



On the unexpected formation of rare earth hydrides in magnesium–rare earth casting alloys

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There have been reports on the presence of rare earth (RE) hydrides in Mg–RE-based alloys either in the as-cast condition or after high-temperature heat treatment. However, the origin of the RE hydrides remains unclear. In this work, the formation of RE hydrides was investigated using binary Mg–La, Mg–Ce and Mg–Nd alloys. It is proposed that the RE hydrides are probably formed as a result of the decomposition of Mg–RE intermetallic phases by hydrogen.

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One of the potential applications of magnesium alloys in the automobile industry is in powertrain components, where significant weight reduction can be achieved. For powertrain applications, an alloy is required to possess both good die-castability and excellent mechanical properties, especially high-temperature creep resistance. Since rare earth (RE) additions have beneficial effects on the high-temperature mechanical properties of Mg, there has been considerable interest in the past decade in adding RE elements to Mg and its alloys for improved performance [1–4].

A cuboid RE-dominated phase is sometimes encountered in as-cast Mg–RE alloys and is observed more frequently after the alloys are subjected to solution treatment or annealing at high temperatures, as summarized in Table 1. In earlier studies [5–8], the RE-dominated phase was shown to have a face-centred cubic (fcc) structure with a lattice parameter of 0.54–0.56 nm, but its identity was not resolved. Later, the fcc phase was identified to be RE hydrides [9–11], especially with the help of electron energy loss spectroscopy

analysis [10]. Zheng et al. [9] reported the formation of cuboid GdH₂ particles in Mg–(6–11)Gd–2Nd–0.5Zr alloys in both as-cast and high-temperature-homogenized states. A greater volume fraction of larger GdH₂ particles was observed in the near-surface region than in the centre of the sample. Zheng et al. suggested that the GdH₂ particles were formed during melting. They also reported that the GdH₂ particles in the as-cast state tended to grow during the homogenization treatment and attributed this growth to the inward diffusion of hydrogen generated from moisture breakdown at the sample surface. Yang et al. [10] observed an NdH₂ phase in Mg–2.5Nd alloy in both the as-cast state and after solution treatment, and listed three possible mechanisms for the formation of the NdH₂ particles: (i) NdH₂ nucleates during the solidification process and then grows when heat treated; (ii) NdH₂ nucleates and grows during the process of solution treatment; or (iii) NdH₂ nucleates and grows during the solidification process and is surrounded by the eutectic phase, which gradually disappears upon heating. According to Yang et al., NdH₂ probably nucleates during the solidification process as a result of the reaction of Nd with hydrogen previously dissolved in Mg matrix, rather than with water vapour in the furnace atmosphere. Different explanations were given by Peng et al. [11] and Gan et al. [12] for the formation of RE hydrides in Mg alloys containing heavy

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Table 1. Summary of RE hydrides observed in Mg–RE-based alloys.

Alloy	Casting and/or heat treatment	Phase identified	Lattice parameter (nm)	Reference
Mg–8Nd	Rapidly solidified, annealed at 400 °C for 1 h	fcc	0.54	[5]
Mg–(5–15)Gd	Squeeze cast	fcc	0.54	[6]
Mg–4Y–4Sm–0.5Zr	Permanent mould cast, solution treated at 525 °C for 8 h	fcc	0.558	[7]
Mg–10Gd–2Y–0.5Zr	Permanent mould cast, solution treated at 480–530 °C for 8 h	fcc	0.56	[8]
Mg–(6–11)Gd–2Nd–0.5Zr	Permanent mould cast, solution treated at 490–525 °C for 4–6 h	GdH ₂	0.56	[9]
Mg–2.5Nd	Permanent mould cast, solution treated at 540 °C for 6 h	NdH ₂	0.55	[10]
Mg–8Gd, Mg–15Gd	Permanent mould cast	GdH ₂	0.53	[11]
Mg–20Dy	Permanent mould cast, solution treated at 520 °C for 24 h	DyH ₂	0.52	[12]
Mg–8La	Sand cast	LaH _{2–3}	0.56	This work
Mg–8Nd	Permanent mould cast	NdH _{2–3}	0.54	This work
Mg–5Ce	High pressure die cast, annealed at 500 °C for 72 h in air	CeH _{2–3}	0.55	This work
Mg–5La	High pressure die cast, annealed at 500 °C for 72 h in air	LaH _{2–3}	0.56	This work

RE elements, i.e. Gd and Dy. They proposed that the heavy RE hydrides are formed during sample preparation at room temperature when hydrogen-containing sources, such as water, are used. According to Gan et al. [12], Mg first reacts with water to produce hydrogen, then hydrogen reacts with Dy to form DyH₂. Peng et al. [11] considered segregation of Gd and mechanical deformation as two factors that could enhance the formation of GdH₂ from external hydrogen.

The formation of RE hydrides is neither expected nor preferred for Mg–RE alloys. REs are normally present as solutes in the Mg matrix or as Mg–RE intermetallic phases in the eutectic, both of which provide strengthening effects. The formation of RE hydrides is considered to have an adverse effect on mechanical properties, as observed in a range of Mg–RE alloys after a hydrogenation treatment [13]. Therefore, it is necessary to get a better understanding of the origin of RE hydrides in Mg–RE alloys so that measures can be taken to prevent its formation.

In this paper, the formation of RE hydrides was investigated in the three most common binary Mg–RE systems, i.e. Mg–Ce, Mg–La and Mg–Nd alloys. The alloys were produced by various casting technologies to evaluate the influence of solidification rate. The alloys were also annealed at 500 °C in different atmospheres to reveal the influence of the environmental atmosphere. Based on the experimental results, a new explanation for the formation of RE hydrides in Mg–RE casting alloys is proposed.

Binary Mg–RE alloys with nominal compositions of Mg–5Ce, Mg–5La, Mg–5Nd, Mg–8La and Mg–8Nd (wt.%) were used in this study. Three casting technologies were used to produce the alloys: sand casting, permanent mould casting and high-pressure die casting (HPDC). Some samples were subjected to further heat treatments by annealing at 500 °C for 72 h in three different atmospheres: air, pure Ar and pure H₂. Microstructures before and after the heat treatments were examined by optical microscopy and scanning electron microscopy (SEM). X-ray diffraction (XRD) and transmission electron microscopy (TEM) were also used for phase identification. For TEM examinations, discs of 3 mm diameter and 0.1 mm thickness were cut from the bulk samples and dimpled, followed by low-angle

ion milling using a Gatan Precision Ion Polishing System. The thin foils were then examined in a Philips CM20 transmission electron microscope, equipped with an Oxford INCA energy-dispersive X-ray (EDX) spectrometer.

The as-cast microstructures of Mg–RE alloys produced by different casting technologies, where the solidification rate varies, are shown in Figure 1. The solidification rate is highest in HPDC, with sand casting being the lowest. The as-cast microstructure is basically characterized by a mixture of primary -Mg dendrites in

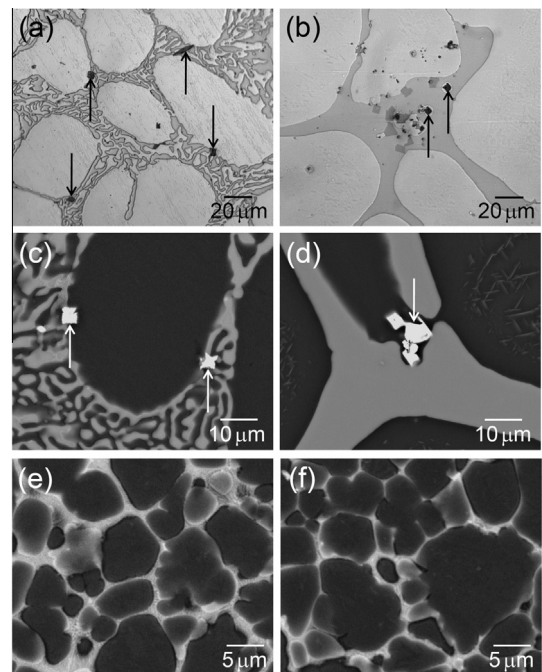


Figure 1. Micrographs showing the as-cast microstructure of selected Mg–RE alloys: (a) sand cast Mg–8La alloy (optical micrograph), (b) permanent mould cast Mg–8Nd alloy (optical micrograph), (c) sand cast Mg–8La alloy (SEM backscattered electron image), (d) permanent mould cast Mg–8Nd alloy (SEM backscattered electron image), (e) HPDC Mg–5La alloy (SEM backscattered electron image) and (f) HPDC Mg–5Ce alloy (SEM backscattered electron image). Note the presence of RE hydrides (arrowed) in the sand cast Mg–8La alloy and the permanent mould cast Mg–8Nd alloy. The RE hydrides were rarely observed in the HPDC Mg–5La and Mg–5Ce alloys.

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