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Effects of precipitates and its interface on thermal conductivity of Mg-12Gd alloy during aging treatment



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ABSTRACT

How to simultaneously improve the thermal conductivity and mechanical properties of Mg alloys is an interesting and important issue topic. In the present study, the effect of aging treatment on the thermal conductivity and micro-hardness of Mg–12Gd alloy was investigated. Results indicated that the thermal conductivity and micro-hardness of the alloy could be significantly improved by aging treatment, and Mg–12Gd alloy exhibited a typical aging hardening response with the increasing aging time. At the early stage of aging (4 h), a large number of fine particles precipitated from α -Mg matrix, but thermal conductivity of alloy increased a little, which was attributed to the coherent interface hindering the increase of thermal conductivity. The thermal conductivity increased to the peak value of 75.7 W/(m-K) at the aging time of 300 h, which was about 2.17 times that of assolutionized alloy. The huge increment in the thermal conductivity of as-aged Mg–12Gd alloy was related to the precipitation of particles and the incoherent interface between precipitates and α -Mg matrix.

1. Introduction

As the lightest structural materials, Mg alloys have high specific strength and stiffness, good thermal conductivity and electromagnetic shielding effectiveness [1-6]. Pure Mg has the best thermal conductivity among various metallic Mg alloys and may potentially help meet increasing demands for heat dispersing materials in modern industries. Unfortunately, the pure Mg with high thermal conductivity generally exhibits poor mechanical properties, which cannot meet the current requirements of the heat dissipation industry. To date, adding various alloying elements is the commonly used method to improve the mechanical properties of Mg alloys [7–10]. However, the experimental results show that microalloying can simultaneously deteriorate the thermal conductivity of Mg alloys [11-13]. Achieving good mechanical properties and receivable thermal performance appears to be contradictory according to the present reports. And the balance between mechanical properties and thermal conductivity of Mg alloys has become a critical concern for expanded engineering applications. Thus, it is vital to develop an effective method to enhance mechanical properties without deteriorating or with enhancing thermal conductivity of Mg alloys.

According to the latest research, aging treatment has a simultaneous influence on the improvement in mechanical properties and physical

properties of materials. Chen et al. [14,15] reported that the EMI shielding effectiveness value of ZK60 alloy increased gradually with the increasing aging time, caused by the increased number density of precipitates during aging treatment. Choi et al. [16] stated that the long needle of the eutectic silicon particles were broken into smaller fragments and then gradually spheroidized after aging treatment and the spheroidizing of particles could improve the tensile strength and thermal diffusivity. Lin et al. [17] reported that the ultimate tensile strength (UTS) and electrical conductivity of Al-Mg-Si alloy can be simultaneously enhanced by pre-aging and then cold rolling followed by re-aging. Yang et al. [18] found that high UTS and high electrical conductivity can be achieved for the Cu-Zr-B alloy by double deformation-aging process, which was attributed to the suitable interactions of strain hardening and precipitation behaviors during double deformation and aging treatments. Recently, the effect of aging treatment on the thermal conductivity of Mg alloys was also investigated. Pan et al. [19,20] has studied the influence of aging treatment on thermal conductivity and micro-hardness of Mg-2Zn and Mg-6Zn-xCu alloys. They found that the aging treatment could improve the electrical conductivity and micro-hardness of studied alloys, and the electrical conductivity increased rapidly and reached the peak value at the beginning of aging, and the electrical conductivity of alloys changed a little with the increasing aging time. Wu et al. [21] studied the

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mechanical properties and thermal conductivity of Mg-2Zn-Zr alloy with mill-annealing and subsequent aging process, and they found that the precipitation of Zn-Zr intermetallics could be promoted by aging treatment, which improves the thermal conductivity and strengths of Mg-2Zn-Zr alloy. More recently, Chen et al. [22] has investigated the thermal conductivity of Mg-5Sn alloy at different aging status, and the results revealed that the electrical conductivity and thermal conductivity of Mg-5Sn alloy aged at 513 K increased linearly with the increasing aging time and the unit cell volume had a significant effect on the thermal conductivity. It mentioned that nanoscale Mg₂Sn particles were coherent with the α -Mg matrix and had an obvious influence on the thermal conductivity of Mg-5Sn alloy. But, how the interface between precipitates and matrix influence thermal conductivity of Mg alloy was not discussed. Clearly the research on the influence of aging treatment on the thermal conductivity of Mg alloys has made some progress. However, the intrinsic mechanism of the effect of phase interface on the thermal conductivity of Mg alloy in the aging process has not yet been discussed intently.

In addition, Mg–Gd system is one of the most promising candidates due to the remarkable age-hardening response [23]. However, Nie et al. [24,25] pointed out that binary Mg–Gd alloys containing less than 10 wt% Gd showed little or no precipitation-hardening response during isothermal or isochronal aging of supersaturated solid solutions of these alloys. Thus, in order to investigate precipitates and its interface on micro-hardness and thermal conductivity of this alloy during aging treatment process, Mg–12Gd alloy was selected as the study subject. Such an investigation aims to provide theoretical basis for the influence of precipitates and its interface on thermal conductivity of Mg alloy during aging treatment, which is helpful to develop Mg alloy with high thermal conductivity and moderate mechanical properties as heat sink materials.

2. Experimental Procedures

Cast billets were prepared from commercial pure Mg (99.95 wt%) and Mg–Gd master alloy (25 wt%). The mixtures were melted at 750 °C in an electric resistance furnace with the protection of $\rm CO_2$ and $\rm SF_6$ and stabilized at 720 °C for 30 min, then pouring into a steel mould preheated to 200 °C. The analyzed composition of the alloys measured by X-ray fluorescence (XRF) was Mg–12.88Gd (wt%) alloy. The samples were solution treated at 798 K for 24 h and then quenched into water at room temperature. Subsequently, the specimens were isothermally aged at 498 K for different times. Micro-hardness of samples was measured on a Vickers hardness testing machine with the loading force and dwelling time of 50 g and 10 s, respectively. For each sample, 10 tests were conducted at least.

The phase of the samples was identified using an X-ray diffraction (XRD, Rigaku D/MAX-2500PC). The microstructures of as-aged samples were observed by using the transmission electron microscopy (TEM, Tecnai G2 F20 S-TWIN) with an operating voltage of 200 kV was conducted. TEM foils were prepared by mechanical polishing to less than $100\,\mu m$, subsequently punched into 3 mm discs and final ion-milling using a Gatan Precision Ion Polishing System (GATAN691).

Samples for thermal diffusivity measurement were cut from samples in the shape of disks with a diameter of 12.7 mm and a thickness of 3 mm. Thermal diffusivity was measured on Netzsch 457 apparatus via laser flash method at 298 K. The room temperature density was obtained by Archimedes method. The specific heat capacities of the samples in this study could be obtained by Neumann-Kopp rule [26,27]. Thermal conductivity (λ) was calculated from specific heat capacity (C_p), thermal diffusivity (α) and density (ρ) via the following equation:

$$\lambda = \alpha \bullet \rho \bullet C_p \tag{1}$$

where α (m²/s) is the thermal diffusivity, ρ (g/cm³) is the density, C_p (J/(g·K)) is the specific heat capacity at constant pressure. The uncertainty for the thermal conductivity was estimated to be less than 9%.

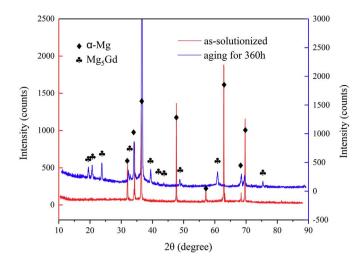


Fig. 1. XRD diffraction patterns of as-solutionized and as-aged Mg-12Gd alloys.

3. Results and Discussion

Fig. 1 illustrates the XRD results of as-solutionized and as-aged Mg–12Gd alloy. It can be seen that the as-solutionized Mg–12Gd alloy only consisted of α -Mg, indicating that the Gd atoms almost dissolved into the α -Mg matrix after solid solution for 24 h at 798 K. After aging for 360 h, the alloy included additional diffraction peaks from a new precipitated Mg₅Gd phase. The XRD results were further consistent with the following TEM analysis.

TEM bright field graphs of Mg-12Gd alloy at different aging time are shown in Fig. 2. It is noteworthy that the precipitation sequence and precipitates of as-aged Mg-Gd alloys have been studied in detail by many scholars [23,28,29]. Therefore, the specific precipitation behavior of as-aged Mg-Gd alloy was not described in this work. It was generally observed that the precipitates tended to grow and coarsen with the increasing aging time, and the length of precipitates increased notably, whereas, the width of the precipitates increased a little, which was consistent with the results in literature [30]. As can be seen from Fig. 2(a) and (d), a high number density of fine lenticular precipitates formed and distributed uniformly in the α -Mg matrix with a length of ~100 nm and a width of ~20 nm after aging for 4 h. As the aging time increased to 24 h, a large number of lozenge shaped precipitates were observed distributed in the α -Mg matrix, as shown in Fig. 2(b) and (e). The size of the lozenge shaped precipitates was bigger than that of the fine lenticular precipitates. After aging for 300 h, the size of precipitates continued to grow, and the morphology of precipitates was plateshaped, as shown in Fig. 2(c) and (f). According to the SAED of the plate-shaped precipitates, this phase was determined to be a face-center cubic structure with a lattice parameter of ~2.2 nm, which was identical to the β phase. And the β phase had a $[110]_{\beta}$ // $[0001]_{\alpha}$, $(-112)_{\beta}$ $//(1-100)_{\alpha}$ and $(1-11)_{\beta}$ $//(11-20)_{\alpha}$ type relationship. This was consistent with the previous reports about the β phase [23,31]. And Fig. 3 further presents the HRTEM image of the interface between precipitates and α -Mg matrix at different aging time. The fine lenticular precipitates were coherent with the α-Mg matrix at the early stage of aging, as can be seen from Fig. 3a. The lozenge shaped precipitates were semi-coherent with α -Mg matrix at the aging time of 24 h as shown in Fig. 3b. Furthermore, the interface between the plate-shaped precipitates and α-Mg matrix was incoherent at the aging time of 300 h in Fig. 3c, which has a transition layer of disorder with 1.5 nm in width. It is implied that the interface between the precipitates and α -Mg matrix transformed from coherent to incoherent with the increasing aging

Fig. 4 presents the variation in micro-hardness values of Mg–12Gd alloy as function of aging time. As seen from Fig. 4, the Mg–12Gd alloy exhibited a typical aging hardening response with the increasing aging

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