositions are shown in Table 1.

Table1: Chemical compositions of A356 and Al-20Si alloys (wt.%)

	Si	Mg	Fe	Cu	Ni	Mn	RE	Al
A356	7.2	0.35	0.19	<0.1		0.05		Bal.
Al-20Si	20.0	0.4	0.29	2.05	1.0	0.5	0.6	Bal.

The equipment used in the experiments includes ultrasonic vibration equipment [8] and an AH-2 type on-line quantitative gas content measurement instrument for the Al alloy melt [12]. The common parameters of ultrasonic vibration are: ultrasound frequency 20 kHz, rest-work ratio during ultrasonic vibration 1:1, diameter of ultrasonic-vibration horn 20 mm, and the horn insertion depth 10 mm. The amount of the treated Al alloy melt was maintained constant at 400 g. The gas content measurement of the melt was taken after ultrasonic vibration, and then the cast samples were solidified under reduced pressure at about 20 kPa. Sample density was measured after solidification by using an Archimedes density-measurement method. The sample was cut along the axis, the section was polished, and then the change of gas holes or shrinkage cavity was observed and analyzed.

The accuracy of the gas content tester is ±0.01 cm^3/100g Al, and measuring time is about 2 to 4 min. The ultrasonic degassing efficiency is defined by equation (1) below:

$$\eta = \frac{C_{Hi} - C_{Hf}}{C_{Hi}} \quad (1)$$

In equation (1), η is the degassing efficiency; C_{Hi} is the gas content in the melt before ultrasonic vibration, cm^3/100g Al; C_{Hf} is the gas content in the melt after ultrasonic vibration, cm^3/100g Al.

Three kinds of experiment were conducted to investigate the influence of ultrasonic vibration on the gas content in melt:

(1) The influence of different slurry temperatures and initial gas contents in the melt on the ultrasonic vibration (UV) effect:

The UV experiments were done at several molten melt temperatures and semi-solid temperatures. All UV time was 90 s, UV power was 1.2 kW, and then the gas content in the melt was measured after UV treatment. The different initial gas contents in the melt were achieved through argon degassing of the molten melt.

(2) The influence of different ultrasonic vibration times:

The UV experiment was made at molten melt or semi-solid state with A356 alloy and Al-20Si alloy, and the UV time of 30 s, 60 s, 90 s, and 120 s was used respectively. The UV power was 1.2 kW. For A356 alloy, the testing temperature at molten melt or semi-solid state was 650 ℃ and 613 ℃, respectively, and for Al-20Si alloy was 710 ℃ and 680 ℃, respectively.

(3) The influence of different ultrasonic vibration powers:

The UV experiment was conducted with Al-20Si alloy at liquid temperature of 710 ℃ and semi-solid temperature of 680 ℃. The UV powers were 400 W, 800 W, 1,200 W, and 1,600 W, for which the corresponding power densities were 2.5, 5, 7.5, 10 $W{\cdot}cm^{-3}$ respectively. The UV time was maintained constant at 90 s.

2 Results

2.1 Influence of melt or semi-solid temperatures on ultrasound-degassing efficiency

Figures 1 and 2 show the relationship between slurry temperature and gas content of the two alloys under four different treatments: no ultrasonic vibration and argon degassing (untreated for short), only ultrasonic vibration, only argon degassing, and combination of ultrasonic vibration with argon degassing.

From Figs. 1 and 2 it can be seen that the gas contents of the two alloys decrease with the decreasing of temperature, but the gas contents under different treatments are totally different. The following conclusions can be drawn from the analysis of the results.

(1) In the semi-solid temperature range of A356 alloy (Fig. 1), the gas content decreases after ultrasonic treatment. As shown in Fig. 1, in the 595 to 615 ℃ range, the two groups of experimental results with UV (curves 2 and 4) show that ultrasonic vibration at semi-solid temperature has the function for degassing. The degassing rate analysis is as follows: with only UV, η is 15% at semi-solid temperature. With the same low initial gas content after argon degassing, η at semi-solid temperature can still be 7.33% (From curves 3 and 4).

For Al-20Si alloy (Fig. 2), with only UV, ultrasound degassing rates η at 710 ℃, 690 ℃, 680 ℃, and 670 ℃ are 30.0%, 28.1%, 25.8%, and 21.0% respectively. Therefore the conclusion is that even at a relatively low temperature of 670 ℃ (24 ℃ below the liquidus line), η can be 21% or higher.

(2) For A356 alloy, η decreases when initial gas content in the melt is small. In Fig. 1, the difference between curve 3 and curve 4 is not as obvious as between curve 2 and curve 1. Analysis from the above data indicates that, at 608 ℃, η reduced from

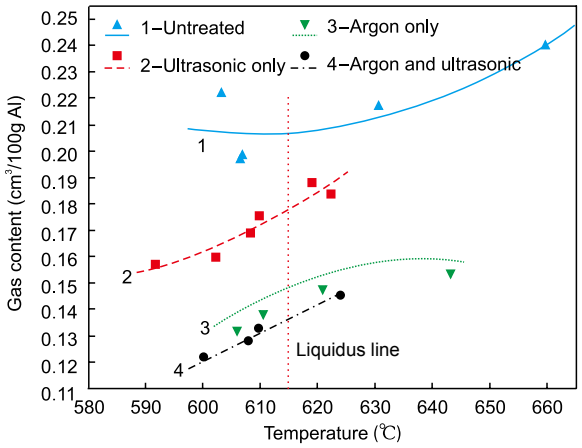

Fig. 1: Relation between gas content in melt and UV temperature for A356 alloy

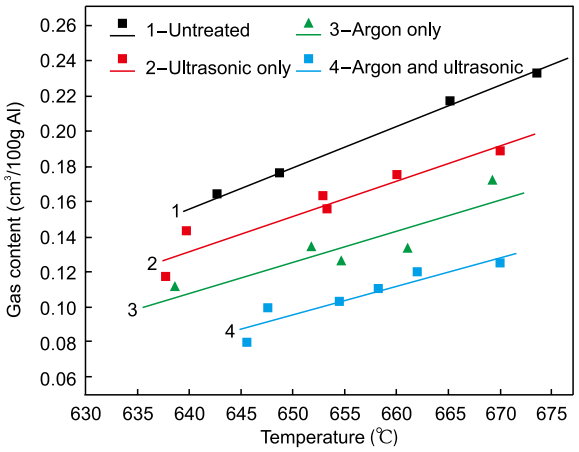

Fig. 2: Relation between gas content and UV temperature for semi-solid Al-20Si alloy slurry

15% to 7% after argon degassing of the melt. After argon degassing treatment, the initial gas content of the slurry decreases, therefore the hydrogen gas which can be removed by UV is reduced.

For Al-20Si alloy, the effect of the initial gas content on ultrasound degassing rate is small. From the four curves in Fig. 2, the distance between curve 1 and curve 2 is nearly equal to the distance between curve 3 and curve 4. The analysis result of degassing rate shows that with no argon degassing treatment η is about 20%; while with argon degassing, η is 18%. The degassing efficiencies under both conditions are similar.

(3) From the positions of the four curves in each of Figs. 1 and 2 and the absolute values of gas content in melt, the best degassing efficiency for the two alloys is under the condition of ultrasonic vibration combined with argon gas degassing. The order of degassing rate from low to high is: ultrasonic vibration only < argon degassing only < ultrasonic vibration after argon degassing.

2.2 Influence of UV time on ultrasound degassing efficiency

The ultrasonic vibration time has a great influence on degassing efficiency. Figure 3 shows the variation of gas content in the melt with the time of UV treatment of the molten melt of the two alloys, respectively, at 650 ℃ for A356 alloy and at 710 ℃ for Al-20Si alloy. Figure 4 shows the variation of gas content in the melt with the time of UV treatment at the semi-solid temperatures of the two alloys, at 613 ℃ for A356 alloy and at 680 ℃ for Al-20Si alloy, respectively.

From Figs. 3 and 4 it is easy to see that the gas content in the melt is decreased after UV for both molten melt and semi-solid slurry. In other words, ultrasonic vibration has a positive effect on degassing for both molten melt and semi-solid slurry. But the trends of gas content variation with UV time are different between these two alloys. For A356 alloy, at molten melt and semi-solid temperatures, the longer the UV time, the lower the gas content. With 120 s vibration, both of the gas contents in melt reach the lowest point at the molten melt temperature of 650 ℃ and semi-solid temperature of 613 ℃, respectively, and η is 48% and 33% respectively. For Al-20Si alloy, at molten or semi-solid temperatures, the gas content drops at first, then goes up with the extension of UV time. The best UV time at molten melt temperature of 710 ℃ or semi-solid temperature of 680 ℃ is 60 s and 90 s, respectively, and η is 48% and 35% respectively. This is because longer UV time can cause the oxidation to increase at the melt surface and absorb gases, resulting in an increase of gas content.

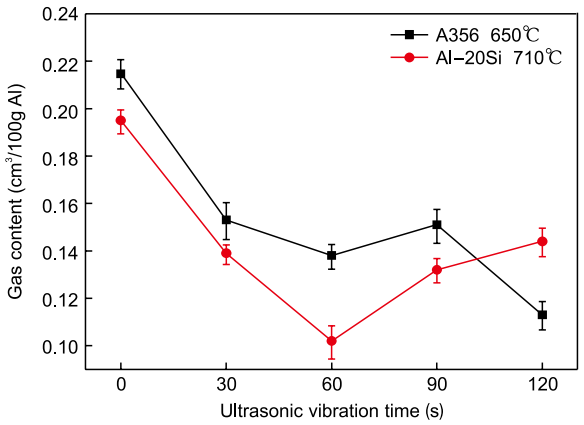

Fig. 3: Relation between gas content in melt and UV time at molten melt temperature

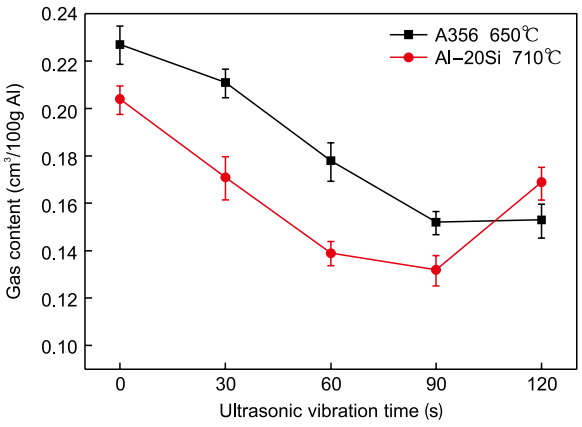

Fig. 4: Relation between gas content in melt and UV time at semi-solid temperature

Comparing the gas contents in molten melt and semi-solid slurry, it can be found that ultrasonic degassing at molten melt temperature is faster than that at semi-solid temperature; and the minimum gas content in molten melt can be smaller than that in semi-solid slurry. When comparing the two alloys, the degassing rate of Al-20Si alloy is bigger than A356 alloy.

Sample density can indirectly reflect the gas content in melt, and it can be used in qualitative analysis of the degassing efficiency. Figures 5 and 6 show the variation of sample density of the two alloys with UV time for molten melt and semi-solid slurry, respectively.

It can be seen from Figs. 5 and 6 that after UV at molten melt temperature or semi-solid temperature, the sample density is obviously increased, and it is also increased with UV time. The variation of sample density indirectly indicates that UV has an obvious effect on degassing for A356 alloy and Al-20Si alloy; and, especially at semi-solid temperature, the UV still has a good effect on degassing, and does not increase the gas content of the alloys.

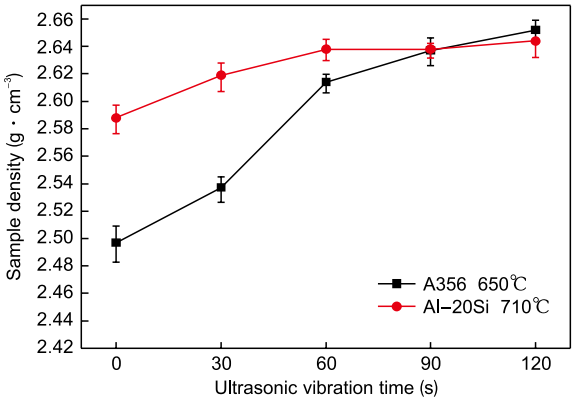

Fig. 5: Relation between sample density and UV time at molten melt temperature

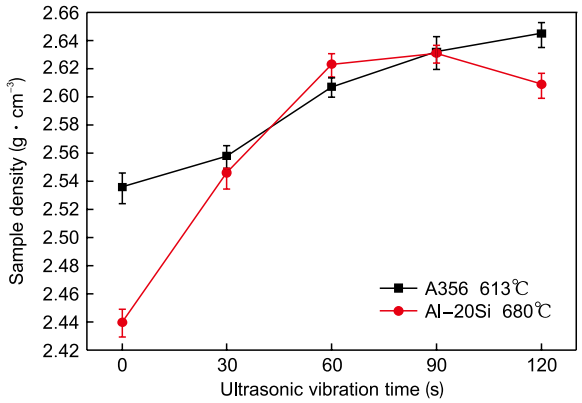

Fig. 6: Relation between sample density and UV time at semi-solid temperature

The gas holes or shrinkage cavities in solidified samples can also indirectly reflect the gas content in melt. Figures 7(a) and 7(c) are sectional photos of samples of A356 alloy and Al-20Si alloy with no UV, and Figures 7(b) and 7(d) are samples of A356 alloy and Al-20Si alloy with 60 s of UV treatment at the semi-solid temperature, respectively. Please note that these samples were solidified under reduced pressure at about 20 kPa.

From Fig. 7 it is clear that, after UV treatment, the gas holes or shrinkage cavities area is decreased greatly.

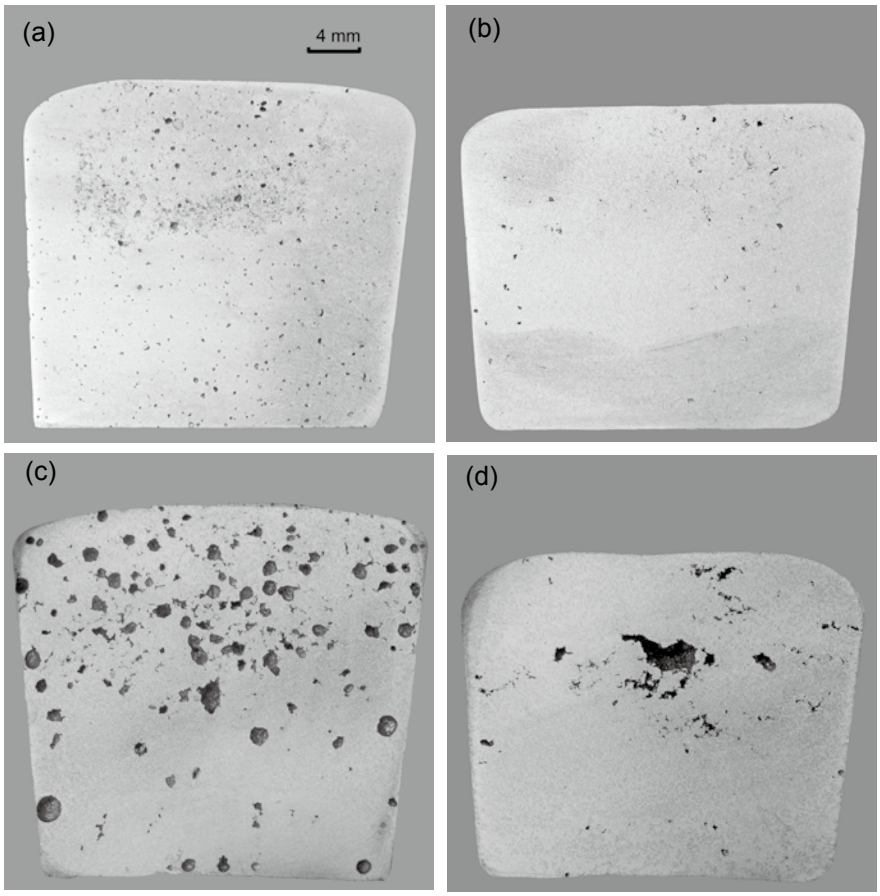

Fig. 7: Gas-holes and shrinkage cavities of samples solidified under reduced pressure: (a) A356 alloy, no UV; (b) A356 alloy, UV for 60 s in semi-solid state; (c) Al-20Si alloy, no UV; and (d) Al-20Si alloy, UV for 60 s in semi-solid state

2.3 Influence of UV power on ultrasound degassing efficiency

Ultrasonic vibration power is directly related to the threshold value of ultrasonic degassing; therefore it has very big effect on degassing efficiency. Figure 8 shows the variation of gas content with UV power for Al-20Si alloy, at molten melt temperature of 710 ℃ and semi-solid temperature of 680 ℃, respectively.

It can be seen from Fig. 8 that for Al-20Si alloy, the trend in variation of gas content with UV power is nearly the same at molten melt and semi-solid temperatures, that is, the gas content gradually decreases at first with the increasing ultrasonic power, it has a minimum value at 1,200 W and then goes up a little. The reason for the going up is mainly because of the violent movement of surface melt under high UV power of 1,600 W. For molten melt or semi-solid slurry, even if only with 400 W power, η can reach 20%.

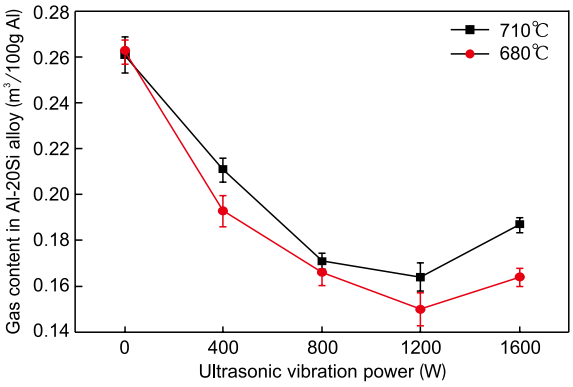

Fig. 8: Variation of gas content with UV power for Al-20Si alloy

The highest value of η is 37% in molten melt, and 35% in semi-solid slurry, at 1,200 W respectively. It is obvious that ultrasound has very good effect on degassing efficiency for Al-20Si whether molten melt or semi-solid slurry. In a range of 0 to 1,600 W, the degassing efficiency is the best in a range of 800 to 1,600 W; or, to put it another way, the degassing efficiency is at maximum in a volume power-density range of 5 to 10 W·cm^{-3}.

From the relationship between the sample density and UV power shown in Fig. 9, it can be seen that whether treated at molten melt temperature of 710 ℃ or semi-solid temperature of 680 ℃, for Al-20Si alloy, the sample density increases with increasing of UV power, and gradually becomes more stable. The density results also indicate that the greater the UV power, the better the degassing efficiency.

3 Discussion

It is considered that the degassing effect is a co-action result of cavitation and acoustic streaming effects of ultrasound in the melt and semi-solid slurry.

The cavitation effect can produce cavitation bubbles at negative-pressure period [13]. After the formation of cavitation bubbles, parts of them rupture quickly during the following

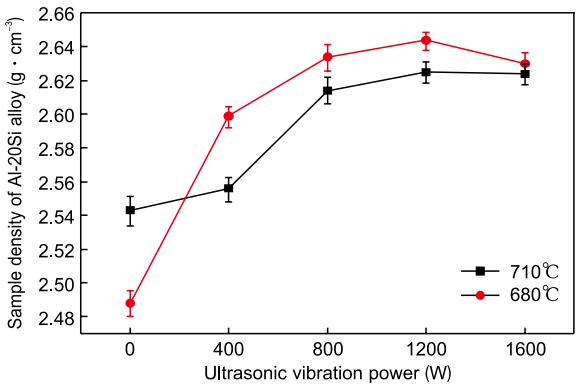

Fig. 9: Relationship between sample density and UV power for Al-20Si alloy

acoustic positive-pressure. The other parts of bubbles adsorb hydrogen molecules under the function of surface tension and the internal negative pressure. Tiny cavitation bubbles constantly gather together and grow larger. Bubbles would float up out of the melt surface when they can overcome resistance, thereby the degassing purpose is reached.

Acoustic streaming effect refers to a fluid jet caused by the sound-pressure gradient generated by the ultrasonic-wave amplitude decay in the fluid [13]. This jet leaves the end surface of the horn directly and leads to a whole circulation, which promotes convection in the melt. In this way, the melt far from the ultrasonic vibration head could behave as if close to the head, thus expanding the range of ultrasonic cavitation, making more hydrogen spread into the bubbles and increasing the degassing efficiency.

The presence of cavitation nuclei would reduce the threshold of ultrasonic cavitation greatly [14]. Impurities are common in metal, like Al_2O_3, which have depressions or cracks on their surface. Because of the effect of surface tension, the aluminum melt could not fill these places completely. Hydrogen gas would spread to such regions and be stable in these gaps, thereby becoming the nuclei of ultrasonic cavitation. From what has been discussed above, on the one hand, hydrogen gas in the melt could promote cavitation bubble nucleation; on the other hand, it could spread into and expand the cavitation bubbles and help their growth.

Some scholars studied the organic material solution under ultrasonic vibration and presented an empirical formula to calculate the cavitation threshold [15]:

$$P_c = 0.8\ [\log(\mu) + 5] \qquad (2)$$

In equation (2), P_c refers to the cavitation threshold and μ is the melt viscosity.

The melt viscosity would affect significantly the attenuation of sound-pressure and the cavitation threshold value. The greater the viscosity, the higher the cavitation threshold value; and vice versa. The greater the solid fraction in the melt, and the more developed the primary dendritic grains, the greater the apparent viscosity of the melt; all of which reduce the region of cavitation [14].

In the study of A356 alloy, when the ultrasound works

in the range of semi-solid temperature, hydrogen solubility in semi-solid state is lower compared with the molten melt. Precipitation of hydrogen gas increases the probability of its entering into the cavitation bubbles. In the semi-solid temperature range, although the viscosity of the melt increases when the temperature decreases, it would be decreased by the melt convection caused by the acoustic streaming. Convection is also beneficial to the movement of the melt and raises the probability of the cavitation bubbles at the bottom floating to the surface. Under the combination of the above factors, ultrasonic vibration in the A356 semi-solid slurry still has the effect of degassing.

Unlike the A356 alloy, when the temperature of Al-20Si alloy is below the liquidus level by no more than 20 ℃, or even if the initial gas content is lower, UV still has a favorable effect of degassing. In cast Al-Si alloy, the higher the silicon content, the better the fluidity of the alloy. When the silicon content in the alloy is at the hypereutectic composition, the liquidus temperature increase is not conducive to improving the fluidity of the alloy, because of the great freezing latent-heat of silicon. However, the silicon phase will release a lot of heat maintaining the melt over-heating during solidification, thereby increasing the fluidity of Al-20Si alloy [15,16]. Therefore during the operation of ultrasonic vibration even at the semi-solid temperature, the Al-20Si alloy slurry still has good fluidity because of this property; so the convection effect caused by the acoustic streaming in the slurry and the effecting area of cavitation is not obviously minimized. That is why the ultrasound degassing rates in liquid state or semi-solid slurry are almost the same for the Al-20Si alloy.

4 Conclusions

(1) The degassing effect of ultrasonic vibration is obvious for molten melt or semi-solid A356 alloy and Al-20Si alloy. The ultrasound degassing rate for A356 semi-solid slurry is 15% at 608 ℃ and 21% for Al-20Si semisolid slurry at 670 ℃.

(2) With the decrease of initial gas content in molten melt or semisolid slurry, the ultrasound degassing rate of A356 alloy decreases. However, the initial gas content has little effect on the ultrasound degassing rate of Al-20Si alloy. At about 650 ℃, the ultrasound degassing rate is stable at about 18% under different initial hydrogen concentrations.

(3) For Al-Si alloy, the gas content in melt or slurry decreases with the extension of vibration time. But at semi-solid temperature, an over-long vibration time can cause an increase of gas holes or porosities. The best vibration time is from 60 to 90 s.

(4) It is considered that the degassing effect is a co-action result of cavitation and acoustic streaming effects of ultrasound in the melt or semi-solid slurry. On the one hand, the hydrogen gas promotes nucleation of cavitation bubbles; on the other hand, hydrogen gas makes cavitation bubbles expand and grow; and these bubbles float up and escape from the surface under the assistance of convection caused by acoustic streaming.

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