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Intermetallics formed at interface of ultrafine grained Al/Mg bi-layered disks processed by high pressure torsion at room temperature

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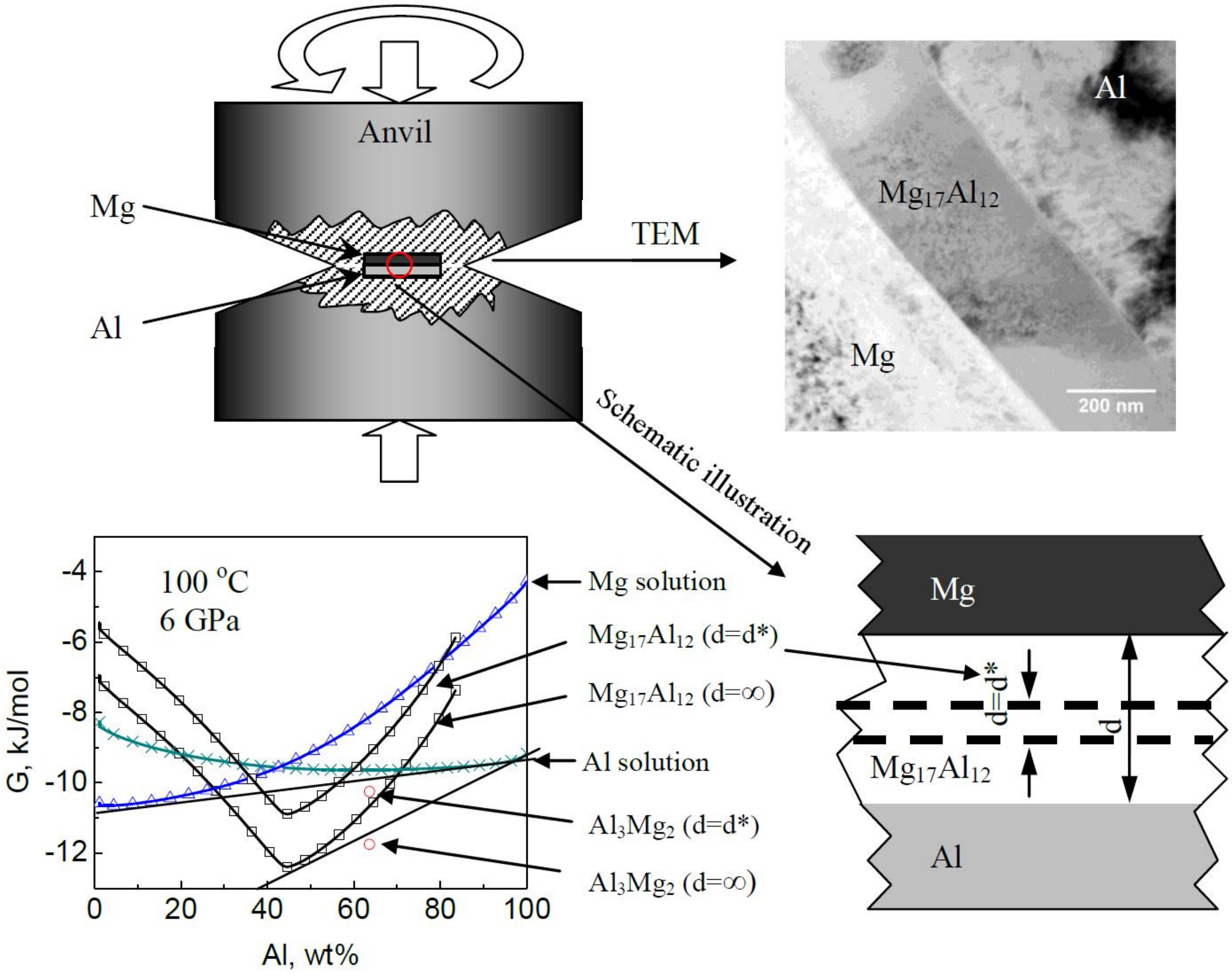
Graphical Abstract



Key words: high pressure torsion, interfacial energy, intermetallics, $Mg1_{17}Al_{12}$, multilayer structure, interfaces

- A Mg/Al bilayered disk is bonded by high pressure torsion at room temperature.
- A very thin layer of γ-$Mg_{17}Al_{12}$ of 260 nm in thickness is formed at the interface.
- Forming γ phase at low temperature is due to high pressure and large strain, etc.
- No other phases were detected, which is attributed to high interfacial energies

Abstract

In this work, Mg/Al bi-layered disks were fabricated by high pressure torsion (HPT) at room temperature. A 0.26 μm layer of γ-$Mg_{17}Al_{12}$ intermetallic formed at the interface of Mg/Al disks. This is very different to the interface of Mg/Al produced by other techniques such as diffusion bonding and friction stir welding where both γ-$Mg_{17}Al_{12}$ and β-Al_3Mg_2 form and the β-Al_3Mg_2 layer is much thicker than the γ-$Mg_{17}Al_{12}$ layer. This unusual phenomenon is attributed to the interfacial energies of intermetallics and the effect of room temperature HPT on diffusion of Al in Mg and the γ-$Mg_{17}Al_{12}$ layer.

1. Introduction

Mg/Al multi-layered composites may achieve an excellent combination of properties derived from the individual metals provided they are fabricated with a sound and reliable bond at the atomic scale. Some Mg/Al products have been applied in industry e.g. the AJ62/AlSi17Cu4Mg engine block fabricated by die casting [1]. Mg/Al composites have been fabricated by friction stir welding [2], twin roll casting [3], roll cladding [4], co-extrusion [5], accumulative roll bonding (ARB) [6] and high pressure torsion (HPT) [7,8]. Amongst the Mg/Al composites fabricated through these techniques, a substantial variation in bonding quality and intermetallics formed at the interface is found. The intermetallics formed at the interface include γ-$Mg_{17}Al_{12}$, β-Al_3Mg_2 or both γ-$Mg_{17}Al_{12}$ and β-Al_3Mg_2, with thickness depending on bonding temperature and time [9,10]. The intermetallic type and its thickness may strongly influence the bonding quality as well as the properties of the composite. Formation of thick intermetallics at the interface is likely to be detrimental to the mechanical properties of the composite (see e.g. Ref [11]), since both γ-$Mg_{17}Al_{12}$ and β-Al_3Mg_2 are brittle and low-melting-point intermetallics [12]. Existing bonding processes are mainly carried out at high temperature and thick brittle intermetallics are unavoidable. We performed HPT on Mg/Al disks and successfully achieved Mg/Al bonds by virtue of the very large pressure imposed on the Al/Mg disks. Additionally, HPT processing leads to a large accumulated strain which causes significant grain refinement in both Al and Mg layers [13]. The main objective of this paper is to evaluate bonding quality and examine the microstructure of the Mg/Al interface of HPT processed bilayer disks.

2. Experimental procedure

This work was carried out on commercially pure magnesium (purity 99.8%) and commercially pure aluminium Al-1050 (purity 99.5%). The Mg was received as an extruded rectangular bar. Disks of 9.8 mm in diameter were machined from the extruded Mg bar with the planar surface parallel to the extrusion direction. The Al-1050 was received as a rolled sheet and was subsequently machined to disks in the same size with the planar surface parallel to the rolling direction. The Al disk and the Mg disk in thickness 0.42 mm were stacked and subsequently processed by HPT at room temperature for 1, 5, 10, 15 and 20 turns. During HPT a pressure of 6 GPa was imposed and the anvil was rotated at a speed of 1 rpm (see Ref 14 for detailed description of the HPT facility). The resulting Mg/Al bi-layered disks possessed a thickness of 0.72 mm-0.76 mm. They were cold mounted, ground and polished. Bonding quality was examined by an Olympus D11 optical microscope (OM). Identification of intermetallics at the interface was carried out using a FEI TECNAI G2 F30 transmission electron microscope (TEM). A TEM sample was prepared by focused ion beam (FIB) milling in a FEI Helios NanoLab 600i.

3. Results

Fig 1 shows optical microstructures of Mg/Al bi-layered disks processed by various turns of HPT at room temperature, taken on cross sections through the disks near the centre of the disks. The dark grey layer is Al and the light grey layer is Mg. After one turn of HPT, the interface appears quite flat and well defined (see Fig 1 (a)), and with increasing HPT turns the Mg and the Al tend to mix at the interface (Fig 1 (b, c, d)).

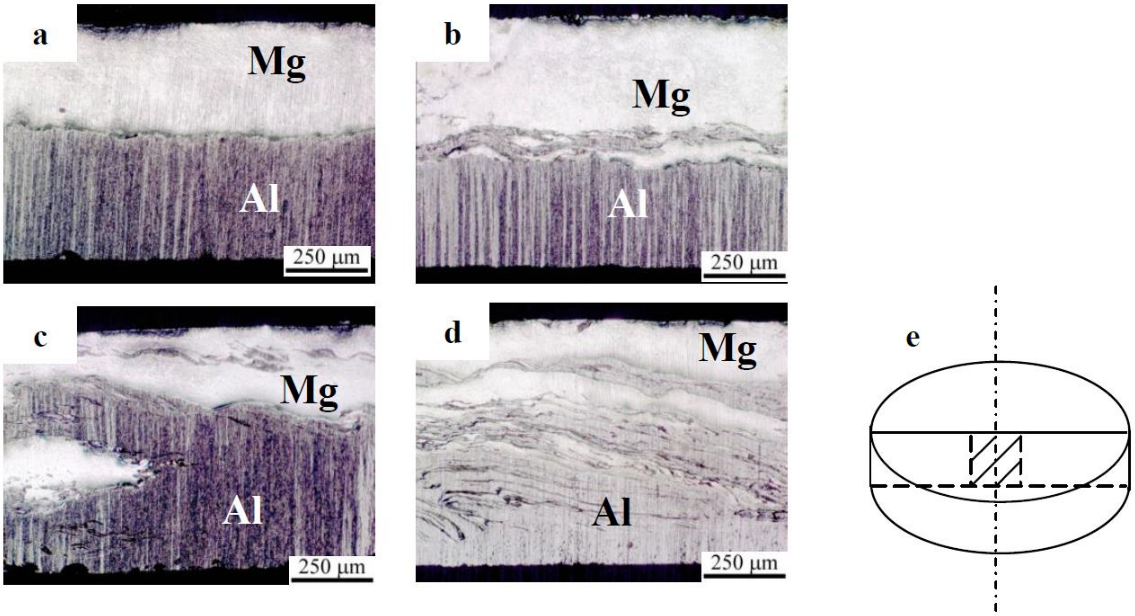


Fig 1 Optical microstructure at the centre of Mg/Al disks processed by (a) 1, (b) 5, (c) 10, and (d) 15 turns of HPT. (e) Schematic illustration of the disk. Images of (a-d) are taken at the shaded area

Fig 2 shows typical TEM microstructures at the interface of an Al/Mg disk processed by 20 turns of HPT. Equiaxed grains are observed in both the Al and Mg layers and the average linear intercept grain size of Al and Mg are about 0.6 μm and 0.8 μm, respectively; see Fig 2 (a). A thin layer of intermetallic phase is observed along the interface (Fig 2 (a)), with a thickness of 0.26 μm (see enlarged image, Fig 2 (b)). From the selected area diffraction pattern (SADP) in Fig 2 (c), the intermetallic is identified as $Mg_{17}Al_{12}$ [15]. Fig 2 (d) shows EDS line scan analyses along the line across the $Mg_{17}Al_{12}$ layer in Fig 2 (b). In the $Mg_{17}Al_{12}$ layer, the Al content decreases on approaching the Mg layer whilst the Mg content increases. In the Mg layer and the Al layer immediately adjacent to the $Mg_{17}Al_{12}$ layer no significantly enhanced solid solution is detected.

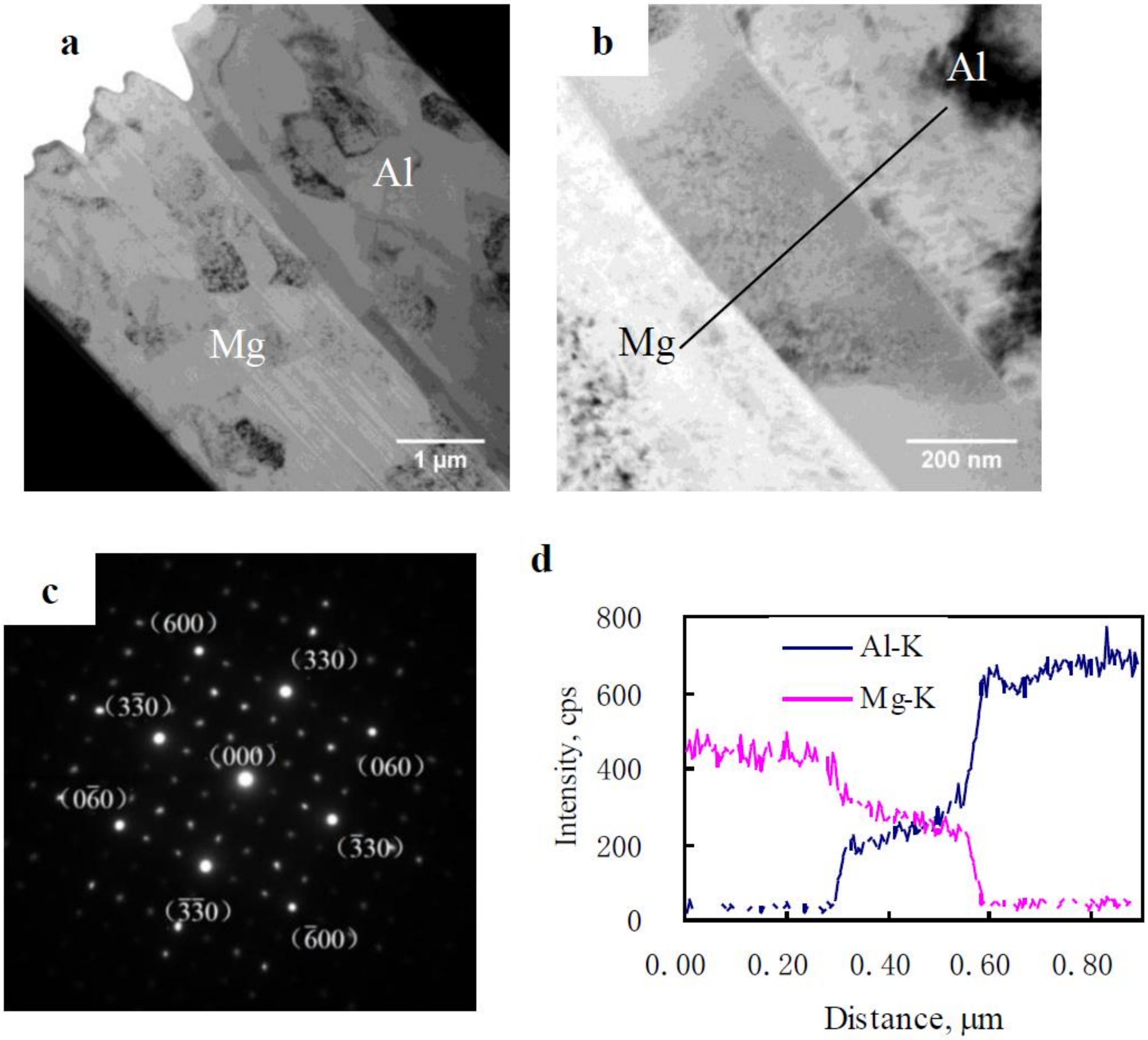


Fig 2 (a) TEM images showing intermetallic formed at interface of Al/Mg/ disks processed by 20 turns of HPT; (b) Enlarged image of showing the intermetallic layer; (c) selected area diffraction pattern of the intermetallic in (b); (d) EDS line scan across the interface along the line in (b).

The molar Gibbs free energies of the Al, Mg, γ-$Mg_{17}Al_{12}$ and β-Al_3Mg_2 phases as a function of composition and temperature were calculated by the thermodynamic software Pandat with the database PanMg2016 and results are shown in Fig 3. Fig 3 (a) shows the molar Gibbs free energy, *G*, of pure Mg, pure Al, γ-$Mg_{17}Al_{12}$ and β-Al_3Mg_2 against temperature at 1 atm. The Gibbs free energies of the two intermetallics are lower than the two pure metals. Fig 3 (b) shows *G* of the various phases against composition at 25 ^oC and at 1 atm. In Fig 3 (c), the *G* for these phases is plotted for 100 ^oC and 6 GPa representing the HPT processing conditions applied here. Fig 3 (b) and (c) indicate that the formation of intermetallics decreases the free energy. The HPT processing condition, i.e. 100 ^oC and 6 GPa, does not change the relative positions of these G-X curves.

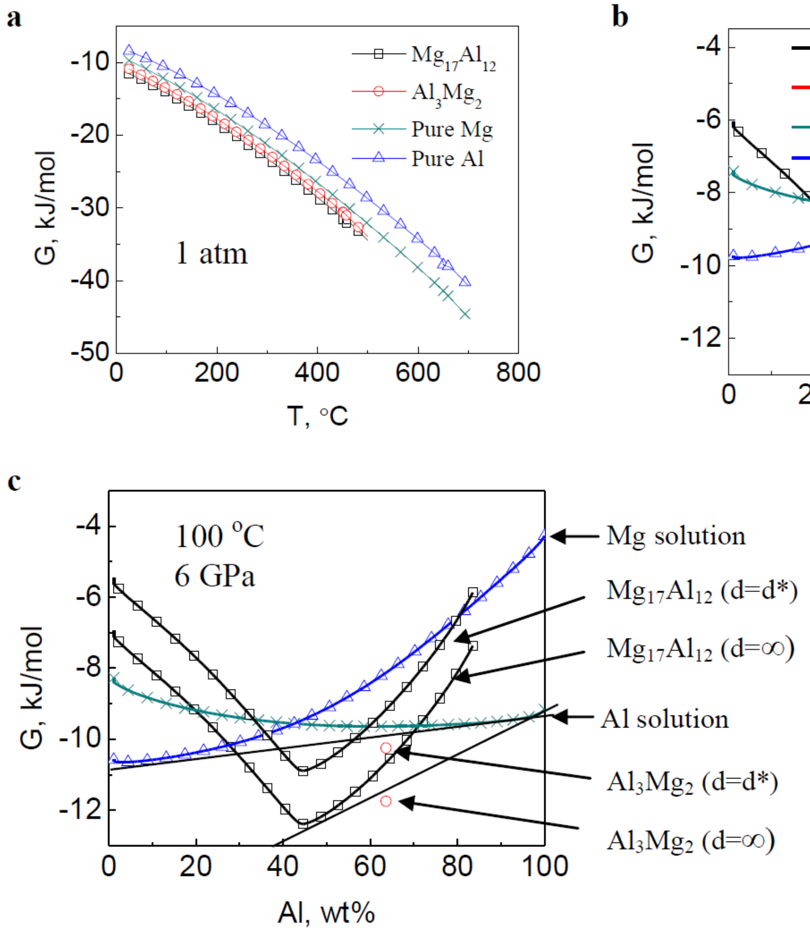


Fig 3 Dependency of molar Gibbs free energy on temperature (G-T) and composition (G-X) in Mg-Al binary system.(a) G-T curves at 25 ^oC and at 1atm, (b) G-X curves at 25 ^oC and 1atm, (c) G-X curves at 100 ^oC and 6 GPa. $Mg_{17}Al_{12}$ (d=d*) and Al_3Mg_2 (d=d*) curves are for schematic illustration only.

4. Discussion

A number of intermetallic phases may be formed in the Al-Mg system including γ, β, R, ε and λ [12]. Amongst these intermetallics, γ-$Mg_{17}Al_{12}$ and β-Al_3Mg_2 are equilibrium phases and they co-exist in Al/Mg interface layers formed during processing at high temperature (see e.g. Refs 6, 9, 10 and 16). Moreover, β-Al_3Mg_2 has been reported to grow faster and is thicker than γ-$Mg_{17}Al_{12}$ [9, 16]. The present unusual formation of intermetallic at the Al/Mg interface (a thin layer of only γ-$Mg_{17}Al_{12}$) is attributed to the unique conditions created during HPT, e.g. an extremely large pressure at low temperature (close to room temperature). The G of γ-$Mg_{17}Al_{12}$ and β-Al_3Mg_2 are both negative at 25 ^oC at 1atm and are -11.6 kJ/mol and -10.9 kJ/mol, respectively, so both γ-$Mg_{17}Al_{12}$ and β-Al_3Mg_2 can form at room temperature, with formation of γ-$Mg_{17}Al_{12}$ being more energetically favorable, see Fig 3 (a). A very thin layer of γ-$Mg_{17}Al_{12}$ was also observed in Mg/Al multi-layered sheets produced by ARB at room temperature [17], but otherwise the formation of intermetallics at the interface of Mg/Al at room temperature by techniques other than HPT has never been reported, presumably because diffusion rates are too low. The present work thus shows that the formation rate of $Mg_{17}Al_{12}$ is remarkably enhanced during HPT, and several factors are considered to be responsible for this. Firstly, HPT imposes a very large pressure on the Mg/Al interface and leads to a good contact all along the Mg and Al disks. Secondly, the thin oxide layers on the surfaces of Al and Mg will be broken down due to the high pressure and strain developed in the HPT process. Thirdly, the temperature of the Al/Mg disks may be somewhat increased because the mechanical work done to the Mg/Al disks during HPT will be mainly converted into heat. Temperature increments of Al, Cu, Ni, Zr and Ti during HPT processing for ten turns were measured as 5 ^oC, 20 ^oC, 35 ^oC, 24 ^oC and 25 ^oC, respectively [18]. The temperature increase in the present Al/Mg samples is expected to be limited because flow strengths of commercially pure Mg and Al are relatively low [19] and heat dissipation during HPT is very considerable due to the large steel anvils. Fourthly, due to the very large strains involved, a high density of defects is created [19], which includes dislocations, vacancies and vacancy clusters. The defects enhance diffusion of Mg and Al through the phases. The good interface contact, broken oxide layers, increased temperature and, in particular, the high density of vacancies will remarkably improve the mobility of Al and Mg atoms. Enhanced diffusion of alloying elements during HPT was observed in other alloys, see Refs. 20, 21 and 22.

Only a very thin layer of γ-$Mg_{17}Al_{12}$ is formed and no other intermetallics are formed in the present work. The absence of a second intermetallic, β-Al_3Mg_2, in our HPT-produced Mg/Al bilayer is thought to be due to the increase in free energy related to the formation of additional interfaces which is dominant at low intermetallic thickness. In particular, the free energy change per unit initial Al/Mg interface area, $\Delta G\ d$, can be approximated as:

$$\Delta G(\gamma)\ d \cong \left(G_{\gamma} - {}^{12}\!/_{29}\, G_{Al} - {}^{17}\!/_{29}\, G_{Mg}\right) d + \gamma_{\gamma/Al} + \gamma_{\gamma/Mg} - \gamma_{Al/Mg} = \Delta G_{vol}(\gamma) d + \gamma_{\gamma/Al} + \gamma_{\gamma/Mg} - \gamma_{Al/Mg}$$

Eq 1

where d is the thickness of the layer of intermetallic, G_{γ}, G_{Al} and , G_{Mg} are the free energy of the γ-$Mg_{17}Al_{12}$ phase, the Al rich phase and the Mg rich phase, respectively. $\gamma_{\gamma,/\mathrm{Al}}$, $\gamma_{\gamma,/\mathrm{Mg}}$ and $\gamma_{\mathrm{Al},/\mathrm{Mg}}$ are the interfacial free energy of the γ/Al ,γ/Mg and Al /Mg interface, respectively. (Here, for the purpose of this estimation of free energy changes, we assume that we can approximate the composition of γ-$Mg_{17}Al_{12}$ as being fixed. We also neglect possible contributions by any strain energy.) We can see that for small d, the interfacial energy terms become important and that every intermetallic phase that is created will cause an additional surface energy term increasing in the free energy, e.g. for the case of two intermetallic phases being present the expression for free energy change is:

$$\Delta G(\gamma,\beta)\, d \cong \Delta G_{vol}(\gamma,\beta)\, d + \gamma_{\beta/Al} + \gamma_{\gamma/Mg} + \gamma_{\gamma/\beta} - \gamma_{Al/Mg} \qquad \text{Eq 2}$$

where $\gamma_{\beta,/\mathrm{Al}}$ and $\gamma_{\gamma,/\beta}$ are the interfacial free energy of the β/Al and the γ/β interface, respectively, $\Delta G_{\mathrm{vol}}(\gamma,\beta)$ is the volume average free energy change in the intermetallics layer excluding the interface contributions, caused by the formation of the two intermetallic phases. Formation of a second intermetallic phase can only be possible if the free energy decreases, i.e. if $\Delta G(\gamma,\beta)$- $\Delta G(\gamma)<0$. Combining the latter two equations then shows that there will be a critical layer thickness below which an intermetallic layer consisting of only one phase will be thermodynamically stable (see Fig 3 c for graphical illustration). Thus, compared to high temperature bonding processes, the reduced diffusion caused by the present low temperature HPT processing, which produces a thin γ-$Mg_{17}Al_{12}$ layer, suppresses the formation of β-Al_3Mg_2.

5. Conclusions

High pressure torsion allows bonding of Mg and Al at room temperature with high-quality interfaces and ultra-fine grained microstructure in the Mg and Al layers. A thin layer of γ-$Mg_{17}Al_{12}$ (thickness of 0.26 μm after 20 turns) is formed and no other intermetallics are formed. Formation of γ-$Mg_{17}Al_{12}$ occurs because the high strain imparted by HPT promotes nucleation of γ-$Mg_{17}Al_{12}$ involving high density of defects enhancing diffusion of Al in Mg. The γ-$Mg_{17}Al_{12}$ layer is very thin and no other intermetallics are formed because growth of γ-$Mg_{17}Al_{12}$ is more energetically favorable than nucleation of β-Al_3Mg_2.

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