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Experimental investigation and simulation of precipitation evolution in Mg-3Nd-0.2Zn alloy

Xiangyu Xia[a,b,*], Amirreza Sanaty-Zadeh[a], Chuan Zhang[c], Alan A. Luo[d], Donald S. Stone[a,b]

[a] Materials Science Program, University of Wisconsin – Madison, 1509 University Ave., Madison, WI 53706, USA
[b] Department of Materials Science and Engineering, University of Wisconsin-Madison, 1509 University Ave., Madison, WI 53706, USA
[c] Computherm LLC, 7 S. Yellowstone Dr, Suite217, Madison, WI 53719, USA
[d] Department of Materials Science and Engineering, The Ohio State University, 116 W. 19th Ave, Columbus, OH 43210, USA

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ABSTRACT

In this research, precipitation evolution and strengthening behavior in Mg-3Nd-0.2Zn alloy during isothermal age treatment at 200 °C are quantitatively studied by a combination of experimental investigation and computational simulation. Microstructure evolution, including volume fraction, number density, and size of precipitates are first measured by TEM and DSC techniques and then modeled using an integrated computational simulation method combining the CALPHAD (CALculation of PHAse Diagram) approach and classical nucleation model. The focus of the analysis is set on the evolution of key strengthening precipitate β'. Critical kinetic parameters such as particle interfacial energy and nucleation site number are obtained computationally by fitting the model to the experimental data. Mechanical properties of a series of Mg-Nd-Zn alloys are predicted using the simulated microstructure evolution as inputs. Good agreement is found between simulated and measured results. Strategies for enhancing the precipitation hardening effect are presented and discussed based on the simulation results. The merit of this study is to use the obtained parameters to extrapolate microstructure evolution and mechanical properties when adjusting alloy composition and/or heat treatment conditions within a certain range, which will be very useful for further development and optimization of multi-component Mg alloys.

1. Introduction

Magnesium (Mg) is of great interest in transportation industries [1–4] because of its potential to lightweight vehicles. However the application of Mg has been limited due to its relatively poor mechanical properties compared to conventional structural materials such as steels or aluminum alloys. Recently, a series of Mg-rare earth (RE) alloys, including Mg-Yttrium (Y), Mg-Cerium (Ce), Mg-Neodymium (Nd), Mg-Samarium (Sm) and Mg-Gadolinium (Gd), have been developed [5–8] with improved mechanical properties. The major strengthening contribution is precipitation hardening of nano-scale Mg-RE precipitates in α(Mg) matrix produced during heat treatment. Thus to develop Mg-RE-based alloys for potential industrial applications, precipitation hardening mechanisms as well as the structure-property relationship must be fully understood.

Neodymium (Nd), as one of the most abundant RE elements, has relatively high solubility (~3.5 wt%) in α(Mg) at the eutectic point (545 °C) and low solubility (< 0.1 wt%) at room temperature, which make it suitable for age hardening [9]. Appropriate addition of Zn further enhances creep resistance and strength of age-hardened Mg-Nd alloys [5,10,11]. The effect of chemical compositions and heat treatment on mechanical properties and microstructure evolution of Mg-Nd-Zn alloys has been the subject of extensive experimental research [3,5,10–17]. An optimum composition of Mg-3Nd-0.2Zn (All compositions in this proposal are in weight percent, unless noted otherwise) has been identified, which exhibits superior combination of strength and elongation and is considered promising candidate for industrial applications [4,13]. Sanaty-Zadeh et al. [10] systematically investigated age-hardening response in Mg-3Nd-0.2Zn alloy during isothermal age treatment at 200 °C by high-angle annular dark-field scanning transmission electron microscopy (HAADF-STEM) and atom probe tomography (APT). The precipitation sequence was identified as: super-saturated solid solution (S.S.S.S.) → Guinier-Preston (G. P.) zone I, II, and III (Mg_3(Nd, Zn), $D0_{19}$, a = 0.642 nm, c = 0.521 nm) → β' (Mg_7(Nd, Zn), orthorhombic, a = 0.642 nm, b = 1.112 nm, c = 0.521 nm) → β_2 (Mg_3(Nd,Zn), orthorhombic, a = 0.642 nm, b = 0.556 nm, c = 0.521 nm) → β_1 (Mg_3(Nd, Zn), FCC, a = 0.74 nm). The maximum hardness was achieved after 8 h aging, at which β' is the

* Corresponding author at: Materials Science Program, University of Wisconsin – Madison, 1509 University Ave., Madison, WI 53706, USA.
E-mail address: xxia.wisc@gmail.com (X. Xia).

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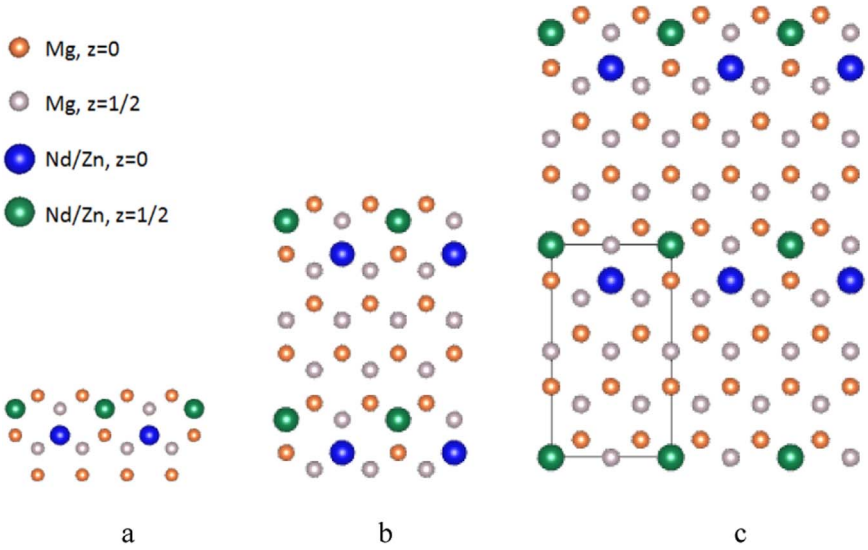

Fig. 1. Atomic models of G. P. zones I (a), II (b), and β' (c) precipitates from $[0001]_{\alpha}$ (visualized using VESTA [65]).

most abundant phase, making β' the key strengthening precipitate in the Mg-3Nd-0.2Zn alloy. Fig. 1 illustrates the crystal structures of G. P. zones I, II, and β'. As seen, both G. P. zones consist of "zigzag" arrangements of RE atoms that are the main components of β'. Therefore, they are considered as pre-β' clusters, or nuclei of β' in this work. G. P. III zone is a pre-cluster of β_2 phase, whose effect on age-hardening is negligible due to relatively low number density [10]. Thus, by investigating evolution of β' and its relationship to the strength of the aged alloy, we are able to assess age-hardening response of Mg-3Nd-0.2Zn alloy for all practical purposes.

Computational simulation such as the CALPHAD (CALculation of PHAse Diagrams) approach simplifies the alloy design procedure and reduces the experimental costs and time. The CALPHAD approach has recently been broadened to a holistic ICME (Integrated Computational Materials Engineering) framework in the design and development of new materials [18–24]. For precipitation-hardening alloys, precipitation kinetic parameters can be modeled by combining measured microstructure data of the alloys with thermodynamic description and atomic mobility developed separately. Mechanical properties such as yield strength upon aging are predicted using these kinetic models. This approach has been successfully applied to simulate the precipitation evolution and strengthening effect in many alloy systems including Ni, Al and Mg [19,21–24].

This paper aims to provide a quantitative description of precipitate evolution and precipitation strengthening behavior in Mg-Nd-Zn alloy through a combination of experimental investigation and CALPHAD-based ICME computational simulation. The changes in volume fraction, number density, and average size of precipitates during aging at 200 °C are first studied quantitatively by differential scanning calorimeter (DSC) and transmission electron microscopy (TEM). Thermodynamic description and atomic mobility of the target system are separately developed using CALPHAD-type methods with the experimental data from this work and literature. The precipitation evolution and hardening effect in the Mg-3Nd-0.2Zn alloy are then modeled using the previously-mentioned integrated simulation approach. Mechanical properties of a series of Mg-Nd-Zn alloys are calculated and compared to literature results to identify the effect of age temperatures and alloy compositions on age-hardening response, and to validate the predictability of current method. The results of this work not only help to quantify our description of the evolution of precipitates in Mg-Nd-Zn alloy, but also provide insights for future optimization of Mg-RE based alloys and explore the application of ICME in novel Mg alloy development.

2. Experimental measurement

The age-hardening response of Mg-3Nd-0.2Zn alloy has been described in earlier publications [5,10,11] and are summarized here. Mg-3Nd-0.2Zn as-cast ingots were prepared and solid solution treated following the same method in ref. [10]. Eight samples were aged for different times (0.5, 1, 2, 4, 8, 16, 32, 64 h) at 200 °C for DSC measurements. The aging times were selected according to age-hardening response, which cover the under-aged, peak-aged, and over-aged conditions. The aged samples were polished and cleaned with ethanol prior to DSC tests, which were performed on a DQ100 differential scanning calorimeter. Standard aluminum pans were used as references and to encapsulate the samples. The samples underwent cyclic thermal treatment (e.g. heating and cooling) at 10 °C/min. The cyclic thermal treatment was repeated 3 times for each sample between 25 °C and 400 °C. The second and third cycles were used to establish a background (over-aged condition).

Three aged samples were selected for TEM analysis (Philips CM200, and Tecnai-TF30): the 2 h (under-aged), 8 h (peak-aged), and 64 h (over-aged) aged samples. For sample preparation, 1000 μm-thick slices were cut from the bulk samples and ground down to ~50 μm, using Allied Multiprep machine with diamond lapping films (15, 6, 3, 1, then 0.1 μm grid size). The polished samples were then ion milled with Fischione 1050, (3.0 kV ion gun energy and 5° milling angle). Finally, 0.6 kV ion gun energy was used to clean up surface. The thickness of the samples was measured by electron energy loss spectroscopy (EELS) on regions where microstructure was analyzed. For each region three EELS runs were performed and the average thickness value was used. The absolute thickness values were extracted by measuring the beam convergence angle and EELS semi collection angle [25].

Fig. 2a–c show representative TEM images taken from alloys aged at 200 °C for 2 h (under-aged), 8 h (peak-aged), and 64 h (over-aged). As was seen earlier in our STEM analysis [10], a combination of β-series phases exist, while in this work it is assumed the quantitative measurements of the multiple phases are identical to those of pure β'. The morphology of β' is plate-like in all beam directions in TEM analysis [10] under different aging times, based on which it is reasonable to induce its shape is round disc. With the diameter as L and the thickness as H, precipitate volume V_p is given by:

$$V_p = \frac{1}{4}\pi L^2 H \tag{1}$$

Volume fractions are estimated by multiplying the average volume, $\overline{V}_p$, by the number density of precipitates, N: