

# Overview of ultrasonic degassing development

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**Abstract** Ultrasonic degassing attracts industrial attention as an alternative to Ar-rotary and Ar-lancing degassing, offering environmentally friendly, economical and efficient technology. This paper gives a brief historical overview of ultrasonic degassing development in the 1960s–2010s, discussed basic principles of cavitation-induced degassing and demonstrates various technological implementations of ultrasonic degassing technology in foundry and cast house. Batch versus continuous, gas- or vacuum-assisted versus purely ultrasonic variant of the technology are considered. Main advantages of ultrasonic degassing include rapid degassing without the use of a carrier gas, significantly reduced dross formation, absence of rotating brittle parts. There are also some technological issues associated with the ultrasonic degassing such as material selection of a sonotrode, requirement to process larger melt volumes with less ultrasonic sources, rapid re-gassing of hydrogen-depleted melt. The paper is illustrated mainly with own results.

Keywords: Ultrasound, Degassing, Aluminum, Hydrogen, Cavitation

## Historic overview

Ultrasonic degassing attracts a great deal of interest nowadays due to its cleanliness, reduced dross formation, absence of brittle moving parts and energy efficiency. At the same time, it is not by any means a new technology. The phenomenon of degassing by acoustic cavitation has been studied since the 1920s and have found some industrial or pilot-scale applications in the 1960-1970s, being in the following decades surpassed by inert gas lancing and rotary degassing due to the simplicity and efficiency of the latter technology. The lack of fundamental knowledge on ultrasonic degassing of metals as well as bulkiness,

low reliability of ultrasonic equipment available in the 1960-1980s played role as well.

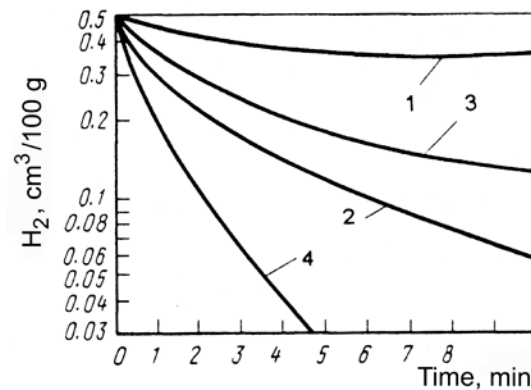
Boyle in 1922 in one of the first studies on cavitation in liquids [1] reported the possible use of ultrasound for the degassing of liquids. Sørensen [2] quantitatively studied the degassing of water at frequencies from 190 to 950 kHz. Krüger was probably the first who applied the ultrasonic degassing to melts, especially to molten glass [3]. He used low-power piezoceramic vibrators and noted the difficulty of finding a stable material for the sonotrode.

To overcome the problem of sonotrode material selection, Eckardt and Eden used high-frequency induction furnace with permanent magnets to induce contactless cavitation [4]. This direction of technology development is currently pursued by scientists in the Universities of Greenwich and Birmingham who apply it to ultrasonic processing of liquid aluminum in a crucible [5].

As early as in the 1940s Esmarch et al. [6] studied the degassing of Al-Mg alloys by sonic vibrations induced by contactless electromagnetic stirring and vibrations in the crucible. Bradfield [7] reported the works of Turner on degassing of molten aluminum and its alloys by direct introduction of ultrasonic oscillations into the melt at 15 kHz and 26 kHz. In 1950 Eisenreich [8] compared vacuum ultrasonic degassing with vacuum degassing, degassing with chlorine lancing, and sonic and ultrasonic degassing. He pointed out the potential of ultrasonic processing but also mentioned related practical difficulties. This experience with combining ultrasonic degassing with gas lancing is currently continued in the Ultra-D Degasser by Southwire [9] where the hollow sonotrode is used for lancing a gas mixture into the melt with the cavitation assisting in fragmenting and dispersing the bubbles. Sergeev [10] noted that despite high potential of ultrasonic degassing there is a challenge in transferring sufficient ultrasonic power to a large mass of liquid metal. This is still a challenge that faces modern zealots of ultrasonic melt processing, and that is addressed in different ways as will be shown below.

The nature of ultrasonic degassing was first revealed on water. Lindström [11] investigated ultrasonic degassing and its relation to the cavitation in water. He suggested that the ultrasonic degassing is due to the diffusion of dissolved gas into the cavitation bubbles, their oscillation and growth and, finally flotation to the surface. The roles of acoustic power (above cavitation threshold), gas concentration and nature of the gas were reported to be not important. Kapustina [12] gave a thorough analysis of ultrasonic degassing mechanisms in water and concluded that the most important role is played by the oscillations of the bubbles in the acoustic field, while ultrasonic cavitation takes the supportive role in intensification of the bubble formation and acceleration of bubble/liquid interfacial diffusion. G. Eskin [13, 14] argued that the cavitation is essential for ultrasonic degassing of metallic melts where the natural gas bubbles are not typically present, unlike those in water. Therefore, the formation and multiplication of bubbles (essential for degassing) in liquid metals can be only achieved by cavitation (unless one used the carrier gas to induce free gas bubbles).

Indeed, early investigations [13, 15] demonstrated that the removal of hydrogen from aluminum alloys depends greatly on the acoustic power transferred to the melt and on the development of cavitation. Figure 1 compares the degassing kinetics for hydrogen in an A356 melt treated with chlorine salts, ultrasound, vacuum, and ultrasound combined with vacuum. These results were recently reconfirmed albeit in small-scale laboratory experiments [16, 17, 18].



**Fig. 1.** Kinetics of hydrogen removal from a 10-kg charge of an A356-type melt for different degassing methods: (1) with chlorine salts; (2) with ultrasound; (3) with vacuum; and (4) with ultrasound in vacuum. (Adapted from [13]).

## Fundamentals of ultrasonic degassing

The main sequence of degassing phenomena can be summarized as follows: (note that ultrasonic intensity larger than the cavitation threshold is required for liquid metals) [8, 12, 13, 19]:

- (1) gas bubbles form on cavitation nuclei (e.g. alumina inclusions with absorbed gas) and grow by rectified diffusion (diffusion from the melt into the bubble with atomic hydrogen recombining into molecular hydrogen) during oscillations in the ultrasonic field (if the liquid contains small bubbles, this stage consists only of their diffusion growth);
- (2) individual bubbles coalesce under the action of the Bjerknes and Bernoulli forces; and
- (3) bubbles float to the surface of the molten metal and release hydrogen into atmosphere.

The degassing efficiency, irrespective of physical and technical means, depends on the concentration of dissolved gas in the liquid. Liquid aluminum and its alloys react actively with gases, either forming non-metallic inclusions or dissolving gas

in the liquid phase. Oxygen in aluminum is an example of the former and hydrogen – of the latter. Hydrogen finds the way to the liquid metal through the interface between the melt and the atmosphere. The main source of hydrogen is water moisture or vapor in the atmosphere. It reacts with liquid aluminum at the surface of the melt and produces alumina and hydrogen. The resultant atomic hydrogen is dissolved in the aluminum and  $\text{Al}_2\text{O}_3$  goes to the melt surface (dross) or is dispersed in the liquid. Hydrogen that is not dissolved or hydrogen that precipitates during degassing or solidification forms molecular hydrogen.

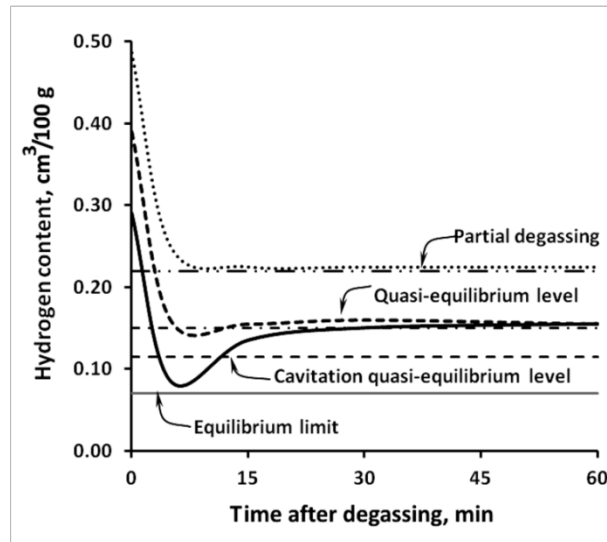
In difference to common metal–metal phase diagrams, the equilibrium between Al and H involves pressure and the solubility of hydrogen in liquid aluminum is not a constant or a fixed number at a given temperature. This solubility depends on the conditions at the interface between the hydrogen-containing medium (atmosphere or bubble) and the liquid metal (surface or bulk). The quasi-equilibrium solubility exists for each combination of the hydrogen concentration in the atmosphere (humidity), in the melt (dissolved hydrogen) and the pressure (air pressure and partial pressure of hydrogen).

The practical importance of dissolved hydrogen in aluminum comes from the sharp decrease of its solubility in aluminum upon solidification: dissolved hydrogen can be measured up to  $0.65 \text{ cm}^3/100\text{g}$  in liquid aluminum just above the melting temperature and just below, the solubility drops down to  $0.034 \text{ cm}^3/100\text{g}$  [20]. During solidification, the excessive hydrogen precipitates and, being trapped between the solid dendrites, forms porosity. Gas porosity combined with shrinkage porosity is detrimental to the mechanical properties of the final products, especially to the fracture toughness, fatigue endurance and ductility [21]. Moreover, hydrogen that has not had time to precipitate and formed supersaturated solid solution with aluminum will precipitate during downstream processing, e.g. homogenization, extrusion or hot rolling, forming delamination and secondary porosity, especially harmful in thin gauge products or surface critical applications.

Thermodynamic analysis [22] shows that the partial pressure of hydrogen is extremely high even at low pressures of water vapor. At  $727 \text{ }^\circ\text{C}$  and a water vapor pressure of  $1.33 \text{ kPa}$  (typical atmospheric value), the equilibrium partial pressure of hydrogen at the liquid–gas interface reaches a huge value of  $8.87 \times 10^6 \text{ GPa}$ , so the hydrogen content of the melt might be as high as  $3.24 \times 10^5 \text{ cm}^3/100 \text{ g}$ . This means that all available hydrogen can be dissolved in liquid aluminum, and that relatively small atmospheric humidity may lead to high hydrogen concentration in the melt. One cubic meter of air contains about  $10 \text{ g}$  of water, which is equivalent to  $1 \text{ g}$  of hydrogen. This one gram of hydrogen being dissolved in a ton of liquid aluminum may produce about 2–3% porosity [22].

In reality the quasi-equilibrium solubility of hydrogen in liquid aluminum is very dynamic and two processes happen simultaneously, i.e. the atomic hydrogen intake from atmosphere (re-gassing) and molecular hydrogen expel from the melt (de-gassing). This quasi-equilibrium can be shifted if the pressure, temperature, humidity or interface conditions change. The general possibilities for the variation

of hydrogen content in liquid aluminum are illustrated in Fig. 2 [23]. It is important to note that degassing process is usually faster than the re-gassing [12].



**Fig. 2.** Possible scenarios of hydrogen levels in the liquid aluminum and corresponding degassing–re-gassing kinetics (after [23]).

For a given charge of liquid aluminum, hydrogen content can be naturally reduced to 0.1–0.2 cm<sup>3</sup>/100 g giving time (up to 1 h) and typical conditions (750 °C, 30% humidity) [20, 22]. However, natural degassing takes long time and is impractical for industrial applications, so forced degassing is a common industrial practice. In most experiments and in industrial practice the degassing process is performed until the desirable concentration of hydrogen (usually about 0.1 cm<sup>3</sup>/100 g) is achieved. After that the melt is cast. It is known that in degassing large volumes, some time is required to finalize the process of degassing by allowing the bubbles to float to the surface. What is much less studied is the process of re-gassing, or what could happen to the degassed melt after the end of the degassing process. The re-gassing is seldom reported but there are some data showing that it is not an unusual phenomenon. Experimentally re-gassing was observed after the end of rotary Ar-assisted degassing [24, 25] and after ultrasonic degassing [26].

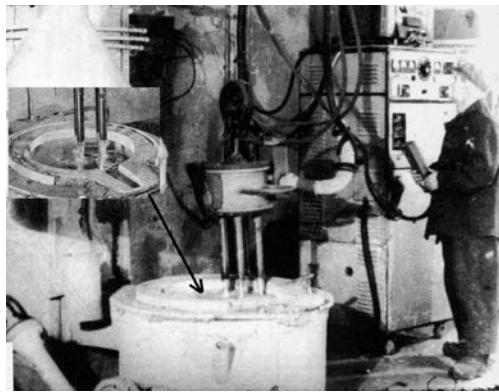
The actual gas solubility in the liquid phase under conditions of cavitation can be lower than the quasi-equilibrium solubility. There exists a limit until which the gas can be extracted from the liquid phase by cavitating bubbles. This limit was estimated to be about 50% of the quasi-equilibrium gas solubility under given environmental conditions. This was first established for degassing water from oxygen [11] and then confirmed for degassing aluminum from hydrogen [21]. This level can be called the cavitation quasi-equilibrium level (see Figure 2).

## Ultrasonic degassing of liquid aluminum

It is well established that the intensity of ultrasonic processing (acoustic power related to the frequency and amplitude of vibrations), time of processing (allowing for the bubbles to form and float), and other conditions such as the alloy composition and cleanliness, melt temperature, air humidity (determining the solubility of hydrogen) are the main process parameters [17, 23, 27, 28, 29].

Let us look at the industrial implementations of ultrasonic melt degassing and the possible ways to tackle the challenges of treating larger melt volumes.

The ultrasonic degassing of aluminum was implemented, already in the 1960s, in foundries for precision investment, sand, die and high-pressure die casting [13]. A special ultrasonic degassing system UZD-200 has been developed in 1959 for degassing up to 250 kg of melt in a ladle (Fig. 3). The installation (in stationary and mobile versions) consisted of a 10 kW generator that fed 4 magnetostrictive transducers working in a sequence with a time gap of 15–20 s. The frequency was 19.5 kHz and the total acoustic power – 1.6 kW. The system was equipped with a time relay that allowed for programmed degassing schedule. The waveguiding system was initially made of steel extension and Ti sonotrode. Later Ti was substituted for a Nb alloy. Table 1 summarizes the results for castings of an Al–Si–Mg alloy in comparison with other degassing methods. It can be easily seen that, ultrasonic degassing significantly increases the density of cast metal and makes it possible to obtain almost pore-free castings (rank 1 in the porosity scale).



**Fig. 3.** Industrial implementation of ultrasonic degassing of casting alloys in the 1960s (courtesy of G. Eskin).

The requirement for processing of large, industrial-scale volumes of melt, especially in large foundries and continuous casting plants shows a limit for batch degassing operations. Another approach needs to be used and the processing of the melt flow seems like a logical and viable possibility. In large melting/casting operations, it is more appropriate to relocate the cleaning of melts from gaseous

and oxide inclusions from the melting or holding furnace/ladle to the zone of metal transfer, somewhere en route from the furnace to the mold. One of the examples of degassing in the melt flow is the combination with vacuum degassing (Fig. 10.3 in [21]). In this scheme vacuum is used in a syphon to transport the melt from the holding furnace to a ladle or mold with the ultrasonic degassing administered in the top part of the syphon. Recently there was a suggestion to combine the ultrasonic degassing with Ar lancing in a vessel through which the melt is constantly flowing [30]. As was already mentioned an Ultra D Degasser by Southwire combines the gas lancing through the sonotrode with ultrasonic vibrations and can be used in both batch and continuous flow operations.

**Table 1.** Comparison of various degassing methods for degassing 250 kg of an Al–Si–Mg alloy [13].

Degassing method	H <sub>2</sub> content, cm <sup>3</sup> /100 g	Density, g/cm <sup>3</sup>	Porosity number	Tensile properties	
				UTS, MPa	El, %
Starting melt	0.35	2.660	4	200	3.8
Ultrasonic degassing	0.17	2.706	1–2	245	5.1
Vacuum degassing	0.2	2.681	1–2	228	4.2
Argon lancing	0.26	2.667	2–3	233	4.0

First industrial trials on ultrasonic degassing in melt flow were performed in USSR in the early 1970s during DC casting of aluminum alloys using a setup similar to that described above for the batch ultrasonic degassing (UZD-200) [13, 21]. The difference was in the arrangement of sonotrodes in line (Fig. 10.4 in [21]). With taking into account that DC casting involves high flow rates and relatively low melt temperatures, an approach of multiple sources was used. A launder had a section of ultrasonic degassing and a section of gas release. Industrial trials were performed for various aluminum alloys cast as flat ingots and round billets [21]. The melt flow varied between 70 and 100 kg/min. The amplitude of the sonotrodes was between 10 and 20  $\mu\text{m}$  and the number of sonotrodes varied between 5 and 11. The results demonstrated that the ultrasonic degassing in the melt flow allowed for 1.5–2 times decrease in hydrogen concentration in the melt. The density measurement of the ingots and billets showed that the density was increased by 15% that corresponded to the 1.5–2

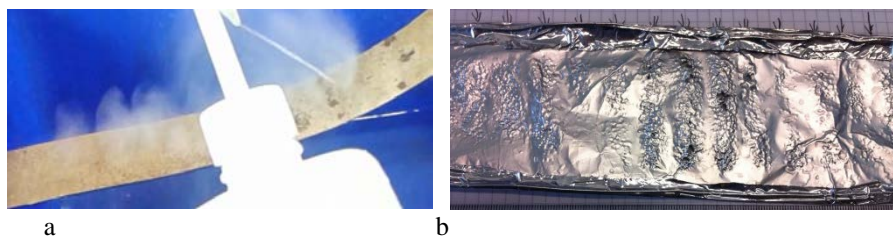
times decreased porosity. The amount of defects (porosity, nonmetallic inclusions) decreased by a factor of 5–8. The mechanical properties were also improved.

The efficiency of this process shows a distinct dependence on the metal flow rate and acoustic power (or the number of sources) conveyed to the melt [21].

In recent years some interesting approaches have been suggested and tested for upscaling if ultrasonic degassing to industrially relevant volumes of melt.

Southwire developed a setup where hollow ceramic tubes are used as sonotrodes connected to a air-cooled piezoelectric transducer and a gas supply [9]. System can be easily scaled up by using several transducers in a row. Although advertised as an ultrasonic degassing, it is clearly the case of combined gas lancing with ultrasonic vibrations. In this case the ultrasonic processing acts not as a source of cavitation but rather as a means to disperse the gas bubbles that collect and evacuate dissolved hydrogen from the melt.

Brunel Centre for Advanced Solidification Technology has tested a principally different scheme of ultrasonic degassing with the sonotrode made of a plate and vibrating in a flexural rather than longitudinal mode. In such a case the wavelength is considerably smaller and the amplitude is considerably higher than for longitudinal oscillations. As a result there are several locations where the cavitation occurs along the length of the plate sonotrode (Fig. 4). With a possibility to place the plate closer to the bottom of a launder or a degassing chamber, the effective degassing area is considerably increased. The lab-scale tests demonstrated 50% increased efficiency of degassing in a batch operation [31]. When used in the melt flow, the degassing efficiency was similar to that achieved by a conventional cylindrical sonotrode in a batch operation. The main challenge in the industrial application of this design is the mechanical connection of the plate to the transducer in a reliable and enduring manner.



**Fig. 4.** Plate sonotrode: (a) demonstration of cavitation fountains at regular intervals and (b) cavitation damage on a foil placed above the plate sonotrode in water.

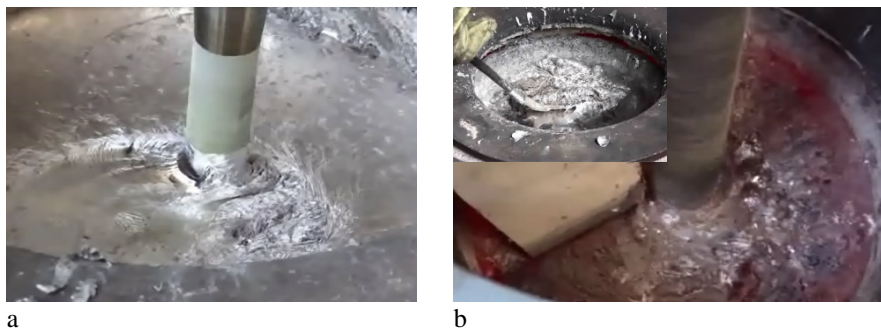
A major effort was done by a consortium of European research and industrial groups on the design and development of ultrasonic degassing systems for foundries [32, 33]. The main challenge was to process considerable melt volumes (up to 500 kg) using only one ultrasonic source and within similar times as a conventional Ar-rotary degasser. As a result a working prototype has been developed, built and tested in two European foundries. The ultrasonic source was



a 5 kW water-cooled magnetostrictive transducer. A combination of a stepped sonotrode made of Nb with its programmed circular movement inside the treated melt volume (facilitated by a robotic arm) resulted in the degassing efficiency (both in terms of the residual hydrogen concentration and the treatment time) similar to that of a conventional Ar-rotary degasser (Fig. 5). The major advantages are more than 5-times reduced (as compared to Ar rotary degassing) amount of dross (Fig. 6) and complete elimination of carrier gas. As degassing requires generation of bubbles and then their flotation to the surface, the circular movement of the cavitation source substitutes for a number of stationary sonotrodes. While the cavitation bubbles are created in one place, the already formed bubbles can still oscillate in the acoustic field (that is not that limited in volume as the cavitation zone) and float to the surface in the wake of the moving sonotrode.



**Fig. 5.** Ultrasonic degassing prototype with a stepped sonotrode and a programmed movement enabled by a robotic arm [33].



**Fig. 6.** A comparison between quiet surface upon ultrasonic degassing (a) and a turbulent melt upon rotary degassing (b) (inset shows dross skimmed from the melt surface after rotary degassing).

These recent examples demonstrate a clear feasibility of ultrasonic degassing for industrial applications. Current efforts are concentrated on understanding the interaction between the melt flow, cavitation field and acoustic streaming via physical and numerical modelling [34].

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### References

- [1] R. W. Boyle, "Cavitation in the Propagation of Sound", *Transactions of Royal Society of Canada*, 3rd Series, 16 (1922) 157–162.
- [2] Ch. Sørensen, "Absorptions-, Geschwindigkeits- und Entgasungsmessungen im Ultraschallgebiet", *Annalen der Physik*, 418 (1936) 121–137.
- [3] F. Krüger, "Über die Entgasung von Glasschmelzen durch Schallwellen", *Glastechnische Berichte*, (7) (1938) 233–236.
- [4] A. Eckardt, E. Eden, "Entgasung optischer Gläser mit Ultraschall", *Glass und Hochvakuumtechnik*, 1(2) (1952) 15–19.
- [5] V. Bojarevics, G.S. Djambazov, and K.A. Pericleous, "Contactless Ultrasound Generation in a Crucible", *Metallurgical and Materials Transactions A*, 46A (2015) 2884–2892.
- [6] W. Esmarch, T. Rommel, and K. Benther, *Werkstoff Sonderheft* (Berlin: W.V. Siemens Werke, 1940) 78.
- [7] G. Bradfield, "Summarized Proceedings of Symposium on Applications of Ultrasonics", *Proceedings of the Physical Society B*, 63 (1950) 305–321.
- [8] H. Eisenreich, *Die Technik*, vol 5 (Berlin: Verlag Technik GmbH, 1950) 310–315.
- [9] V. Rundquist and K. Manchiraju, "Ultrasonic Degassing and Processing of Molten Aluminum" *Light Metals 2013*, ed. B. Sadler (Warrendale, PA: TMS/Wiley, 2013) 949–955.
- [10] S.V. Sergeev, *Physical and Chemical Properties of Liquid Metals*, (Moscow: Oborongiz, 1952) 172.
- [11] O. Lindström, "Physico-chemical aspects of chemically active ultrasonic cavitation in aqueous solutions", *Journal of the Acoustical Society of America*, 27, (1955) 654–671.
- [12] O.A. Kapustina, *Physical Principles of Ultrasonic Technology*, ed. L.D. Rozenberg (Moscow: Nauka, 1970) 253–336 (Translated by New York, Plenum, 1973).
- [13] G.I. Eskin, *Ultrasonic Treatment of Molten Aluminum* (Moscow: Metallurgiya, 1965).
- [14] G.I. Eskin, "Cavitation mechanism of ultrasonic melt degassing", *Ultrasonics Sonochemistry*, 2 (1995) 137–141.
- [15] M.B. Altman et al., "On the impact of elastic oscillations of ultrasonic frequency on the degassing process in aluminum alloys", *Izvestiya Akademii Nauk SSSR, Otdelenie Tekhnicheskikh Nauk*, (9) (1958) 25–30.
- [16] H. Xu, T.T. Meek, and Q. Han, Effects of ultrasonic field and vacuum on degassing of molten aluminum alloys", *Materials Letters*, 61 (2007) 1246–1250.
- [17] H. Xu, Q. Han, and T.T. Meek, "Effect of ultrasonic vibration on degassing of aluminum alloys", *Materials Science and Engineering A*, 473 (2008) 96–104.
- [18] H. Puga et al., New Trends in Aluminium Degassing –A Comparative Study (Paper presented at the Fourth International Conference on Advances and Trends in Engineering

- Materials and their Applications, AES – ATEMA' 2009, Hamburg, Ottawa, AES-Advanced Engineering Solutions, 1-4 September 2009) 101–106.
- [19] G.I. Eskin, *Ultrasonic Degassing of Molten Metal* (Moscow: Mashinostroyeniye, 1968).
- [20] J. Campbell, *Castings*, 2nd revised edition (Oxford, UK: Butterworth-Heinemann Ltd, 1993).
- [21] G.I. Eskin, *Ultrasonic Treatment of Light Alloy Melts* (Amsterdam: Gordon and Breach OPA, 1998).
- [22] V.I. Dobatkin et al., *Gases and Oxides in Wrought Aluminum Alloys* (Moscow: Metallurgiya, 1976) 65–90.
- [23] N. Alba-Baena and D.G. Eskin, “Kinetics of ultrasonic degassing of aluminum alloys”, *Light Metals 2013*, ed. B. Sadler (Warrendale, PA: TMS/Wiley, 2013) 958–962.
- [24] P.D. Waite, “Improved metallurgical understanding of the Ican compact degasser after two years of industrial implementation in aluminum casting plants”, *Light Metals 1998*, ed. B. Welch (Warrendale, PA: TMS, 1998) 791–796.
- [25] L. Zhang, “A Kinetic Study of Hydrogen Absorption and Degassing Behaviour of DURALCAN Composites” (MSc Thesis, University of Quebec at Chicoutimi, 1996).
- [26] D. Eskin et al., “Ultrasonic Degassing of Aluminium Alloys: Basic Studies and Practical Implementation”, *Materials Science and Technology*, 31 (2015) 79–84.
- [27] T.T. Meek, Q. Han, and H. Xu. “Degassing of Aluminum Alloys using Ultrasonic Vibrations”, *Report ORNL/TM-2006/61* (U.S. Department of Energy, 2006).
- [28] H. Puga et al., “The influence of processing parameters on the ultrasonic degassing of molten AlSi9Cu3 aluminium alloy”, *Materials Letters*, 63 (2009) 806–808.
- [29] J. Li et al., “Application of ultrasonic treating to degassing of metal ingots”, *Materials Letters*, 62 (2008) 4152–4154.
- [30] Q. Han, H. Xu, and T.T. Meek, “Degassing of molten alloys with the assistance of ultrasonic vibration”, *US Patent 7682556 B2*, 23.03.2010.
- [31] D.G. Eskin, K. Al-Helal, and I. Tzanakis, “Application of a plate sonotrode to ultrasonic degassing of aluminum melt”, *Journal of Materials Processing Technology*, 222 (2015) 148–154.
- [32] [www.ultragassing.eu](http://www.ultragassing.eu)
- [33] [www.doshormat.eu](http://www.doshormat.eu)
- [34] G.S.B. Lebon et al., “A high-order acoustic cavitation model for the treatment of a moving liquid metal volume”, *CFD Modeling and Simulation in Materials Processing*, ed. L. Nastac et al. (Warrendale, PA: TMS/Wiley, 2016) 135–142.