DISSOLUTION STUDIES OF SI METAL IN LIQUID AI WITH GAS INJECTION

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ABSTRACT

This study investigates the role of nitrogen gas injection in the dissolution rate of Si into liquid Al. A unique revolving liquid metal tank was used, with a capacity of 50 kg of liquid Al. Experiments without gas injection have shown that the dissolution of Si increases as the bath superheat and the tangential velocity of the liquid Al increase. When injecting nitrogen gas at flow rates between 0-5 lit/min in the vicinity of the solid Si, results so far clearly show greater dissolution. In addition, results on the role of the gas flow rate relative to the position of the nozzle are presented and analyzed.

INTRODUCTION

In liquid metals processing operations, low assimilation rates of additions affect the production cost. The assimilation of a solid metal into a liquid metal can be distinguished as melting or dissolution: the former takes place through heat transfer, while the latter occurs when the solid comes in contact with liquid at a temperature below the melting point of the solid. This study focuses specifically on the dissolution of solid Si into liquid Al. Al-Si alloys have attractive properties for the aerospace and automotive industries. However, the production process is very slow, because the time required for Si to dissolve into liquid Al is long. Industrially, at Novelis, chunks of Si are added to molten Al, and stirred for up to 30 minutes. During that time, dross forms on the top of the melt that is skimmed off. The longer the alloying process, the more dross forms, therefore, we are seeking to accelerate the dissolution process to reduce material loss and energy consumption, by reducing the tap to tap time. Significant savings would be achieved by only a 10% decrease in the dissolution time [1-2].

When an addition (initially at room temperature) is introduced into a liquid metal, the addition and the liquid quickly reach a thermal equilibrium, and then the solid begins to dissolve and its size gradually decreases. Dissolution of a solid addition in a metal bath takes place in two steps [3]:

1. Interface reaction at the solid-liquid interface, $C_{Si} \rightarrow C_{Sat}$

Atoms migrate from the solid phase into the melt. The concentration of the dissolving species in the liquid at the interface is given by the liquidus curve on the Al-Si phase diagram [3], at the melt temperature.

2. Transport of dissolved species from the interface to the bulk liquid metal, $C_{\text{Sat}} \rightarrow C_{\text{b}}$

The dissolved species travels from the solid-liquid interface to the edge of the concentration boundary layer, and then from the edge of the boundary layer to the bulk liquid Al.

When the rate of dissolution is determined by the rate of mass transfer across the concentration boundary layer, flow in the vicinity of the addition will enhance the mass transfer coefficient. Injecting nitrogen gas within the melt can be used to agitate the flow, and therefore promote the dissolution rate.

In the case of heat transfer only (i.e. solid Al melting into liquid Al), Sismanis and Argyropoulos [4] showed that gas injection flow increases the convective heat flux in liquid Al. As heat transfer is analogous to mass transfer, it would seem that mass transfer rates can be similarly increased by injecting gas. Independent support for this proposition can be obtained from work carried out with fluids other than liquid metals. Kim and Fruehan [5] conducted experiments to measure the mass transfer coefficient of solid benzoic acid in water, at various positions when stirred by gas injected. Results showed that for the gas flow rates tested, mass transfer coefficients increased with increasing gas flow rates, regardless of the position of the sample. Mazumdar et al. [6] examined a similar system and found that mass transfer rates are higher in the gas stirred plume compared to a region outside of the plume. Iguchi et al. [7] measured the mass transfer coefficient of a solid sphere and a flat plate immersed in a cold gas-stirred bath. Measurements were made for a wide range of turbulence intensity values, and correlations for Sherwood numbers were derived, that confirmed a significant effect of gas agitation on mass transfer rates.

Gas agitation changes the turbulence intensity of the moving liquid Al. This change has the potential of increasing the dissolution rate of an addition within the Al bath. The current study seeks to accelerate the dissolution of Si into liquid Al by changing the turbulence intensity of the Al bath. Previously, the mechanical stirring of the melt was addressed [1]. The current work establishes a quantitative comparison of the dissolution rate with and without gas injection.

EXPERIMENTAL SETUP

Electric Resistance Furnace and Revolving Liquid Metal Tank

The melting of the Al metal took place inside a Revolving Liquid Metal Tank (RLMT). This (RLMT) is inside a resistance furnace and is made from mild steel as shown in Figure 1. In an effort to minimize the dissolution rate of steel crucible into the Al bath, the walls of RLMT, which were in touch

with liquid Al, were painted with boron nitride. The central heating area, which houses the (RLMT), totals 0.018 m^2 . Two K-type thermocouples located in the furnace hot zone were used to control the temperature.



Figure 1 - The electrical resistance furnace including the revolving liquid metal tank showing the Al charge prior to melting

The cylindrical steel tank has a capacity of approximately 50 kg (20 l) of Al. The interior radius of the Revolving Liquid Metal Tank (RLMT) is 19.5 cm, and the height is 32.3 cm. A heavily insulated lid is used to avoid heat loss through the top of the furnace. The lid has two holes, as shown in Figure 2; one is used to immerse Si samples; another to insert a titanium tube to inject nitrogen gas into the melt. Si samples are immersed at position (A), shown in Figure 2, unless otherwise noted.



Figure 2 - Schematic of the revolving liquid metal tank showing the holes for gas tube and Si sample inserting

Si Cylinders

The Si additions were made of metallurgical grade Si, which is available in large pieces. From these large pieces, cylinders of 18.75 mm diameter were manufactured using a core drill. The length of the cylinders was approximately 10 cm, of which 8 cm was immersed into the liquid metal while the rest was used to hold the cylinder. Both ends of each cylinder were trimmed on a diamond wheel saw. Before each experiment, the cylinders were cleaned with acetone to remove any grease or dirt.

Al Bath

The Al was of commercial purity (99.87% Al, 0.04% Si and 0.09% Fe) and was received as ingots. The temperature of the Al bath is measured with a K-type thermocouple with the tip positioned at the center of the bath, about 5 cm below the free surface.

Experimental Procedure

The RLMT was filled with solid pieces of Al, and heated slowly to melt. When the melt temperature was stable, nitrogen gas was injected into the melt downwards by a 4.37 mm ID and 6.25 mm OD titanium tube. The tip of the gas tube was immersed 3 cm beneath the melt surface. Then, a Si cylinder of known weight was attached to the holder and immersed into the liquid using an immersion apparatus shown in Figure 3. After a specified time, the cylinder was withdrawn and detached from the holder. Any solidified Al was removed from the cylinder using 38 vol pct HCl at room temperature. This solution dissolved the Al while the Si remained intact [8]. Subsequently, the cylinder was weighted again.



Figure 3 – Immersion apparatus showing the holder with an attached Si sample

Figure 4 illustrates the Si samples following different immersion times. In this case, the fraction of each cylinder that was dissolved after being immersed for 2, 3, and 3.5 min was measured to be 0.29, 0.43 and 0.44, respectively.



 $\begin{array}{cc} 2 \text{ min} & 3 \text{ min} & 3.5 \text{ min} \\ \text{Figure 4 - Si samples after immersion for different times (gas flow rate = 3 lit/min, Al bath temperature = 738°C)} \end{array}$

EXPERIMENTAL RESULTS AND DISCUSSION

Using the experimental procedure described above, the dissolution of a Si addition into liquid Al was studied. The bath temperature was set at 738 °C, which corresponded to a superheat of 78°C.

The gas flow rate of nitrogen was set at 0, 2, 3, 4, 5 lit/min. These gas flow rates were chosen to create vigorous agitation within the melt, yet not so strong as to splash liquid metal on the furnace coils.

Effect of Gas Agitation

Figure 5 depicts the effect of gas agitation on the dissolution rate of a cylindrical Si addition at a superheat of 78°C. As shown, the gas agitation enhances the dissolution rate. For example, at an immersion time of 2 minutes, the dissolved fraction is approximately 5 times higher when the gas flow rate is 5 lit/min compared to the case of no gas agitation.



Figure 5 - The effect of gas agitation on the fraction of a cylindrical Si addition dissolved into Al at 78°C superheat

Effect of Nozzle Position Relative to the Si Sample

As shown in Figure 2, the relative position of the gas injection tube and the sample was changed to investigate its effect on the dissolution of samples. The dissolved fractions after 2 minutes of immersion are presented in Table 1.

Table 1 - The dissolved fraction of samples at different positions relative to the gas injection tube, after 2 minutes immersion time (T = 738°C and gas flow rate of 4 lit/min)

	D 1		
	Position A	Position B	Position C
Dissolved fraction	0.61	0.29	0.10

As can be seen, as the sample is moved further away from the gas injection, the dissolved fraction decreases significantly. This indicates that in order to accelerate the mass transfer rate, the nozzle must be placed close to the specimen. As the distance between the nozzle and the specimen increases, there is

limited fluid agitation in the vicinity of the dissolving sample, resulting in a reduction in the amount of Si dissolved.

Comparison of Mechanical Stirring and Gas Agitation

The enhancement of mass transfer rate due to gas injection was compared to the enhancement of forced convection induced by rotating the RLMT. The forced convection increases the mass transfer rate at the interface by decreasing the mass transfer boundary layer thickness. Figure 6 compares the effect of gas agitation to forced convection at a constant bath temperature.



Figure 6 - Comparison of the use of gas injection and forced convection on the dissolved fraction of Si samples at 78°C superheat

The experimental results show that both mechanical stirring and gas agitation increases the dissolution rate. Moreover, it is evident that blowing gas at the flow rate of 5 lit/min has a similar effect on the rate of dissolution as does the tangential velocity of 0.36 m/s under forced convection, at the point of immersion.

CONCLUSIONS

The dissolution of solid Si in molten Al with gas agitation was studied. The preliminary results presented in this paper allow us to conclude that:

- An increase in nitrogen gas flow rate increases the dissolution rate of the Si.
- The position of the Si sample relative to the gas nozzle strongly affects the dissolution rate of the sample. The larger the distance, the lower the dissolution rate.
- Injecting gas at 5 lit/min, has a similar impact in terms of increasing the dissolution rate as rotating the tank at 0.36 m/s.

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REFERENCES

1. M. Seyed Ahmadi, S. A. Argyropoulos, M. Bussmann and D. Doutre "Dissolution Studies of Si Metal in Liquid Al Under Different Forced Convection Conditions", Light Metals, Ed., S. J. Lindsay, 2011-809-14.

2. D. Doutre, private communication, Novelis, Kingston, 26 August 2010.

3. D. Mazumdar, J. W. Evans, Modeling of Steelmaking Processes, Boca Raton London, New York: CRC Press, Taylor & Francis Group, 2009.

4. P.G. Sismanis, S. A. Argyropoulos, "Convective Heat-transfer Measurements in Liquid Metals Under Different Fluid Flow Conditions", Metallurgical Transactions B, Vol. 19B, 1988, 859-70.

5. J. K. Kim, R. J. Fruehan, "Physical Modeling of Liquid/liquid Mass Transfer in Gas Stirred Ladles", Metallurgical Transactions B, Vol. 18B, 1987, 381-90.

6. D. Mazumdar, S.K. Kajani, A. Ghosh, "Mass Transfer Between Solid and Liquid in Vessels Agitated by Bubble Plume", Steel Research, Vol. 61(8), 1990, 339-46.

7. M. Iguchi, K. Tomida, K. Nakajima, Z. Morita, "Mass Transfer from a Solid Body Immersed in a Cylindrical Bath with Bottom Gas Injection", ISIJ Int., 1993, 728-34.

8. W. Zulehner, B. Neuer, G. Rau, Silicon, in Ullmann's Encyclopedia of Industrial Chemistry, Wiley, 2005.