High Strength and Fracture Toughness Balances in Extruded Mg-Zn-RE Alloys by Dispersion of Quasicrystalline Phase Particles

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Six kinds of Mg-2.5 at%Zn-0.5 at%RE (RE: rare earth element, Y, Gd, Tb, Dy, Ho and Er) alloys with grain size of 1~2 μm and containing a quasicrystalline icosahedral phase were prepared by casting and extrusion. These alloys had high strength and high fracture toughness balances, due to the synergetic effect of grain refinement and the dispersion of quasicrystalline phase particles. Microstructural observations showed a ductile fracture pattern, and that the origin of void nucleation was not the quasicrystalline phase particles but the conventional precipitates and the W-phase particles, because of the difference in the interface structure between the matrix and the particles.

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1. Introduction

Magnesium alloys have a high potential for application as structural materials, since they are the lightest among all structural alloys in use. For use in structural applications, their mechanical properties must satisfy both reliability and safety requirements. A method of ensuring that is to investigate their fracture toughness. Although several reports exist on the fracture toughness of magnesium and magnesium alloys,¹⁻³⁾ the value of fracture toughness in magnesium alloys has generally been reported to be lower than that in aluminum alloys. Thus, methods for improving the fracture toughness of magnesium and magnesium alloys have been investigated.

Fracture toughness in magnesium⁴⁾ and magnesium alloys⁵⁾ have been shown to improve with grain refinement. From the microstructure observations of fracture toughness tested samples in Mg-Al-Zn alloys, it is observed that grain refinement helps in (i) prevention of the formation of deformation twins, which are the origin of micro-cracks, (ii) activation of dislocations on basal as well as non-basal planes, and (iii) grain boundary sliding to release the accumulated strain at the grain boundary.^{5,6)} In addition, the dispersion of fine spherical shaped precipitates enhances the fracture toughness by pinning the dislocation movements around them.^{7,8)} Controlling the grain refinement and/or the presence of precipitates in the matrix are clearly effective methods for improving the fracture toughness in magnesium and magnesium alloys.

As-cast Mg-Zn-RE ternary alloys (where, RE is a rare earth element) are known to contain a quasicrystalline icosahedral phase,⁹⁾ and the formation of this phase has been confirmed for RE, RE = Y, Gd, Tb, Dy, Ho and Er, through various experiments.^{10,11)} The icosahedral phase possesses a five fold symmetry and has a quasi-periodic structure, which is very different from the conventional crystalline phases. This phase occurs in equilibrium with the magnesium phase,¹²⁾ showing a definite orientation relationship and with a strong interface,¹³⁾ and has been studied as the strengthening phase in magnesium alloys.¹⁴⁻¹⁹⁾ The

quasicrystal phase has been dispersed as fine particles in the matrix by severe plastic deformation such as rolling^{14,15)} or extrusion.¹⁶⁻¹⁹⁾ These alloys exhibit high ductility with good strength at room temperature and a highly stable microstructure at elevated temperatures. In addition, by using a well known Mg-Zn-Y alloy, our recent study shows that the dispersions of quasicrystalline phase particles are effective in enhancing the fracture toughness of magnesium alloy.²⁰⁾ On the other hand, in general, the particles have been known to affect the fracture toughness, because they become the void nucleation site due to the creation of a large strain field around them. The dispersion of a large size²¹⁾ and/or a high volume fraction of particles²²⁾ are not effective, and the alloys containing these particles cause lower fracture toughness. Thus, understanding the role of the particles is important for improving the fracture toughness. However, (i) the detailed effect of dispersion of quasicrystalline phase particles on the fracture mechanism, *i.e.*, understanding the origin of fracture such as the void nucleation site, and (ii) the fracture toughness of other kinds of Mg-Zn-RE (RE: Gd, Tb, Dy, Ho and Er) alloys containing quasicrystalline phase particles have not been investigated yet. Therefore, the fracture toughness and the origin of void nucleation were examined using several extruded Mg-Zn-RE alloys in this study.

2. Experimental Procedure

Six kinds of cast Mg-2.5 at%Zn-0.5 at%RE (RE: Y, Gd, Tb, Dy, Ho and Er) alloys were used in the present study. The cast alloys were solution treated at a temperature of 673 K for 24 hours, and then were extruded with a reduction ratio of 18 at a temperature of 473~483 K to refine the microstructure. The microstructures of the extruded alloys were observed by transmission electron microscopy (TEM) and the phases were identified by X-ray diffraction (XRD) using Cu-K α radiation.

The tensile test was performed at an initial strain rate of $1 \times 10^{-3} \text{ s}^{-1}$ at room temperature. The tensile specimen with a gauge length of 10 mm and a gauge diameter of 2.5 mm was machined from the extruded bar to make the tensile axis parallel to the extrusion direction.

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The fracture toughness test was carried out to obtain the value of fracture toughness and to observe the fracture feature of the extruded alloy according to two kinds of ASTM methods: E399²³⁾ and E1820-99a.²⁴⁾ The specimen that was used for investigating the fracture toughness consisted of a three point bending sample with a width of 10 mm and a thickness of 5 mm. The V-notch normal to the extrusion direction exhibited higher fracture toughness according to a previous report, which described the effect of cutting position on fracture toughness.²⁵⁾ The value of the plane-strain fracture toughness, K_{IC} and J_{IC} , was obtained by the J -integral method based on ASTM E1820-99a. The fracture surface after the fracture toughness test based on ASTM E399 was examined by scanning electron microscopy (SEM). The particle, which is the origin of void formation, was analyzed on the fracture surface in at least 15 positions by SEM in an instrument equipped with an Energy Dispersive X-ray Spectroscopy (EDX).

3. Results and Discussion

The XRD patterns of three of the extruded alloys are shown in Fig. 1. Three phases, *i.e.*, the precipitate particle (β -phase: Mg_4Zn_7), the quasicrystalline icosahedral phase

Fig. 1 XRD patterns of the extruded Mg-Zn-Dy, -Tb and -Y alloys.

Fig. 2 Typical microstructures of the extruded alloys: (a) Mg-Zn-Y, (b) Mg-Zn-Gd, (c) Mg-Zn-Tb, (d) Mg-Zn-Dy, (e) Mg-Zn-Ho and (f) Mg-Zn-Er alloys.

particle: $\text{Mg}_3\text{Zn}_6\text{RE}_1$ and the W-phase: $\text{Mg}_3\text{Zn}_1\text{RE}_2$, and α -Mg were confirmed by the XRD patterns in all the extruded alloys. Typical TEM images of all the extruded alloys are shown in Fig. 2: (a) Mg-Zn-Y, (b) Mg-Zn-Gd, (c) Mg-Zn-Tb, (d) Mg-Zn-Dy, (e) Mg-Zn-Ho and (f) Mg-Zn-Er alloys. The average grain size, d , was about $1\sim 2\mu\text{m}$. No deformed structure was observed in the grains of any alloy because of full recrystallization. Typical selected area diffraction pattern (inset in Fig. 2(a)) shows that the quasicrystalline phase, indicated by the black arrows, exists in the matrix. The quasicrystalline phase has been reported to be observed in Mg-Zn-Y system alloys by the wrought process,^{15–17} and that the quasicrystalline icosahedral phase is formed in the matrix when the Zn/Y ratio is $2\sim 7$.²⁶ The present Zn/Y and Zn/RE ratio is 5, which falls in this range. Fine precipitates of the β -phase also exist in the matrix. The formation of the β -phase precipitate is assumed to occur during the extrusion process, because the alloys were solutionized to dissolve the Mg-Zn precipitates in the present solution treatment.²⁷ The tendency for the formation of the precipitates during severe plastic deformation has already been observed in several kinds of Mg-Zn based alloys.^{7,22,28,29}

Nominal stress and strain curves in the extruded alloys are shown in Fig. 3. All the alloys have yield strength and elongation-to-failure of over 300 MPa and 13%, respectively, and show high strength and high ductility balance. The results of the tensile test of the present extruded Mg-Zn-RE alloys are summarized in Table 1.

Fig. 3 Nominal stress and strain tensile curves of the extruded Mg-Zn-RE (RE = Y, Gd, Tb, Dy, Ho and Er) alloys.

Table 1 Results of the tensile test and fracture toughness test in the extruded Mg-Zn-RE alloys.

Material	tensile test			fracture toughness test	
	σ_{ys} , MPa	σ_{UTS} , MPa	δ , %	J_{IC} , MPam	K_{JIC} , MPam ^{1/2}
Mg-Zn-Y	303	337	17.9	0.0211	32.8
Mg-Zn-Gd	317	346	16.4	0.0165	29.0
Mg-Zn-Tb	315	345	13.6	0.0172	29.6
Mg-Zn-Dy	305	338	19.5	0.0160	28.6
Mg-Zn-Ho	304	346	16.1	0.0203	32.3
Mg-Zn-Er	305	341	17.9	0.0162	28.8

where σ_{ys} is the yield strength, σ_{UTS} is the ultimate tensile strength, δ is the elongation-to-failure, J_{IC} is the fracture toughness obtained by the J -integral method and K_{JIC} is the fracture toughness calculated by eq. (1).

Fig. 4 Typical J -integral and crack extension curve of the extruded Mg-Zn-Gd alloy.

A typical J - Δa curve in the extruded Mg-Zn-Gd alloy is shown in Fig. 4. The value of J was obtained to be 1.65×10^{-2} MPam, which is acceptable as the value of J_{IC} according to the ASTM E1820-99a.²⁴ The results of the fracture toughness tests are also listed in Table 1. The values of J_{IC} and K_{JIC} are closely related as follows;²⁴

$$K_{JIC} = \{E \times J_{IC} / (1 - \nu^2)\}^{1/2} \quad (1)$$

where E is Young's modulus and ν is the Poisson ratio (44.8 GPa and 0.35,¹ respectively, for the wrought magnesium alloy). This table includes the value of K_{JIC} calculated by eq. (1). The fracture toughness of the Mg-Zn-Y alloy is the highest among the present extruded alloys. The value of K_{JIC} , which was obtained by the J -integral method, in the extruded Mg-2.4 at%Zn binary alloy with fine grain sizes is reported to be $26.8 \text{ MPam}^{1/2}$.²² The present Mg-2.5 at%Zn-0.5 at%RE alloys are found to have a higher fracture toughness than that of the Mg-2.4 at%Zn alloy. Since (i) the additional zinc content is almost similar and (ii) the Mg-2.4 at%Zn alloy contains the same type of precipitate particles without any quasicrystalline phase particles, the dispersion of the quasicrystalline phase particles is apparently effective for improving the fracture toughness of magnesium alloys.

Typical SEM micrographs of the fracture surface after the fracture toughness test based on ASTM E399 are shown in Fig. 5: (a) Mg-Zn-Ho and (c) Mg-Zn-Er alloys, respectively. The pre-crack direction and the stretched zone, which is created ahead of the fatigue pre-crack, are marked in the figures. When a load is applied to the specimen having a sharp pre-crack, the pre-crack itself blunts and the crack-tip moves forward normal to the tensile axis, producing a stretched zone (SZ). In addition, this figure shows that there are many dimples, which are typical of a ductile fracture pattern, and is indicated by the white small arrows on the fractured surface. The fracture features of the formations of the stretched zone (SZ), and the dimple pattern have previously been observed in the wrought magnesium alloys.³⁰

The dimple formation is progressed by the process of void nucleation, growth and coalescence. The void nucleation generally originates from the particles such as inclusions, dispersoids or precipitates, and the fracture occurs at the interface between the matrix and the particles or in the

Fig. 5 Typical SEM micrographs of the fracture surface after the fracture toughness test based on ASTM E399: (a) Mg-Zn-Ho and (b) Mg-Zn-Er alloys.

particle-itself. This process takes place because these particles prevent dislocation motion during plastic deformation and then the pinned dislocations create a large strain field. Three kinds of precipitates/particles in the matrix are confirmed by the present microstructure observations in Figs. 1 and 2. The fracture features in Fig. 5 show that many particles, which affect the void nucleation site, as marked by the white arrows, exist on the fracture surfaces. The analyses of EDX on the particles are shown in Fig. 6 for (a) Mg-Zn-Y and (b) Mg-Zn-Dy alloys, respectively. The particles, which were analyzed by EDX, were composed of mainly Mg-Zn (Mg-60 at%Zn-3 at%Y) in the Mg-Zn-Y alloy, and had about twice the amount of RE than Zn (Mg-15 at%Zn-37 at%Dy) in the Mg-Zn-Dy alloy. The quasicrystalline icosahedral phase, $\text{Mg}_3\text{Zn}_6\text{Y}_1$, in Mg-Zn-Y alloy is reported to be composed of Mg-(56–62)at%Zn-(8–10)at%Y,³¹⁾ and the quasicrystalline icosahedral phase in the others Mg-Zn-RE (RE: Gd, Tb, Dy, Ho and Er) alloys is also assumed to be Mg-(55–65)at%Zn-(8–12)at%RE due to consisting of $\text{Mg}_3\text{Zn}_6\text{RE}_1$.¹⁰⁾ Compared to the present microstructure observations, the particles of void nucleation site are mainly Mg-Zn (β -phase) and/or Mg-Zn-RE (W-phase). The quasicrystalline icosahedral phase particle is not the origin of fracture. This result is obtained from examining the inter-

face structure between the matrix and the particles. The quasicrystalline phase particle has a strong and matching interface with the matrix, *i.e.*, a coherent interface, on all sides.^{18,32)} On the contrary, some parts of the interface are incoherent in the precipitates³³⁾ and the W-phase.³⁴⁾ The dislocation movement is easily interrupted, as does a conventional second-phase particle, regardless of the interface structures. In the fracture toughness test, recent studies have also shown that the quasicrystalline phase particle²⁰⁾ and the precipitate particle⁷⁾ have a role of pinning the dislocation movements. However, the interface structures affect the nucleation site for void formation when the strains are increased: the fracture occurs at the interface between the matrix and the particle or in the particle itself, for the particle consisting of an incoherent interface. On the other hand, the quasicrystal phases interact quite easily with many dislocations, but could also prevent a void nucleation at the interfaces. In the fatigue test, the crack was not propagated at the interface between the quasicrystalline phase particle and the matrix in the cast Mg-Zn-Y-Zr alloy.³⁵⁾ Therefore, controlling the interface structure is effective for the prevention of the void nucleation site, and is important for improving the fracture toughness.

The results of the fracture toughness tests in Table 1 show

Fig. 6 The results of EDX investigations on the particles: (a) Mg-Zn-Y and (b) Mg-Zn-Dy alloys.

that the fracture toughness is influenced by particular RE elements: the Mg-Zn-Y alloy has the highest toughness while the Mg-Zn-Dy alloy is the lowest toughness among the present alloys. This is assumed to be related to the existence of the W-phase. Since the size of the W-phase tends to be larger than that of the quasicrystalline icosahedral phase,³⁴⁾ the W-phase easily becomes the origin of void formation. Lee *et al.* reported that the alloys containing the W-phase produce a lower ductility in the Mg-Zn-Y alloys.²⁶⁾ The volume fraction of a phase is proportionate to the integral intensity, which is obtained by the XRD measurement.³⁶⁾ The XRD results in Fig. 1 show that the intensity of the W-phase varies—the highest intensity is in the Mg-Zn-Dy alloy: the amount integral intensity of the W-phase in the Mg-Zn-Dy alloy is about five times larger than that in the Mg-Zn-Y alloy. The tendency for the intensity of the W-phase is close to the result of the fracture toughness test, *i.e.*, the present alloy with the high intensity of the W-phase is lower fracture toughness. The W-phase is formed at a higher temperature, such as during casting. Thus, the heat treatment or the choice of the RE element (valances of RE and Zn) is assumed to be necessary for making further improvements in the fracture toughness of Mg-Zn-RE (RE: Dy, Gd and Er) alloys. However, the effect of the W-phase should also be investigated in more detail, in the future.

4. Summary

The fracture toughness and the role of quasicrystalline icosahedral phase particles were investigated by using six

kinds of extruded Mg-2.5at%Zn-0.5at%RE alloys with RE = Y, Gd, Tb, Dy, Ho and Er. The following results were obtained.

- (1) In the extruded microstructures, the average grain size was about 1~2 μm, and the precipitates, the quasicrystalline icosahedral phase particles and the W-phase existed in the extruded alloy.
- (2) The results of the mechanical property tests showed high-strength, -ductility and -fracture toughness balances in the present alloys. These mechanical properties in the alloys containing quasicrystalline icosahedral phase particles were superior to those in the conventional wrought magnesium alloys.
- (3) The fracture surface observations showed a dimple feature in all the extruded alloys. The origin of void nucleation was not the quasicrystalline phase, due to its strong interfaces with the matrix, but was found to be the precipitates (β -phase) and the W-phase.

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