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Review

# Microstructure evolution and mechanical properties of magnesium alloys containing long period stacking ordered phase



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#### ABSTRACT

Magnesium alloys containing rare earth (RE) in conjunction with other alloying elements, such as Zn, have drawn increasing interest due to their high strength, high ductility and high creep-resistance with the existence of long period stacking ordered (LPSO) phase. Although the LPSO phase has been studied over a decade, the question remains how it impacts the mechanical properties of Mg-RE-X alloys, where X represents a metallic element with its atomic radius smaller than Mg. This paper provides a tutorial review of the current status on the role of LPSO phase in manipulating the mechanical properties in Mg-RE-X based alloys, where some outstanding issues are also highlighted and discussed.

#### 1. Introduction

The long period stacking ordered (LPSO) phase was firstly identified as an X-Mg<sub>12</sub>ZnY phase in a Mg-7.2Y-1.2Zn-0.48Zr (wt%) alloy by Luo et al. [1] in 2000, but considerable attentions have not been paid until it was found in the Mg97Y2Zn1 (at. %) RS P/M (rapidly solidified powder metallurgy) alloy, where the yield strength can be enhanced up to ~600 MPa [2,3]. Since then, precise characterization at atomic level using high resolution transmission electron microscopy (HRTEM) has confirmed that such an LPSO phase can exhibit different polytypes, including 6H (now clarified to be a building block and a segment of 18R) [4–6], 10H [7–9], 14H [8–21], 15R [22], 18R [8-11,18-20,23-25], and 24R [9], which refer to different stacking sequences along the normal to the  $(0002)_{\alpha-Mg}$  basal plane. These polytypes can be classified into two categories: Type I LPSO phase forms along grain boundaries as a secondary phase during solidification, whereas Type II LPSO phase precipitates from the  $\alpha$ -Mg matrix during heat treatment at elevated temperatures after casting [26].

With the presence of LPSO phase, ternary Mg-RE-X (X represents a metallic element with an atomic radius smaller than Mg) alloys usually exhibit excellent mechanical properties, such as high strength [2,14-16,23,26-32], high ductility [16,17,23,26,28,29,32] and high creep-resistance [27,33-35]. However, the underlying relationship between the LPSO phase and the superior mechanical properties has not

been fully clarified.

It is widely accepted that intragranular precipitates strengthen the matrix via impeding the movement of dislocations, and the magnitude of strengthening effect on the matrix strongly depends on the size, morphology and crystallography of precipitates. Nie et al. [36,37] provide a criterion for selecting key strengthening constituents in Mg alloys, where plate-shaped precipitates that form on prismatic plane of the matrix result in a much higher yield strength compared to those that form on the basal plane. Since the LPSO phase is essentially periodic stacking faults that usually form on the basal plane, it is very difficult to achieve a significant improvement in strength by merely introducing dispersive LPSO precipitates. Through composition modulation and heat treatment alternation, our previous work [38,39] has confirmed that the effect of LPSO phase on age-hardening response is negligible, and the plate-shaped  $\beta'$  phase (*c*-axis base-centered orthogonal, a = 0.642 nm, b = 2.224 nm, c = 0.521 nm) that forms on prismatic plane of  $\alpha$ -Mg matrix is the key strengthening precipitate during artificial ageing.

Given the fact that the LPSO phase usually precipitates at grain boundary and grows towards grain center, we believe that the LPSO precipitates can act as inhibitors retarding grain boundary sliding and grain growth at elevated temperatures. It has been proved that the LPSO phase can not only effectively retard the grain growth at elevated temperatures [40], it can also retard the coarsening of  $\beta$ ' phase during

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ageing [39]. Based on the literature review as well as our recent work, the present paper aims to provide a new perspective for the design of advanced Mg-RE-X alloys incorporating the LPSO phase. The proposed synergistic mechanism between the LPSO and  $\beta$ ' phase can be a potentially effective approach to develop new Mg-RE-X alloys with high strength, high ductility and high creep-resistance.

## 2. Formation of LPSO Polytypes

As aforementioned, the LPSO phase can exhibit multiple polytypes in different Mg-RE-X alloys. So far, it is well acknowledged that the LPSO polytypes are distinguished by their different arrangements of the periodically RE/X enriched ABCA-type building blocks that have a local face-centered cubic (fcc) stacking sequence on the close-packed planes [9,18,19,25]. It is worth mentioning that the Ramsdell notation (such as 14H or 18R, where the Arabic numeral indicates the number of layers in one period and the letter behind stands for hexagonal or rhombohedral symmetry) cannot unambiguously distinguish different variants in one polytype [41]. For example, the stacking sequence of 18R LPSO polytype was reported to be ABABABCACACABCBCBC... [1], ABABCACACABCBCBCAB... [10] and ABACBCBCBACACACBAB... [18] etc. With the help of Zhdanov (Жданов) notation [41], these 18R-LPSO stacking sequences can all be described as  $[(3, \overline{1})(1, \overline{1})]_3$  (ABCACACA-BCBCBCABAB...), in which the numbers with and without an overbar represent the layer numbers of reverse and normal stacking sequences in order, respectively. The subscript outside the parentheses indicates the total number of cycles within one period. Then, each LPSO polytype can be unambiguously described through the combination of Ramsdell notation and Zhdanov notation if there are different variants.

Saal et al. [42] predicted the stabilities for 14H and 18R LPSO polytypes with density functional theory (DFT) in many ternary Mg-RE-X alloys, using a new structure model based on the inclusion of a Mg interstitial atom. They found that favorable mixing enthalpy between Mg and RE on the local fcc lattice and the size mismatch together serve as excellent criteria determining LPSO formation. Fig. 1 shows the prediction of possible LPSO forming ternary alloys, of which 12 have been reported with LPSO formation (including Mg-Y-Al system that was reported after Saal et al.'s publication [19,40], as marked in Fig.1). In addition, such a prediction provides a chance for designing new Mg alloys that are free of RE elements but still contain LPSO phase for achieving improved mechanical properties with a lower cost. However, further study is required to figure out how to form the four-layer ABCAtype building block that provides near perfect matching with the  $(0002)_{\alpha-Mg}$  basal plane in Mg alloys without RE addition.



## Ternary Mg-X<sub>L</sub>-X<sub>S</sub> LPSO Stability

It is commonly accepted that the formation mechanism of LPSO phase usually contains four steps [11,18,19,25,43]: (1) enrichment of RE and X atoms around newly formed stacking faults; (2) in-plane longrange ordering of RE and X atoms so as to locally and periodically form the ABCA-type building blocks; (3) growth of each LPSO polytype occurs by the generation and propagation of growth ledges, with a displacement vector of  $a/3 < 10\overline{10} > \alpha$ ; (4) different LPSO polytypes are controlled by different  $\alpha$ -Mg atomic layers separating the building blocks. The transformation between different LPSO polytypes requires segregation of RE and X atoms and the rearrangement of  $\alpha$ -Mg atomic layers via the movement of Shockley partial dislocation on the neighboring plane. Fig. 2 shows the characteristic stacking features for the unit cells of 14H, 18R and 24R LPSO polytypes in the same Mg<sub>97</sub>Y<sub>2</sub>Zn<sub>1</sub> (at.%) alloy annealed at 500 °C for 2.5 h, indicating various metastable LPSO polytypes can coexist during heat treatment. Kim et al. [25] systematically examined the irregular arrangements of ABCA-type building blocks for both the  $\alpha$ -Mg matrix with LPSO plates and the interdendritic LPSO phase in a Mg97Y2Zn1 alloy. As shown in Fig. 3, the thermodynamically most stable LPSO polytype is 14H, where both thick 14H plates embedded inside the  $\alpha$ -Mg matrix and interdendritic 14H LPSO phase with thin  $\alpha$ -Mg bands are formed after long-time annealing.

#### 3. LPSO Phase and Mechanical Properties

Although the LPSO phase exhibits different polytypes, its effect on mechanical properties should be almost identical due to the same morphology and similar crystallography so far reported in different Mg-RE-X alloys. In order to clarify the intrinsic effect of LPSO phase on mechanical properties, we deliberately designed experiments to illustrate the variations of specific mechanical properties between different Mg-RE based alloys, where either the composition or microstructure is similar with different distribution and volume fraction of LPSO phase.

## 3.1. Strength

The perspective that the LPSO phase plays a significant role in strengthening Mg-RE-X alloys stems from the excellent mechanical properties achieved by warm extrusion of gas-atomized Mg<sub>97</sub>Y<sub>2</sub>Zn<sub>1</sub> powders, where the Mg grains are 100-200 nm in diameter with the volume fraction of LPSO phase about 20-25% [3,12]. In order to clarify whether the LPSO phase improves the strength of Mg-RE-X alloys or not, Mg-9.86Y-5.32Gd-1.54Zn-0.36Zr (WGZ1051K, wt%) and Mg-12.56Y-4.88Gd-1.30Zn-0.33Zr (WGZ1251K, wt%) were designed [38,39]. After solution treatment (denoted as T4 state hereinafter), the

> Fig. 1. DFT-predicted stability of 14H and 18R LPSO polytypes for all Mg-X<sup>L</sup>-X<sup>S</sup> ternary systems (atomic radius  $r_{X^L} > r_{Mg} > r_{X^S}$ ). The blue squares without an "x" indicate systems where Saal et al. predict as-yet-unobserved LPSO polytypes to be stable [42]. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

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