

Gas Fluxing of Molten Aluminum: An Overview

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Abstract

The aluminum industry is under continual pressure to improve metal quality, while at the same time reduce costs. Although a reasonably mature technology, there has been a continual evolution in degassing equipment over the years. A detailed review and theoretical analysis is given of the chemical and kinetic factors which control the metal quality after gas fluxing, and the evolution of degassing technology in Alcoa is summarized. Particular emphasis is placed on hydrogen removal, minimization of chlorine use, reduced operating costs and minimization of environmental emissions. Considerations related to inclusion removal are also discussed briefly.

Introduction

Aluminum is an extremely reactive metal. When it comes in contact with moist air the water vapor decomposes and releases hydrogen into the melt. It has been well documented that excessive quantities of dissolved gas have a detrimental effect on the mechanical properties of aluminum castings [1, 2]. In order to produce good quality castings, one must degas before casting.

The scientific basis for effective gas removal has been well established [3-5]. Any degassing process is subject to two fundamental laws. The first is the thermodynamic relationship between gas dissolved in metal and the pressure of hydrogen in purge gas. The second concerns the rate of hydrogen diffusion into ascending gas bubbles. Both are considered briefly, so that important practical considerations may be understood.

Experiments have shown that only hydrogen gas dissolves in molten aluminum. The gas solubility has been measured carefully in a number of studies. These results have been reviewed and summarized in [5]. The solubility follows Sievert's law. That is,

$$1/2 \text{ H}_2 (\text{v}) = H (\text{in Al}) \quad (1)$$

The diatomic hydrogen molecule decomposes to form two atoms, which dissolve in molten metal as a monatomic species. From the law of mass action the pressure of gas in equilibrium with a melt follows the relationship:

$$\sqrt{P} = \frac{H}{S \cdot C} \quad (2)$$

where P is the pressure of hydrogen vapor in atmospheres and H is the amount of gas dissolved in the melt, usually given in standard cc/100 g. The solubility, S , is the gas content in pure aluminum at a hydrogen gas pressure of one atmosphere. It is given by the equation [5]:

$$\log S = -2692/T + 2.726 \quad (3)$$

where T is the thermodynamic temperature (in degrees Kelvin). At 1000 K (727 C) the hydrogen solubility is 1.081 cc/100g.

An alloy correction factor, C , is employed to account for the change in gas solubility in any particular alloy, relative to pure aluminum. Equations are given in [5] which may be used to calculate correction factors for any alloy composition. Table 1 lists values for some common alloys.

Table 1. Correction Factors for Aluminum Alloys

Alloy	1100	1200	1350	2011	2024	2218
C	0.96	0.96	0.98	0.64	0.82	0.71
Alloy	3003	3004	4032	5050	5052	5154
C	0.78	0.92	0.63	1.15	1.40	1.65
Alloy	6061	6063	7001	7075	7150	8006
C	1.05	1.06	0.99	1.00	0.94	0.71

It should be noted that the solubility in equation (3) is an exponential function of temperature. The gas content (S) doubles for each 110C (200F) increase in temperature. This is why gas problems become much worse at higher furnace temperatures.

Another important conclusion may be drawn from equation (2). Our ability to degas can be no better than thermodynamic laws permit. We admit purge gas bubbles into the melt. They collect hydrogen as they float towards the surface. The best possible situation is when bubbles are saturated with hydrogen gas as they leave the surface of the melt. In this case, the degassing process efficiency is 100% from the thermodynamic point of view.

However, as the gas content drops so does the pressure of hydrogen in the bubbles. This means that more purge gas is required to remove a specific volume of hydrogen from the metal. This is best seen by defining a gas removal ratio, R , which is the volume of inert purge gas needed to remove one liter of hydrogen from the metal. At one atmosphere pressure the removal ratio is given by the relation:

$$R = \frac{1-P}{P} = \left(\frac{S \cdot C}{H} \right)^2 - 1 \quad (4)$$

The gas removal ratio, R , is shown in Figure 1 for pure aluminum at 750 C. It can be seen that as the gas content drops below 0.1 cc/100g, the ratio R becomes more than 200. This limits our ability to degas to extremely low levels.

As noted above, the solubility increases exponentially with temperature, but $(S)^2$ appears in equation 4. In other words, a temperature increase of 110 C doubles the solubility, S , but the gas removal ratio is increased by four times. Thus, keeping metal temperature low is even more important for degassing efficiency.

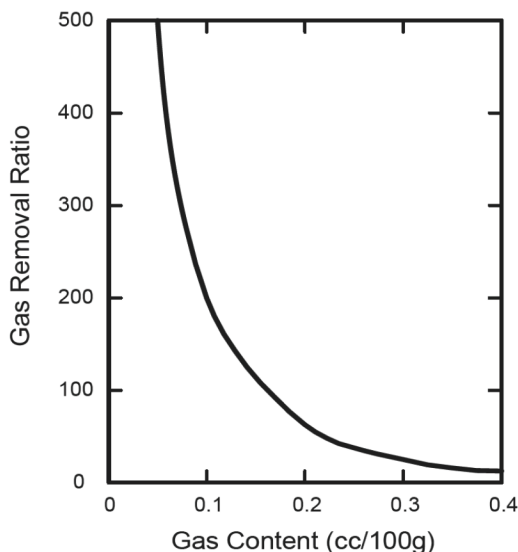


Figure 1. Gas Removal Ratio in Aluminum at 750 C

We now consider the kinetics of hydrogen removal. This subject was studied in detail by Sigworth and Engh [3]. They considered bubbles floating up towards the surface and developed equations for gas removal. A 'dimensionless hydrogen concentration' was used to characterize the approach to thermodynamic equilibrium. The practical implications of their result are best seen by an example calculation given in [4], where the degassing efficiency in a 250 kg (550 lb.) crucible furnace is shown as a function of bubble diameter. For this case the bubbles ascend through approximately 0.7 meter of metal. The results are reproduced in Figure 2. Note that a high degassing efficiency (> 80 %) occurs when the bubble size is less than about 5 millimeters. When one halves the bubble size, the surface area of bubbles increases by a factor of four. This increases the rate of hydrogen delivery to the bubble, since it is proportional to surface area. But the rise velocity of the bubble also decreases. Smaller bubbles take longer to float out, so they have more time to reach thermodynamic equilibrium. The two effects are additive, and so the process efficiency increases sharply as bubbles become smaller.

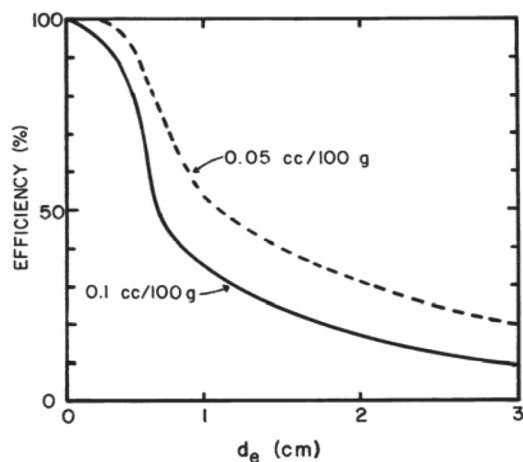


Figure 2. Thermodynamic Degassing Efficiency vs. Bubble Size (Results are for two different gas contents)

This fact should be communicated to your casthouse personnel and procedures should be established to inspect the bubble patterns in a degassing box every time it is skimmed to remove accumulated dross. If large bubbles are observed at the surface, actions need to be taken to restore degassing efficiency. (Checking the inert gas flow rates, replacing worn impeller heads, etc.)

The importance of having small bubbles cannot be overemphasized, since it has been the essential impetus behind the commercial development of improved degassing processes during the last fifty years. If one places a lance into a melt, the bubbles produced are several centimeters in diameter and give a thermodynamic efficiency of only 10 or 20%. The use of a porous plug improves the situation greatly compared to a simple lance, but even in this case the efficiency is typically only about 40 or 50%. The next significant improvement came forty years ago, when Szekely used mechanical stirring in the SNIF (Spinning Nozzle Inert Flotation) process [6]. Not only were fine bubbles produced by this means, but they were dispersed throughout the melt, instead of rising in a plume (as with a porous plug). The original SNIF process worked extremely well, but the impeller was complex and costly, being a modified flotation cell machined out of graphite and immersed in liquid metal. In subsequent years, many companies (including Alcoa) have developed more simple means of producing the desired fine bubbles.

From Figure 2 we understand that any process which produces small bubbles will yield a high degassing efficiency. From this point of view, nearly all equipment using rotating impeller heads should give equivalent results. After all, once a process efficiency close to 100% is attained, it is not possible to make further improvements. This simple conclusion is borne out in detailed simulation studies which compared different head designs [7]. Consequently, the casthouse manager must make his decision primarily on the reliability of equipment and cost of operation. Ease of maintenance and the lifetime of consumable lances and impellers are particularly important.

Another practical implication also becomes obvious. Most industrial degassing processes produce bubbles that are on the order of 5 mm in diameter or less. This means that, in most cases, one is operating at a degassing efficiency reasonably close to thermodynamic equilibrium [5]. This simplifies greatly the equations that describe gas removal.

For a batch degassing process, it can be shown that the hydrogen content in the melt as a function of time is given by the relation:

$$\frac{1}{H} - \frac{1}{H_o} = \frac{\dot{V}}{10M(CS)^2} t \tag{5}$$

where the inert gas flow (\dot{V}) is given in cc/sec, the mass of metal in the crucible or furnace (M) is given in kg, the solubility (S) is given in cc/100g, and the time (t) is given in seconds. The instantaneous hydrogen concentration (H) and the starting original hydrogen content (H_o) are also given in cc/100g.

It is also common to have metal flowing through an in-line treatment box. Because of the square root in equation (2), a quadratic equation is used to describe hydrogen removal for this case. The gas content of the metal as it exits the treatment box is given by:

$$H_e = -\beta/2 + 1/2\sqrt{\beta^2 + 4\beta H_i} \tag{6}$$

where

$$\beta = \frac{\dot{M}P_{inert}C^2S^2}{100 \cdot \dot{G}} \tag{7}$$

and where

- \dot{G} is the flow rate of inert gas (l/min)
- \dot{M} is the mass flow of metal through the box (kg/min)
- P_{inert} is the pressure of inert gas (nominally 1 atm.), and
- H_i is the gas content of metal going into the box

The term β has the same units as the gas content (cc/100g) and describes the ratio of the two flows in the process; metal and gas; and their relative capacity to carry the hydrogen in or out of the system.

Equation (6) was used to calculate the percent hydrogen removed by the in-line degassing treatment, for five values of β . The results are plotted in Figure 3.

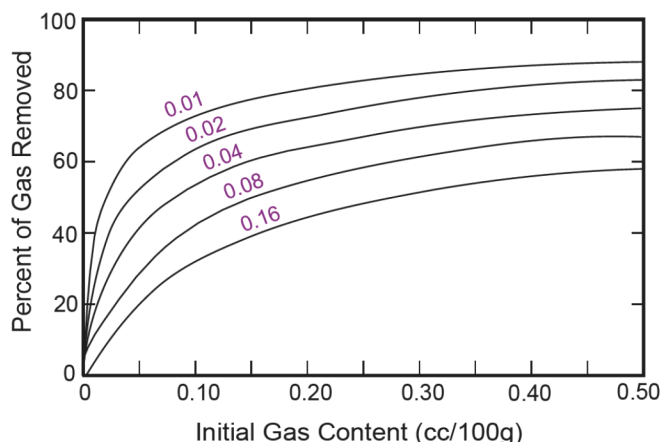


Figure 3. Hydrogen Removed During In-Line Degassing

Discussion

There are a number of interesting and useful observations that can be made from the preceding analysis.

Equations 5-7 all contain the term $(CS)^2$. The gas solubility, S , is an exponential function of temperature. This means that S^2 will double with a temperature increase of 60 C. In the case of a batch degassing process, this means it will take twice as long to reach the desired hydrogen level.

The effect of higher temperatures on the results obtained during in-line treatments are equally dramatic. This is best seen from the curves in Figure 3. A 60 C increase in temperature changes β by a factor of two. A significant drop in degassing efficiency is the result.

For some reason, it has not been widely recognized that in-line degassing results depend on the original gas level. There is only one reference to this effect in the literature. (See Figure 1 of reference [8].) This is surprising, since the limitations on the degassing process arise largely from thermodynamic equilibrium when small bubbles are present.

The equations given above can also be used to predict the performance of multi stage in-line degassing systems. In this case, the calculated exit gas concentration in the metal from the first stage becomes the gas concentration entering the second stage; and so on, if there are more than two stages. The results of these calculations for a two stage system are shown below in Figure 4. The corresponding curves for a single stage unit, having the same total gas flow (and the same value of β) as a two stage unit are also plotted for comparison. As noted before [8], the effect of staging a unit is not significant.

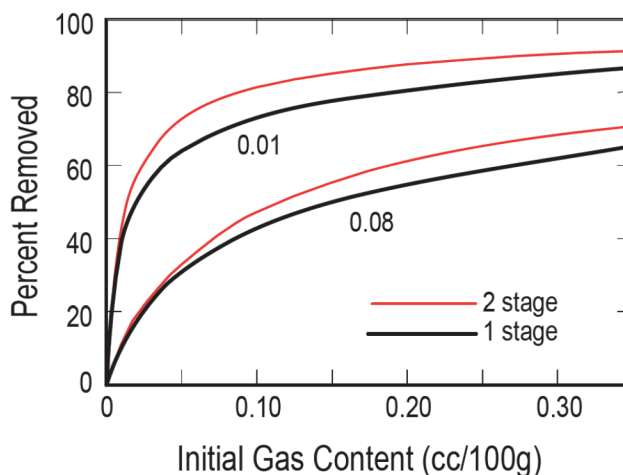


Figure 4. Degassing Efficiency of Two Stages Compared to a Single Stage

Of course, one reason to add an additional stage is to be able to use more gas in the fluxing process. Any rotor head will be able to deliver only a certain amount of fluxing gas. If you are operating at this maximum with a single stage unit, and need additional degassing capacity, one possible solution is to add a second stage. This will allow you to use more gas, and to move to smaller values of β in Figures 3-4. Thus, the degassing performance (percentage of gas removed) will increase.

There is another possibility, of course. It is also possible to modify the rotor design, so that it will deliver larger quantities of gas. This solution is a great deal more convenient, and less costly, than adding a second stage. For this reason, a great deal of research and development has gone into improving rotor head design.

It is beyond the scope of this paper to offer a history of rotor head design, or to present a detailed comparison of the different heads which have been used. Nor are we prepared to explain the hydrodynamic theory related to the subject. For the reader who wishes to pursue this area of technology in more depth, there are several excellent publications which present a great deal of information [7, 9-11]. It will, however, be possible to offer some important general observations here.

Other things being equal, the degassing performance of an in-line rotor head improves when the stirring energy is increased. The effect of stirring on bubble size and bubble distribution has been studied in great detail. The most appealing explanation, because of its simplicity, is a theory offered by our Norwegian colleagues

[9]. They examined the forces acting on bubbles in a stirred melt. The surface tension force on the bubble holds it together. Viscous forces acting on the bubble, because of turbulent shear produced by stirring, tend to break it up. Calculating and balancing the two types of forces, they showed that the maximum bubble diameter that can exist in a stirred melt is:

$$d = \left(\frac{\sigma}{\rho} \right)^{0.6} \left(\frac{M}{E} \right)^{0.4} \quad (8)$$

where

σ is the surface tension of the metal

ρ is the density of molten aluminum

M is the mass of metal in the chamber, and

E is the energy of stirring (watts/m³)

This simple relation was sufficient to allow them to calculate the bubble sizes observed in a large number of water model experiments with the HYCAST rotor. It also allowed them to deduce that the bubble size in liquid metal will be just over twice that observed in water model studies.

Of course, as the stirring intensity increases, the designer has to deal somehow with the increased amount of rotational energy placed into the metal. Otherwise a great deal of splashing, vortexing, or an gas/metal emulsion are produced. Various design solutions have been placed into service, either singly or in combination:

- using baffle plates
- using rectangular boxes
- placing the rotor off center in the box
- alternating rotor direction in multi-stage units
- using 'pumping' heads, to control and direct metal flow
- using rotor designs, having a high localized shear where gas enters the melt
- reversing rotation of the rotor assembly

In the last method the rotation of the rotor shaft is reversed periodically at 5-10 second intervals. In this way the rotational momentum introduced into the melt during one cycle is canceled by the following reverse rotation.

Recent Developments at Alcoa

Alcoa has several types of traditional degassing systems operating in our casthouses: SNIF, Alpur, and Alcoa's own 622 process. As noted above, the technology is relatively mature at this point, so further process development does not appear to be promising. Instead, we have focused on improving equipment reliability and reducing operating costs. For several years we have focused on developing a simplified and standardized version of the 622 process, which can be supplied in off-the-shelf designs. The units also employ several improvements:

- higher power electric motors
- a 'quick-disconnect' design to change carbon shafts, and
- a new design of rotary gas union and mounting shaft, which allow the 'straight-through' passage of flux gas

The last feature is particularly important as we consider the possible replacement of chlorine gas by salt flux injection.

Continual improvements have also been made in improving the operating life of carbon consumables. Rotor shafts and heads are made of high quality, oxidation-resistant grades of carbon, and the shafts are coated with a thin refractory sheath to reduce wear and oxidation at the melt line. We have also taken steps to seal our

degassing boxes, so that air cannot enter the box. This was accomplished by using a 'submerged' design for the metal entrance and exit ports, and by placing a refractory seal between the box and the top (which contains the degassing motors and shafts). This improvement ensures that the box is filled with the inert gas used for degassing (argon), and reduces oxidation inside the box.

The use of 'inert' degassing boxes has several operating advantages. The most significant is perhaps the reduction in particulate matter (PM) and chloride emissions. A previous study [12] has shown that chloride emissions from an inert degassing box is reduced to less than one-half found in an 'open' box. Particulate emissions are reduced by almost a factor of ten. Use of inert degassers has allowed Alcoa to meet SMACT particulate emission standards in the U.S. where chlorine fluxing is required. The inert degassers also produce less dross--10% to 20% of the skim generated in an 'open' degasser. The reduction of oxygen content inside the box also means that graphite life is longer in the inert boxes, as there is less carbon burning, and less oxide skim in the box to erode the shaft.

Alcoa also has installed the Alcan Compact Degasser (ACD) at several locations. The primary advantage of this system is that there is no large box to drain when an alloy change is required. It is also attractive in crowded casthouses, when floor space is at a premium. Since its introduction about ten years ago [13, 14] it has found widespread use. Currently more than 125 units are operating in 20 different countries, and an inerted version of the process has been developed [15].

The early information on the ACD suggested that its performance was nearly equivalent to the large in-line boxes it was designed to replace. At first glance this is surprising. With the ACD the metal residence time can be measured in seconds, not in minutes. One way to look at the degassing process is to compare the metal residence time in the reactor to the mixing time. This ratio is a dimensionless Stanton number, which can be used to describe the minimum level of circulation required in the melt for effective mass transport. However, a recent analysis [16] suggests that the shorter residence time in the ACD should not limit its performance.

The same may not be said, however, for the bubble residence time. Because the metal height in the ACD is less, the bubble will be in contact with liquid metal a shorter time. Consequently, one would expect to find a lesser thermodynamic efficiency for gas removal with the ACD. It will be useful, therefore, to look at some production data comparing the Alcoa 622 and ACD processes.

Table 2 shows the degassing performance of the Alcoa 622 and the ACD in-line systems when casting the same alloy—a high Mg variant of AA5182. The correction factor for this alloy is 2.0. The metal temperature was 720 C. The metal level in the ACD during that cast was 0.22 meters. Other details regarding the process conditions are given in Table 3. It can be seen that the metal and inert gas flow rates are nearly the same, and the degassing process parameter (β , equation 7) differs by only 5%. So, this represents a good comparison for the relative thermodynamic efficiency of the two systems.

Table 2. Comparison of Degassing Performance

Degasser	Hydrogen In	Hydrogen Out	Gas Removed
A622	0.432	0.155	64.1 %
A622	0.473	0.156	67.0 %
A622	0.545	0.162	70.0 %
ACD	0.503	0.230	54.3 %

Table 3. Degassing Process Conditions

	A622	ACD
metal flow	710 kg/min	650 kg/min
total gas flow	280 l/min	270 l/min
gas flow/head	70 l/min	45 l/min
chlorine flow	3 l/min	0.5 l/min
β for degasser	0.109	0.103

If one uses equation (6) or Figure 3 for the entire degassing system (and ignores the effect of the stages) one finds that the theoretical hydrogen removal efficiency for the A622 and ACD are 63% and 63.7% respectively. The A622 results are somewhat better than predicted by this calculation, which may be expected considering that four stages are employed in the degasser. With the ACD unit, however, the actual hydrogen removal is 54.3%, not the theoretical 63.7%. The lower efficiency could be expected, since the metal height (and time the bubble is in contact with the metal) is less.

It should be noted that this example was chosen to provide a direct comparison of the thermodynamic efficiency of the ACD versus more traditional degassing processes. For this reason the total inert gas flow (and β) in both systems was the same. In commercial practice the slightly lower efficiency of the ACD is easily compensated for by using more inert gas (and/or additional stages).

One should also keep in mind that the above comparison represents an extreme case. The correction factor for this alloy was equal to 2, which means it is four times harder to degas than pure aluminum. The gas content entering the degasser was also high. In 'easier' alloys, and in heats having less gas, the performance of the ACD will be closer to traditional degassing systems.

Inclusion Removal

In the above we considered only degassing performance, but a degassing station can also remove significant amounts of inclusions suspended in the metal [8, 17]. A theoretical analysis of the processes involved suggests that flotation of inclusions is most important [18]. The most compelling experimental fact in favor of this interpretation is that inclusion removal is sensitive to the composition of the purge gas [18-19]. Additions of small amounts of chlorine improve inclusion removal. This is especially true for oxide films suspended in the melt.

Inclusion measurements via LIMCA are complicated by the fact that there are high concentrations of micro-bubbles after the degassing station. One has to look carefully to see them. They come to the surface in the launder, as far as 10 or more meters

from the degasser, as little 'pin pricks' on the surface. It is necessary to scrape away the dross, to get a fresh metal surface, to see them. Because one cannot distinguish between these small bubbles and other discontinuities in the metal with LIMCA, the PODFA test is a more reliable measurement technique. Table 4 shows the inclusions concentrations before and after the A622 degasser found in a heat of 8111 alloy. These results were found using a small amount of chlorine gas injected into a single stage A622 degasser. The inclusion concentrations are given in mm^2/kg , except for the oxides. From these results we find that about 90% of the inclusions were removed by the degasser.

Table 4. Inclusion Removal in A622 Degasser

Sample Location	Taphole	After A622
Total Inclusions	0.132	0.008
TiB ₂	0.087	0.003
Al ₄ C ₃ (< 3 μm)	0.036	0.001
Al ₄ C ₃ (> 3 μm)	0.007	0.004
MgO	0.003	---
MgAl ₂ O ₄ (spinel)	---	---
Potential Chlorides	trace	trace
Oxide Films (#/kg)	3	0

We have made PODFA tests comparing inclusion removal in the A622 and ACD degassers. Any difference is not statistically significant, when compared to the relatively large sampling and measurement errors inherent in PODFA tests. However, at one of our plants we have had both an A622 degasser and an ACD operating in front of deep bed filters. These filters are used to produce critical products and remove nearly all inclusions present in the metal. In this plant the life of the deep bed filter with ACD is half that found when using the A622 degasser. This means the inclusion loading with ACD is twice that with the A622. Aside from filter life, this difference is not as great as it first appears: If the A622 removes 90% of the inclusions, then the ACD removes 80% on average. This is still an acceptable performance for most applications. In addition, it is possible to employ practices to compensate for this difference. For example, one may use a settling time in the furnace before casting to remove inclusions.

Environmental Considerations

The alkali metals Na, and to a lesser extent Li and Ca, are found as undesirable impurities in aluminum. These must be removed to low levels for acceptable product quality in most alloys. The traditional way to refine the metal is by adding chlorine to the inert gas during degassing. Chlorine is a toxic gas, however, so Alcoa has devoted considerable attention to minimization or elimination of its use.

The reaction of chlorine with alkaline earth elements dissolved in aluminum was the subject of several recent experimental and theoretical studies [20-22]. The most important practical observation from this work is this: The amount of chlorine that can react to remove Na (and Li or Ca) is proportional to the concentration of alkaline earth elements in the metal. Thus, the highest concentration of chlorine (typically no more than about 0.5 to 1%) goes into the first stage of the degassing box. It is here that the concentration of Na is highest. If chlorine is also required in subsequent stages of degassing, it is used at smaller concentrations (0.1 to 0.5%), because the amount of Na to be

removed is less. Staging the chlorine use in this way maximizes the use of chlorine, so that nearly all is used to remove alkaline earth impurities, and chlorine emissions are minimized.

When these procedures are followed, the efficiency of sodium removal is also high. This issue is especially important in high Mg alloys, since Na contents as low as 5 ppm can embrittle the casting. In one of our plants producing 5xxx alloys, 96% of the Na is removed on average by a four rotor A622. In the same plant using an 8 rotor ACD, 89% of the Na was removed by the degasser when producing the same alloys.

Alcoa is also now working to eliminate chlorine gas by degassing with a mixture of argon and reactive salts. Numerous trials have been conducted, with positive results. As a consequence, Alcoa's new smelter in Iceland will use salt injected into an ACD system. This equipment has been described in [23]. Elimination of chlorine gas by use of salt injection is presently a major focus area of development for our company. Recent developments in this area are outlined in a companion publication [24].

Concluding Remarks

In this paper we gave an overview of degassing and provided an introduction to important scientific principles which underlie the process. One of these is the thermodynamic degassing efficiency. This concept has not been treated to any appreciable extent in the past literature and deserves to be more widely understood. It is hoped that the understanding offered herein will help cast house engineers to improve their melt treatment.

We did not try to establish which degassing process is 'best'. Many important factors were not considered, such as: capital cost, ease of maintenance, and operating expense. It was noted above that the ACD operates at lower thermodynamic efficiency than the A622, but when the ACD is allowed to use more flux gas both systems provide the same hydrogen removal. Thus, there will be situations where the ACD is the preferred choice. In other plants a traditional in-line station (A622, SNIF, Alpur) will be used. The choice made often depends on a number of factors not considered in this paper.

Acknowledgements

The author gratefully acknowledges Alcoa Primary Metals, whose support has made this study possible.

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