

DISSOLUTION STUDIES OF Si METAL IN LIQUID Al UNDER DIFFERENT FORCED CONVECTION CONDITIONS

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Abstract

The dissolution of solid cylinders of Si in a revolving tank of molten Al was studied, under both natural and forced convection conditions. The experiments were carried out at bath superheats ranging from 21°C to 78°C. The dissolution of Si increased with increasing bath superheat, and was accelerated by the forced convection. The increase of the mass transfer coefficient with increasing tank rotation rate clearly shows that the rate controlling factor in the dissolution of Si into Al is mass transfer across the concentration boundary layer, for the range of rotation rates tested.

Introduction

The assimilation rates of additions affect the production cost of alloys. 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 focused specifically on the dissolution of solid Si into liquid Al. Al-Si alloys have attractive properties for the aerospace and automobile 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, which is then stirred for up to 30 minutes. During that time, dross forms on the top of the melt that is skimmed off; the longer the alloy making 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].

When an addition (initially at room temperature) is introduced to a molten bath, 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 as two steps [2]:

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 phase diagram, at the operating temperature, see Figure 1.

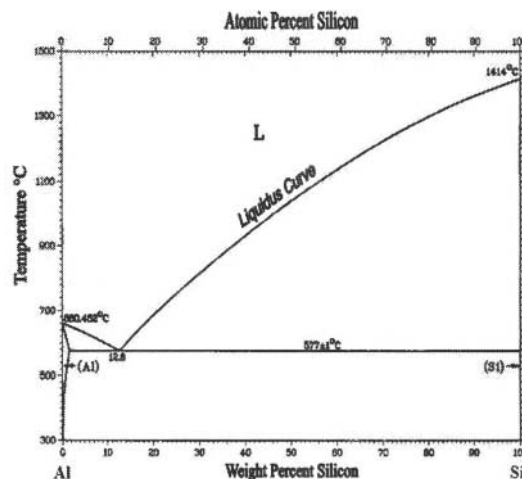


Figure 1. Al-Si phase diagram showing the liquidus curve [3].

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

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. The solute concentration profile next to a dissolving addition is shown in Figure 2.

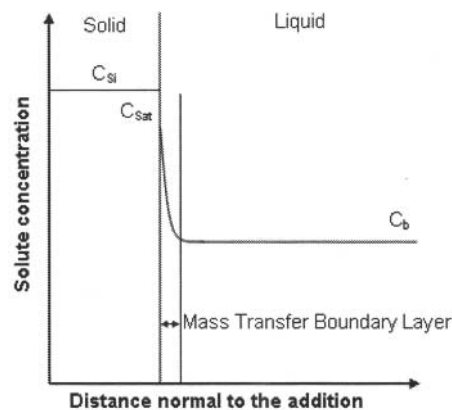


Figure 2. Solute concentration profile in the solid addition, at the solid/liquid interface, and in the liquid metal.

When the rate of dissolution is determined by the rate of mass transfer across the concentration boundary layer, the flow in the vicinity of the addition will have a significant influence through the mass transfer coefficient associated with that flow.

Examples of dissolution limited by the second step include the natural convection work of Gairrola et al.[4], who studied the dissolution of a vertical Ni cylinder in liquid Al, Shoji et al. [5], who studied the effect of natural convection on the dissolution of solid Cu in molten Sn-Pb alloys, and Niinomi et al. [6], who studied the rate of dissolution of ferrous alloys into molten Al assuming natural convection mass transfer. Their results showed that Fe-Cr, Fe-Cu and Fe-Ni dissolution is controlled by mass transfer within the boundary layer. On the other hand, the same authors showed that the first step, the interface reaction, to be rate limiting for the Fe-C alloy and that both steps (the so-called "mixed effect") affects the dissolution rate for Fe-Si, commercially pure iron, and Fe-Mn.

Turning to forced convection, Shoji et al. [7] studied the dissolution of a Cu cylinder in molten tin-lead alloys. The velocity ranged from 0 to 0.754 m/s; the temperature was fixed at 673K. It was shown that the dissolution rate increases under forced convection.

To be best of the authors' knowledge, the effect of flow conditions on the dissolution rate of Si into Al has not been studied. The only study related to the Al alloying process using Si is that of Balusis et al. [8], who showed that the use of Si granules over Si chunks decreases the dissolution time of Si. It was found that when the diameter ratio of lumpy to granular is about 5, the required time for dissolution increases by a factor of between 2 and 3.

The current study seeks to accelerate the dissolution of Si into liquid Al, thus decreasing the cost of producing Al alloys with high Si levels, by quantifying the impact of agitation of the liquid Al using mechanical stirring (i.e. forced convection). To address this objective experiments have been conducted for different velocities of the liquid Al. This work provides a quantitative comparison of the dissolution rate under natural and forced convection conditions.

Experimental Setup

Electrical Furnace and Revolving Liquid Metal Tank:

An electric resistance furnace was used to melt the Al. The furnace was constructed of refractory brick within a steel enclosure; the central heating area for the crucible totaled 0.018 m². Two K-type thermocouples located in the furnace hot zone were used to control the temperature.

The cylindrical stainless steel tank that rotates within the furnace is shown in Figure 3. The Revolving Liquid Metal Tank (RLMT) is connected to a DC motor that controls the speed to within 1 RPM. The interior diameter of the RLMT is 43.2 cm, the height is 19.1 cm, and it has a capacity of approximately 50 kg (20 l) of Al. 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 3; one (diameter of 7.9 cm) is used to insert a thermocouple into the center of the Al bath; another (diameter of 6.3 cm) is used to immerse Si samples. The center of this second hole is located 16.7 cm from the center of the RLMT.

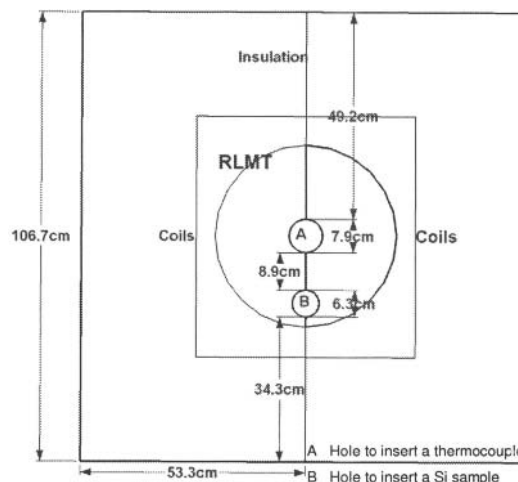


Figure 3. Schematic of the electrical resistance furnace including the revolving liquid metal tank showing the holes with corresponding dimensions

Si Cylinders

The Si additions were made of metallurgical grade Si; the nominal composition is presented in Table 1.

Table 1. Nominal composition of the metallurgical grade Si

Si (%)	Fe (%)	Al (%)	Ca (%)	Ti (%)
>98.0	<1.0	<0.25	<0.1	<0.1

The Si we use arrives as large pieces, from which cylinders of 18.75 mm diameter were cut. The length of the cylinders is approximately 10 cm, of which 8 cm is immersed into the liquid metal while the rest is used to hold the cylinder. Both ends of each cylinder are trimmed on a diamond wheel saw. Before each experiment, the cylinders are cleaned with acetone to remove any grease or dirt.

Al Bath

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

Experimental Procedure

To initiate an experiment, the electrical resistance furnace is heated from room temperature; the RLMT contains approximately 46 kg of Al. It takes about 12 hours to melt the Al. When the melt temperature is stable, the target rotating speed is set. Then, a Si cylinder of known weight is attached to the holder and immersed into the liquid. The immersion speed is approximately 5 cm/s. After a specified time, the cylinder is withdrawn and detached from the holder. Any attached Al is removed using 38 vol pct HCl at room temperature. This solution dissolves the Al while the Si remains intact [9]. Then, the cylinder is weighed again. Figure 4 (a) and (b) show the Si samples after immersion for different times, with and without the attached Al.

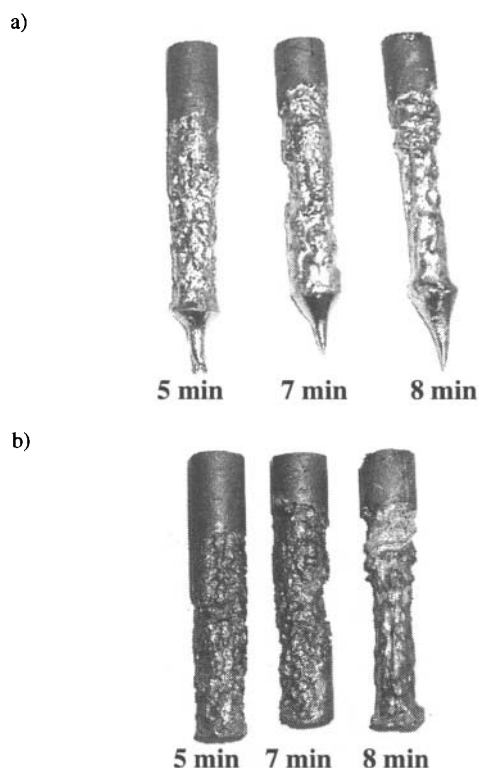


Figure 4. Si samples after immersion for different times, with and without attached Al, under natural convection condition.

Experimental Results and Discussions

Using the experimental procedure described above, the dissolution of a Si addition into liquid Al was studied. The bath temperature was set at 681, 703, 715 and 738 °C for different experimental runs. These temperatures correspond to superheats of 21, 43, 55 and 78°C, respectively.

The RLMT was rotated at $\omega = 0, 10$ and 20 rpm for each melt temperature. The tangential velocity of liquid metal at the center of the Si cylinder was calculated using the following equation,

$$u_{\theta} = \frac{2\pi}{60} \times r_{\text{rot}} \times \omega \quad (1)$$

where $r_{\text{rot}} = 16.7$ cm is the distance from the center of the tank to the point of immersion,. These rotational speeds correspond to tangential velocities of 0, 0.18 and 0.36 m/s, respectively.

Reproducibility of the Experimental Results

We begin by assessing the variability of the results. An experiment was run to measure the dissolved fraction (the ratio of dissolved mass to the initial immersed mass of the cylinder) of cylinders at the same superheat (78 °C), rotational speed (10 rpm) and immersion time (2 min.). The test was performed for 9 cylinders and the results are presented in Table 2. The average dissolved fraction and the standard deviation are 0.39 and 0.023, respectively.

Table 2. Variation in the dissolution of identical samples under the same experimental conditions (T = 738°C and rotational speed = 10 rpm)

0.39	0.43	0.37	0.45	0.39	0.47	0.29	0.43	0.31
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The uncertainty associated with the ratio of the dissolved mass to initial mass can be calculated as

$$U_{m_d/m_i} = \left[\left(\frac{U_{m_d}}{m_i} \right)^2 + \left(\frac{-m_d}{m_i^2} U_{m_i} \right)^2 \right]^{1/2} \quad (2)$$

where m_i (kg) is the initial mass of the addition and m_d is the dissolved mass. The value of U_{m_d/m_i} is 0.074 based on these experimental results.

The source of scatter in the data can be largely attributed to the surface of the samples. The Si samples have different macroscopic structure depending on the chunk of Si from which they were cored. Some of the samples have very small pores, some larger and some have directional scratches, related to the method of casting of the Si. For example, the pores are generated due to trapped gas bubbles during the solidification of Si. Nevertheless, these variations reflect the reality of the dissolution process. Put another way, industry uses Si in the same form and so will experience similar variability in the dissolution of Si. It should be noted that the same uncertainty is assumed for all of the data presented here, because it is not possible to repeat a test for each data point several times given the difficulty of the experiments and the required resources. Therefore, the results presented from now on all correspond to a single measurement.

Effect of Bath Superheat on the Dissolution Rate

The first objective of this work is to study the effect of bath superheat on the dissolution rate of a Si addition in Al.

As shown in Figure 5, the superheat of the bath strongly affects the rate at which the sample dissolves. The higher superheat increases the saturation concentration of Si in liquid Al (according to the liquidus curve on the Al-Si phase diagram), which in turn provides a greater driving force for mass transfer.

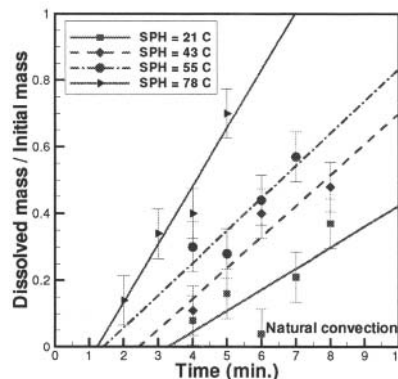


Figure 5. The effect of superheat on the dissolution rate of a cylindrical Si addition in Al, under natural convection conditions.

In this study a range of bath superheats (from 21 to 78°C) was investigated. At the same immersion period ($t = 5$ min.), Figure 5

shows a 4.5 times increase in the dissolution rate when the degree of superheat increases from 21 to 78°C.

Extrapolating the experimental data (linear regression of the experimental data points) also reveals that the higher superheat causes the dissolution to begin sooner. As shown in Figure 5, when the superheat is just 21°C, it takes approximately 3 minutes for the sample to start to dissolve in the bath, while this time decreases to about 1 minute at 78°C superheat. The reason could be the higher heating rate of the sample (from room temperature upon immersion to the bath temperature at higher superheat), to reach equilibrium with the bath, and retaining the saturation concentration at that temperature at the solid/liquid interface.

The superheat of the bath has the same effect on the dissolution rate when the RLMT rotates (i.e. forced convection). As shown in Figure 6, for both rotation rates, it takes less time for an addition to dissolve at higher superheats. An increase of superheat from 43 to 55°C yields a 4.5 times faster dissolution of the addition after 3 minutes of immersion at 10 rpm. The same increase in the superheat increases the dissolved mass by a factor of 2.3 at 20 rpm after immersion for 3 minutes.

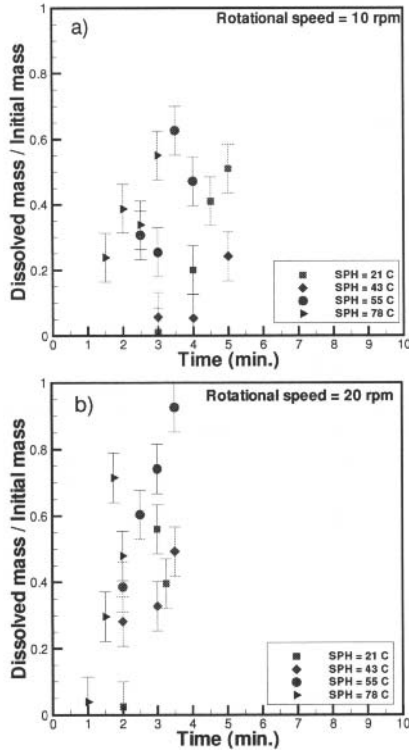


Figure 6. The effect of superheat on the dissolution rate of a cylindrical Si addition into Al under forced convection conditions, when rotating at a) 10 and b) 20 rpm.

Effect of Rotating Bath

The rotation of the RLMT introduces forced convection near the immersed sample. It is known that forced convection increases the mass transfer rate at the interface by decreasing the mass transfer boundary layer thickness. As a result, higher dissolution rates are expected when the bath rotates (at a constant liquid Al temperature).

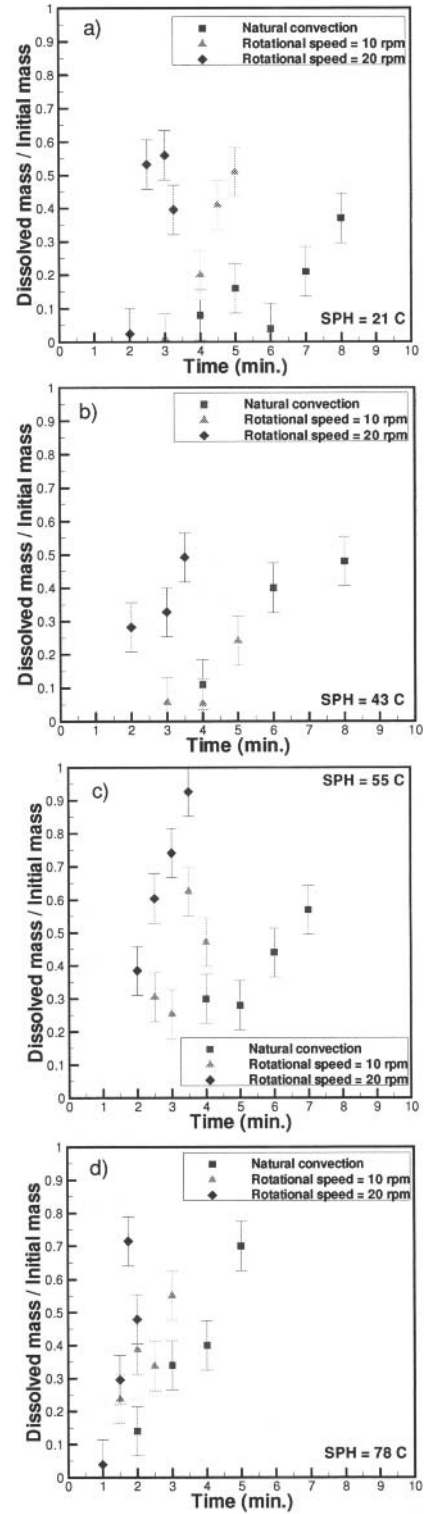


Figure 7. The effect of forced convection on the dissolution rate of a cylindrical Si addition into Al at a) 21, b) 43, c) 55 and d) 78 °C superheat.

Figure 7 depicts the effect of forced convection on the dissolution rate of a cylindrical addition at different superheat values. As shown, when the bath rotates the addition dissolves more quickly than when the bath is still. This clearly indicates that the mass transfer across the boundary layer is the rate controlling factor in the dissolution of Si into Al, for these rotational speeds.

Experimental Mass Transfer Coefficient

The dissolution of Si into Al is a mass transfer process, that is usually quantified by a mass transfer coefficient that is a function of operating conditions. Invoking some simplifying assumptions the mass transfer coefficient for the dissolution of Si in Al is calculated using the experimental data presented here.

An average mass transfer coefficient k_m (m/s) can be calculated by the following equation

$$k_m = \frac{m_{ave}^*}{C_{Sat} - C_b} \quad (3)$$

where C_{Sat} (kg/m³) is the saturation concentration of the addition in the liquid at temperature T (°C), and C_b (kg/m³) is the bulk concentration.

The average mass flux m_{ave}^* (kg/s/m²) from the solid addition to the liquid from $t = 0$ (the moment the cylinder is fully immersed into the bath) to time t (when the cylinder is removed from the bath) can be calculated as

$$m_{ave}^* = \frac{m_d}{tA_{ave}} \quad (4)$$

In the above equation m_d (kg) is the dissolved mass of the Si sample and A_{ave} (m²) is the average solid/liquid interface area, expressed as

$$A_{ave} = \frac{A_i + A_f}{2} \quad (5)$$

where A_i (m²) is the initial surface area of the immersed part, and A_f (m²) is the final surface area. A_f can be calculated assuming the following:

- The density of the Si cylinder is constant (i.e. the variability in the density due to different pore distributions can be neglected).
- The bottom surface area is small relative to the side area. Based on the cylinder dimensions in our current experiments, the initial ratio is 1.86% which justifies this assumption.

Therefore,

$$A_f = A_i \times \frac{d_i}{d_f} \times \frac{m_f}{m_i} \quad (6)$$

where d_f is the average value calculated from 5 measurements of the diameter of the cylinder. As shown in Figure 4, the dissolution of Si is not uniform; that is, the diameter of the cylinder varies along its length. Therefore, an average diameter was used to estimate the surface area of a cylinder after removing it from the bath.

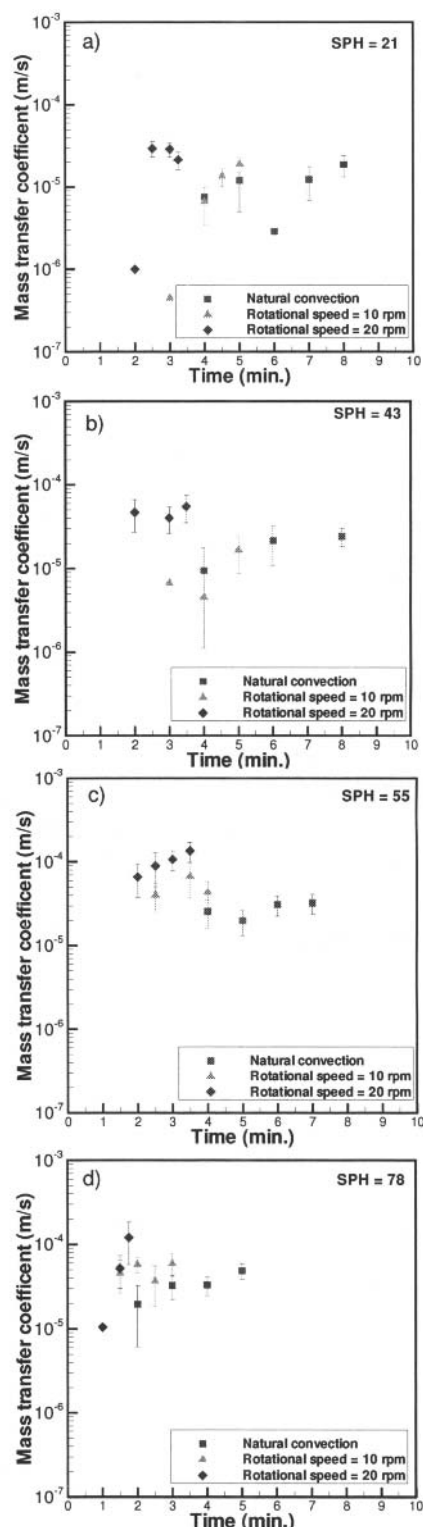


Figure 8. Estimated mass transfer coefficients for different flow conditions, using experimental data at a) 21, b) 43, c) 55 and d) 78°C superheat.

Figure 8 shows the calculated average mass transfer coefficients at different bath superheats. The mass transfer coefficient ranges 2.9×10^{-6} to 4.8×10^{-5} m/s for natural convection, 1.3×10^{-6} to 9.4×10^{-5} m/s at 10 rpm, and 1.78×10^{-6} to 1.4×10^{-4} m/s at 20 rpm, depending on the bath superheat. It should be noted that the error bars shown in the graphs are associated with the calculated mass transfer coefficient using the following equation,

$$U_{k_m} = \left[\left(\frac{1}{t A_{ave} \Delta C} U_{m_d} \right)^2 + \left(\frac{-m_d}{t^2 A_{ave} \Delta C} U_t \right)^2 + \left(\frac{-m_d}{t A_{ave} \Delta C} U_{A_{ave}} \right)^2 + \left(\frac{-m_d}{t A_{ave} \Delta C^2} U_{\Delta C} \right)^2 \right]^{1/2} \quad (7)$$

For the data points without error bars the calculated uncertainty was higher than the value itself, because the amount of dissolved mass was less than the uncertainty associated with the immersed mass of the cylinder.

As can be seen, the mass transfer coefficient increases as the rotational speed increases. This clearly shows that the forced convection causes a faster dissolution of the specimen within the bath.

Finally, the mass transfer coefficient can be defined as

$$k_m \equiv \frac{D_{Si/Al}}{\delta_m} \quad (8)$$

where $D_{Si/Al}$ is the binary diffusion coefficient of Si into liquid Al and δ_m (m) is the mass boundary layer thickness. This equation in conjunction with the obtained experimental results indicates that the boundary layer thickness decreases when the fluid is agitated in the vicinity of the sample. A decrease in the mass transfer boundary layer thickness implies a lower resistance to transport of species from the solid/liquid interface to the bulk of the liquid, and therefore the dissolution rate increases.

Conclusions

The dissolution of solid Si in molten Al was studied under natural and forced convection conditions at different superheat values.

- An increase in bath superheat causes faster initiation of the dissolution process, as well as faster dissolution of the Si.
- Forced convection causes faster dissolution of Si within the Al, which indicates that mass transfer across the boundary layer controls the dissolution process, for tangential velocities from 0 (stationary tank) to 0.36 m/s (the highest tangential velocity tested).
- The maximum mass transfer coefficient associated with 78°C superheat increased from 4.8×10^{-5} to 1.4×10^{-4} m/s as the tangential velocity increased from 0 to 0.36 m/s. This clearly indicates that in order to dissolve silicon faster some type of agitation must be induced within the bath.

Acknowledgments

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