Effects of Y content on microstructures and thermal expansion behavior of Mg-Al-Y alloys

Hongbin Ma, Zhuohua Li and Jinhui Wang

Qinghai Provincial Key Laboratory of New Light Alloys, Qinghai Provincial Engineering Research Center of High Performance Light Metal Alloys and Forming, Qinghai University, Xining 810016, People’s Republic of China

* Author to whom any correspondence should be addressed.

E-mail: ahui1004@163.com

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Abstract

Low thermal expansion alloys play an important role in the applications of high-precision instruments because of their excellent dimensional stability under thermal shock. Hence, the effect of Y content on coefficient of thermal expansion (CTE) of Mg-1Al-xY (x = 4, 6, 8, wt. %) alloys were systematically studied. The results showed that the volume fraction of second phase increased and the grain size of the Mg alloys decreased with the increases of Y content, resulting in the CTE of Mg-1Al-xY alloys decreased with the increases of Y content. The grain size of the Mg-1Al-6Y alloy was more suitable and the second phase distributed uniformly, which lead to the lowest CTE after multiple cycles. The experimental value and model predictions of CTE of alloys differ greatly at lower temperature and coincide well at high temperature. The XRD and EBSD results proved the existence of residual stress in the three alloys, which reduced the CTEs of the magnesium alloy. While the rotation of crystal grains reduces the residual stress in multiple thermal cycles, increasing the CTEs and stability of thermal cycling of Mg alloys. The pole diagram of Mg alloys revealed that number of thermal cycling has a great effect on the intensity of the texture.

1. Introduction

Magnesium alloys are the lightest commercial metal structural materials due to their advantages of high specific strength, excellent electrical and thermal conductivity, which make it widely used in aerospace, vehicles, electronic device field [1–8]. Studies on Mg alloys mainly focus on their mechanical properties. There were few reports on thermal expansion properties of Mg alloys. Under the action of temperature, if the coefficient of thermal expansion (CTE) of the interfaces of materials is mismatched, it will cause excessive stress in the components and shorten the service life of the devices [9–12]. Therefore, it is important to develop new magnesium alloys having low CTE to widen the utilization of Mg alloys in the aforementioned applications.

Several researchers have studied the thermal expansion behavior of Mg alloys [13–16]. A. Rudajevo et al found that the coefficient of thermal expansion (CTE) of the Mg-xLi binary alloys increases with increasing lithium content in the alloys [13]. T. Guo et al reported that the CTE of Mg–Si alloys decrease sharply with the increase of Si content, which is decreased by 30.8%, from 26.00 × 10–6/K of pure Mg to 17.98 × 10–6/K of Mg–4Si alloy at room temperature [14]. Yasuda et al confirmed that the CTE of Mg58Zn49Y3 containing long-period ordered stacking phase (LPSO) was smaller than that of Mg [15]. The CTE values of pure magnesium decreases with additions of Al and La were obtained by Amit Kumar et al [16]. These studies indicated that the CTE of the magnesium alloy can be diminished by the addition of alloying elements having low CTE. The rare earth element Y is a good choice to add into Mg alloys as it leads to increase the strength whilst retaining ductility of Mg alloys [17–21]. The researches on Mg-1Al-xY alloys mainly focuses on the mechanical properties, there were few studies on the effect of Y content on the microstructure and thermal expansion behavior of Mg alloys.

In this work, the microstructures and thermal expansion behaviors of Mg-1Al-xY (x = 4, 6, 8, wt. %) alloys were investigated. The effect of Y content on the CTE of as-extruded Mg-1Al-xY alloys with the belief that the
information provided will be useful in the selection of the Mg alloys in applications where stringent dimensional stability is a critical design criterion.

2. Experimental

The Mg-1Al-\(x\) \((x = 4, 6, 8)\) alloys were fabricated by melting and casting the raw materials, namely pure magnesium ingot (99.9%), pure aluminum ingot (99.9%), and Mg-20Y master alloy (88.5 wt% Mg + 19.5 wt% Y). The as-extruded Mg-1Al-\(x\) \((x = 4, 6, 8)\) alloys were obtained by using four-column type hydraulic press (IM-Y300). The extrusion temperature was 500 °C, the extrusion ratio was 13:1, and the extrusion rate was 0.2 mm/min. All samples were heat treated into a box-type resistance furnace to homogenize the structure. The
specimens for x-ray diffraction (XRD) and thermal expansion experiments were processed into appropriate sizes by using CABINET FW2UP wire EDM.

The XRD was used to analyze the phases of Mg alloys. The microstructures and elemental distribution of alloys were obtained by using scanning electron microscopy (SEM, Zeiss Merlin Compact) equipped with energy dispersive x-ray (EDX, Oxford Instruments). The thermal expansion performance of samples with a cylinder of \( \Phi 6 \times 25 \text{ mm} \) was measured within a temperature range from 30 \( ^\circ \text{C} \) to 300 \( ^\circ \text{C} \) by using a thermal dilatometer Netsch DIL402 PC. Thermal expansion tests of Mg alloys were carried out under argon protection with a gas flow rate of 70 ml min\(^{-1}\). The number of thermal cycling of the samples is 6, 12, and 18 times, respectively. The heating and cooling rate is 5 K min\(^{-1}\). The evolution of deformation texture was characterized by electron backscatter diffraction (EBSD), with the corresponding data analyzed by the Oxford HKL channel 5 software.

### 3. Results and discussion

Figure 1 shows the diffraction patterns of as-extruded Mg-1Al-xY (x = 4, 6, 8) alloys without thermal cycling. It was found that only \( \alpha \)-Mg and second phase \( \text{Al}_2\text{Y} \) can be detected in Mg-1Al-xY (x = 4, 6, 8) alloys, and the \( \alpha \)-Mg phase was the main phase.

The microstructures and elemental distributions of as-extruded Mg-1Al-xY (x = 4, 6, 8) alloys were acquired by SEM. The elemental distribution map of Mg, Al, and Y elements are shown in figure 2. The EDX mapping results of three alloys indicated that an amount of white particle \( \text{Al}_2\text{Y} \) presented a weak streamline distribution phenomenon due to the extrusion. It was consistent with the experimental result of XRD. The content of the \( \text{Al}_2\text{Y} \) phase gradually increases with the increase of Y content, which can effectively pin the movement of dislocations and play a role of dispersion strengthening during the deformation process. However, the second phases of the Mg-1Al-8Y alloy appeared coarseness and slight aggregation due to the excessive addition of Y, which is unfavorable to the thermal expansion properties of the Mg alloys.
The volume fraction of second phase increased and the grain size of the alloys decreased with the increases of Y content. Figure 3 is the EBSD gray scale image and the average grain sizes diagram of as-extruded Mg-1Al-xY alloys. The addition of Y content has an important influence on the microstructure of the magnesium alloys. From the grayscale images, the number of fine grains increased with the increases of Y content. That is because the number of Al2Y phases increases with increasing Y content. The second phases have a pinning effect during the extrusion deformation, which makes the grains broken and refined, so the fine grains are mainly distributed near the coarse second phases. From the comparison chart of average grain sizes, the average grain sizes of the alloys have decreased from 14.61 μm of Mg-1Al-4Y to 8.10 μm of Mg-1Al-8Y. It indicated that the average grain size of the alloy shows a downward trend with the increases of Y content. By observing the grain size of the EBSD grayscale image, the grain size distribution of Mg-1Al-6Y alloy is more uniform than that of Mg-1Al-8Y alloy.

The linear CTE is one of the main thermal properties of materials and an important index to evaluate the thermal stability of materials. The extruded materials are anisotropic, the size of the extruded bar is limited, therefore, this experiment mainly focuses on the linear CTE. The basic calculation is shown as follows:[22–24]

\[
\alpha = \frac{L_2 - L_1}{L_0(T_2 - T_1)} = \frac{\Delta L}{L_0\Delta T} \quad (T_2 > T_1)
\]

where \(\alpha\) is the CTE, \(L_1\) and \(L_2\) represent the length of the specimen at the temperature of \(T_1\) and \(T_2\), respectively. It should be noted that \(\alpha\) is not a constant, which is related to temperature, and usually increases with increase of the temperature. When comparing the CTE of alloy materials with different alloys, the average temperature range is usually selected to express its size [25]. The CTEs of the Mg-Al-Y alloy increased with increasing of temperature. According to the temperature-strain rate curve, the average CTE of each temperature range were calculated. The calculation formula of the material linear CTE is as follows:

\[
\alpha_{T,T_0} = \frac{L_T - L_0}{L_0(T - T_0)} = \frac{dL}{L_0dT}
\]

where \(\alpha_{T,T_0}\) is the average CTE, \(L_0\) is the length of the sample at temperature \(T_0\) (\(T_0\) is 30 °C), \(L_T\) is the length of the sample at temperature \(T\).

Figure 4 shows the linear average CTE-temperature curves of alloys with different Y content. It was found that the CTE of three alloys increased with the temperature elevated. The linear average CTE of alloy increase fast at low temperature, while increase slowly at high temperature. Comparing the three curves, the CTE of the materials show a decreasing trend with the increase of Y content. The number of Al2Y phases increased and the grain size of Mg-Al-Y alloys decreased with the increasing of the Y content. Meanwhile, the CTE of Al2Y is lower than that of Mg, which lead to a decreasing the CTEs of the Mg-Al-Y alloys. The largest CTE was acquired around 300 °C for all as-extruded samples. The overall thermal expansion behaviors of the alloys show a synergistic effect between the Mg matrix and the second phase at close to room temperature, thus, the CTE of alloys increase nearly uniformly [26]. While the temperature is higher than 150 °C, the vibration amplitude of each atom in the alloy material became larger, and the tensile stress of the original turned into compressive stress. The hard Al2Y phases hinder the thermal expansion behavior of the matrix due to the difference in the
CTE of the two phases. Therefore, the increase of the CTE of alloys slows down with the increasing of temperature.

It can be known from the rule of mixtures (ROM) that the CTE of the composites is approximately equal to the sum of the CTE of each phase in the alloy multiplied by its volume fraction. This rule is applicable to both composites and alloys [27–29]. According to the CTE calculation formula of alloy, the CTE of Mg-Al-Y alloys with different Y content can be calculated by following formula:

\[
\alpha_{\text{alloy}} = \alpha_{\text{mix}} + C = \sum \left( \frac{K_i}{E_i} \times i \% \right) + C
\]

where \(E_i\) is the bond energy of the base metal or alloy element, the unit is kcal/mol; \(K_i\) is the empirical constant related to the configuration of pure metal; \(C\) is the empirical constant related to the matrix, which magnesium alloy is 3.1 [30]. The parameters are shown in table 1.

The CTE of Mg-1Al-\(x\)Y (\(x = 4, 6, 8\)) alloys can be calculated according to the formula and related parameters, which is 26.68 \(\times 10^{-6}\) K\(^{-1}\), 26.44 \(\times 10^{-6}\) K\(^{-1}\) and 26.20 \(\times 10^{-6}\) K\(^{-1}\), respectively. The experimental value and model predictions of CTE of alloys differ greatly at lower temperature and coincide well at high temperature. The theoretical predictions of CTE decrease with the increase of Y content, which is consistent with the experimental results. The reason for the large deviation in the low temperature region is that the theoretical model cannot reflect the influence of the size and distribution of the second phase on the CTE of the alloys, and do not consider the change of solid solubility with temperature.

Figure 5(a) shows the thermal strain rate-time relationship curves of Mg-1Al-xY (\(x = 4, 6, 8\)) alloys, represented by the 18 thermal cycles curve. Figure 5(b) is obtained by selecting the lower end of the loop curve from the 14th to the 17th in figure 5(a) which marked with the red box. It can be clearly seen from the figure 5(b) that red strain rate curve of the Mg-1Al-6Y is the lowest, indicating that the Mg-1Al-6Y alloy has the best dimensional stability at temperature close to room temperature after multiple thermal cycles. The grain size of the alloys becomes smaller with the increase of Y content. The grain sizes of Mg-1Al-6Y and Mg-1Al-8Y are roughly the same. However, the Al\(_2\)Y with high hardness and low CTE is uniformly distributed in the Mg-1Al-6Y alloy and agglomerated in the Mg-1Al-8Y. Thus, Mg-1Al-6Y has better dimensional stability after multiple thermal cycles. The difference in CTE between Al\(_2\)Y and the Mg matrix results in a thermal compression stress at the interface of Al\(_2\)Y, which can effectively limit the thermal expansion performance of the matrix, thereby reducing the overall CTE of the alloys and increasing the dimensional stability of the materials.

| Table 1. Summary of calculation parameters of CTE of Mg-Al-Y alloys. |
|------------------------|-----------------|--------|
| \(K_i\) (Kcal/mol) | \(E_i\) (Kcal/mol) |
| Mg        | 850             | 35.3   |
| Al        | 1600            | 76.9   |
| Y         | 1200            | 97.6   |

Figure 5. Mg-1Al-xY (\(x = 4, 6, 8\)) alloy strain rate-time relationship curve. (a) Mg-1Al-xY (\(x = 4, 6, 8\)) alloys strain rate-time relationship curve; (b) Enlarged picture in red frame.
Table 2 is the calculation of the strain value statistics and cumulative strain of Mg-1Al-4Y, Mg-1Al-6Y, and Mg-1Al-8Y alloys at 50 °C for 6, 12 and 18 cycles, respectively. It was found that the accumulated residual strain value of Mg-1Al-6Y alloy is lower than that of the other two alloys after multiple cycles. Since the residual strain value of Mg-1Al-6Y alloy is relatively low, it can be judged that the dimensional stability of Mg-1Al-6Y alloy is better than that of the other two alloys. Comparing the cumulative strain of three alloys, it was found that the cumulative residual strain value of Mg-1Al-6Y alloy is the lowest, and the cumulative residual strain values of Mg-1Al-4Y and Mg-1Al-8Y are relatively close at different cycles. This is because the grain size of the Mg-1Al-6Y alloy was more suitable and the second phase distributed uniformly, while the element Y of the Mg-1Al-8Y alloy is excessively added, which leads to more serious segregation and aggregation of the second phase in alloys, resulting in thermal expansion properties of the alloys decreased. Compared the number of cycles of alloys, the cumulative residual strain of 12 cycles is the lowest, and the values of alloys’ 6 and 18 thermal cycles are relatively close.

To verify the existence of residual stress, calculate the deviation of the diffraction peak of XRD. It is extremely important to accurately determine the position of the diffraction peak. A comparison diagram of XRD

![Figure 6. Strain-temperature curves of Mg-1Al-6Y alloy at 6, 12, and 18 cycles.](image-url)

| Table 2. Cumulative strain values of Mg-1Al-4Y (x = 4, 6, 8) alloys. |
|---------------------------------------------------------------|
| Strain (×10⁻⁴)                                               |
| | Mg-1Al-4Y | Mg-1Al-6Y | Mg-1Al-8Y |
| 1st Cycle | 4.20      | 4.76      | 4.66      |
| 6th Cycle | 5.28      | 4.35      | 5.34      |
| 12th Cycle | 3.54     | 3.08      | 4.29      |
| 18th Cycle | 5.42     | 4.32      | 5.48      |
| 6 Cycles | 1.08     | −0.41     | 0.68      |
| 12 Cycles | −0.66    | −1.68     | −0.37     |
| 18 Cycles | 1.22     | −0.44     | 0.82      |
diffraction peaks of Mg-1Al-\(x\text{Y}\) (\(x = 4, 6, 8\)) alloy under different thermal cycling was showed in figure 7. It was found that only \(\alpha\)-Mg and second phase \(\text{Al}_2\text{Y}\) can be detected in Mg-1Al-\(x\text{Y}\) (\(x = 4, 6, 8\)) alloys after thermal cycles, and the \(\alpha\)-Mg phase was still the main phase. It can be seen from the enlarged diagram that the graph line goes from bottom to top. As the number of cycles increases, the x-ray diffraction peak of the \(\text{Al}_2\text{Y}\) has a slight shift to the left. According to the Bragg diffraction equation: \(2\sin \theta = n\lambda\), the x-ray wavelength \(\lambda\) does not change. When the angle \(\theta\) shifts to the left, the value of \(\sin \theta\) will decrease. Therefore, the value of interplanar spacing \(d\) increases, which means that the lattice constant increases. The greater degree of lattice distortion indicates that multiple thermal cycling will cause the accumulation of residual stress in the material. This phenomenon occurs in the three alloys. The existence of residual stress in the three alloys, which reduced the CTEs of the magnesium alloy. While the rotation of crystal grains reduces the residual stress in multiple thermal cycles, increasing the stability of thermal cycling of Mg alloys. Figure 8 presented the EBSD images of Mg-1Al-6Y.
alloy with different thermal cycling. From the perspective of grain orientation distribution, the arrangement of grains of Mg alloys were still streamlined. The average grain size of the alloy gradually increases with the increase of the number of thermal cycling, which because of the growth of grains in alloy during the thermal cycling. Due to the hindrance of the second phase, the large-sized grains did not continue to grow, but the small-sized grains gradually grew into equiaxed crystals. The average grain size of alloys after 12 thermal cycling and 18 thermal cycling were almost same, but the grain size distribution after the 12th cycle was relatively more uniform, which is also beneficial to the dimensional stability of the Mg-1Al-6Y.

**Figure 8.** Grain orientation distribution map of Mg-1Al-6Y alloy under different thermal cycling (a) 0 cycles, (c) 6 cycles, (e) 12 cycles, (g) 18 cycles; Grain size distribution map of Mg-1Al-6Y alloy under different thermal cycling: (b) 0 cycles, (d) 6 cycles, (f) 12 cycles, (h) 18 cycles.
Figure 9. Stress distribution of Mg-1Al-6Y alloy. (a) Extruded and annealed; (b) 6 cycles; (c) 12 cycles; (d) 18 cycles.

Figure 10. Pole diagram of Mg-1Al-6Y alloy after 0, 6, 12, and 18 thermal cycling. (a) 0 times; (b) 6 times; (c) 12 times; (d) 18 times.
The internal stresses of the Mg-1Al-6Y alloy were obtained by EBSD technique. The stress distributions of Mg-1Al-6Y alloy with different thermal cycling were shown in figure 9. The internal stresses of the Mg-1Al-6Y alloy were significantly reduced with the increase of number of thermal cycles. The reason was that the grains of alloy grow and rotate as the number of cycles increases, which ultimately leads to the release of residual stress.

The evolution of micro-textures of the as-extruded Mg-1Al-6Y with different thermal cycling along the extrusion direction were studied by pole diagram. The texture has very important effects on thermal expansion behaviors of Mg alloys, which is related to anisotropy of crystals [31, 32]. According to the pole figure in figure 10(a), as-extruded Mg-1Al-6Y alloy had a typical extruded fiber texture with a max texture intensity of 9.29. It was found that the intensity of the texture decreased after multiple thermal cycles. The intensity of the texture increased first and then decreased with the increase of the number of thermal cycling, which means the number of thermal cycling has a great influence on the intensity of the texture. It was considered that the crystal grains have rotated during the thermal cycling.

4. Conclusion

The Mg-1Al-xY (x = 4, 6, 8) alloys were fabricated by melting and casting method, the microstructures and thermal expansion behaviors of Mg-1Al-xY (x = 4, 6, 8) alloys were studied. It was found that the CTE of three alloys increased with the temperature elevated. The overall thermal expansion behaviors of the alloys show a synergistic effect between the Mg matrix and the Al2Y phase at close to room temperature. The CTE of Mg-1Al-6Y alloy were significantly reduced with the increase of number of thermal cycling. The reason was that the grains of alloy grow and rotate as the number of cycles increases, which ultimately leads to the release of residual stress. This effect was especially significant at close to room temperature. The CTE of three alloys increased with the temperature elevated. The overall thermal expansion behaviors of the alloys show a synergistic effect between the Mg matrix and the Al2Y phase at close to room temperature. The CTE of three alloys were calculated by ROM, which coincide well with experimental result at high temperature. The results of thermal cycling showed that the grain size of the Mg-1Al-6Y alloy was more suitable and the second phase distributed uniformly, resulting in good thermal stability properties. The accumulation of residual stress occurs in the three alloys after the multiple thermal cycling. Hence, the Y content has a great influence on the thermal expansion behaviors of Mg-Al-Y alloys.

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Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

ORCID iDs

Hongbin Ma https://orcid.org/0000-0003-4208-8891
Jinhui Wang https://orcid.org/0000-0003-3658-360X

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