Microstructural stability and creep of rare-earth containing magnesium alloys

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Abstract

The creep behavior of die cast magnesium alloys is examined for the high temperature alloys AE42 and MEZ. Creep behavior in these fine-grain die castings is dependent on the stability of the near grain boundary microstructure and is improved by rare-earth element additions and reductions in aluminum content.

Keywords: Magnesium; Die casting; Microstructure; Plastic; Creep

1. Introduction

High-pressure die cast magnesium alloys are increasingly used in automotive applications because of their low density. Mg–Al alloys such as AM60B and AZ91D are used extensively since these alloys exhibit superior die castability and a good balance of strength and ductility [1]. However, the application temperature of these alloys is limited to about 120 °C, above which there is a rapid degradation in mechanical properties, especially creep resistance. Studies to understand the reasons for the poor mechanical behavior of die cast Mg–Al alloys at elevated temperatures have focused on the near grain boundary regions where nonequilibrium interdendritic solidification produces a divorced eutectic microstructure of Mg17Al12 and α magnesium [2]. Early work attributed the poor creep resistance of Mg–Al alloys to the presumed poor elevated temperature properties of Mg17Al12 [3,4]. More recent studies have shown that discontinuous precipitation of the Mg17Al12 (β) intermetallic at elevated temperatures is more likely responsible for deterioration of the creep properties of fine grained die cast Mg–Al alloys [5,6]. This latter view has been supported by observations of dynamic precipitation of β during creep in AM60B [7].

While the role of near-grain boundary microstructural evolution on creep behavior remains a subject of debate, it is apparent that the role of aluminum is critical and that alloy design to increase microstructural stability, especially in the near-grain boundary regions, is an important consideration in improving the temperature capabilities of die cast Mg alloys. For example, recent alloy development has sought to increase the stability of near-grain boundary phases in Mg17Al12 and α magnesium [2].
aluminum-containing alloys with rare-earth additions and decreased aluminum contents (such as AE42) [8] or by removing aluminum completely, as for the magnesium elektron alloy, MEZ [1]. These alloys, while having lower yield and tensile strengths than alloys with higher aluminum contents, exhibit considerable improvement in creep resistance, especially above 150 °C. In this paper, the creep behavior of high-pressure die cast MEZ and AE42 alloys are examined in an attempt to better understand the role of grain boundary microstructural stability and the possible role of aluminum on the creep behavior of die cast magnesium alloys.

2. Experimental

The alloys MEZ (Mg–1.92wt.%RE–0.33Zn–0.26Mn) and AE42 (Mg–3.7Al–2.69RE–0.21Mn) were melted in a 450T B&T cold chamber high-pressure die casting machine at Mag-Tec Casting Corporation, Michigan, USA. Rare-earth additions were achieved by adding misch metal with a nominal composition of 53%Ce, 25%La, 17%Nd, and 5%Pr. Cylindrical specimens for tensile and creep tests were produced by high-pressure die casting using four-cavity dies. Details of the processing are provided elsewhere [9,10].

As-cast and post-crept microstructures were investigated using a Philips XL30 FEG SEM. The compositions of different phases were analyzed by electron probe microanalysis. Foils for transmission electron microscopy were prepared by jet polishing in a perchloric acid solution and subsequently ion milling to remove surface oxide layers. A Philips CM12 transmission electron microscope was used for the examination of microstructure before and after creep.

Tensile and creep (compressive and tensile) tests were conducted at temperatures of 125, 150 and 175 °C in accordance with ASTM procedures and the details of the test procedures can be found in reference [10]. Tensile and compressive creep tests were performed to determine if differences in loading mode are observed, similar to that reported previously for Mg–Al alloys [7]. Post-crept specimens were examined by transmission electron microscopy to investigate structural changes that occurred during creep.

3. Results

Die cast samples of both alloys exhibited an equiaxed microstructure with an average grain size ranging from 10 to 20 μm. The as-cast microstructure of AE42 contains a fine, two-phase aggregate at the grain boundaries (Fig. 1a), while a semi-continuous grain boundary phase dominates microstructure of MEZ, as shown in Fig. 1b. The near-grain boundary microstructure of AE42 alloy, which has been examined in detail [11], is a mixture of α Mg and the intermetallic Al₁₁(RE)₃. The grain boundary phase in die cast MEZ alloy has been identified by transmission electron microscopy as the Mg₁₂Ce type intermetallic (Mn₁₂Th prototype) [9]. This phase has also been reported by Bettles and coworkers [12] in the sand cast version of this alloy. The grain boundary phase appears to be heavily faulted and contains sub-boundaries [9]. Also, small particles of α magnesium are observed at the grain boundaries and the occasional occurrence of ring diffraction patterns indicates the presence of very fine oxide particles [9]. The near-grain boundary microstructures of post-crept AE42 and MEZ are shown in Fig. 1c and d, respectively. No change in microstructure morphology after creep exposure is observed in MEZ, while significant changes are observed for AE42.

The tensile properties of both alloys as a function of temperature are shown in Table 1. Yield strength and ultimate tensile strength decrease with increasing temperature, while ductility reaches a maximum at around 150 °C. At a given temperature, the tensile properties of AE42 are always superior to those of MEZ, which is expected considering the presence of 4% aluminum in AE42. Typical creep strain versus time curves for both the alloys are shown in Fig. 2 for both tension and compression. At 150 °C and 70 MPa, AE42 exhibits lower primary creep in both tension and compression and a lower total creep strain for the test times investigated, as shown in Fig. 2. The difference in total creep strain is somewhat greater.
for the compressive loading condition at this stress level. However, as the stress is increased at 150 °C, the total creep strain accumulation in AE42 rapidly exceeds that seen in MEZ, as shown in Fig. 2b for compressive loading. A similar trend occurs with increasing temperature, as illustrated in Fig. 2a. At 175 °C and 80 MPa, the primary creep strain in AE42 is significantly less than that observed for MEZ, but the total creep strain accumulation exceeds that for MEZ after only 100 h.

Table 1
Tensile properties of MEZ and AE42 alloys at different temperatures

<table>
<thead>
<tr>
<th>Temperature (°C)</th>
<th>MEZ</th>
<th>AE42</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>0.2% YS (MPa)</td>
<td>UTS (MPa)</td>
</tr>
<tr>
<td>25</td>
<td>97.6</td>
<td>134.8</td>
</tr>
<tr>
<td>125</td>
<td>83.6</td>
<td>115.9</td>
</tr>
<tr>
<td>150</td>
<td>78.0</td>
<td>110.0</td>
</tr>
<tr>
<td>175</td>
<td>73.5</td>
<td>99.7</td>
</tr>
</tbody>
</table>

Minimum tensile creep rates as a function of stress at 150 and 175 °C for the two alloys are shown in Fig. 3a. At low stresses, the stress exponent ranges from 3 to 7, and exponents in this range are associated with creep processes controlled by dislocation glide and climb. It should be noted that the very low creep rates observed at low stresses make the determination of the stress exponents problematic. At higher stresses, a transition to significantly higher stress dependence
occurs. This may be attributed to power law breakdown, the onset of mechanical damage during creep, or a combination of the two. Initial studies of microstructure from the samples crept in the high stress regime do not, however, indicate obvious accumulation of damage. At 150 °C, a higher minimum creep rate is observed for AE42 at all stress levels, although, as shown in Fig. 2, primary creep dominates the total creep strain accumulation at intermediate to low stresses. At 175 °C, MEZ shows significantly lower minimum creep rates than those observed for AE42 only at stresses exceeding the transition in stress dependence. Tensile and compressive creep rates for MEZ at 150 °C as a function of stress are compared in Fig. 3b. The transition in stress dependence observed in tension is absent in compression and the mechanism for this behavior is unknown at this time. Below the transition stress in tension, no significant differences in creep behavior between tension and compression are noted. However, a transition in stress dependence (not shown here) occurs in both tension and compression for AE42. The general differences in 150 °C creep behavior between MEZ and AE42 that are observed in tension (Fig. 3a) are also observed in
compression, as shown in Fig. 3c. The notable difference is, again, the absence of a transition in stress dependence at higher stresses for MEZ in compression and the presence of such a transition for AE42.

4. Discussion

The major differences in the microstructure of AE42 and MEZ arise from the presence of aluminum in AE42. While the predominant phase in both alloys remains \( \alpha \) Mg, the grain boundary microstructure varies significantly. In AE42, the grain boundary consists of a fine eutectic mixture of \( \alpha \) Mg and \( \text{Al}_{11}(\text{RE})_3 \) intermetallic. The \( \alpha \) Mg present in the eutectic region is supersaturated in aluminum, as has been observed in other Mg-Al alloys without rare-earth additions [6]. MEZ, on the other hand, exhibits a predominantly semi-continuous single phase \( \text{Mg}_{12}(\text{RE}) \) intermetallic at the grain boundaries where final eutectic solidification takes place. The presence of \( \alpha \) Mg in the eutectic region is extremely low in MEZ suggesting a dissolved eutectic reaction which is typical of die-cast magnesium alloys and is attributed to the high cooling rates achieved [6,9,13]. The fine eutectic in AE42 and a predominantly single-phase structure in MEZ alloy at the grain boundary appear to play a predominant role in the observed differences in creep behavior.

At all temperatures, AE42 exhibits a higher yield strength than the MEZ alloy, which can be attributed to solid solution strengthening by aluminum [2]. The \( \alpha \) Mg in MEZ has only traces of manganese, zinc and rare-earth elements [9], making it a relatively weaker phase in comparison to the \( \alpha \) Mg phase in AE42. The lower primary creep strain observed in AE42 for tensile and compressive creep (Fig. 2) can be attributed to the higher yield strength of AE42. However, at least at the intermediate temperature of 150 °C, creep resistance, measured as minimum creep rate, is lower in the aluminum containing AE42 for the entire stress range examined. Previous studies have supported the concept that degradation of creep resistance with increasing aluminum content is caused by microstructural instabilities in the grain boundary regions, such as discontinuous precipitation of \( \text{Mg}_{17}\text{Al}_{12} \) from supersaturated \( \alpha \) Mg during creep [6]. Additional studies [11] have shown that in aluminum containing alloys, \( \text{Al}_{11}(\text{RE})_3 \) readily decomposes at high temperature to \( \text{Al}_2(\text{RE}) \) and \( \text{Mg}_{17}\text{Al}_{12} \). In the present study, as shown in Fig. 1, changes in near-grain boundary morphology can be observed after creep exposure for AE42, while no such changes are observed in MEZ. This indicates the potential importance of microstructural stability on creep resistance in these fine-grained die cast alloys. The significant asymmetry between tensile and compressive creep rates that has been observed for die-cast alloys with significant aluminum content [7,14] has not been observed for either AE42 or MEZ. While it is possible that the role of near-grain boundary microstructure and microstructural instability during creep may account for these observations, the stress dependence of creep would suggest that dislocation glide plus climb are still dominant. To address this issue, the nature of creep deformation in the near-grain boundary region and its contribution to overall creep is being investigated by the use of a high resolution strain mapping technique.

The general observations reported here indicate that near-grain boundary microstructure is a controlling parameter in fine-grained die castings when little intragranular strengthening is derived from solid solution or from fine homogeneous precipitation in grain interiors. Because of this, the control of grain boundary microstructure is critical in the design of new high temperature alloys. It is also important to consider additional matrix strengthening processes and further improvements in elevated temperature grain boundary strength and stability if significant high temperature uses for magnesium alloys are to be realized.

5. Conclusions

1. The higher tensile strength of AE42, which can be attributed in part to solid solution strengthening by aluminum, does not translate to higher long-time creep resistance when compared to MEZ.
2. Microstructural instability in the near-grain boundary regions significantly influences creep behavior in fine-grain die castings such as AE42 and MEZ.

3. The design of improved high temperature magnesium alloys must address both grain boundary strength and stability as well as improved creep resistance by solid solution and precipitation strengthening.

Acknowledgements

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References