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TEM OBSERVATION OF AGE-HARDENING PRECIPITATION IN Mg-Gd-Y ALLOYS AS DIFFERENT Gd/Y RATIO

ZASTOSOWANIE MIKROSKOPII TEM DO OBSERWACJI WYDZIELEŃ PO UTWARDZANIU WYDZIELENIOWYM W STOPACH Mg-Gd-Y O RÓŻNYCH STOSUNKACH Gd/Y

In this study, the early stage of aging in Mg-Gd-Y alloys has been observed by transmission electron microscopy (TEM), high angle annular dark field – scanning transmission electron microscopy (HAADF-STEM) and calculations of images and electron density and bond overlap population (BOP) by first principal to understand the origin of precipitation in this alloy. The small hexagon of 0.37 nm is the first precipitate in this alloy, and this is the evidence of short range ordering of D0₁₉ structure. This is referred as the pre β'' -phase. In the peak aged condition, β' phase with bco structure was mainly observed.

Keywords: Mg-RE alloy, precipitation behavior, TEM, HAADF-STEM

W pracy przedstawiono mikroskopowe obserwacje TEM wczesnego etapu starzenia stopów Mg-Gd-Y. Ponadto badania prowadzono z użyciem skaningowej transmisyjnej mikroskopii elektronowej. Wykorzystano zjawisko niekoherentnego rozpraszania w ciemnym polu i materiał obrazowano przy pomocy detektora pierścieniowego, który zbiera elektrony rozproszone pod dużymi kątami (HAADF-STEM). Przeprowadzono także analizę obrazów, obliczenie gęstości elektronowej i ilości nakładających się wiązań w celu zrozumienia źródła wydzielenia w ww. stopie. Pierwszym wytrąceniem w stopie jest mały sześciokąt o wielkości 0,37 nm, a dowodem tego jest uporządkowanie krótkiego zasięgu struktury D0₁₉.

1. Introduction

The magnesium alloy containing rare earth element is known to show good heat resistance. Particularly, it was reported that Mg-Gd alloys containing above 10mass% Gd are show a clear age-hardening at high temperature [1]. Mg-Gd-Y alloys have been developed for practical Mg alloys by Kamado et. al. to reduce the density of alloy and these alloys have a good creep resistance, even 523-573 K. In our previous study, Mg-Gd-Y alloys show the mono-layer structure has been discovered before β'' phase with D0₁₉ structure in the aged sample at 473 K, and the β'' and β' phases with bco structure co-existed at the peak aged condition [2]. Recently, Nishijima et.al. detailed examinations on the precipitation behaviours of Mg-Gd and Mg-Y alloy by high angle annular dark field-scanning transmission electron microscopy (HAADF-STEM) technique [3,4]. They concluded that the arrangement of bright dots indicates the short range ordered state and the β' phase nuclide in the short range ordered structure. And they have a doubt for the existence of the β'' phase with D0₁₉ structure. Moreover, they presented a new structure model for the β' phase, an Mg₇RE-type structure different from the previously proposed Mg₁₅RE-type. However, the precipi-

tation behavior of the Mg-Gd-Y alloys at early stage of aging after quenching was not understood clearly. In this study, the early stage of aging in Mg-Gd-Y alloys has been observed by transmission electron microscopy (TEM), HAADF-STEM and calculations of images and electron density and bond overlap population (BOP) by first principal to understand the origin of precipitation in this alloy.

2. Experimental procedure

Experimental alloy was a Mg-2.9%Gd-0.8%Y alloy (at.%, hereinafter called Gd:Y=3:1 alloy), Mg-2.0%Gd-1.7%Y alloy (Gd:Y=1:1 alloy) and Mg-1.0%Gd-2.3%Y alloy (Gd:Y=1:3 alloy). The ingot was homogenized at 773 K for 43.2 ks, and then hot rolled at 773 K to 1 mm thickness. The sample was capsulated into pyrex glass tube with argon gas. Then, it was solution heat-treated at 773 K for 3.6 ks and quenched in hot water at 353 K, and aged in a silicone oil bath at 473 K. TEM specimens were cut from plate samples and thinned by the twin-jet electro polishing technique using an electrolyte of 10 percent perchloric acid-ethanol solution at about 243 K. HRTEM observation

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was performed by using a TOPCON EM-002B type, operated at 120kV. HAADF-STEM observation were performed by using a JEOL ARM-200F operated at 200 kV. HRTEM and HAADF-STEM images were calculated using the multi-slice method by Mac Tempus and the electron density and BOP value was calculated using the DV-X α program [5].

3. Results and discussion

Fig. 1 shows TEM bright field images in (a) as-quenched and (b) peak aged conditions of Gd:Y=3:1 alloy. The morphology of β' phase can not be seen in the bright field images of Fig. 1(a), and the typical contrast of the platelet β' phase with base centered orthorhombic (bco) structure has appeared in the peak-aged sample of Fig. 1(b). The Gd:Y=3:1 alloy aged for 7.2ks included the β' phase because of split of diffraction spots in the its SAED pattern. Fig. 2 shows HRTEM image of Gd:Y=3:1 alloy aged at 473 K for 7.2 ks. HRTEM image of the β'' phase with D0₁₉ structure and β' phase with bco structure. In the specimen in early stage of aging after quenching, the monolayer and β'' with D0₁₉ structure phase were observed. In the peak aged specimen, β' phase with bco structure were formed dominantly.

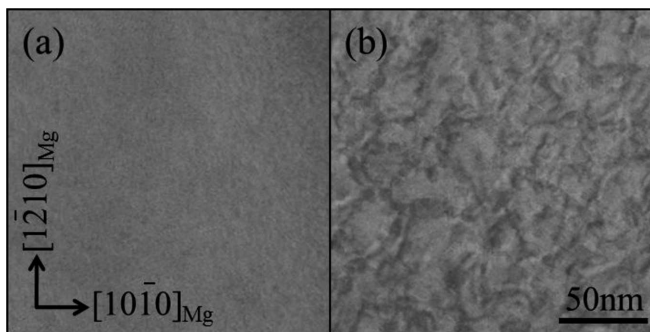


Fig. 1. TEM bright field images obtained for Gd:Y=3:1 alloy aged at 473K for some conditions; (a) as quenched and (b) peak aged

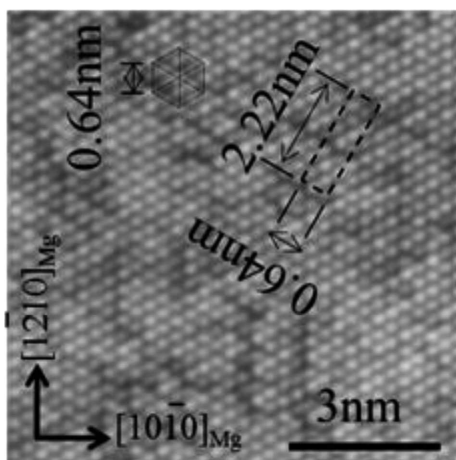


Fig. 2. HRTEM image of Gd:Y=3:1 alloy aged at 7.2ks for 473K

4. Conclusions

TEM and HAADF-STEM observation was performed to clarify the early stage of precipitation in Mg-Gd-Y alloys as different Gd/Y ratio.

1. The diffuse scattering was obtained in as-quenched samples in all alloys by SAED pattern.

2. The small hexagon of 0.37 nm is the first precipitate, and this is the evidence of short range ordering close to D0₁₉ structure.

3. We concluded that the proposed precipitation sequence is as follows; S.S.S.S. \rightarrow pre β'' phase having SRO \rightarrow β'' \rightarrow β' .

REFERENCES

- [1] S. Iwasawa, Y. Negishi, S. Kamado, Y. Kojima, R. Ninomiya, J. Japan Institute of Light Metals. **44**, 506-511 (1994).
- [2] T. Kawabata, D. Nakagawa, S. Saikawa, J. Nakamura, S. Ikeno, K. Matsuda, Materials Transactions **54**, 225-230 (2013).
- [3] M. Nishijima, K. Hiraga, Materials Transactions **48**, 10-15 (2007).
- [4] M. Nishijima, K. Yubuta, K. Hiraga, Materials Transactions **48**, 84-87 (2007).
- [5] H. Adachi, Y. Owada, I. Tanaka, H. Nakamatsu, M. Mizuno, Introduction of DV-X α Method, 3rd ed., (Sankyo Shuppan, Tokyo, 2001).