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The Effect of Ultrasonic Treatment on Microstructural and Mechanical Properties of Cast Magnesium Alloys

Yeong-Jern Chen[1,*1], Wen-Nong Hsu[2,*2] and Jhih-Ren Shih[2,*2]

[1]*Department of Mechanical Engineering, De Lin Institute of Technology, Tu-Cheng, Taiwan 236, R. O. China*
[2]*Department of Mechanical Engineering, National Central University, Chung-Li, Taiwan 32054, R. O. China*

Magnesium alloys are important light metals. In recent years, they have been widely applied in the aerospace and automotive industries, and in the manufacture of communication devices, consumer-electronics appliances and computer products. However, cast magnesium and magnesium alloys are subject to problems due to gas pores, inclusion particles, oxide films, and so on. How to reduce the harm caused by these defects and refine the structure to improve casting quality has become an important topic. In this study, we evaluate the effect on casting quality of an ultrasonic method for treating the melt. The method is based on the generation of cavitation bubbles from ultrasonic treatment of the melt, which induces dispersion and degassing action. Analysis of the microstructure and determination of the mechanical properties of the resultant castings are the basis for identifying the quality of the magnesium and magnesium alloys. The microstructure was evaluated using an optical microscope and scanning electron microscopy (SEM). The elemental constituents of the inclusion particles and oxide films were identified using scanning electron microscopy in conjunction with an X-ray energy dispersive spectrometer and electron probe microanalyzer (EPMA). Finally, the mechanical properties of the magnesium and magnesium alloys, including the tensile strength, the elongation and the hardness, were also determined and discussed. In addition, variations in mechanical properties of cast aluminum and magnesium alloys by ultrasonic treatment are also discussed. [doi:10.2320/matertrans.MER2008273]

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Keywords: *ultrasonic treatment, cast magnesium alloys, cavitation bubbles, oxide films, inclusions*

1. Introduction

Magnesium alloys are important light metals that have a high specific strength and excellent machining and radiation characteristics. They have therefore been widely used in the aerospace and automotive industries and in the manufacture of communications devices, consumer-electronics appliances and computer products in recent years. Their future application potential should not be neglected. Most magnesium alloy products are produced using die casting methods. However, gas pores, oxide films and inclusion particles can exist in thin shell castings, seriously impairing the casting quality. Therefore, how to reduce these defects and thus improve the quality of magnesium alloy castings is an engineering problem that is in urgent need of a solution. Nowadays degassing and fluxing treatments are used during the melting process to reduce casting defects in magnesium alloys. However, the results achieved have not yet been ideal. In this study, we propose ultrasonic treatment of the melts and evaluate the effects of this treatment method on the casting quality. The ultrasonic treatment method is principally based upon the behavior of cavitation bubbles caused by the ultrasound in the liquid. A piezoelectric transducer stimulates the propagation of an ultrasound through the liquid. This ultrasound generates cyclic alternating acoustic pressures of compression and tension. If the acoustic pressure is high enough, the liquid will be subjected to tensile force during the negative cycle of ultrasound, and the molecules of the liquid can be pulled away from one another to form cavitation bubbles.[1–5] Sequentially, during the next compressive cycle, the cavitation bubbles violently collapse forming micro-jets and shock waves. The bubbles cluster to form streaming patterns within the acoustic field. Some of these cavitation bubbles grow rapidly under the influence of the alternating acoustic pressure and rectified diffusion of dissolved gas (almost always hydrogen) from the liquid to the cavitation bubbles. These large bubbles coagulate and float to the surface of the liquid.[6] This is called ultrasonic degassing. In addition, the streaming patterns and shock waves induce the dispersion of the ultrasound.[4,7] The magnitude of the acoustic pressure is the principal factor determining whether the ultrasound can produce cavitation bubbles in the liquid. For a pure liquid, the acoustic pressure required to destroy the tensile strength of the liquid is able to produce cavitation bubbles in the liquid. Impurities in the liquid not only reduce the liquid's tensile strength, but also provide surfaces and crevices that act as nucleation sites for the formation of cavitation bubbles, thus making the nucleation of cavitation bubbles easier.[5,8–10]

According to Eskin,[11,12] studies of the ultrasonic treatment of aluminum alloy melts have demonstrated that tiny impurities exist in aluminum alloy melts. In aluminum alloys, these impurities are mainly oxides, such as alumina and spinel. Cracks or crevices can normally be found on the surfaces of these impurities. These defects become sites for adsorbing hydrogen in the aluminum melt. In general casting work, this hydrogen will diffuse away in air bubbles and slowly dissolve in the aluminum melt. The aluminum melt solidifies gas pores are formed. When the aluminum melt is treated with ultrasonic vibration, the volume fraction of hydrogen on the surface of an impurity particle is very small (less than 0.01%), but acoustic pressure is sufficient for developing cavitation bubbles on the surfaces of the impurity particles. These bubbles then become the nuclei for degassing and solidification in the melt, which can be used to refine the microstructure, reduce segregation, and improve secondary phase formation and distribution.[12]

[*1] Corresponding author, E-mail: jern@dlit.edu.tw

[*2] Graduate student, National Central University

Table 1 Chemical composition of magnesium and magnesium alloys.

Materials	Element (mass%)											
	Al	Zn	Mn	Cu	Ni	Fe	Si	Be	Zr	Ca	Pb	Mg
Mg	0.0019		0.0130	0.0005	0.0004	0.0008						99.99
AM60	6.0132	0.0113	0.3568	0.0016	0.0007	0.0022	0.0251	0.0006	0.0012	0.0001	0.0004	93.5868
AZ91	8.9069	0.6136	0.3047	0.0017	0.0006	0.0018	0.0302	0.0009	0.0007			90.1397

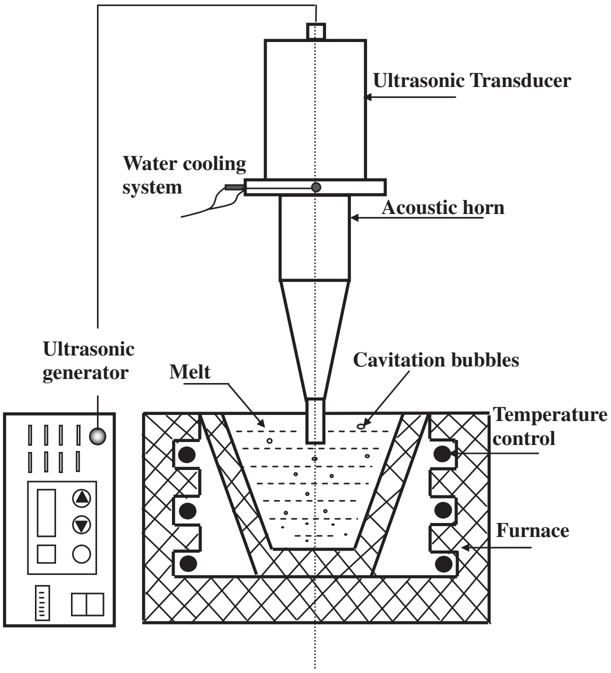


Fig. 1 Experimental setup for ultrasonic treatment of magnesium alloy melts.

The efficacy of ultrasonic treatment of aluminum alloy melts to improve their quality has been confirmed worldwide.[11–15] Although different casting processes are used for magnesium alloys and aluminum alloys and the solidification characteristics are not the same, approximately the same acoustic cavitation mechanism occurs in the melts. Therefore, we propose using an ultrasonic treatment method for treating these melts and evaluate the effects of this treatment on the casting quality. We can predict that the processing of molten magnesium and their alloys by ultrasonic treatment methods can be used to improve the casting process in the future.

2. Experimental Procedures

This study seeks to investigate the influence of ultrasonic treatment on the quality of cast magnesium and magnesium alloys. We control the acoustic frequency, acoustic power and the treatment time to analyze the variations of the mechanical properties and the microstructure of cast magnesium and magnesium alloys. The experimental setup is shown in Fig. 1. The ultrasonic system consisted of a 1.0 kW acoustic generator and two water-cooling transducers made of piezoelectric lead zirconate (PZT) operating at frequencies of 20 kHz and 15 kHz. The setup included an ultrasonic radiator made of titanium alloy (Ti-6Al-4V), 20 mm in diameter and 245 mm in length. Three different materials were tested, commercial magnesium, and AZ91 and AM60 magnesium alloys. The chemical compositions of the alloys are listed in Table 1. Magnesium and magnesium alloy ingots were melted in an electric resist furnace in conjunction with a stainless steel crucible and using protective gases of dried air mixed with SF_6 and CO_2. The dimension of the crucible is 130 mm in diameter and 200 mm in depth, and 3 kg of magnesium alloys for each melting experiment. The molten metals were heated to 650°C then held at this temperature for 10 minutes prior to the ultrasonic treatment. The preheated temperature of an ultrasonic radiator about 150°C was inserted into the melt 15 mm below the surface of the liquid metal. Ultrasonic treatment was then applied in the melts for specified periods of time, 180 s, 300 s and 600 s, respectively. The molten metals were poured into chilled copper molds to obtain chilled samples 40 mm in diameter and JIS-5202 boat-type tested bar. These specimens were used for analysis of the microstructure and mechanical properties.

Ultrasonic-vibration treatment was used to diagnose the oxide films entrapped in the sample.[16–18] A polished specimen was set on the bottom of the vessel. The vessel was filled with distilled water to a volume of 500 to 600 ml. A short 10 s ultrasonic-vibration treatment was applied at 46 kHz. White foggy marks with differently shaped lumps, flakes, scripts or spots gradually appeared on the shiny surface of the specimen as the treatment time increased. The foggy marks could be used to identify the area of oxide film erosion on the samples. The mechanical properties of the samples including hardness, tensile strength and elongation, were determined with a digital Vickers tester and a universal testing machine, respectively. The microstructure was observed using a microscope and scanning electron microscopy (SEM). Specimens were etched with a solution of acetic picral, 5 ml acetic acid, 6 g picric acid, 10 ml water and 100 ml ethanol. Image analysis software was employed to determine the grain sizes and the counts of inclusion particles. The elemental constituents of the inclusion particles and oxide films were identified by scanning electron microscopy in conjunction X-ray energy dispersive spectrometry (EDS) and an electron probe microanalyzer (EPMA).

3. Results and Discussion

3.1 Mechanisms for ultrasonic treatment of the melts

The nucleation of a cavitation bubble is mainly dependent on the acoustic pressure of the ultrasound and the surface tension or tensile strength of the melt. For nucleating a homogeneous cavitation bubble, the acoustic pressure should be high enough to overcome the ambient pressure and the surface tension of the melt. In addition, the melt always contains some tiny particles such as inclusions, impurities and intermetallic compounds, which reduce the tensile stress of the metals and can provide heterogeneous nucleation sites for the formation of cavitation bubbles. The cavitation bubbles provide nuclei for hydrogen bubbles to coalesce and flow out of the melt induced by degassing. Figure 2 illustrates the nucleation type of ultrasonic treatment of cast magnesium alloy melts. For homogeneous nucleation, the cavitation bubbles absorb heat energy from the melt, resulting in undercooling on the bubble surface, which promotes the formation of tiny atom clusters that form the solidification nuclei, as illustrated in Fig. 2(a).[19] For heterogeneous nucleation during ultrasonic treatment of the melt, the crevices or cracks on particle surfaces are filled with the melt, thus ensuring the transformation of these nonwettable particles to active solidification nuclei, as shown in Fig. 2(b). The formed nuclei can be dispersed homogeneously in melts by ultrasonic treatment. Thus, the microstructure and grain sizes of magnesium and magnesium alloys can be refined by ultrasonic treatment.

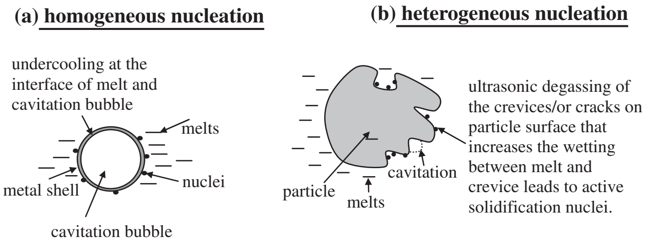
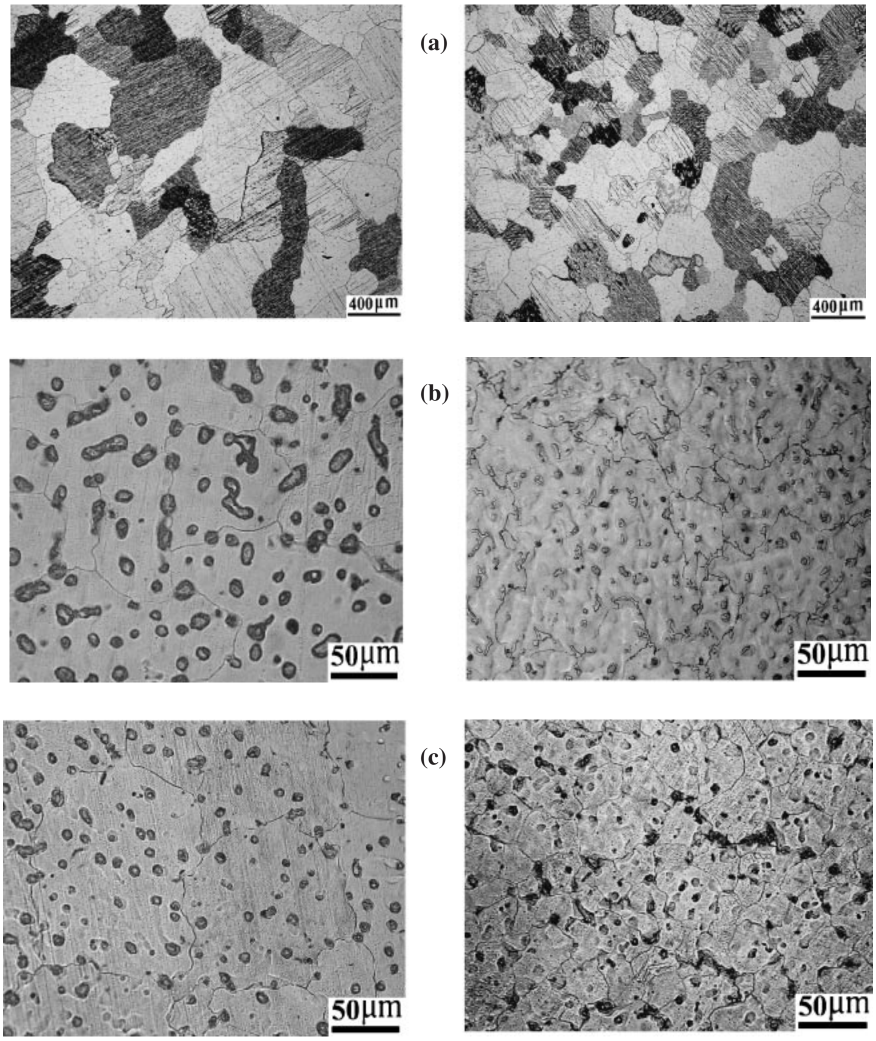


Fig. 2 Nucleation of solidification nuclei in the melts: (a) homogeneous nucleation; (b) heterogeneous nucleation.

3.2 Effects of ultrasonic treatment on the microstructure of cast magnesium and magnesium alloys

The use of ultrasonic treatment will enhance degassing action due to the development of acoustic cavitation bubbles in a molten metal, and will also accelerate nucleation and the formation of an excess amount of solidification sites ahead of the solidification front, which promotes the microstructural refinement. Figure 3 compares the microstructure of cast magnesium and magnesium alloys with and



Without ultrasonic treatment

With ultrasonic treatment at a frequency of 20 kHz for 180s

Fig. 3 Microstructure of magnesium and magnesium alloys: (a) magnesium; (b) AZ91; (c) AM60.

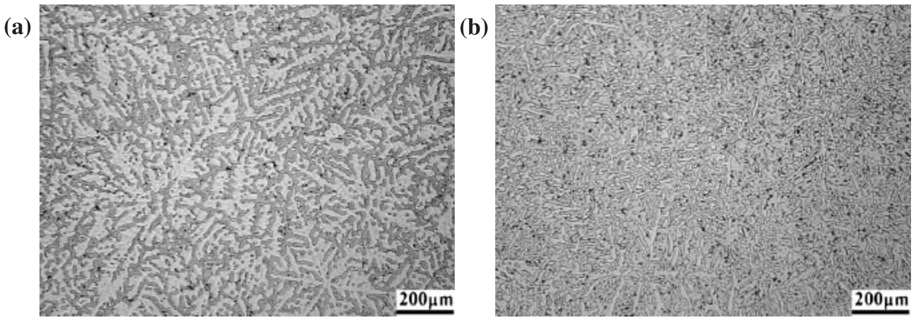


Fig. 4 Microstructure of AZ91with and without ultrasonic treatment of the melts: (a) without; (b) vibrated at a frequency of 20 kHz for 180 s.

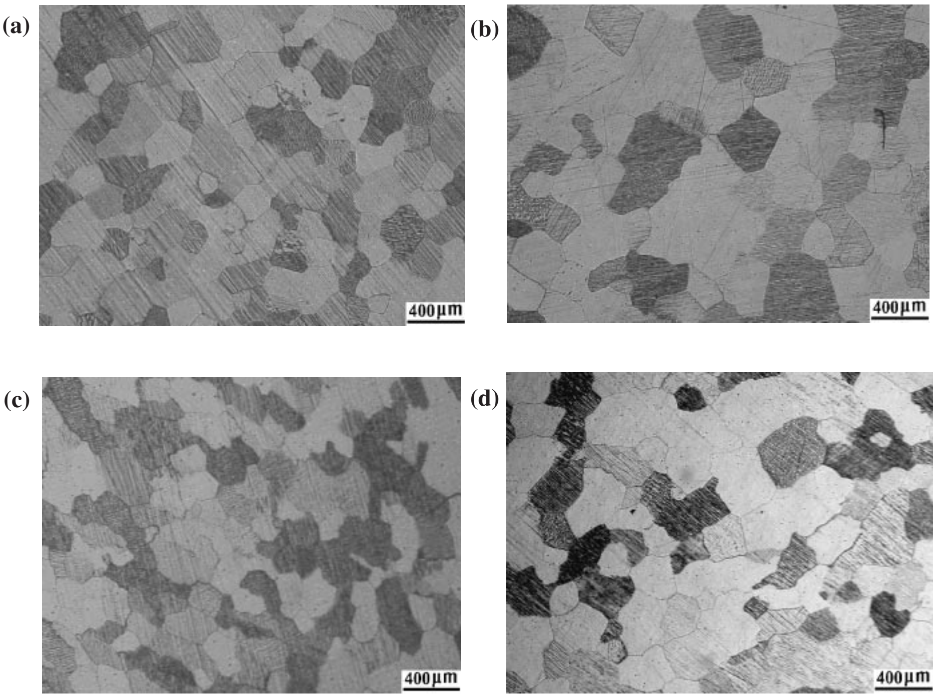


Fig. 5 Microstructure of magnesium with ultrasonic treatment at a given frequency and time: (a) at a frequency of 15 kHz for 300 s; (b) at a frequency of 15 kHz for 300 s, followed by a rest of 600 s; (c) at a frequency of 20 kHz for 300 s; (d) at a frequency of 20 kHz for 300 s, followed by a rest of 600 s.

without the application of ultrasonic treatment at a frequency of 20 kHz. The grain sizes appear to be reduced by the ultrasonic treatment, and hence the microstructure of the alloys without ultrasonic treatment exhibit larger grain sizes. Ultrasonic treatment disperses the particles in the magnesium melts. In this condition, it is difficult to grow the eutectic phase of $Mg_{17}Al_{12}$ due to interference by these particles. Therefore, they appear tiny in $Mg_{17}Al_{12}$ after ultrasonic treatment (Fig. 4). When the frequency is increased from 15 to 20 kHz, the period needed for the growth of cavitation bubbles in magnesium melts becomes shorter. Small cavitation bubbles have a stronger degassing ability for fine cavities on tiny particle surfaces. Therefore, they can provide many nucleation sites in magnesium melts leading to the formation of fine grain sizes (Figs. 5(a) and 5(c)). If the magnesium melts are allowed to rest for a period of time after the ultrasonic treatment, prior to the casting work, the grain-refining effect on the castings is gradually lost with increasing settling time. The grain sizes of the castings will increase gradually with the settling time, eventually approaching the original grain size without

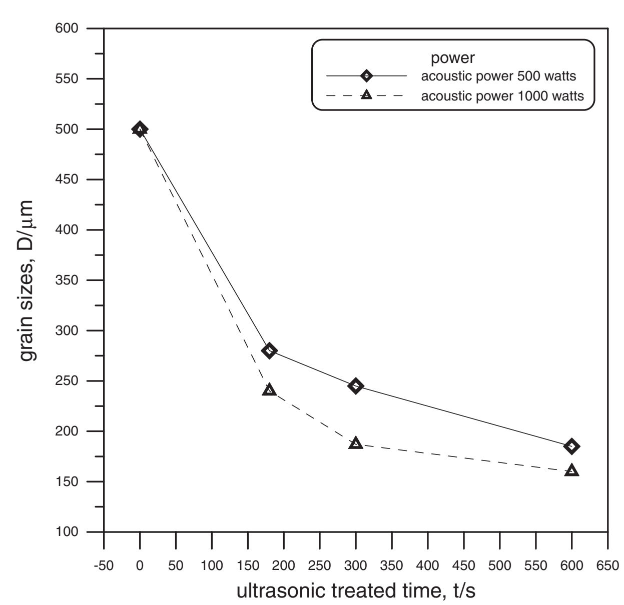


Fig. 6 Influence of acoustic power and treatment time on grain sizes of cast magnesium.

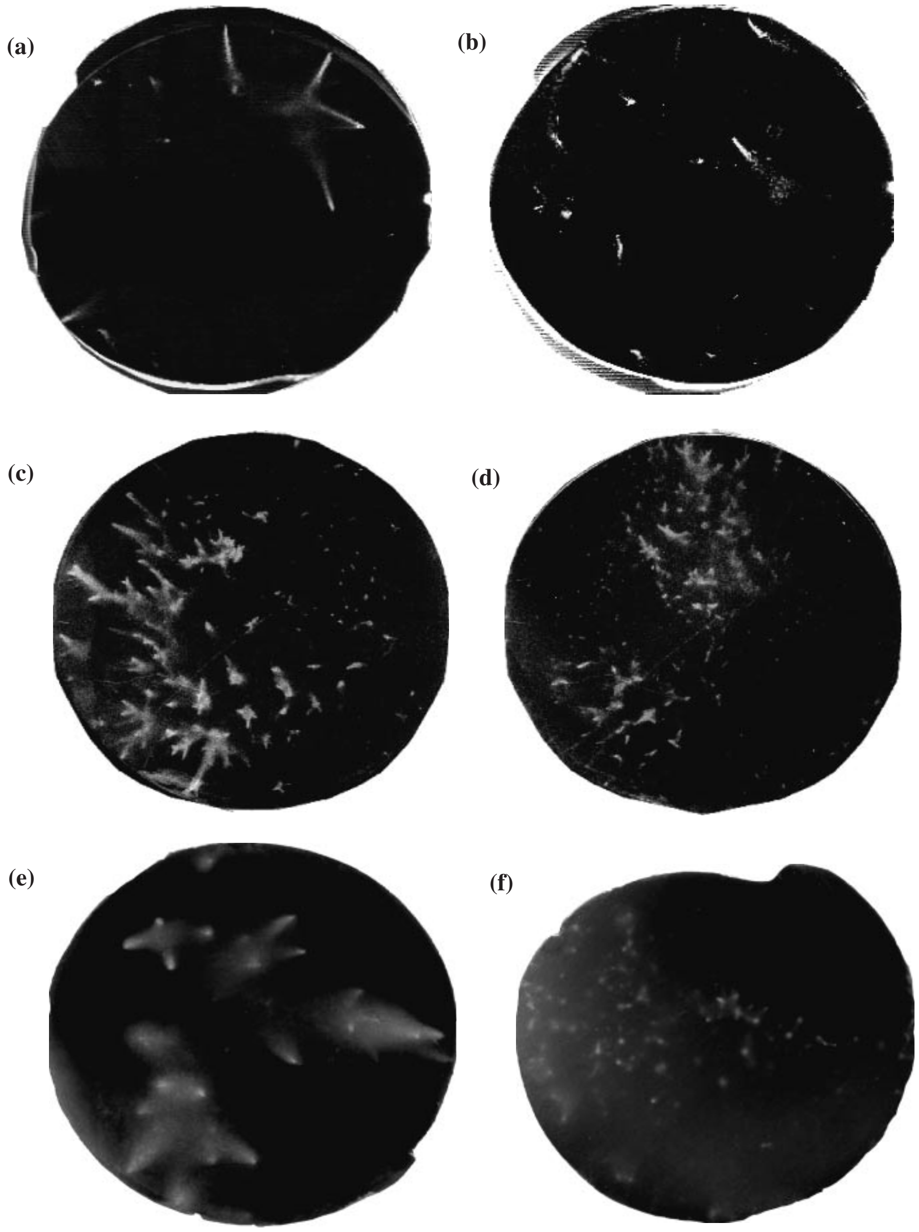


Fig. 7 Foggy marks on the polished sample surface after ultrasonic-vibration treatment for 180 s at 46 kHz: (a) magnesium; (b) magnesium after ultrasonic treatment of the melt for 300 s; (c) AZ91 alloy; (b) AZ91alloy after ultrasonic treatment of the melt for 300 s; (e) AM60 alloy; (f) AM60 alloy after ultrasonic treatment of the melts for 300 s.

treatment (Figs. 5(d) and 3(a)). The displacement amplitude at the end of the vibrating rod is dependent on the power output of the ultrasound; specifically the displacement amplitude increases with increasing power output of the ultrasound. Therefore, a larger displacement amplitude leads to more homogeneously nucleated cavitation bubbles and promotes the dispersion of compound particles in the melts. Ultrasonic treatment was applied to magnesium melts at powers of 1000 watts and 500 watts. As the acoustic power and vibrating time increase, the fineness of the particles changes and the distribution becomes more homogeneous. These particles can serve as grain nucleation sites to induce finer grains of castings (Fig. 6). After ultrasonic treatment for 600 s, the grain sizes produced by the two acoustic powers tend to be similar since the effect of ultrasonic dispersion has reached the maximum limit.

3.3 Effects of ultrasonic treatment on oxide films and inclusions in cast magnesium and magnesium alloys

In the experiments, polished chilled magnesium specimens were immersed in distilled water and then subjected to ultrasonic-vibration treatment for 10 s. Several obvious damage marks or stripes (foggy marks) appeared on the polished surface of the magnesium, AM60 and AZ91 magnesium alloys (Figs. 7(a) to 7(f)). The foggy marks were observed by scanning electronic microscopy, and the elemental constituents of the foggy marks were studied with an electron x-ray probe microanalyzer. The results of the EPMA analysis of the foggy marks on the cast magnesium indicate that the area was rich in oxygen, confirming the existence of oxide films (Figs. 8(a) to 8(c)). The foggy marks appeared on the cast magnesium samples mostly as long stripes. In the AM60 alloy sample, the foggy marks appeared

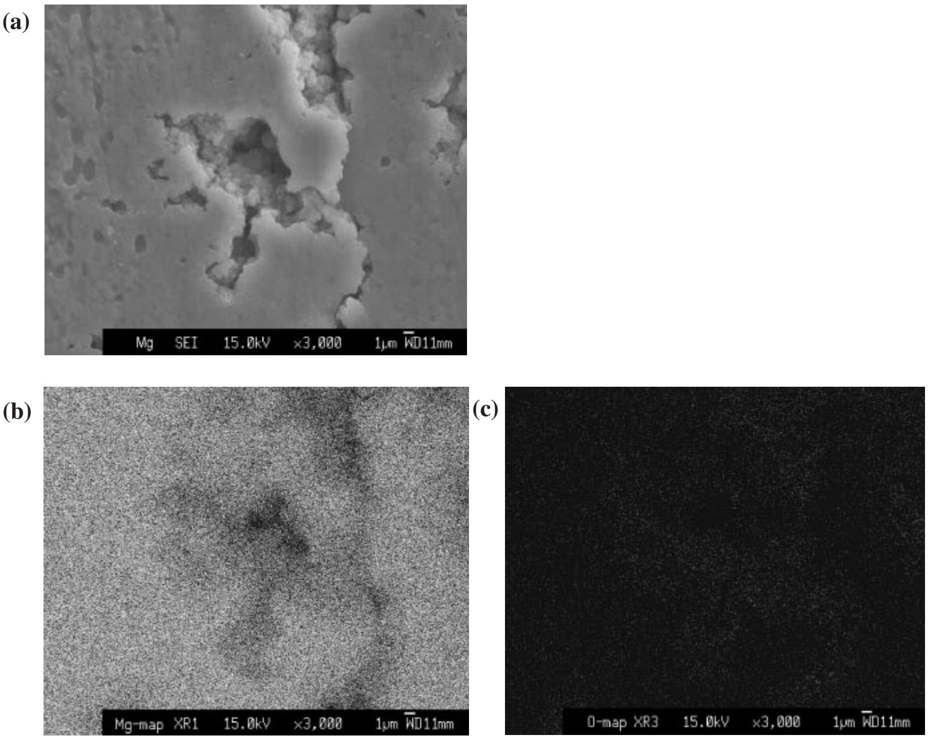


Fig. 8 EPMA analysis of oxide films on cast magnesium: (a) SEM photograph; (b) Mg mapping; (c) O mapping.

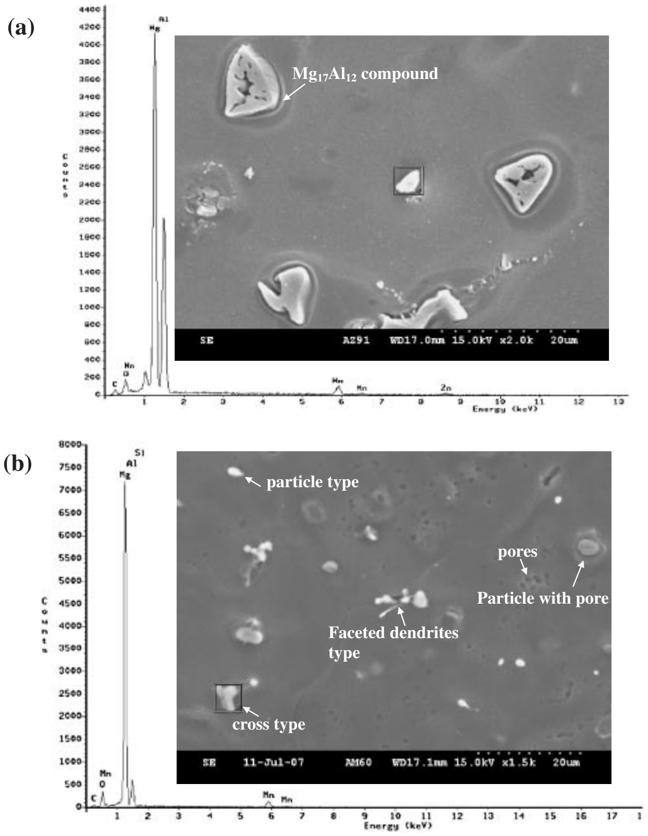


Fig. 9 EDS analysis of inclusion particles in cast magnesium alloys: (a) AZ91; (b) AM60.

as shiny spots or lumps, and in the AZ91 alloy, they appeared as fine, long strips. There was great variation in the area fraction of the foggy marks on the different samples since the oxide film was not evenly distributed.

Cavitation bubbles in the melt can occur due to homogeneous or heterogeneous nucleation during ultrasonic treatment. The cavitation bubbles will grow to form bubble clouds. These bubble clouds then collapse and generate large intense micro-jets and shock waves that impact the oxide film, resulting in their fragmentation. Therefore, the oxide films that appear after ultrasonic treatment of magnesium, AM60 and AZ91 alloy melts are uneven and disrupted in shape (Fig. 7(b), 7(d) and 7(f)). The disrupted oxide film can reduce the impairment of casting quality. In addition, we examined the ultrasonic treatment of magnesium melts. The ultrasound forces dispersion, disrupting large inclusion particles into finer particles more homogeneously dispersed in the magnesium melts. These tiny particles may provide sites for cavitation bubble nucleation and further promote the degassing of the melts. These tiny particles become heterogeneous nucleation sites for grains during the solidification process of the magnesium melt, and tend to further obstruct grain growth, leading to grain-refining of the magnesium alloys. For the AZ91 magnesium alloy, $Mg_{17}Al_{12}$ intermetallic compound appeared as lumps or long stripes. They mainly exist near the grain boundaries, with the minority within the grain. The formation of concave around the particle is mainly due to the different volume shrinkage between $Mg_{17}Al_{12}$ compound particle and magnesium matrix during solidification. EDS analysis of the particles demonstrated that they mainly consist of Al-Mn compound (Fig. 9(a)). For the AM60 magnesium alloy, the EDS analysis of the particles demonstrates that they are also mainly Al-Mn compound which appear in two shapes (Fig. 9(b)). When the cooling rate is high, the cross type is present; when the cooling rate is low, the particle or faceted types appear. For medium cooling rate, three types of particles may have existed (Fig. 9(b)). These results are

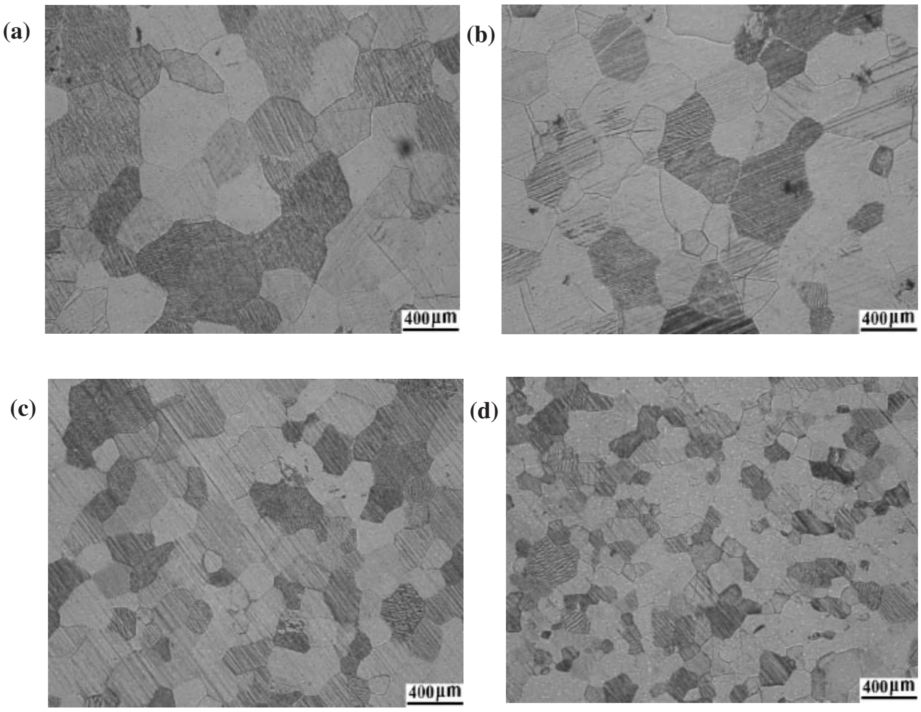


Fig. 10 Microstructure of magnesium: (a) without ultrasonic treatment; (b) without ultrasonic treatment but after adding MgO to the melt; (c) with ultrasonic treatment at 15 kHz for 300 s; (d) with ultrasonic treatment at 15 kHz for 300 s, after adding MgO particles to the melt.

Table 2 Mechanical properties of magnesium and magnesium alloys before/after ultrasonic treatment at a frequency of 20 kHz for 600 s.

Materials	Ultrasonic treatment (1000 watts)	Average grain sizes (μm)	Mechanical properties			Inclusion counts (counts/mm^2)
			Tensile stress (MPa)	Elongation (%)	Hardness (HV)	
Mg	○	175	103	6.9	37	395
	×	500	91	6.3	35	305
AZ91	○	125	156	2.5	80	812
	×	205	170	3.7	72	376
AM60	○	90	175	5.4	67	814
	×	200	183	5.6	54	537

consistent with those of Lun Sin *et al.*20) These particles can act as nucleation sites for grains during solidification, as well as prevent grain growth to achieve grain refinement. Inclusion particles play an important role in the grain refinement of magnesium and magnesium alloys during ultrasonic treatment. In this study, MgO particles of about 5 μm in diameter were added to the melts prior to ultrasonic treatment. The results indicated that there was a refining effect on the magnesium grains after ultrasonic treatment, and grain sizes can achieve about of 100 μm (Fig. 10).

3.4 Effects of ultrasonic treatment on mechanical properties of cast magnesium and magnesium alloys

Variations in grain sizes and mechanical properties of magnesium and magnesium alloys after using ultrasonic treatment are shown in Table 2. Grain-refining effects are achieved in magnesium, AZ91 and AM60 alloys after ultrasonic treatment. The mechanical properties including tensile strength, elongation and hardness of the cast magnesium are also enhanced. For AZ91 and AM60 alloys, Al-Mn particles can serve as nucleation sites for cavitation bubbles, made possible by gas or vapor bubbles. These bubbles form gas pores or accompany particles to form air pockets that become entrapped in the matrix of the alloy during solidification (Figs. 9(b) and 11). Therefore, the tensile strength and elongation of AZ91 and AM60 alloys were reduced by ultrasonic treatment. Elemental zinc also has a high vapor pressure (about 10000 Pa) at the melting point of magnesium alloys, so it can intensify the formation and growth of cavitation bubbles during ultrasonic treatment of the melt. Some bubbles initiated at particle surfaces were captured and remained in the matrix of the alloy during solidification. The resultant larger gas pores and air pockets would seriously reduce the tensile strength and elongation in the cast AZ91 alloy. Inclusion particles become smaller and disrupted after ultrasonic treatment due to the behavior of ultrasonic dispersion, so the particle counts were greater in AZ91 and AM60 alloys than in magnesium after ultrasonic treatment.

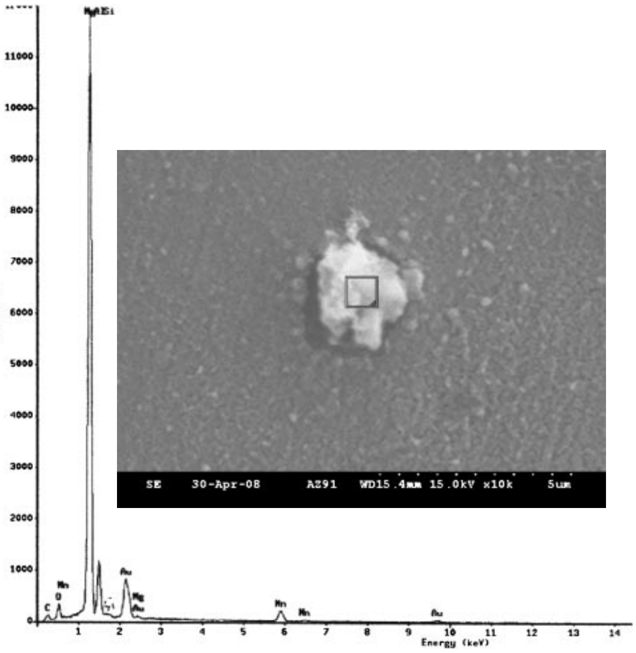


Fig. 11 Al-Mn particle along with pore in cast AZ91 magnesium alloy after ultrasonic treatment.

3.5 Variations in mechanical properties of cast aluminum and magnesium alloys by ultrasonic treatment

Grain-refining effects are achieved in aluminum and magnesium alloys by ultrasonic treatment, but variations of mechanical properties of the two alloys are quite different. Ultrasonic treatment to the melt induces dispersion and degassing action. The effect of cavitation degassing plays an important role for improving the mechanical properties of the alloys. The nucleation of a cavitation bubble mainly depends on the surface tension and the vapor pressure of the melts.[8] Cavitation bubbles can readily be formed in the melts with a smaller surface tension and a larger vapor pressure. The surface tension and vapor pressure are 0.871 N/m and 0.000012 Pa, and 0.577 N/m and 1000 Pa at melting point for aluminum and magnesium, respectively.[21] As a result, magnesium is much easier than aluminum to form vapor bubbles in the melts by ultrasonic treatment. Numerous vapor bubbles like supersaturated gases dissolved in magnesium melts are not completely degassed by ultrasonic treatment. Some of these bubbles can form gas pores or air pockets that become entrapped in the matrix of magnesium alloys during solidification. Therefore, the mechanical properties of magnesium alloys were reduced after ultrasonic treatment. On the other hand, vapor bubbles are not likely to be induced by ultrasonic treatment of aluminum melt. Therefore, ultrasonic treatment is very effective in aluminum melt degassing, which can minimize gas pores and promote the mechanical properties of aluminum alloys.[11–15]

4. Conclusions

Ultrasonic treatment of the melt is a new approach for magnesium and magnesium alloy casting. The fundamental basis is the generation of cavitation bubbles during ultrasonic treatment of the melt which induces dispersion and degassing action. The eutectic phase is refined, oxide films are disrupted and inclusion particles are made smaller and more numerous after the ultrasonic treatment of magnesium and magnesium alloys. Ultrasonic treatment has a grain refining effect on cast magnesium and AZ91 and AM60 magnesium alloys, but only improves the mechanical properties of cast magnesium. The effects of grain refinement increase with increasing acoustic frequency, acoustic power, and treatment time. The results of grain refinement gradually vanish with increased settling time of the melts after ultrasonic treatment. The addition of fine MgO particles can provide nucleation sites for grain formation during solidification, and prevent grain growth, resulting in the refinement of the grains in cast magnesium.

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REFERENCES

1) E. A. Neppiras: Ultrasonics **22** (1984) 25–28.
2) E. Apfel: Ultrasonics **22** (1984) 167–173.
3) E. A. Neppiras: Ultrasonics **18** (1980) 201–209.
4) K. S. Suslick: Sci. Am. **80** (1989) 80–86.
5) A. Crum: Appl. Sci. Res. **38** (1982) 101–105.
6) H. Xu, X. Jian, T. Meek and Q. Han: Mater. Lett. **58** (2004) 3669–3673.
7) T. J. Mason: Phil. Trans. R. Soc. Lond. A **357** (1999) 355–369.
8) E. Apfel: J. Acoust. Soc. Am. **48** (1970) 1179–1186.
9) K. S. Suslick, S. J. Doktycz and E. B. Flint: Ultrasonics **28** (1990) 280–290.
10) B. Pugin: Ultrasonics **25** (1987) 49–55.
11) G. I. Eskin: Ultrason. Sonochem. **1** (1994) 59–63.
12) G. I. Eskin: Ultrason. Sonochem. **2** (1995) 137–141.
13) X. Jian, H. Xu, T. T. Meek and Q. Han: Mater. Lett. **59** (2005) 190–193.
14) J. Campbell: Int. Met. Rev. **2** (1981) 71–108.
15) G. I. Eskin: Ultrason. Sonochem. **8** (2001) 319–325.
16) Y. J. Chen, L. W. Huang and T. S. Shih: Mater. Trans. **44** (2003) 327–335.
17) Y. J. Chen, L. W. Huang and T. S. Shih: Mater. Trans. **44** (2003) 1190–1197.
18) T. S. Shih, L. W. Huang and Y. J. Chen: Int. J. Cast Metals Res. **18** (2005) 301–308.
19) Z. Q. Zhang, Q. C. Le and J. Z. Cui: Mater. Sci. Forum **546–549** (2007) 129–132.
20) S. Lun Sin, D. Dube and R. Tremblay: Mater. Charact. **58** (2007) 989–996.
21) J. J. Jasper: J. Phys. Chem. **1** (1972) 841–1010.