Cavitation mechanism of ultrasonic melt degassing

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The real melt always contains non-wettable fine inclusions which are potential nuclei for cavitation and degassing. This paper deals with the nature of ultrasonic degassing and the industrial application of a relevant technology.

Keywords: acoustic cavitation; cavitation and degassing nuclei; ultrasonic degassing

The degassing of liquids and low-melting melts under action of ultrasound was among first effects revealed in the $1930s^{1-4}$. We started own investigations on the mechanism of ultrasonic degassing of light alloy melts and began the industrial application of ultrasonicinduced degassing in the $1960s^{3.5}$. These investigations showed that hydrogen could be efficiently removed from Al- and Mg-based melts only when the ultrasonic treatment is accompanied by developing cavitation. It was shown that the ultrasonic degassing of liquid metals differed essentially from that of aqueous solutions and organic liquids. This is due to the different nature of cavitation nuclei and, hence, different conditions required for the origination and development of acoustic cavitation.

In the case of water and organic liquids, the cavitation nuclei are represented by solid inclusions and very fine gaseous bubbles. In contrast, only fine solid particles (mainly oxides, e.g. Al_2O_3 in aluminium melts) can act as cavitation nuclei in metallic melts.

Cavitation and degassing nuclei

Owing to its nature, any metallic melt always contains a suspension of submicroscopic particles that are non-wettable by the melt and containing a gaseous phase in surface defects. This 'plankton' produces potential cavitation nuclei. The proportion of free hydrogen on the surface of these particles is very small, less than 0.1%. Nevertheless, this amount is sufficient to initiate cavitation. The transformation of cavitation bubbles into gaseous bubbles depends on the dynamics of cavitation bubbles and the diffusion-induced penetration of dissolved hydrogen into pulsing cavitation bubbles. Hydrogen bubbles thus formed can coarsen and, on reaching a certain size, float up to the surface of a liquid bath.

Earlier, we have shown^{5,6,8-10} that real melts always contain oxide particles that are non-wettable by the melt and adsorbed hydrogen in surface defects of these particles. Under ultrasonic treatment, the amount of these particles and the content of adsorbed hydrogen become sufficient for simultaneous cavitation and ultrasonicinduced degassing.

Diffusion growth of cavitation bubbles

The diffusion growth of bubbles in an ultrasonic field is one of the most interesting features of acoustic cavitation in metallic melts. In the absence of the acoustic field, the gaseous bubble should slowly dissolve owing to the gas diffusion from the bubble to the melt^{5,6}. The situation changes dramatically when the surface of the bubble pulsates.

Under cavitation ultrasonic treatment, the diffusion of gas occurs in a unique direction, from the liquid to the bubble. This directed diffusion, being superimposed with the usual static gas diffusion from the bubble to the liquid, can greatly improve the gas motion to the bubble, especially when the sound pressure is over the cavitation threshold.

Directed diffusion can be considered on the basis of the following three effects of cavity pulsation:

- 1 The gas diffusion from the bubble to the liquid is directed when the cavity is compressed and the content of gas inside is increased.
- 2 During cavitation pulsation, the surface of the expanded bubble is much higher than that of the compressed bubble. Hence the amount of gas that enters the bubble during its extension is higher than the amount of gas leaving the bubble upon its compression.
- 3 The diffusion is controlled by the thickness of a diffusion layer that is formed in liquid enveloping the bubble. When the bubble is compressed, this layer grows and the concentration gradient decreases. Upon extension, the layer becomes thinner and the

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concentration gradient increases. In this case, the flow-rate of gas to the bubble also increases.

The behaviour of bubbles during cavitation should be considered in terms of the changing amount of gas inside bubbles on their extension and collapse. In the general case, this process can be described using a complex system of equations.

We tried to simplify the problem^{5,6}. Assuming that the bubble pulsates in non-condensable liquid, the time dependence of the bubble radius R(t) can be expressed in terms of the Rayleigh (Noltingk–Neppiras) equation. Hence, the variation in amount of gas inside the bubble or the internal gas pressure P(t) can be given by a non-linear dependence on the radius.

Assuming that the amount of gas inside the bubble is $M = 4\pi R^3 m\gamma/3$ (where γ is the gas density inside the bubble and *m* is the molecular weight of the gas), the variation in the amount of gas is

$$(4/3\pi)d(\gamma R^3)/dt = 4\pi R^2 i(t) \tag{1}$$

where i(t) is the density of gas flow through the bubble surface. If the gas inside the bubble is ideal, then $P = \gamma kT$ (where T is the absolute temperature of the gas in the bubble, which depends on time, and k is the Boltzmann constant).

Assuming that the cavitation occurs under isothermal conditions, Equation (1) becomes

$$(R/3kT)\partial P/\partial t + P\partial R/(\partial tkT) = i(t)$$
⁽²⁾

The gas flow into the bubble can occur by several mechanisms, but diffusion is the major one. The following expression was proposed by Boguslavskii⁷ to describe the density of gas flow into the bubble:

$$i = (8/3)\sqrt{5\pi}Z/\rho D^{1/2}C_0 t^{3/2}$$
(3)

where $Z = 0.8P_s$ (P_s is the amplitude of sound pressure),

 ρ is the density of liquid aluminium, D is the diffusion coefficient of hydrogen in liquid aluminium, C_0 is the content of hydrogen in liquid aluminium and t is the period of bubble pulsation.

Let the surface area of the bubble $S = 4\pi R^2$ be the unknown in the equations of bubble pulsation and gas diffusion into the pulsating bubble. In this case, the system of equations that describes the bubble dynamics and gas diffusion into the pulsating bubble is as follows:

$$\begin{cases} \partial^2 S/\partial t^2 = -(1/4)S(\partial S/\partial t)^2 + 2\pi/\rho [P(t) - P_0 - P_s \sin \omega t] \\ -4\sigma \pi^{3/2}/S^{1/2} - 4\mu \dot{S}/S \\ 3S^{3/2}/\sqrt{\pi}kT(\partial P/\partial t) + PS^{1/2}/2\sqrt{\pi}kT(\partial S/\partial t) = i \end{cases}$$
(4)

where ρ , σ and μ are the density, surface tension and viscosity of the aluminium melt.

This system of equations was solved using numerical methods and computer software. $S(0) = S_0$, $P(0) = P_0$ and $\dot{S}(0) = 0$ were used as the initial conditions at $C_0 = 0.2$ cm³ per 100 g and D = 1 cm² s⁻¹ for various starting radii of the cavitation bubble R_0 . Bubbles 1–10 μ m in radius were examined. These values reflect real dimensions of cavitation nuclei (non-wettable particles of alumina).

Figure 1 shows the variations in the relative radius R/R_0 and in the hydrogen pressure inside the pulsating bubble P_g (bubble radius $R_0 = 10 \ \mu$ m) at three applied pressures P_s : 0.2 MPa (no cavitation occurs), 1.0 MPa (the cavitation threshold) and 10 MPa (developed cavitation). Note that the flow of hydrogen into the bubble increases with cavitation development and sound pressure build-up. The bubble pulsation before cavitation changes the gas pressure only slightly (Figure 1a). At the cavitation threshold (Figure 1b), the hydrogen pressure inside the bubble changes by three orders of magnitude. Finally, the diffusion of hydrogen under cavitation pulsation



Figure 1 Dynamics of cavitation bubbles in aluminium melt during ultrasonic treatment of various intensity. Initial bubble radius $R_0 = 10 \ \mu m$; sound pressure $P_s =$ (a) 0.2, (b) 1.0 and (c) 10 MPa. Calculations were performed (1) taking account of hydrogen diffusion into the bubble and (2) without it

increases the gas pressure by almost six orders of magnitude.

We can conclude that the ultrasonic treatment changes dramatically the behaviour of fine hydrogen bubbles in liquid aluminium. Without applied ultrasound, these bubbles dissolve owing to the diffusion flow of hydrogen out of the bubbles to the melt^{5,6}. However, the alternating sound pressure applied to the melt along with the cavitation facilities the growth of gaseous bubbles. The diffusion-induced growth of bubbles depends on the intensity of ultrasound treatment, in other words, on the magnitude of sound pressure, gas content and other parameters of the liquid metal.

Ultrasonic degassing of light alloy melts

Experimental sstudies of cavitation development in light alloy melts and the effect of cavitation on the kinetics of hydrogen removal during ultrasonic treatment confirm the essential connection between cavitation and degassing.

Figure 2 demonstrates the effect of ultrasound on commercial aluminium melt over a wide range of sound intensity. Three typical regions occur in this dependence. The formation of these regions can be considered in terms of cavitation development. Region I reflects the precavitation treatment; only slight degassing can be observed. Within region II, the degassing increases sharply and then stabilizes; this is the region of developing cavitation. Region III is the region of optimum treatment modes with active cavitation; the degassing efficiency increases linearly with increasing ultrasound intensity. In other words, the transition to cavitation modes provides for the efficient removal of hydrogen from the melt.

Figure 3 shows kinetic curves of ultrasonic degassing. One can compare the efficiency of ultrasonic treatment (1-2 kg charge of a liquid Al-Si-Mg alloy) with other known techniques of degassing. Note that the ultrasonic treatment (2) is more appropriate than vacuum treatment (3). It is noteworthy that mutual vacuum and ultrasonic treatment (4) considerably facilitates hydrogen removal from the melt.



Figure 2 Variation of the efficiency of ultrasonic degassing of (1) commercial A7 grade aluminium and (2) an industrial AI–6% Mg alloy with the intensity of ultrasonic treatment (oscillation amplitude of ultrasound source, A): I, no cavitation occurs; II, cavitation threshold; and III, developed cavitation



Figure 3 Kinetics of hydrogen removal from an industrial Al–Si–Mg alloy during (1) processing with chlorine salts, (2) ultrasonic treatment, (3) vacuum treatment and (4) combined ultrasonic and vacuum treatment

We can distinguish the following regularities of degassing occurring under developed cavitation:

- 1 The nucleation of hydrogen bubbles occurs on the surface of non-wettable oxide particles in the sites of gas adsorption. This becomes possible only at sound pressures above the cavitation threshold.
- 2 Hydrogen bubbles grow owing to the directed diffusion of gas to the bubbles. The growth rate depends on the size of the initial nucleus, the starting content of hydrogen in the melt, the amplitude of sound pressure (i.e. ultrasound intensity and, hence, cavitation development) and the period of ultrasonic treatment.
- 3 Individual pulsating bubbles coagulate to form coarse macrobubbles owing to the action of the Bjerknes force and the development of acoustic microflows that are formed in the vicinity of pulsating bubbles.
- 4 The ultrasonic degassing results in the floating up of coarse hydrogen bubbles to the surface of the liquid bath. This process occurs due to the action of Stokes forces and with the aid of acoustic flows.

It should be noted that the bubble-aided hydrogen removal is accompanied by oxide flotation. In this case, the cavitation generates and develops on the non-wettable surface of these oxide particles. This effect can be easily observed when studying the fluidity of an Al–Si–Mg alloy melt⁵, the fluidity (length of spiral) being 500 mm without treatment, 550 mm with argon blasting and 670 mm with ultrasonic degassing.

Efficiency of ultrasonic degassing

The efficiency of ultrasonic-induced hydrogen removal from light alloy metals was thoroughly examined under industrial conditions. We also compared the ultrasonic degassing with other known techniques. *Table 1* shows the results obtained during different treatments of a liquid foundry Al–Si–Mg alloy. The highest efficiency achieved with ultrasonic degassing can be easily seen.

Industrial experiments performed during continuous casting of an Al-6% Mg alloy (ingot section 300×1700 mm; ultrasonic treatment of melt flow) showed that the cavitation treatment halves the content of hydrogen, from 0.6 to 0.3 cm³ per 100 g (*Figure 4*). Similar results

Table 1 Comparative efficiencies of industrial degassing techniques (150-300 kg charge; Al-Si-Mg alloy)

Technique	H ₂ (cm ³ per 100 g)	Density (10 ³ kg m ⁻³)	Porosity number	UTS (MPa)	El (%)
Ultrasound	0.17	2.706	1–2	245	5.1
Vacuum	0.20	2.681	1–2	228	4.2
Argon blasting	0.26	2.667	2–3	233	4.0
Hexachloroethane	0.30	2.663	2–3	212	4.5
Flux	0.26	2.660	3–4	235	4.0
_	0.35	2.660	4	200	3.8



Figure 4 Efficiency of ultrasonic degassing performed in a melt flow of an industrial Al–6% Mg alloy (flat ingots 300×1700 mm in section and 6 m long). Ultrasound power: (1) 9 and (2) 11 kW



Figure 5 Distribution of hydrogen across the section of a flat $(550 \times 165 \text{ mm})$ ingot of an industrial Mg–Al–Zn alloy: (1) without ultrasonic treatment and (2) after ultrasonic treatment (shaded bars)

 Table 2
 Effect of ultrasonic degassing performed in a melt flow on the content of hydrogen and quality of 10 mm hot-rolled sheets of an Al–6% Mg alloy

	Techniques of degassing			
Property	Conventional	Ultrasonic		
H ₂ (cm ³ per 100 g)				
Melt	>0.60	0.30-0.33		
Ingot	0.33-0.37	0.20-0.25		
Sheet	0.30-0.34	0.18-0.22		
Density number	2–3	1		
Ultimate tensile strength (MPa):				
As-rolled	193–235	239247		
Annealed at high temperature	69-80	170-206		
Fatique endurance (×10 ⁵				
cycles to fracture) ($\sigma_{max} =$		ĺ		
160 MPa)	0.97-2.61	1.96-6.45		
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were obtained for magnesium alloys. *Figure 5* shows the distribution of hydrogen across the section of a flat Mg-Al-Zn alloy before and after ultrasonic treatment. The cleaning of the melt to remove gaseous and solid impurities during ultrasonic treatment improves considerably the quality of as-cast and deformed metal.

Table 2 shows the effect of ultrasonic degassing on the content of hydrogen and the mechanical properties of an industrial Al-6% Mg alloy.

In recent years, we have developed a technique for combined cleaning of melt to remove both hydrogen and fine oxides (Usfirals process)¹¹. This technique is based on a sonocapillary effect and capturing of solid inclusions in capillary channels of multilayer screen filters.

Conclusions

The thorough investigation of ultrasonic melt degassing allowed us to establish an essential relationship between hydrogen removal and cavitation development. The developed cavitation increases the efficiency of ultrasonic degassing by 30-60%. Under optimum conditions, the ultrasonic degassing permits the hydrogen content in ingots and castings to be more than halved, improves the density and plasticity of as-cast metal with retained strength and increases the service reliability of deformed semifinished items.

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