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Degassing of Aluminum Alloys Using Ultrasonic Vibration

June 2006

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Abbreviations and Acronyms

American Foundry Society
American Society for Metals
U.S. Department of Energy
General Motors
Industrial Materials for the Future (DOE)
Industries of the Future (DOE)
Industrial Technologies Program (DOE)
North American Die Casting Association
Oak Ridge National Laboratory
piezoelectric lead zirconate titanate
radio frequency
reduced pressure test
The Minerals, Metals, and Materials Society
University of Tennessee, Knoxville

1. Executive Summary

1.1 Research and Development

The project described in this report was conducted in response to a call for proposals under the Aluminum Industries of the Future Program (IOF), Industrial Technologies Program (ITP), of the U.S. Department of Energy (DOE). The research was intended to lead to a better fundamental understanding of the effect of ultrasonic energy on the degassing of liquid metals and to develop practical approaches for the ultrasonic degassing of alloys.

Hydrogen precipitates as porosity during the solidification of aluminum alloys and is detrimental to the mechanical properties of aluminum alloy castings, heavy section plates, and forgings. Reducing porosity during casting is still a challenging problem in the aluminum industry. The most effective way to do so is degassing. Two types of degassing methods are currently in use. One of these, vacuum degassing, is used primarily in the steel industry and thus not generally used in the aluminum industry. The second method, generally employed in the aluminum industry, is rotary degassing, which uses finely dispersed argon, chlorine, and various salts. However, the use of chlorine creates environmental problems. Thus, a clean and inexpensive technique for degassing should be useful for the aluminum industry.

Ultrasonic degassing uses high-intensity ultrasonic vibrations to generate oscillating pressures in molten aluminum. In the region of minimum pressure, cavitation occurs in the melt, and fine bubbles are produced. The bubbles produced during cavitation could provide nuclei for hydrogen bubbles to coalesce and flow out of the melt. There is a considerable amount of documentation in the Russian literature on the application of ultrasonic energy to materials processing in general, and to melts and solidifying alloys in particular [1, 2]. However, little systematic work has been reported in the public literature on the application of ultrasonic energy to the degassing of aluminum alloy melts. Most of the data is empirical, and only general phenomenological studies have been conducted. Currently, ultrasonic energy is not used commercially for degassing liquid metals.

The goals of the project described here were to evaluate core principles, establish a quantitative basis for the ultrasonic degassing of aluminum alloy melts, and demonstrate the application of ultrasonic processing during ingot casting and foundry shape casting.

The project focused on validating the feasibility of ultrasonic degassing in molten aluminum, exploring the possibility of combining ultrasonic processing with other technologies such as vacuum degassing and argon degassing, and investigating the mechanisms of ultrasonic degassing. The work was carried out as a collaborative effort between the University of Tennessee at Knoxville (UTK) and Oak Ridge National Laboratory (ORNL). Industrial support was provided by Ohio Valley Aluminum Co., a high-quality aluminum extrusion billet manufacturer, and Secat, Inc., a consortium of aluminum companies.

The achievements of this project are as follows:

• **Designed and built a number of experimental systems for testing ultrasonic degassing** Several experimental systems have been designed and built for the degassing of aluminum using ultrasonic vibration under various conditions such as in air, under vacuum, and in combination with argon degassing. These systems, located at ORNL, are available for use by the aluminum industry through ORNL's User Facilities program (see http://www.ornl.gov/adm/tted/technology_comm/collaborations/user_facilities.shtml), which is sponsored by DOE.

• Demonstrated ultrasonic degassing of aluminum alloys

Ultrasonic degassing was tested in aluminum melt under various conditions including degassing in air, under vacuum, and in combination with an argon purge. The experimental results indicate that degassing can be achieved within a few minutes of ultrasonic vibration, much faster than the traditional degassing methods. The limitations of ultrasonic degassing have been ascertained.

• Investigated the mechanisms of ultrasonic degassing

Experiments were carried out in water to observe cavitation and the evolution of cavitation bubbles during high-intensity ultrasonic vibration. The conditions under which cavitation occurs and the survival of the cavitation bubbles were examined.

• Developed new technology for ultrasonic degassing of molten aluminum alloy

Based on the understanding of the evolution of cavitation bubbles derived from this research, a new method was validated for using a small amount of purge gas to increase the efficiency of ultrasonic degassing. The method has the potential of significantly reducing the degassing time and dross formation during degassing. Initial experimental results show that a reduction of more than 50% of dross formation can be achieved using this new technology. The technology can be further optimized for improved results.

The new technology offers several advantages:

- No moving/rotating parts in the degassing system No rotary graphite parts are used in this technology, so the system is more robust.
- Fast degassing The bubbles formed in this technology are much smaller than those in the conventional degassing system.
- Less use of argon and no use of chlorine The cavitation bubbles are formed within the metal, so the bubble/melt surface is free from oxidation. As a result, no chlorine is needed.
- Minimal capital investment /operating cost
- Less dross formation The melt surface is not disturbed during ultrasonic degassing.

The results can be scaled up for continuous degassing of aluminum alloys and magnesium alloys.

Energy and environmental benefits from the results of this project can occur through a number of avenues:

- Energy benefits accrue from less dross formation.
- Productivity increases occur due to the shorter processing time.
- Environmental benefits accrue from the elimination of fluorine gas from the process.

The U.S. aluminum industry produces more than 23 billion pounds of aluminum metal annually. The industrial average of dross formation during degassing of molten metal is about 0.2%. Assuming a 50% reduction in dross formation during degassing, full-scale industrial implementation of the technology developed during this project would lead to energy savings in excess of **2 trillion Btu** by the year 2015 considering an annual market growth of 2% and a likely technology market share of 90%.

1.2 Technology Transfer

As a result of this research effort, a U.S. patent application has been filed (see Sect. 6.1). Several U.S. companies have signed nondisclosure agreements with ORNL and are evaluating the technology.

1.3 Commercialization

This study focused on aluminum alloys and demonstrated the application of ultrasonic processing for molten aluminum degassing. Project participants have used several mechanisms to inform industries of the research results and to advance commercialization. These have included conducting review meetings at industrial sites and at ORNL; making presentations at national meetings organized by the Minerals, Metals, and Materials Society (TMS), the American Society for Metals (ASM), and the American Foundry Society (AFS); and making presentations at industrial locations and for organizations such as the North American Die Casting Association (NADCA) and AFS.

Companies or organizations that have expressed interest in the technology during this endeavor include Feseco Metallurgical, Inc., Lunt Manufacturing, the North American Die Casting Association (NADCA), Ohio Valley Aluminum, Pyrotek, and Secat.

1.4 Recommendations

The new approach developed in this project — ultrasonic degassing with the assistance of argon bubbling — has the potential to be used by the aluminum industry in processing molten aluminum alloys and by the metalcasting industry in processing other nonferrous alloys that are prone to porosity due to dissolved gases in molten alloys.

While this technology was briefly tested and validated in this project, the limited scoped did not include testing and validation in melt on an industrial scale. Companies interested in the technology developed in this project would need to evaluate it on a large scale before making a commitment to it. Therefore, it is recommended that further effort be made to scale up the new methodology and evaluate it in industrial environments.

2. Introduction

This project was conducted in response to a call for proposals under DOE's IOF program. The goals of the project were to evaluate core principles, establish a quantitative basis for the ultrasonic degassing of aluminum alloy melts, and demonstrate the application of ultrasonic processing during ingot casting and foundry shape casting. The results of the work could impact a wide range of aluminum alloy processing tasks, including direct chill casting and foundry shape casting.

The Russian literature contains a considerable amount of documentation on the application of ultrasonic energy to materials processing in general, and to melts and solidifying alloys in particular [1, 2]. However, very little systematic work has been reported in the public literature on the application of ultrasonic energy to the degassing of aluminum alloy melts. Most of the data is empirical, and only general phenomenological studies have been conducted. Currently, ultrasonic energy is not used commercially for degassing liquid metals. The project described here was intended to provide a better fundamental understanding of the effect of ultrasonic energy on the degassing of liquid metals and the development of practical approaches for the ultrasonic degassing of alloys.

The important issues that were addressed and solved by this project included the coupling of the ultrasonic transducer to the melt, the effective transmission and distribution of ultrasonic vibrations in the melt, determining the effective level of ultrasonic vibration intensity, and protection of the melt surface. The project developed laboratory-scale equipment for ultrasonic degassing, studied the effect of process parameters, and identified the range of applicable process parameters for commercial implementation of the technology.

The work was carried out as a collaborative effort between UTK and ORNL. Industrial support was provided by Ohio Valley Aluminum Co. and Secat, a consortium of aluminum companies. Ohio Valley Aluminum has been a manufacturer of high-quality aluminum billets for the aluminum extrusion industry since 1955, utilizing highly skilled employees and the latest melting and casting technology.

The project carried out systematic research on the application of ultrasonic energy to the degassing of aluminum alloy melts. The results provide a quantitative basis for hydrogen removal during degassing of aluminum alloy melts. A new approach has been proposed using ultrasonic vibration to break up large argon bubbles into small bubbles for more efficient degassing of aluminum melt. This new approach is described in the patent application cited in Sect. 6.1.

Commercial application of this new technology would provide energy benefits and cost savings by reducing dross formation during the degassing process and by reducing the use of argon for the degassing of aluminum alloys. The U.S. aluminum industry produces more than 23 billion pounds of aluminum metal annually. Dross formation during degassing of molten metal is about 0.2%. Assuming a 50% reduction in dross formation during degassing, full-scale industrial implementation of the technology would lead to energy savings in excess of 2 trillion Btu by the year 2015.

3. Background and Technical Approach

3.1. Background

Porosity is one of the major defects in aluminum alloy castings because it can be detrimental to the mechanical properties and the pressure tightness of a casting. Porosity occurs in castings because gas precipitates from solution during solidification or because the liquid metal cannot feed through the interdendritic regions to compensate for the volume shrinkage associated with solidification. Hydrogen is the only gas that is appreciably soluble in molten aluminum [3, 4]. Thus, the removal of the dissolved hydrogen from the molten aluminum alloy is critical for the production of high-quality castings. Several methods are currently in use to degas aluminum [5–7]. These methods include the use of nitrogen or argon, or a mixture of either of these with chlorine, as a purge gas [5, 6]. Other techniques include tablet degassing by use of hexachloroethane (C_2Cl_6) tablets [5, 6], vacuum degassing [8–10], and ultrasonic degassing [11–15].

Ultrasonic degassing, an environmentally clean and relative inexpensive technique, uses highintensity ultrasonic vibrations to generate oscillating pressures in molten aluminum. Degassing requires introducing into the melt acoustic energy of a sufficient intensity to set up a pressure variation that will initiate cavitation [1, 12]. A minimum acoustic intensity of 10 w/cm² at frequencies of 20 kHz is required for cavitation to occur in the liquid form of most materials. The maximum and minimum pressures caused in the melt are given by the following equations [16, 17]:

$$p_{\max} = p_0 + \sqrt{2\rho cI}, \qquad (3.1)$$

$$p_{\min} = p_0 - \sqrt{2\rho c I} , \qquad (3.2)$$

where p_0 is the atmospheric pressure; ρ and c are the density and the wave velocity of the melt, respectively; and I is the wave energy density in the melt. Thus, the application of ultrasonic energy to a melt results in instantaneous variation in the local pressure from the minimum to the maximum. The low pressure during cavitation creates tiny bubbles. At high pressures, the bubbles collapse and produce shock waves. The ultrasonically induced cavitation can be used for degassing.

3.1 Project Goal and Scope

The goal of the project was to evaluate the ultrasonic degassing technology, with the aim of helping the aluminum industry reduce dross formation during degassing and achieve significant energy savings. The research evaluated core principles and established quantitative bases for ultrasonic degassing of aluminum alloy melts, and demonstrated the application of ultrasonic vibration for the degassing of aluminum melts. Important issues to be studied and resolved included the coupling of the ultrasonic transducer to the melt, the effective transmission and distribution of ultrasonic vibrations in the melt, determining the effective level of ultrasonic vibration intensity and frequency, and protection of the melt surface. The project led to the development of equipment systems for ultrasonic degassing, an understanding of the effect of processing parameters, and identification of the range of applicable process parameters for commercial implementation of the technology.

This project evaluated the effect of acoustic energy of varying phonon energies introduced in a melt during degassing. The variables were acoustic power and melt temperature. Previous work has employed acoustic frequencies of 20 kHz or less and pulsed power of up to a few kilowatts. This

study used power densities more than an order of magnitude higher than previously used. The results of this research are expected to impact a wide range of aluminum alloy processing methods, including direct chill casting and foundry shape casting.

3.2 Project Objectives and Approach

The objectives of the project were (a) to obtain a better fundamental understanding of the effect of ultrasonic energy on the degassing of liquid metals during melting and (b) to develop practical approaches for the ultrasonic degassing of alloys.

To achieve these objectives, we approached the project with the following tasks:

• Task 1. Experimental systems

Experimental systems were designed and assembled for the investigation of the effect of ultrasonic energy on the degassing of aluminum alloys. The experimental systems included a radio-frequency (RF) generator coupled to an ultrasonic transducer, affixed to a crucible of molten alloy, and thermocouples for monitoring temperatures in the melt.

• Task 2. Degassing of aluminum alloy melts

The experimental systems assembled in Task 1 were used to study the degassing of aluminum alloy melts. A number of variables and process variations were investigated. The efficacy of degassing was measured by density measurements and LecoTM hydrogen analysis. Subtasks included

- Transmission of ultrasonic vibrations into the melt
- Effect of humidity
- Effect of melt temperature
- Effect of melt volume
- Vacuum degassing with ultrasonic vibration
- Argon degassing with ultrasonic vibration
- **Task 3. Determination of fundamental mechanisms during ultrasonic processing** In this task, the mechanisms of ultrasonic degassing of alloy melts were determined. Based on the fundamental understanding obtained, optimum parameters for ultrasonic degassing were proposed.
- **Task 4. Industrial applications of results** Based on the results of laboratory studies, a new approach was developed for the application of ultrasonic degassing to large melts.

• Task 5. Reports and publications The results of this project have been compiled in reports to DOE and published in the open literature.

4. Experimental Systems and Methods

4.1 Experimental Systems

Three experimental systems were designed and built during this project: one for ultrasonic degassing in air, one for ultrasonic degassing under reduced pressure, and one for ultrasonic degassing with a purging gas. These systems were used successfully to test ultrasonic degassing under various conditions. The three experimental systems, located at ORNL, are available for use by private industrial companies and research institutions through various user programs at ORNL that are funded by DOE.

Most of the experiments described here were carried out at ORNL and UTK. Industrial partners provided aluminum alloys and performed hydrogen measurements of the ultrasonically processed samples. The aluminum alloy used in this project was the A356 alloy: its composition is shown in Table 4.1.

····· , · ··· · ·			
Element	Wt. %	Element	Wt. %
AI	92.5	Fe	0.1
Si	7.2	Ti	0.1
Mg	0.35	Mn	0.05
Cu	0.1	Zn	0.05

 Table 4.1. Chemical composition of aluminum

 alloy A356

4.1.1 Ultrasonic System for Degassing in Air

Figure 4.1 shows the experimental system for ultrasonic degassing under normal atmospheric pressure. This system consisted of a 20-kHz ultrasonic generator, an air-cooled converter made of piezoelectric lead zirconate titanate (PZT) crystals, a booster, a probe, an acoustic radiator to transmit ultrasonic vibration into aluminum melt, and a furnace in which the aluminum melt was held. The transducer was capable of converting up to 1.5 kW of electric energy at a resonant frequency of 20 kHz. The amplitude of the ultrasonic vibration could be continuously adjusted from 30 to 100% of 81 μ m, which is the maximum amplitude of the unit.

In this experimental system, ultrasound was injected into the aluminum melt by using a cylindrical radiator made of titanium alloy Ti-6Al-4V. The aluminum alloy was held in a graphite crucible and melted in the electric furnace. The temperature of the melt was controlled within an accuracy of $\pm 10^{\circ}$ C. After the melt was heated to a predetermined temperature, a preheated ultrasonic radiator was inserted in molten metal. Ultrasonic vibration was then applied in the molten metal for specified periods of time before samples were taken for density or hydrogen concentration measurements.

One of the difficulties encountered in designing these systems was designing an ultrasonic radiator that is resonant with the system. During the degassing of an aluminum alloy, the radiator is inserted into the molten metal. The temperature in the end of the radiator that is immersed into molten metal is high while the temperature at the other end, attached to the booster, is low. As a result, the radiator needs to be specially tuned.



Fig. 4.1. Experimental system for ultrasonic degassing of aluminum in air. *Key:* (1) ultrasonic generator; (2) controller for electric furnace; (3) melt temperature indicator; (4) pneumatically operated device; (5) air inlet; (6) transducer; (7) booster; (8) horn; (9) radiator; (10) electric furnace.

4.1.2 Ultrasonic System for Degassing under Reduced Pressure

Figure 4.2 shows the vacuum chamber used for vacuum degassing with the assistance of ultrasonic vibrations. The crucible inside the electric furnace can hold molten aluminum alloys weighing up to 800 g. The minimum remnant pressure of this vacuum chamber is 50 mtorr. In this vacuum system, it takes a few seconds for the vacuum chamber to reach 100 torr and 10 torr, about 1 min to reach 1 torr, and 20–30 min to reach 0.1 torr.

One of the features of this system is that it allows the use of a cover gas for aluminum processing. A gas can be filled into the vacuum chamber, through the gas inlet shown at the top left in Fig. 4.2, for protection or processing of an alloy during melting. In the meanwhile, ultrasonic vibration can be injected into the alloy.

4.1.3 System for Ultrasonic Degassing Combined with Argon Degassing

A third experimental system was designed and fabricated in order to investigate the possibility of combining ultrasonic vibration with argon degassing. Apart from the usual components shown in Figs. 4.1 and 4.2, an acoustic probe with a gas purging capability was fabricated. Tests were carried out in A356 melt and in water. The furnace used to melt the A356 alloy can hold up to 6 kg of melt. Degassing was carried out in the melt held in the furnace. A flowmeter was used to control and monitor the flow rate of argon. Ultrasonic vibration was injected from the top of the melt.



Fig. 4.2. Vacuum chamber used for vacuum degassing with the assistance of ultrasonic vibration.

In order to determine the mechanism of argon degassing when combined with ultrasonic vibration, specially designed radiators with a gas purging capability were used to treat the water. The interaction of the air bubbles with high-intensity ultrasonic vibrations was recorded with a video camera, and the data were analyzed. The location and orientation of the radiator with respect to the water container were varied to determine the optimal effect on breaking up argon bubbles and to distribute the fine bubbles uniformly in the water.

4.2 Experimental Methods

Ultrasonic degassing was first carried out in aluminum A356 melt in air. The metal was melted and held at desired temperatures for a half an hour before degassing. The parameters that affect ultrasonic degassing were then studied. These parameters included the humidity of the air, the temperature of the melt, and the volume/size of the melt.

Vacuum degassing was then carried out in the experimental system shown in Figure 4.2. After the solid metal charged into the furnace was melted and held at a desired temperature for a half an hour, a vacuum was drawn. The process parameters studied under vacuum degassing were the remnant pressure and degassing time. After the vacuum degassing characteristics were determined, ultrasonic degassing under the reduced pressure was carried out to compare the degassing rates. For the experiments involving ultrasonic degassing under reduced pressure, ultrasonic vibration was applied to the melt as soon as the vacuum pump was started.

Ultrasonic degassing combined with argon degassing was also carried out in air. Argon was blown through the center of an ultrasonic radiator. In order to reveal the mechanism of degassing in liquids, the system was tested in water with the radiator in the container holding water in varying locations and orientations. A video camera was used to capture the behavior of air bubbles under ultrasonic vibrations.

The molten metal that had been processed using these degassing systems was then analyzed to determine metal density and hydrogen content. The density of the specimens was determined using the reduced pressure test, which is the industrial standard for measuring the density of aluminum castings. The hydrogen in the melt was measured by the use of the LecoTM hydrogen analysis. Details of these two methods are discussed in the following sections.

4.2.1 Density Measurements

The most common method used by foundrymen to monitor the degassing process in order to estimate the hydrogen level accurately in the melt is the reduced pressure test (RPT). The test specimen can be evaluated by visually examining the top surface. A more exact evaluation of the test specimen can be made by apparent density measurements, which yield a semi-quantitative estimate of hydrogen content.

For these experiments, RPT was used to determine the porosity level of the melt. Figure 4.3 shows the RPT system. Molten alloy (~120 g) was poured into a preheated, thin-walled iron cup and allowed to solidify under a reduced pressure of 50 mm of mercury. (Pressures of 50–100 mm are usually used for RPT [18, 19]). The RPT specimens were sectioned vertically in the middle and polished to reveal the extent of the hydrogen porosity. The density of RPT specimens was measured by using the apparent density measurement method [6]. The specimens were weighed in air and in water. The density, D, of the specimen is given by the following equation:

$$D = \frac{W_a}{W_a - W_w} , \qquad (4.1)$$

where W_a and W_w are the weights of the specimen measured in air and water, respectively.

In the experiments, the specimens for density measurements ($\sim 1 \text{ cm}^3$ each, here referred to as the small samples) were cut from the center of RPT specimens. All the density results discussed in Sect. 5 were obtained from the small samples.

To understand the error introduced by the use of the small sample, density measurements were also carried out using half of the RPT specimens (here referred to as the large samples). Thirty large samples were measured, and the results were compared with the results for the small samples.

Figure 4.4 shows the relationship between the measured density for the small samples and the large samples. The density values obtained for the large samples were higher than those obtained for the small samples. This result occurred because the solidified metal near the mold/metal surface was denser than the metal near the center of the RPT ingot. Readers who intend to compare their results with the results of this project by using half of a RPT specimen for density measurement should use Fig. 4.4 for converting the data from large specimens to small specimens.





Fig. 4.3. Reduced pressure test (RPT) system used in this project for measuring the density of aluminum alloys processed by various degassing methods.

Fig. 4.4. Comparison of the measured density from small sample and large sample.

4.2.2 Hydrogen Measurements

Although the RPT method has been widely used in the aluminum foundry industry because of its rapidity, simplicity, and economy, it does not measure absolute hydrogen levels. The Leco hydrogen analysis is an accurate method for measuring the hydrogen content. However, it is difficult and expensive to use as a degassing control technique.

In this project, the degassing effect of the various methods was evaluated using RPT due to funding constraints. Hydrogen content was evaluated using a calibration curve for converting the density data into hydrogen content data. The calibration curve is shown in Fig. 4.5. The curve was obtained using more than 40 specimens made from molten metal of various hydrogen contents. A Ransley mold — a metal mold especially designed to solidify molten metal to form a preferred specimen for Leco hydrogen analysis — was used for making the specimens for hydrogen measurements. RPT type specimens were also prepared from the same melts.

The correlation between the hydrogen content determined by the use of the Leco hydrogen analysis and the density as determined from PRT is shown in Fig. 4.5. The curve was obtained using the method of least squares (linear regression analysis). As was expected, the measured density of an RPT sample decreases as the measured hydrogen content increases. The measured hydrogen content varied from 0.5 ppm to <0.1 ppm, while the measured density varied from 1.75 g/cm³ to ~2.65 g/ cm³. Extrapolating the hydrogen content to zero using the calibration curve, the corresponding density of a specimen is close to a theoretical density of 2.68g/ cm³. The data points plotted in Fig. 4.5 were used for converting the measured density data to hydrogen content.



Fig. 4.5. Correlation between hydrogen content from the measured hydrogen data (using the Leco hydrogen analysis method) and measured density using PRT.

5. Results and Discussion

The experimental systems and methods described in Sect. 4 were used to evaluate degassing in molten aluminum. Experiments were first carried out in air to test degassing using ultrasonic vibration alone, and the limitations of this method were noted. Further experiments were then performed under reduced pressures and in combination with purging by argon gas. This section describes these results in detail. At the end of this section, the mechanisms of ultrasonic degassing are discussed.

5.1 Ultrasonic Degassing in Air

Ultrasonic degassing was carried out for an aluminum alloy A356 melt under three variable conditions:

- 1. *Humidity*. The humidity of the air was varied from 40 to 60%.
- 2. *Temperature of the melt*. Four melt temperatures (620, 660, 700, and 740°C) were tested.
- 3. Volume or size of the melt. The weight of the melt was either 0.2, 0.6, or 2 kg.

The effects of each of these variables is discussed below.

5.1.1 Effect of Humidity on Bulk Hydrogen Content in Aluminum Melt

The humidity was varied in order to examine its effect on the initial hydrogen content of the melt. These are important baseline data for evaluating the degassing efficiency of various methods.

The atmosphere is one of the most important sources of water vapor. Most of the hydrogen atoms dissolved in molten aluminum come from the dissociation of water vapor at the surface of the liquid aluminum, according to the reaction [9]

$$2Al_{(l)} + 3H_2O_{(g)} = Al_2O_{3(s)} + 6H.$$
(5.1)

The hydrogen content in melts varies across a wide range and depends considerably on atmospheric humidity, and the time and temperature at which the melt is held.

Figure 5.1 shows the porosity levels in the RPT specimens using melts prepared at 740°C under humidities of 60% and 40%. More pores occurred in the specimen made from melt prepared at 60% humidity than in that prepared at 40% humidity.

Figure 5.2 shows the measured densities from the specimens made from melts prepared at different humidity levels. The porosity level increases with an increase of humidity. As a result, the density of the specimen decreases with increasing humidity. Obviously, the initial hydrogen content in the melt increases with increasing humidity at a given temperature and a melting time. The measured density data can be converted into hydrogen content using the calibration curve shown in Figure 4.5. The hydrogen content as calculated using this calibration curve were about 0.45, 0.37, and 0.35 ppm when the humidity levels were 40, 50, and 60%, respectively.



(a) 60% humidity

(b) 40% humidity

Fig. 5.1. Porosity in the RPT specimens using the melt prepared at 740°C at 60 and 40% humidity.



Fig. 5.2. Correlation between density and humidity for the RPT specimens.

Figures 5.3 and 5.4 show the ultrasonic degassing rates in molten A356 alloy prepared at 740°C at 40 and 60% humidity or with differing initial hydrogen concentrations The experiments were carried out using a crucible containing 0.2 kg of aluminum melt. Without ultrasonic vibration, the density of the specimen cast under a humidity of 60% was much lower than that cast under a humidity of 40%. With ultrasonic vibration, the density of the specimen increased rapidly with increasing ultrasonic processing time during the first minute and then reached a plateau density, which corresponds to the steady-state hydrogen concentration in the melt at 740°C. This trend was true for specimens cast under both humidity levels. The results shown in Figure 5.3 suggest that degassing in this small aluminum melt was extremely fast. No matter what the initial hydrogen concentrations were, degassing was achieved within one minute. Humidity had little effect on the time required for degassing using ultrasonic vibration.







Fig. 5.4. Hydrogen content as a function of ultrasonic processing time in melts prepared at different humidities.

The hydrogen content in the melt processed for various ultrasonic processing times is shown in Fig. 5.4. Before ultrasonic treatment, the initial hydrogen concentrations were 0.45 and 0.36 ppm at humidity levels of 40 and 60%, respectively. The hydrogen content decreased sharply with increasing ultrasonic processing time. The hydrogen content at the plateau density shown in Fig. 5.4 was 0.14 ppm, although the data scatter is much larger than that shown in Figure 5.3.

The porosity levels in the RPT specimens are illustrated in Figure 5.5. Within just 1 min of ultrasonic vibration, the hydrogen level in the melt was so low that few pores were found on the polished surface taken at the center of the RPT specimen [Fig. 5.5(b)]. In fact, Fig. 5.5(b) represents the porosity level in the specimen when the steady-state plateau density had been reached. Increasing the ultrasonic processing time did not change the porosity level in the specimen significantly.

The mechanism of ultrasonic degassing is closely related to the phenomenon of cavitation in the melt. An ultrasonic wave propagating through a liquid metal generates alternate regions of compression and rarefaction. The alternating pressure above the cavitation threshold creates a large number of small cavities in the liquid. In the rarefaction phase, the surface area of a pulsating bubble is many times greater than its area in the compression phase. Therefore, the gas diffusion flow toward the bubble during the rarefaction phase exceeds the gas diffusion flow from the bubble during the compression phase. Because of the one-way gas diffusion toward the cavity, the pulsating cavity enlarges, resulting in degassing in the melt. Since the cavitation bubbles are quite



(a) 0 min



(b) 1 min



(c) 4 min

Fig. 5.5. Porosity in the RPT specimens at various ultrasonic processing times. The melt was processed at 740°C at 60% humidity.

small and their number is large, the early stage of ultrasonic degassing is extremely fast.

The kinetics of ultrasonic degassing suggests that as the gas is being removed from the liquid, the rate of ultrasonic degassing slows down. In the meantime, hydrogen is still being absorbed and dissolved into the melt at the melt surface according to Eq. (5.1). When the hydrogen removal rate equals the hydrogen absorption rate, a steady-state hydrogen concentration is established in the melt. This steady-state hydrogen concentration should not be affected by the initial hydrogen concentration in the melt.

5.1.2 Effect of Melt Temperature on Ultrasonic Degassing

Figure 5.6 shows the measured density of RPT specimens at different melt temperatures and 60% humidity. At a given atmospheric humidity, the density of a specimen decreases with increasing temperature. This trend can be explained by the fact that the solubility of hydrogen in molten aluminum increases with increasing temperature.



Fig. 5.6. Variation of density for the RPT specimens at various melt temperatures at a humidity of ~60%.

Figure 5.7 and 5.8 show the efficiency of ultrasonic degassing in A356 alloy melts under various melt temperatures. The results were obtained in a crucible containing 0.2 kg of the aluminum alloy. As illustrated in Fig. 5.7, when the melt was ultrasonically processed at 700 or 740°C, the melt reached a steady-state density plateau after about 1 min. The processing time required to degas the melt (to reach the steady-state density plateau) increased with decreasing melt temperature in the temperature range of 620 to 700°C. It took almost 10 min to degas the melt held at 620°C, much longer than in the melt held at 740°C than that held at 700°C. This indicates that degassing efficiency decreases with decreasing processing temperature.

The hydrogen content in the melt prepared at various temperatures and processed at various ultrasonic processing times is shown in Fig. 5.8. The trend for degassing times is identical to that shown in Fig. 5.7. It took a much longer time to degas a melt held at 620°C than one held at 700°C.

There are various reasons for the decrease in degassing efficiency with decreasing temperature. The temperature of the melt has a significant effect on the efficiency of ultrasonic degassing. The lower the melt temperature, the higher the viscosity of the melt. When the melt temperature is below 700°C, the high viscosity will hamper the pulsation of the cavitation bubbles and their coagulation and floating. In the meantime, the diffusion coefficient of hydrogen in liquid metals decreases with decreasing temperature, thus decreasing the rate of one-way diffusion of hydrogen from the solution to bubbles. These two effects contribute to the decreased efficiency of ultrasonic degassing at low temperatures.



Fig. 5.7. Measured density of the RPT specimen as a function of ultrasonic processing time in melt degassed at different processing temperatures.



Fig. 5.8. Hydrogen content as a function of ultrasonic processing time in melt degassed at different processing temperatures.

The data plotted in Fig. 5.7 also indicate that plateau density is not sensitive to the processing temperature in the range between 620°C and 740°C. A longer processing time is needed to reach plateau density when the processing temperature is low, but once the plateau density is reached, the porosity levels in the RPT specimens are identical. The hydrogen concentration corresponding to the plateau density is in the range of 0.1 to 0.2 ppm.

5.1.3 Effect of Melt Volume on Ultrasonic Degassing

In order to obtain a quantitative evaluation of degassing speed for each ultrasonic radiator in molten aluminum, the weight (volume) of the melt was varied. These experiments were carried out in melt at 700°C with a humidity of 60%. The weights of the melts were 0.2, 0.6, and 2.0 kg. As illustrated in Fig. 5.9, the ultrasonic processing time required to reach the steady-state plateau density increases with increasing weight (or volume) of the melt. Degassing times were as follows: 0.2-kg melt — 1 min; 0.6-kg melt — 4 min; 2.0-kg melt — almost 7 min. Degassing speed in a larger-volume melt is much slower than in a smaller melt. However, it is clear that the steady-state plateau density is not sensitive to the volume of the melt. Fast degassing of a large-volume melt can be obtained by use of multiple ultrasonic radiators to inject ultrasonic vibrations into the melt.

Figure 5.10 shows hydrogen content as a function of ultrasonic processing time in melts of different weights (sizes). The hydrogen content was estimated from the density data shown in Fig. 5.9 using the calibration curve shown in Fig. 4.5. The hydrogen content corresponding to the plateau density is $\sim 0.1-0.2$ ppm. As it was expected, it took much longer time to degas a larger-volume melt than a smaller-volume melt.

Figure 5.11 shows the porosity levels in a 2.0-kg aluminum melt degassed at various times using ultrasonic vibration. The humidity during the testing was 60%, and the melt temperature was 700°C. The porosity level in the specimens taken within the first 2 min of ultrasonic processing was still high. After 4 min, the porosity level had been reduced substantially. In fact, the porosity level shown in Fig. 5.11(c) is almost identical to that shown in Fig. 5.5(b), indicating that it takes longer to reach the steady-state hydrogen concentration in a larger-volume melt than in a smaller-volume melt.

The following conclusions can be drawn about using ultrasonic vibration alone as a method for the degassing of molten aluminum in open air:

- Ultrasonic degassing is an efficient way of degassing a small-volume melt. The early stage of ultrasonic degassing is extremely fast. It takes only a few minutes to degas a small-volume melt and reach a steady-state plateau density in an RPT specimen.
- The humidity and initial hydrogen concentration have little effect on the degassing efficiency of ultrasonic vibration.
- The melt temperature has a significant effect on the efficiency of ultrasonic degassing. The rate of degassing in the temperature range of 700 to 74°C is faster than that in the temperature range of 620 to 660°C.
- Neither the processing temperature nor the atmospheric humidity changes the steady-state hydrogen concentration achievable using ultrasonic degassing in a melt.
- The ultrasonic degassing rate in a large-volume melt is obviously lower than that in a small-volume melt. However, the steady-state density of RPT specimens made from ultrasonically processed melts does not change with changing volume of the melt.

These results clearly suggest that ultrasonic vibration alone is efficient for degassing a small volume of melt. Potential applications might be degassing a shallow aluminum trough for transporting aluminum from a melting furnace to a casting machine. Often, multiple radiators would have to be used for degassing a large volume of molten aluminum using ultrasonic vibration only.



Fig. 5.9. Measured density of the RPT specimen as a function of ultrasonic processing time in melts of different sizes.



Fig. 5.10. Hydrogen content as a function of ultrasonic processing time in melts of different sizes.



(c) 2 min

(d) 4 min



5.2 Ultrasonic Degassing under Reduced Pressures

Ultrasonic degassing under reduced pressure was also evaluated in this project. Experiments were initially carried out to assess vacuum degassing alone (with an ultrasonic assist) using an experimental system designed and built in this project. Vacuum degassing data were used as baseline data for evaluating ultrasonic degassing under reduced pressures.

Vacuum degassing was carried out in 0.6-kg aluminum melt samples. The process parameters studied under vacuum degassing were remnant pressure and degassing time. Remnant pressure was varied from 0.1 to 760 torr, and degassing time was varied from 1 to 45 min. For remnant pressures of 10 and 100 torr, the experiments were conducted at temperatures of 720°C with a humidity of ~60%. For remnant pressures of 0.1 and 1 torr, humidity was maintained at ~50%.

Vacuum degassing with the assistance of ultrasonic vibration was performed under two different remnant pressures: 1 and 100 torr. The experiments were carried out with 0.6-kg melts at 720°C with a humidity of 50%.

5.2.1 Vacuum Degassing

The hydrogen level in a melt can be decreased by creating a vacuum above the melt surface. Even a partial vacuum has been found to be effective in reducing the hydrogen level in the melt. Figure 5.12 shows the measured densities of an RPT specimen as a function of treatment time under different remnant pressures. As is indicated in the figure, the density of a specimen increased with increasing holding (vacuum degassing) time. The degassing rates at lower remnant pressures (0.1 and 1 torr) were much higher than at higher pressures (10 and 100 torr). It was evident that degassing rates were

slow when vacuum degassing was used. For instance, with the vacuum degassing technique, 20–30 min were required for a 0.6-kg melt to reach a steady-state plateau density, while the ultrasonic vibration technique required <4 min to achieve the same result, as shown in Fig. 5.9. Figure 5.13 shows hydrogen content as a function of degassing time under various remnant pressures. The hydrogen contents were converted using the calibration curve from the density measurement data shown in Fig. 5.12.

The data points plotted in Figs. 5.12 and 5.13 also indicate that steady-state plateau density or hydrogen content is strongly affected by the remnant pressure. The plateau density or the hydrogen content at the steady state is much higher at lower than that at higher pressures.

The results shown in these two figures can be explained by considering the hydrogen/water vapor partial pressure in the degassing chamber. The equilibrium hydrogen content in the melt is proportional to the partial hydrogen pressure in the atmosphere above the melt. When the pressure in the chamber is reduced, the partial pressure of hydrogen is also reduced. As a result, hydrogen dissolved in the melt evaporates at the melt surface, leading to a decrease in the bulk hydrogen concentration in the melt. Since hydrogen evaporation occurs only at the melt/air interface, the hydrogen atoms in the melt have to diffuse to the melt surface before evaporating into the atmosphere. Thus, vacuum degassing is a diffusion-controlled, slow process. This explains the 20–30 min needed to reach the steady-state plateau density during vacuum degassing. Also, since the equilibrium hydrogen content is proportional to the partial pressure of hydrogen in the atmosphere, the steady-state plateau density and the degassing rate should increase with decreasing remnant pressure. These trends are clearly shown in Figs. 5.12 and 5.13.

Assuming that the equilibrium hydrogen concentration in the melt was maintained for 45 min during vacuum degassing, the equilibrium density and hydrogen concentration can be plotted in Figs. 5.14 and 5.15 under remnant pressures of 0.1, 1, 10, 100, and 760 torr. As shown in Fig. 5.14, the density of the RPT specimen increases with the decrease of the remnant pressure. The corresponding hydrogen content decreases with decreasing remnant pressure.



Fig. 5.12. Measured densities of the RPT specimen as a function of treatment time under different remnant pressure levels.



Fig. 5.13. Hydrogen content as a function of degassing time under different remnant pressure levels.



Fig. 5.14. Measured density of the RPT specimen as a function of remnant pressure. The melts were held at reduced pressure for 45 min before RPT.



Fig. 5.15. Hydrogen content as a function of remnant pressure. The melts were held at various reduced pressures for 45 min before casting into a Ransley mold.

5.2.2 Ultrasonic Degassing under Reduced Pressure

Having established the baseline of vacuum degassing in the experimental system designed and built in this project, the research was then focused on ultrasonic degassing of aluminum melt under reduced pressure. During these experiments, ultrasonic vibration was induced into the melt after the remnant pressure reached 100 and 1 torr. Several seconds were required for the remnant pressure to reach 100 torr, and ~1 min to reach 1 torr.

Figure 5.16 shows the porosity levels in the RPT specimens using melt prepared at 720°C and 50% humidity. Figure 5.16(a) shows the porosity level in the RPT specimen without ultrasonic degassing; Figs. 5.16(b) and (c) show the porosity levels after 30 sec and 2 min of ultrasonic degassing under a reduced pressure of 1 torr. With just 30 sec of ultrasonic processing, the hydrogen level in the melt was so low that few pores could be seen on the polished surface taken at the center of the RPT specimen.

The measured density and the calculated hydrogen content as a function of processing time under various conditions are shown in Figs. 5.17 and 5.18. Data points marked by filled squares indicate the efficiency of ultrasonic degassing under two remnant pressure levels (100 and 1 torr). As these figures indicate, the plateau density was attained much faster through the use of the combination of ultrasonic degassing and vacuum degassing than by use of either ultrasonic degassing or vacuum degassing alone. For comparison, Figs. 5.17 and 5.18 also display data obtained using ultrasonic vibration











(c) 2 min



under normal pressure (in air) (marked on the figures with triangles) and data obtained using vacuum degassing (marked with filled circles). As is illustrated by these two figures, the most efficient degassing method is ultrasonic degassing under reduced pressure and the slowest is vacuum degassing.

The processing times required to reach the steady-state plateau density for the three methods shown in Figs. 5.17 and 5.18 were as shown in Table 5.1.

various degassing methods			
Degassing method	Processing time required		
Ultrasonic	4 min		
Vacuum	20 min		
Ultrasonic under reduced pressure	1 min		

Table 5.1. Processing time for steady-state plateau density using various degassing methods



Fig. 5.17. Measured density as a function of processing time for different degassing techniques, with remnant pressure at 100 and 1 torr.



Fig. 5.18. Calculated hydrogen content as a function of processing time for different degassing techniques and remnant pressures of 100 and 1 torr.

In summary, the density of the RTP specimen increases or the hydrogen content in a melt decreases with decreasing remnant pressure using vacuum degassing, but the vacuum degassing rate is slow because it is a diffusion-controlled process. The degassing rate of vacuum degassing is much slower than that of ultrasonic degassing. The combination of ultrasonic degassing and vacuum degassing results in much faster degassing than does the use of ultrasonic degassing alone. Even under a partial vacuum condition such as 100 torr, the efficiency of ultrasonic degassing could be increased by using ultrasonic degassing under a reduced pressure.

Nevertheless, ultrasonic degassing under reduced pressure has the inherent limitations of ultrasonic degassing. The method can only be used for degassing a small-volume melt. Other methods need to be explored for the degassing of a large volume of molten aluminum.

5.3 Ultrasonically Assisted Argon Degassing

Due to their inherent limitations, methods such as ultrasonic degassing and ultrasonic degassing under reduced pressure cannot be used for a fast degassing of a large-volume aluminum melt. This is partly because there is a large attenuation of ultrasonic vibration in liquids. As a result, the intensity of ultrasonic vibrations decreases sharply with increasing distance from the ultrasonic radiator. On the other hand, degassing with argon is also a relatively slow process due to the large size of argon bubbles. If, however, ultrasonic vibration is applied to the melt, the pressure induced by acoustic waves can break up the large argon bubbles into numerous smaller bubbles, increasing the area of the bubble surface in the melt substantially. This should allow for more efficient degassing. Based on such considerations, ultrasonically assisted argon degassing was tested in this project.

Experiments were first carried out in water to examine the feasibility of using ultrasonic vibration to break up air bubbles and to investigate the conditions under which ultrasonic vibration would be effective in breaking up bubbles injected into the liquid through an acoustic probe with a gas purging capability. Observations were carried out with different air flow rates and vibration amplitudes. The critical amplitude of the ultrasonic vibration above which air bubbles can be broken up was determined by varying the amplitude from a minimum to maximum and observing the bubble size change under a given air flow rate. Ultrasonic vibrations were introduced from the top and the bottom of the melt.

Based on what was learned in the water experiments, ultrasonic vibrations were injected from the top of the molten aluminum alloy to test the efficiency of ultrasonically assisted argon degassing using a vibration amplitude high enough to break up the argon bubbles. A furnace with the capacity to hold 6 kg of molten aluminum was used to melt and hold the A356 alloy. Degassing was carried out in melts of about 5 kg. A flowmeter with a range from 0 to 6 ft³/h was used to control the flow rate of argon. An argon flow rate of 2 ft³/h was used in the experiments. Experimental results show that ultrasonically assisted argon degassing is much faster than argon degassing alone. More importantly, dross formation during ultrasonically assisted degassing is only ¹/₈ that generated during argon degassing.

5.3.1 Water Experiments

The experiments in water were used to reveal the effect of ultrasonic vibration in breaking up air bubbles. Tests were carried out by injecting ultrasonic vibrations from the bottom of the water. Parameters that were investigated included the air flow rate and the amplitude of the ultrasonic vibrations. Air was introduced into water through a 3-mm hole drilled at the center of the ultrasonic radiator. During the experiments, the amplitude of ultrasonic vibration was varied from 30 to 100% of

the maximum amplitude of the ultrasonic unit at a given air flow rate. The size of the air bubbles near the ultrasonic radiator was monitored. The critical vibration amplitude was determined as the point at which a further increase of amplitude resulted in a sharp decrease in the injected air bubble size. Figure 5.19 shows the critical amplitude of ultrasonic vibration as a function of air flow rate. The air bubbles can be broken up easily when the air flow rate is low. As the air flow rate increases, the critical amplitude of ultrasonic vibration has to be increased in order to break up the bubbles. When the air flow rate is below 0.5 ft³/h, the critical amplitude of ultrasonic vibration is about 30% of the maximum amplitude of the ultrasonic unit. When the air flow rate is 6 ft³/h, which is the upper limit of the flowmeter reading, the vibration amplitude needed to break up the air bubble is about 80%.



Fig. 5.19. The relationship between critical ultrasonic vibration amplitude and air flow rate. The critical ultrasonic vibration amplitude is defined as the one above which the injected air bubbles at a certain flow rate can be broken up into smaller bubbles.

5.3.2 Degassing in Molten Aluminum

The results obtained in the water experiments suggested that air bubbles injected at a flow rate of 0-6 ft³/h can be broken up when the amplitude of the ultrasonic vibration is higher than 80% of the maximum amplitude of the unit. Based on this observation, ultrasonically assisted degassing was carried out at the maximum ultrasonic amplitude to ensure the fragmentation of the injected argon bubbles. Because of the constraints of the experimental apparatus, the ultrasonic vibrations were injected into the top of the melt although it was not the preferred method for degassing. (The funding level of this project was not high enough to afford the manufacture of a specially designed degassing system for ultrasonically assisted argon degassing.)

Figure 5.20 shows the density of RPT specimens as a function of processing time in 5-kg melts processed using argon degassing and ultrasonically assisted argon degassing. Figure 5.21 illustrates the hydrogen content of the melt as a function of processing time for both degassing methods. As illustrated in Fig. 5.20, ultrasonically assisted argon degassing is more efficient for degassing than argon degassing. When ultrasonically assisted argon degassing was used, the aluminum melt reached



Fig. 5.20. Measured density of 5-kg melts as a function of processing time with argon degassing and ultrasonically assisted argon degassing. Plateau density is reached within 4–5 min with ultrasonically assisted argon degassing.



Fig. 5.21. Hydrogen content in 50-kg melts as a function of processing time with argon degassing and ultrasonically assisted argon degassing.

the steady-state plateau density in 5 min, whereas more than 10 min were required when argon degassing alone was used. The efficiency of ultrasonically assisted argon degassing is almost two times that of argon degassing.

Despite the much greater efficiency of the ultrasonically assisted argon degassing technique, in our experiments there were still limitations to this method. Because the ultrasonic vibration was injected from the top of the melt, the bubbles released from the ultrasonic radiator could not penetrate deep into the melt due to the large viscous drag force on the bubbles in molten aluminum. As a result, the volume of the molten metal that the argon bubble passed through was limited. An increase in the vibration amplitude or the argon flow rate can increase the volume of molten aluminum being degassed.

The benefits of combining argon degassing with ultrasonic vibration also include a reduction of dross formation during the degassing process. In our experiments, when the large argon bubbles were injected into the molten metal, the top surface of the melt became turbulent as the large bubbles escaped from the melt. The oxide layer on the melt top surface was broken so the molten aluminum was exposed to the air, forming more oxides. The use of ultrasonic vibration broke these large argon bubbles up into tiny bubbles and made the melt surface less turbulent. Furthermore, the shorter degassing time with the use of ultrasonic processing meant that much less dross was formed.

Figure 5.22 shows the amount of dross formed on top of the melt as a function of processing time for argon degassing and ultrasonically assisted argon degassing. The line at the top of the graph illustrates the weight of dross formed during argon degassing of a 5-kg aluminum melt. The bottom line was obtained using ultrasonically assisted argon degassing.

Table 5.2 lists the measured weight of dross formed during degassing using argon degassing without and with ultrasonic processing. At a processing time of 15 min, the amount of dross formed with argon degassing was 39.4 g but only 6.4 g with ultrasonically assisted degassing. Thus, six times more dross formed when argon degassing was used without the ultrasonic processing. And taking into consideration the much shorter degassing time with the ultrasonically assisted argon degassing was much (5 min vs 10 min), dross formation during ultrasonically assisted argon degassing was much less than one-sixth of that produced during argon degassing. As can be seen in Table 5.2, for instance, during a 5-min ultrasonically assisted argon degassing, dross formation was only 1.8 g; by contrast, during a 10-min argon degassing can achieve a reduction of about 92% of dross formation over the use of argon degassing method in degassing of molten aluminum alloys.

It should be noted that our experimental data on dross formation during argon degassing were higher than the industry average. Using our experimental data, if one assumes that argon degassing of 5 kg of molten aluminum has been completed in 10 min, the percentage of dross formation is 0.46% (22.8/5000), as compared to the industrial average of about 0.2%. However the average data for industry were obtained using equipment capable of processing 60,000–140,000 lb/h of molten aluminum. It is reasonable to assume that dross formation in a small-volume melt is much higher than that in a large volume during a turbulent argon degassing process. It is interesting to note that the dross formation during ultrasonically assisted argon degassing time. Dross formation during ultrasonically assuming a 10-min degassing time. Dross formation during ultrasonically assisted degassing of a small melt is much less than the industrial average of 0.2%.



Fig. 5.22. Dross formation as a function of processing time during argon degassing and ultrasonically assisted argon degassing. Much less dross was formed during the ultrasonically assisted argon degassing process.

	Degassing time (min)					
Degassing method	2	5	10	15	20	25
Argon degassing alone	NA	16.6	22.8 ^a	39.4	49.5	65.1
Argon degassing with ultrasonics	0.9	1.8ª	5.2	6.4	NA	NA

Table 5.2. Amount (grams) of dross formed at various degassing times using argon degassing and ultrasonically assisted argon degassing

^a Bold italic indicates the point at which degassing was completed.

In summary, ultrasonically assisted argon degassing is a more efficient method than argon degassing alone, which is a method widely used by the aluminum industry. For a 5-kg aluminum melt, a steady-state density plateau was reached in only 5 min using ultrasonically assisted degassing, compared to more than 10 min using argon degassing. More importantly, dross formation during ultrasonically assisted argon degassing is much less than that during argon degassing

5.4 Mechanisms of Ultrasonic Degassing

The injection of ultrasonic vibration in molten aluminum causes alternating pressure in the melt. While pressure can affect the solubility of hydrogen in molten aluminum, alternating pressure, especially pressure that is alternating symmetrically at high frequencies, cannot affect solubility. However, a sound wave with pressure above the cavitation threshold propagating through a liquid metal generates cavitation bubbles in the melt and drastically alters the mass transfer of gas from the solution to the bubbles.

Cavitation consists of the formation of tiny discontinuities or cavities in liquids, followed by their growth, pulsation, and collapse. Cavities appear as a result of the tensile stress produced by an acoustic wave in the rarefaction phase. If the tensile stress (or negative pressure) persists after the cavity has been formed, the cavity will expand to several times the initial size. (During cavitation in an ultrasonic field, many cavities appear simultaneously at distances less than the ultrasonic wave-length.) In this case, the cavity bubbles retain their spherical form. The subsequent behavior of the cavitation bubbles is highly variable: a small fraction of the bubbles coalesce to form large bubbles, but most are collapsed by an acoustic wave in the cavitation bubbles through the bubble/melt interfaces during the nucleation and growth stage of the cavitation bubbles. Some of the dissolved gases escape when cavitation bubbles at the molten surface collapse, thereby resulting in degassing.

Generally, ultrasonic degassing can be divided into three stages: (1) nucleation of cavitation bubbles on nuclei (usually solid inclusions containing cavities) and growth of the bubbles due to the diffusion of hydrogen atoms from the surrounding melt to the bubbles; (2) coalescence of bubbles to form large bubbles; and (3) floatation of large bubbles to the surface of the molten metal and escape of the bubbles at the top of the melt surface.

The first stage of ultrasonic degassing—nucleation and growth of cavitation bubbles—can be easily achieved using high-intensity ultrasonic vibration. The ultrasonic unit used in this project has a power density much higher than the critical power density for creating cavitation. As a result, cavitation can be readily achieved when the unit is operated at amplitudes higher than 30% of the maximum amplitude. In fact, cavitation was observed in the water experiments described in Sect. 5.3.1. Due to the high attenuation of ultrasonic vibration in the liquid, cavitation forms mainly near the ultrasonic radiator, and the cavitation bubbles are transported to the bulk liquid by ultrasonic streaming. Since the cavitation bubbles are small (in the range of microns) and the interfacial area of the bubble/melt interfaces is large, degassing is extremely fast if most of the cavitation bubbles escape from the melt. This is what happens when ultrasonic vibration is used for degassing a small volume of melt. Due to flow produced by acoustic streaming, the cavitation bubbles formed near the ultrasonic radiator are quickly transported to the melt surface, bringing with them the dissolved gases, which then escape the melt. Use of the reduced pressure of a vacuum facilitates the escape of the gasses at the melt surfaces. This accounts for the fact that ultrasonic degassing and ultrasonic degassing under reduced pressure are two of the methods which enable fast degassing of a small-volume melt.

One critical issue in ultrasonic degassing is the survival of cavitation bubbles in the melt, a phenomenon that affects degassing efficiency. During the initial stages of the nucleation and growth of the cavitation bubbles, the dissolved gases diffuse into the bubble and form gas molecules. As these bubbles travel away from the ultrasonic radiator or as the tensile stress become less tensile or even compressive, the gas molecules can decompose at the bubble/melt interfaces and dissolve back into the melt. The gases in the cavitation bubbles will also dissolve into the melt when the cavitation bubbles collapse. In fact, most of the cavitation bubbles cannot survive long enough to reach the melt surface. The water experiments carried out in this project confirmed that only a small portion of visible cavitation bubbles escaped from the top surface of the water.

Based on this understanding of the mechanism of ultrasonic degassing, a novel approach for ultrasonic degassing has been developed in this project. The new approach involves the use of a small amount of purging gas (argon) for ultrasonic degassing. The idea is to use this purging gas to extend the survival time of the cavitation bubbles, which contain dissolved gas, as the bubbles nucleate and grow. In practice, the purging gas is introduced through the ultrasonic radiator. High-intensity ultrasonic vibration is fed through the radiator to break up the purging gas bubbles as well as to create large numbers of cavitation bubbles. The small purging gas bubbles can survive in the melt forever because argon will not dissolve into the melt. As the purging gas bubbles travel from the radiator to the bulk melt, they collect the cavitation bubbles containing dissolved gases. Acoustic streaming helps to distribute the small purging gas bubbles uniformly throughout the melt, improving the degassing efficiency further. Experiments carried out in water validated the uniform distribution of the purging gas bubbles and the survival of more cavitation bubbles. Experimental results described in Sect. 5.3 of this report show that the new degassing method is more efficient than the traditional purging gas methods.

The use of ultrasonically assisted argon degassing has several benefits:

- 1. This method provides fast degassing. In the experiments the samples reached a steady-state density plateau twice as fast as with argon degassing alone.
- 2. Ultrasonically assisted argon degassing may be an excellent technique for the degassing of a large-volume melt, where bubbles can survive in the melt.
- 3. This method results in less dross formation because the bubbles are much smaller than those that occur in the traditional method. As a result, the melt surface is less turbulent.
- 4. The new method makes much less use of purging gas than does the traditional rotary degassing method.

6. Accomplishments

Several experimental systems were built for the degassing of aluminum using ultrasonic vibration under various conditions: in air, in a vacuum, and in combination with argon degassing. These systems, located at ORNL, are available for use by the aluminum industry through ORNL's User Facilities program (see http://www.ornl.gov/adm/tted/technology_comm/collaborations/ user facilities.shtml).

Ultrasonic degassing was tested under various conditions including ultrasonic degassing alone, ultrasonic degassing under reduced pressures, and the combination of ultrasonic vibration with argon purging degassing. The experimental results, obtained in small volume melts, indicate that degassing can be achieved within a few minutes of ultrasonic vibration, much faster than the traditional degassing methods. The combination of vacuum degassing and ultrasonic vibration will help to promote efficiency in vacuum degassing. Ultrasonic degassing—either using ultrasonic vibration alone, or with vacuum degassing, or in combination with using an argon purging gas—was found to be much more efficient in degassing a small-volume melt than the traditional argon degassing method.

The research in this project has led to an understanding of the mechanism of ultrasonic degassing. As a result, a three-stage degassing procedure proposed (Sect. 5.4 above). The critical issues for ultrasonic degassing have been identified. These issues include ultrasonically induced cavitation and the survival of the cavitation bubbles. Methods that promote cavitation bubble formation and survival increase the degassing capability of ultrasonic degassing.

Based on the understanding of the degassing mechanism, a new approach for ultrasonic degassing has been developed for degassing a large-volume melt. The new approach uses a small amount of purging gas and ultrasonic vibration to break up the purging gas bubbles into tiny bubbles. These tiny bubbles can survive indefinitely and collect the cavitation bubbles as they are distributed throughout the bulk melt by acoustically induced streaming. By coalescing with the purging gas bubbles, more cavitation bubbles survive in the melt. Dissolved gas absorbed into these bubbles can be released at the surface of the melt, leading to degassing. Since the tiny bubbles can survive in the melt indefinitely, the method can be used for degassing a large-volume melt. This approach has been validated in molten aluminum in this project.

The benefits of the method proposed here include the following:

- Fast degassing The degassing rate is at least 50% faster than the traditional argon degassing method.
- Minimized dross formation The dross formation is less than 0.1% using the new method for degassing a 5-kg melt, which is more than 90% less than the dross formation using argon degassing for the same size melt and lower than the industrial average, 0.2, obtained for degassing in large-volume melt.

6.1 Patents

A patent application entitled "Degassing of Molten Alloys with the Assistance of Ultrasonic Vibrations," by Q.Han, H. Xu, and T. T. Meek (Docket No. 920976.00027), has been filed.

6.2 Publications and Presentations

6.2.1 Publications

- Hanbing Xu, Xiaogang Jian, Thomas T. Meek, and Qingyou Han, "Degassing of Molten Aluminum A356 Alloy Using Ultrasonic Vibration," *Materials Letters* 58, no. 29 (2004): 3669–3673.
- Hanbing Xu, Xiaogang Jian, Thomas T. Meek, and Qingyou Han, "Investigation of Ultrasonic Degassing in Molten Aluminum A356 Alloy," pp. 731–735 in *Light Metals 2004*, ed. A. T. Tabereaux (Warrendale, PA: The Minerals, Metals & Materials Society, 2004).
- Hanbing Xu, Xiaogang Jian, Thomas T. Meek, and Qingyou Han, "Ultrasonic Degassing of Molten Aluminum under Reduced Pressure," pp. 915–920 in *Light Metals 2005*, ed. Halvor Kvande (Warrendale, PA: The Minerals, Metals & Materials Society, 2005).
- Hanbing Xu, Thomas T. Meek, and Qingyou Han, "Effects of Ultrasonic Field and Vacuum on Degassing of Molten Aluminum Alloy," submitted to *Materials Letters* on July 26, 2005.

6.2.2 Presentations

- Hanbing Xu, Thomas T. Meek, and Qingyou Han, "Ultrasonic Degassing in Aluminum Alloys," MS&T 05 Conference, Pittsburgh, September 25–28, 2005.
- Hanbing Xu, Thomas T. Meek, and Qingyou Han, "Ultrasonic Degassing of Molten Aluminum under Reduced Pressure," TMS 2005 Annual Meeting, San Francisco, February 13–17, 2005.
- Hanbing Xu, Thomas T. Meek, and Qingyou Han "Investigation of Ultrasonic Degassing in Molten Aluminum A356 Alloy," TMS 2004 Annual Meeting, Charlotte, NC, March 14–18, 2004.
- Hanbing Xu, Thomas T. Meek, and Qingyou Han "Ultrasonic Degassing in Aluminum A356 Alloy," Third International Conference on Light Materials for Transportation Systems (LiMAT 2003), Honolulu, November 2–6, 2003.
- Hanbing Xu, Thomas T. Meek, and Qingyou Han "Ultrasonic Degassing in Aluminum A356 Alloy," Pyrotek, Inc., Canastota, NY, August 18, 2004.
- Hanbing Xu, Thomas T. Meek, and Qingyou Han "Ultrasonic Degassing in Aluminum A356 Alloy," Oak Ridge National Laboratory, Oak Ridge, Tenn., December 8, 2004.

6.3 Technology Transfer

As noted above, a patent application has been filed as a result of this research.

Project participants have used several mechanisms to inform industry of the research results and to advance commercialization. These efforts have encompassed

- conducting review meetings at industrial sites and at ORNL;
- making presentations at national meetings organized by the Minerals, Metals, and Materials Society (TMS), the American Society for Metals (ASM), and the American Foundry Society (AFS); and
- making presentations at industrial locations and for organizations such as the North American Die Casting Association (NADCA) and AFS.

Several U.S. companies have signed nondisclosure agreements with ORNL and are evaluating the technology. Companies or organizations who have expressed interest in the technology during this endeavor include Feseco Metallurgical, Inc.; Lunt Manufacturing; the North American Die Casting Association (NADCA); Ohio Valley Aluminum; Pyrotek; and Secat.

7. Summary and Conclusion

Three experimental systems were designed and built during this project: one for ultrasonic degassing in air, one for ultrasonic degassing under reduced pressure, and one for ultrasonic degassing with a purging gas. These systems were used successfully to test ultrasonic degassing under various conditions. The three experimental systems, located at ORNL, are available for use by private industrial companies and research institutions through various user programs at ORNL that are funded by DOE.

Ultrasonic degassing was carried out in molten aluminum alloy A356 in open air with three variable parameters: humidity (varied from 40 to 60%), melt temperature (620, 660, 700, and 740°C), and volume/size of the melt (0.2, 0.6, and 2 kg). The results show that ultrasonic degassing is an efficient way of degassing a small-volume melt. The early stage of ultrasonic degassing is extremely fast. It takes only a few minutes to degas such a melt and to reach steady-state plateau hydrogen content. The humidity and initial hydrogen concentration have little effect on degassing efficiency when ultrasonic vibration is used. The melt temperature, however, does have a significant effect on the efficiency of this technique. Degassing rates in the temperature range between 700 and 740°C are faster than those in the temperature range between 620 and 660°C. Neither the processing temperature nor atmospheric humidity changes the steady-state hydrogen concentration in the melt. The ultrasonic degassing rate in a large-volume melt is lower than that in a small-volume melt. However, the steady-state hydrogen content of ultrasonically processed melts does not change with changing volume of the melt.

Vacuum degassing was carried out in a 0.6-kg aluminum melt. The parameters studied for the vacuum degassing process were remnant pressure and degassing time. The remnant pressure was varied from 760 to 0.1 torr, and the degassing time was from 1 to 45 min. Vacuum degassing combined with ultrasonic vibration was performed under two different remnant pressures: 100 torr and 1 torr. The results show that under vacuum degassing the hydrogen content in the melt decreases with a decrease of remnant pressure. With this technique, the degassing rate is much lower than that of ultrasonic degassing. A combination of ultrasonic degassing with vacuum degassing makes degassing more efficient than degassing using ultrasonic vibration alone or vacuum degassing alone.

The mechanism of ultrasonic degassing is now fully understood. A three-stage degassing processing has been proposed. The critical issues for ultrasonic degassing have been identified. These issues include ultrasonically induced cavitation and the survival of the cavitation bubbles. Methods that promote cavitation bubble formation and survival increase the degassing capability of ultrasonic degassing.

Based on the understanding of the degassing mechanism, a new approach for ultrasonic degassing has been developed for degassing a large-volume melt. The new approach utilizes a small amount of purging gas and ultrasonic vibration to break the purging gas bubbles into tiny bubbles. These tiny bubbles can survive indefinitely and collect the cavitation bubbles as they are distributed throughout the bulk melt by acoustically induced streaming. The melt surface is also less turbulent because the bubble sizes are much smaller than those present with traditional argon degassing. As a result, the new method is much faster in degassing and produces much less in dross than the traditional argon degassing.

Energy and environmental benefits from the findings of this project can occur through a number of avenues: (1) energy benefits accruing from less dross formation, (2) productivity increases due to the shorter processing time, and (3) environmental benefits through the elimination of fluorine gas from

the process. The U.S. aluminum industry produces more than 23 billion pounds of aluminum metal annually. The industrial average for dross formation during the degassing of molten metal is 0.2%. Assuming a 50% reduction in dross formation during degassing, full-scale industrial implementation of the project results would lead to energy savings in excess of 2 trillion Btu by the year 2015 considering an annual market growth of 2% and a likely technology market share of 90%.

8. Recommendations

The results of this research could impact the aluminum industry in the processing of molten aluminum alloys and the metalcasting industry in the processing of other nonferrous alloys that are prone to porosity formation due to dissolved gases in molten alloys. The new methodology that has been developed for ultrasonically assisted argon degassing has a good potential to be used by these industries. This technology was briefly tested and validated in this university-led project. However, the limited scope of the project meant that the technology has not been tested and validated in quantities of molten aluminum large enough for industrial production. Companies interested in the project results would need to evaluate the new technology on a large scale before making commitment to it. Therefore, it is recommended that the new approach be scaled up and evaluated in industrial environments.

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Appendix: Publication

H. Xu, X. Jian, T. T. Meek, and Q. Han, "Degassing of Molten Aluminum A356 Alloy Using Ultrasonic Vibration, *Materials Letters* 58 (29) (2004): 3669.