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Intermetallic compounds and antiphase domains in Al/Mg compound casting

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ABSTRACT

Interfacial microstructure of Al/Mg light metals couples prepared by compound casting process was studied. The interface consists of three different layers. The layers adjacent to the Al and Mg base metals are mainly composed of Al_3Mg_2 intermetallic compound and ($Al_{12}Mg_{17}+\delta$) eutectic structure, respectively, and the middle layer is mainly composed of $Al_{12}Mg_{17}$ intermetallic compound. A network of periodic antiphase domains (APDs) was detected within the $Al_{12}Mg_{17}$ intermetallic compound at the middle layer. The size of the APDs was not constant throughout the layer containing this intermetallic compound and was depended on the amount of deviation from the stoichiometric proportion (SP) of the $Al_{12}Mg_{17}$ intermetallic compound. It was found that by increasing the deviation from the SP the size of the APDs of the $Al_{12}Mg_{17}$ intermetallic compound decreases and the density of the antiphase boundaries (APBs) increases.

1. Introduction

Increasing industrial interests for using cost-effective routes for processing combined structures has led to the developing of some economic methods, such as compound casting and twin roll casting. Recently, Bae et al. [1] have used twin roll casting for production of lightweight Al/Mg sheet metals by simultaneous casting, cladding, and rolling processes. Compound casting is an economic process which enables forming the shape of the product and bonding other parts with complex structures to castings at the same time. In this process two metallic materials – one in solid state and the other liquid – are brought into contact with each other. In this way, a diffusion reaction zone between the two materials and thus a continuous metallic transition from one metal to the other is formed [2]. Although many researchers have studied the interfacial microstructure of Al/Mg joint in different joining and bonding methods [3–7], formation of the interface of Al/Mg couples in the compound casting process is still a relatively unexplored area. Al_3Mg_2 and $Al_{12}Mg_{17}$ are reported to be the main intermetallic compounds formed at Al/Mg interface in different joining and bonding processes of these light metals [3,6,7]. These intermetallic compounds and their structures determine the performance of the joint. Despite many researchers have studied the interfacial microstructure of the Al/Mg joint in different joining processes, few works have been conducted to study the submicron structures of Al–Mg intermetallic compounds formed at the Al/Mg interface. APBs are one of the planar crystal imperfections which usually occur in ordered structures [8,9]. The APBs which are a special type of stacking fault [9,10] separate domains with opposite relative displacements of atoms and otherwise identical physical properties named APDs [8]. APBs constitute a network of structural defects of a crystal and because of the surface tension of the interfaces (surface free energy), they are in global disequilibrium with the crystal [9]. The APDs are usually formed as a result of deformation or ordering in the intermetallic compounds [9–13]. The later is usually referred as thermal APDs [13]. This paper deals with the formation of the Al–Mg intermetallic compounds and APDs at the interface of the Al/Mg couples prepared by the compound casting process. The reason for this work is to report on the microstructure of the interface, and on the relation between domain size and chemical composition.

2. Experimental procedures

In order to prepare the Al/Mg couples by the compound casting process commercially pure Al (99.58 wt.%) and commercially pure Mg (99.85 wt.%) were used. Cylindrical inserts with 20 mm diameter and 100 mm height were machined from Al ingots. Their

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surfaces were ground with silicon carbide papers up to 1200 grit, then rinsed with acetone and placed within a cylindrical cavity of a CO_2 sand mold with 30 mm diameter and 80 mm height. Mg ingots were melted in a steel crucible under the Foseco MAGREX 36 covering flux, to protect Mg melt form oxidation. The molten Mg was cast around the Al inserts at 700 °C under normal atmospheric conditions (Fig. 1).

After casting process, microstructure characterization of the Al/Mg joint was carried out on cross-section perpendicular to the cylindrical Al insert using a JEOL JSM-7000F field emission gun scanning electron microscope (FE-SEM) equipped with an ATW2 Oxford Instrument energy dispersive X-ray spectroscopy (EDS) detector. Specimens for SEM investigations were prepared with a final oxide polish (OP-S) and etched by a 1% HF distilled water solution on the Al side and a 1% HNO_3 alcohol solution on the Mg side. Thin foils for the TEM observations were prepared by ion milling using a GATAN-691 Precision Ion Polishing System. The presence of APBs was verified by means of two-beam dark field imaging technique in a JEOL JEM-2100F transmission electron microscope (TEM) operating at 200 kV. Quantitative analysis was carried out to determine the APD sizes at different areas of the intermetallic compounds formed at the interface.

3. Results and discussion

Fig. 2a shows a typical SEM micrograph and its corresponding EDS line scan from the interfacial microstructure of the Al/Mg joint in the compound casting process. As can be seen a relatively uniform interface without any visible defect has been formed along the Al/Mg interface.

The microstructure of the interface is composed of three different layers. According to the EDS line scan in Fig. 2a, the layers adjacent to the Al and Mg base metals are rich in Al and Mg, respectively. In addition a concentration gradient of Al and Mg elements can be seen at the middle layer. The elemental analysis conducted in the different layers formed at the interface, showed that the layer adjacent to the Al base metal consists of about 60 at.% Al and 40 at.% Mg. The SP of Al to Mg is 3:2, such a composition corresponds to the Al_3Mg_2 intermetallic compound. The layer adjacent to the Mg base metal has a eutectic structure composed of about 27 at.% Al and 73 at.% Mg. Combining with the Al–Mg binary phase diagram (Fig. 3a), this layer was estimated to consist of the $Al_{12}Mg_{17}$ intermetallic compound and Mg solid solution (δ). Existence of such constituents at the interface has been proved by using X-ray diffraction (XRD) in our previous study [14].

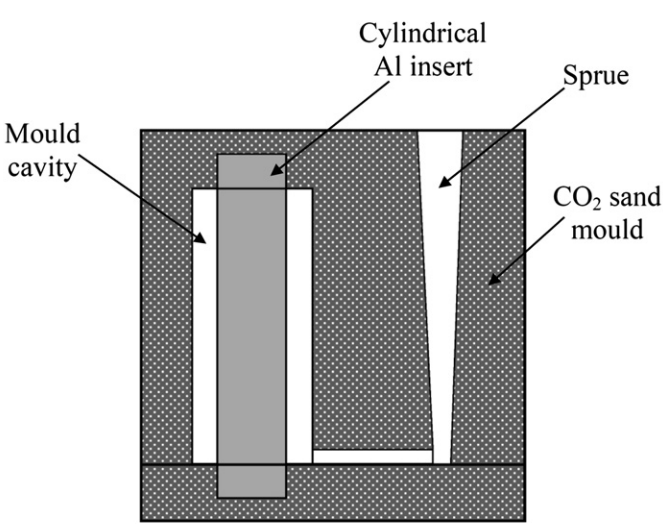

Fig. 1. Schematic sketch of the mold used for the casting process.

Unlike the layers adjacent to the Al and Mg base metals, which almost have constant chemical compositions, the concentrations of the Al and Mg elements for different areas of the middle layer are not constant and vary from about 49 at.% Al–51 at.% Mg to 41 at.% Al–59 at.% Mg at the left and right side of this layer, respectively. Considering the relatively wide range of the composition for the $Al_{12}Mg_{17}$ intermetallic compound in the Al–Mg phase diagram (45–60 at.% Mg) shown in Fig. 3a and non-equilibrium condition for formation of the interface in the compound casting process, it seems that the middle layer is mainly composed of the $Al_{12}Mg_{17}$ intermetallic compound.

More detailed observations by the FE-SEM (Fig. 2b–d) and TEM (Fig. 4a,b) in high magnifications, revealed a submicron structure for the middle layer of the Al/Mg interface, which seems to be related to the APDs formed during the non-equilibrium solidification of the interface. HRTEM image in Fig. 4c recorded from the area between two neighbor domains, shows atomic scale structure of an APB in the $Al_{12}Mg_{17}$ intermetallic compound.

Selected area diffraction patterns (SADs) shown in Fig. 4d,e which are taken at different diffraction lengths from a single domain and multiple domains in Fig. 4a, respectively, confirm the submicron structure in the middle layer is related to the body-centered cubic $Al_{12}Mg_{17}$ intermetallic compound with lattice parameter of 1.054 nm. Fig. 4f which is a higher magnification view of the Fig. 4e indicates splitting of the diffraction spots of the SAD obtained from multiple domains shown in Fig. 4a. The splitting of the spots in SAD is attributed to the formation of APBs within the submicron structure of the $Al_{12}Mg_{17}$ intermetallic compound. Several researchers have reported spot splitting of diffraction patterns due to presence of APBs in the ordered structures [17–19].

On the other hand according to the SEM micrographs shown in Fig. 2b–d, the size of the APDs is not constant and is depended on the chemical composition of the $Al_{12}Mg_{17}$ intermetallic compound at different areas of the middle layer. As a result, by increasing the proportion of Mg:Al of the $Al_{12}Mg_{17}$ intermetallic compound from the left to right side of the middle layer, the size of the APDs increases and consequently the density of the APBs decreases. Based on the linear quantitative analysis and measuring the mean intercept length of the APD boundaries (APBs) [20], the average APD sizes for the SEM micrographs shown in Fig. 2b–d, are 47, 106 and 130 nm respectively.

This is in agreement with the TEM micrographs shown in Fig. 4a,b. As can be seen in these figures, the size of the APDs for the TEM micrograph shown in Fig. 4b with higher proportion of Mg:Al (Fig. 4h) is considerably more than that is for the TEM micrograph shown in Fig. 4a with lower proportion of Mg:Al (Fig. 4g). Comparing the range of chemical composition for the $Al_{12}Mg_{17}$ intermetallic compound formed at the middle layer (about 49 at.% Al–51 at.% Mg to 41 at.% Al–59 at.% Mg) with the stoichiometric composition of the $Al_{12}Mg_{17}$ intermetallic compound (41.38 at.% Al–58.62 at.% Mg), and considering the concentration gradient of the Al and Mg elements at the middle layer of the interface (Fig. 2a), it can be concluded that the density of the APBs is affected by deviation from the SP of the $Al_{12}Mg_{17}$ intermetallic compound. The density of the APBs at the right side area of the $Al_{12}Mg_{17}$ intermetallic compound at the middle layer (Fig. 2d) which has a chemical composition near the SP (about 41 at.% Al–59 at.% Mg, i.e. 0.65% deviation from the SP) is considerably lower than the left side of this layer (Fig. 2b), with a chemical

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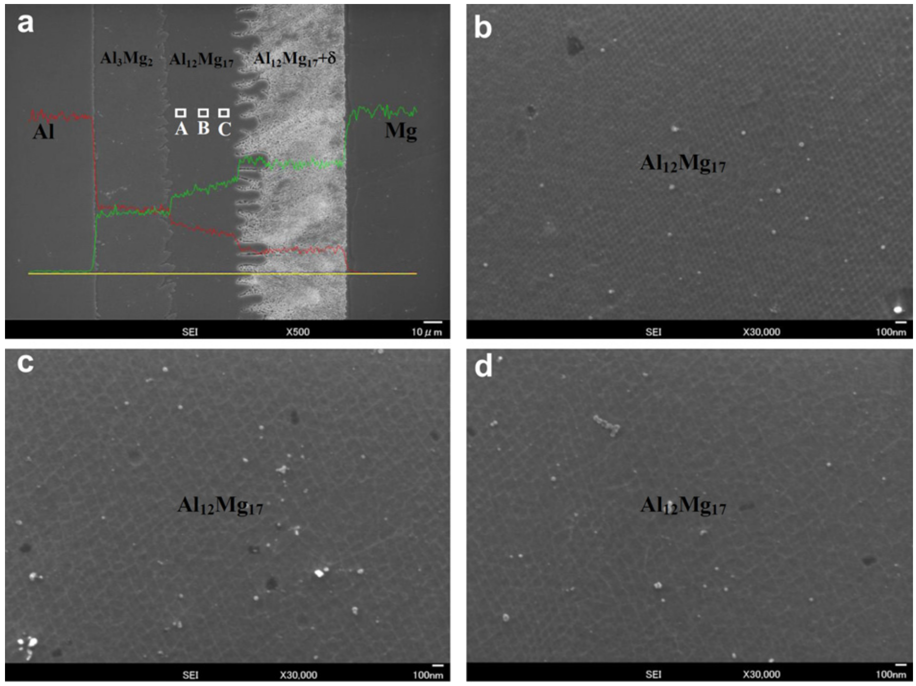

Fig. 2. SEM micrographs from the (a) interfacial microstructure of the Al/Mg couple prepared by the compound casting process and its corresponding EDS line scan, (b) area marked A in Fig. 2a, (c) area marked B in Fig. 2a, (d) area marked C in Fig. 2a.

composition of (about 49 at.% Al–51 at.% Mg, i.e. 12.99% deviation from the SP). The $Al_{12}Mg_{17}$ intermetallic compound has a relatively complex crystal structure with a $Fd\overline{3}m$ space group [16]. The structure of this intermetallic compound is based on a body-centered packing of Friauf polyhedra with Al on the T sites and Mg on the D sites (Fig. 3b) [16]. Entering more than SP of the Al atoms with different atomic radius than the Mg element into the crystal structure of the $Al_{12}Mg_{17}$ intermetallic compound, can bring about increasing the internal energy and instability of the crystal structure. In order to meet the equilibrium condition, a submicron structure composed of APDs and APBs is formed within the $Al_{12}Mg_{17}$ intermetallic compound during the solidification. In other words, based on the conservation of energy, increasing the internal energy of the crystal due to extra Al atoms in the crystal structure will be spent by surface free energy of the APBs surrounding the APDs.

Studying the $Al_{12}Mg_{17}$ intermetallic compound within the eutectic structure formed due to $L \xrightarrow{437\,^{\circ}C} Al_{12}Mg_{17} + \delta$ eutectic transformation [15] adjacent to the Mg base metal revealed no submicron structure same as that seen for the $Al_{12}Mg_{17}$ intermetallic compound at the middle layer. This indicates that, the deviation from SP is the main reason for formation of the APDs in the $Al_{12}Mg_{17}$ intermetallic compound formed at the middle layer. Furthermore for the Al_3Mg_2 intermetallic compound which has a relatively narrow chemical composition range in the Al–Mg binary phase diagram (Fig. 3a) and almost is formed at constant chemical composition adjacent to the Al base metal (Fig. 2a), no submicron structure was detected.

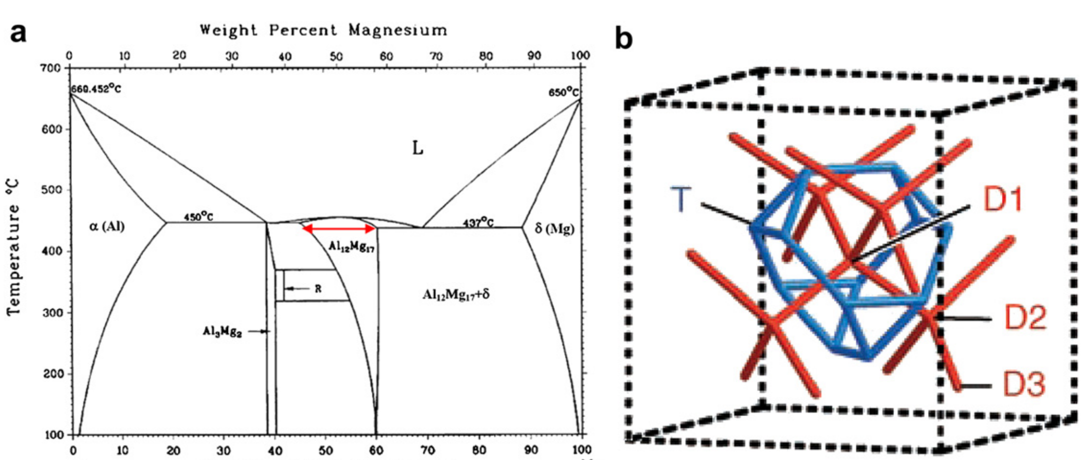

Fig. 3. (a) Al–Mg binary phase diagram [15], (b) Crystal structure of the $Al_{12}Mg_{17}$ intermetallic compound [16].

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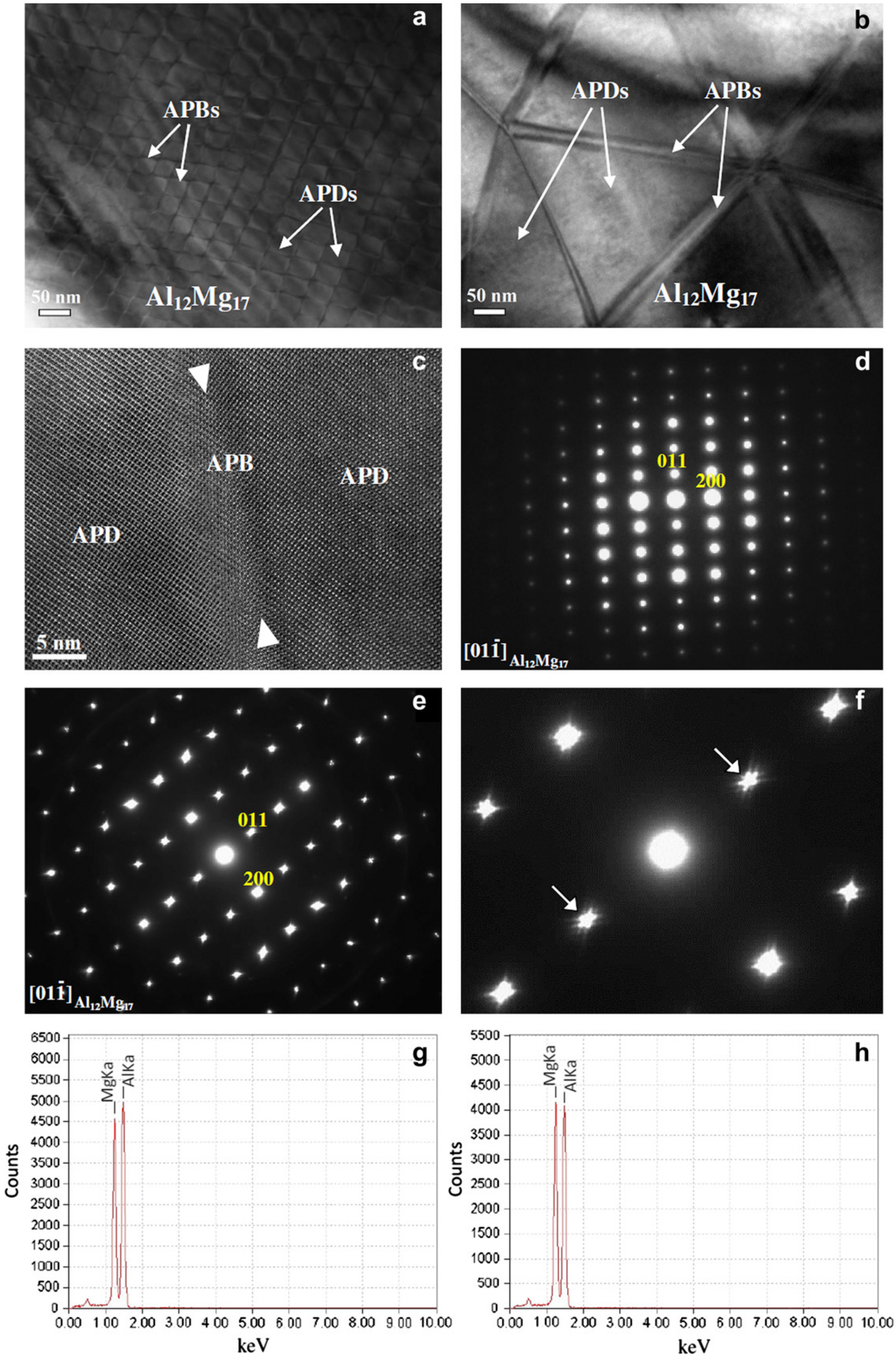

Fig. 4. (a,b) Bright-field TEM micrographs of the $Al_{12}Mg_{17}$ intermetallic compound in two different chemical compositions, (c) HRTEM image, showing the atomic scale structure of an APB in $Al_{12}Mg_{17}$ intermetallic compound, (d,e) SADs from a single domain and multiple domains in Fig. 4a respectively, (f) Magnified view of the SAD shown in Fig. 4e, (g,h) EDS analysis corresponding to the TEM micrographs shown in Fig. 4a,b respectively.

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4. Conclusions

Interfacial microstructure of the Al/Mg light metals couples prepared by the compound casting process consists of three different layers. The layers adjacent to the Al and Mg base metals are mainly composed of the Al_3Mg_2 intermetallic compound and the $(Al_{12}Mg_{17}+\delta)$ eutectic structure, respectively, and the middle layer is mainly composed of the $Al_{12}Mg_{17}$ intermetallic compound. A submicron structure composed of APDs and APBs is formed within the $Al_{12}Mg_{17}$ intermetallic compound at the middle layer due to deviation from the SP. Increasing the deviation from the SP brings about increasing the size of the APDs and decreasing the density of the APBs formed within the $Al_{12}Mg_{17}$ intermetallic compound.

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References

[1] Bae JH, Prasada Rao AK, Kimc KH, Kim NJ. Cladding of Mg alloy with Al by twin-roll casting. Scripta Mater 2011;64:836–9.
[2] Volder JP. An overview of a number of casting processes. Aluminum 1993;4: 11–6.
[3] Liu LM, Wang HY, Zhang ZD. The analysis of laser weld bonding of Al alloy to Mg alloy. Scripta Mater 2007;56:473–6.
[4] Liu L, Tan J, Liu X. Reactive brazing of Al alloy to Mg alloy using zinc-based brazing alloy. Mater Lett 2007;61:2373–7.
[5] Liu L, Liu X, Liu S. Microstructure of laser-TIG hybrid welds of dissimilar Mg alloy and Al alloy with Ce as interlayer. Scripta Mater 2006;55:383–6.
[6] Kostka A, Coelho RS, dos Santos J, Pyzalla AR. Microstructure of friction stir welding of aluminium alloy to magnesium alloy. Scripta Mater 2009;60: 953–6.
[7] Dietrich D, Nickel D, Krause M, Lampke T, Coleman MP, Randle V. Formation of intermetallic phases in diffusion-welded joints of aluminium and magnesium alloys. J Mater Sci 2010;46:357–64.
[8] Umantsev A. Thermal drag of the antiphase domain boundary motion. Acta Mater 1998;46:4935–9.
[9] Allen SM, Thomas EL. The structure of materials. 1st ed. New York: John Wiley & Sons; 1998.
[10] Morniroli JP. CBED and LACBED analysis of stacking faults and antiphase boundaries. Mater Chem Phys 2003;81:209–13.
[11] Heggen M, Feuerbacher M, Lange T, Urban K. Microstructural analysis of plastically deformed icosahedral Zn–Mg–Dy single quasicrystals. J Alloy Compd 2002;342:330–6.
[12] Lou BY, Zhang X, Liu M, Mao Z. TEM observation of DO_3 structure in Fe_3Al alloy with Mn addition. J Mater Sci 1999;34:1025–30.
[13] Venkateswaran SP, Nuhfer NT, De Graef M. Anti-phase boundaries and magnetic domain structures in Ni_2MnGa-type Heusler alloys. Acta Mater 2007;55:2621–36.
[14] Hajjari E, Divandari M, Razavi SH, Emami SM, Homma T, Kamado S. Dissimilar joining of Al/Mg light metals by compound casting process. J Mater Sci 2011; 46:6491–9.
[15] Alloy phase diagrams, ASM Handbook. 9th ed., vol. 3. Materials Park, OH: ASM International; 1995.
[16] Fredrickson DC, Lee S, Hoffmann R. Interpenetrating polar and nonpolar sublattices in intermetallics: the $NaCd_2$ structure. Angew Chem Int Ed 2007; 46:1958–76.
[17] Zhu J, Cowley JM. Microdiffraction from antiphase domain boundaries in Cu_3Au. Acta Cryst 1982;A38:718–24.
[18] Donnadieu P, Proult A, Ricolleau C. The role of defects in the crystal/quasicrystal transformation. Bull Mater Sci 1997;20:509–17.
[19] Lizzit S, Baraldi A, Grütter Ch, Bilgram JH, Hofmann Ph. The surface phase transition and low-temperature phase of α-Ga (010) studied by SPA-LEED. Surf Sci 2009;603:3222–6.
[20] Rase CL, Mikkola DE. Effect of excess Au on antiphase domain growth in Cu_3Au. Met Trans A 1975;6:2267–71.

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