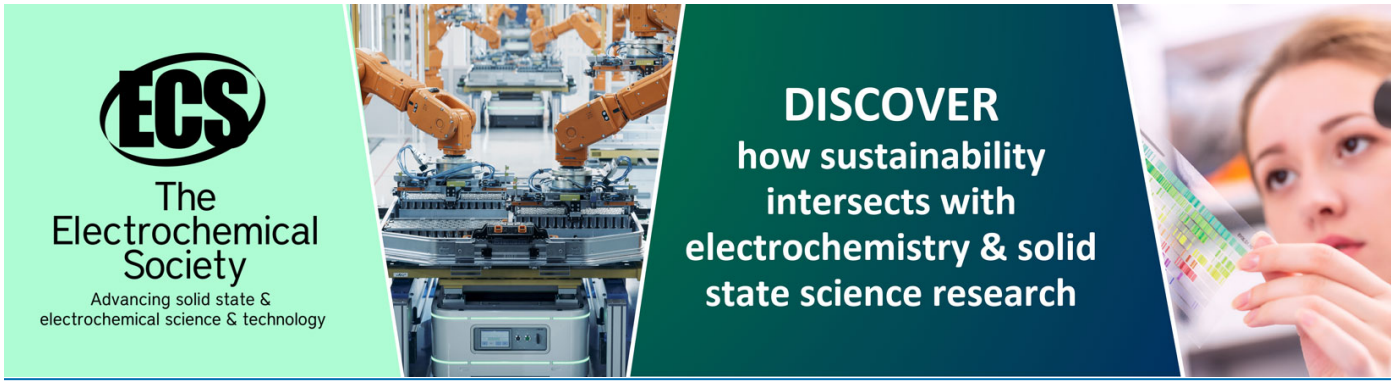
Microstructural changes near the interface of eleven-layered AZ31/AA1050 composites fabricated by single-shot explosive welding



Microstructural changes near the interface of eleven-layered AZ31/AA1050 composites fabricated by single-shot explosive welding

S Puchlerska[1,*], H Paul[2], M M Miszczyk[2], P Petrzak[2] and M Prażmowski[3]

[1]Faculty of Non-Ferrous Metals, AGH University of Science and Technology, Poland
[2]Institute of Metallurgy and Materials Science, Polish Academy of Sciences, Poland
[3]Faculty of Mechanical Engineering, Opole University of Technology, Poland

[*]Corresponding author: spuchler@agh.edu.pl

Abstract. The evolution of the microstructure of reaction regions in as-welded and annealed states was thoroughly investigated by SEM and TEM in the eleven-layered AZ31/AA1050 composite plates fabricated *via* a single-shot explosive welding process. With the detonation velocities used, only the first and second interface is wavy, while the others are flat. Near all interfaces, local melting and rapid solidification processes lead to the formation of reaction regions composed of phases of different chemical compositions and structures. Even though 2 equilibrium phases of γ-$Mg_{17}Al_{12}$ and β-Mg_3Al_2 have been found a large part of the solidified melt region consists of non-equilibrium phases that exhibit an amorphous or ultrafine-grained structure. Further heat treatment of the multilayer composite resulted in rapid nucleation and growth of the γ-$Mg_{17}Al_{12}$ and β-Mg_2Al_3 phases in the form of hard layers. Within the pre-existing reaction regions, the systematic transformation of phases of different chemical compositions into one β-Mg_2Al_3 phase was observed. It was found that using a pressure of 3 MPa is very important in preventing delamination of the clad during heating, but it is not able to block the formation of the linear cracks in the β-Mg_3Al_2 phase, which crucially reduces the composite formability.

1. Introduction

Magnesium alloys seem to be an excellent alternative to materials conventionally used in the aerospace and automotive industries due to features such as low density and high specific strength [1]. However, low ductility and low resistance to corrosion and wear limit their use in industrial practice [2]. An opportunity to improve the workability and other properties of Mg alloys may be the introduction of aluminium layers to create a multilayer Al-Mg composite [3]. There are scientific premises that the joining of Mg and Al by explosive welding (EXW) [4] or by friction stir welding (FSW) [5] can lead to the fabrication of a composite structure or bi-metallic structures combining the excellent properties of these two metals.

EXW is a metal bonding technique that involves carrying out a controlled detonation of an explosive charge on the surface of a so-called flyer plate made of metal with special properties, to bond to a base plate made of a covered metal [6]. The authors indicate that EXW is a promising technique for the production of multi-layered composites [7], among other things, by the possibility of getting rid of the oxide compounds from the surfaces of the contacting sheets/plates. This results in more efficient atoms migration through the interface during heat treatment after joining the sheets. Through long-term

annealing, further modification of the structure is possible, leading to the development of hard intermetallic layers [8-12].

In the literature, there are a small number of reports on microstructural changes occurring near the interface of Al/Mg composites produced by EXW. One of the first works focused on the observation of morphological and microstructural changes at the interface Al-Mg explosive welded composite was [13]. They observed localized zones of solidified melt near the corrugated interface. These areas were characterized by a complex microstructure. Authors [14] studied the morphology and structure of the Al/Mg contact zone bonded by a magnetic pulse. EDS analysis in the middle of the Al/Mg interface zone showed large fluctuations in chemical composition, indicating insufficient mixing of the components. They concluded, that the chemical composition and the sharp transition zone are not typical of the diffusion bonding process. Furthermore, it was found that melting and rapid solidification occur in a narrow zone at the interface which creates the possibility of intermetallic phase formation. In a later work [4], attempts were made to characterize the microstructure of the interface in Al/Mg composites produced by the EXW method. However, the authors did not find any reaction areas at the Al/Mg interface. They concluded that adhesive bonding occurred at the Al/Mg boundary. Yan et al. [15] did not notice the occurrence of intermetallic compounds at the interface of the joined materials. They found that the 'metallurgical bond' is the result of local diffusion in a layer 3.5 μm thick. The authors [16-17] drew similar conclusions, pointing to the metallurgical bonding at the Al/Mg interface. In turn, the authors [18] found that there is a heterogeneity of chemical composition in the weld of Al/Mg explosively welded composite. In addition, the existence of intermetallic compounds was not observed, but the presence of a solid solution with a chemical composition changing in the direction from the weld to the base materials was found. The authors [19-22] studied the effect of annealing on the formation of intermetallic phases in the structure of Al/Mg composites produced by the EXW method. They found that heat-treatment leads to the development of the intermetallic phases: β-Mg_2Al_3 and γ-$Mg_{17}Al_{12}$.

Despite the extensive knowledge of the production of multilayer composites using dynamic bonding methods, slight attention has been focused on the production of the composites based on Al and Mg. This work determined the essential features of the mechanisms responsible for reaction region formation in multi-layered Al/Mg composites using single-shot EXW. Analyses were made to define the factors affecting the structure and chemical composition of the reaction regions during the EXW process, and the impact of annealing on the phase transformations inside reaction regions and in layers near the interfaces was determined. To assess the mechanical properties, microhardness tests were performed.

2. Methodology

For the manufacturing of eleven-layer Al/Mg composites, sheets of technical purity aluminium AA1050 (Al) and magnesium alloy AZ31 (Mg) with a thickness of 1 mm and dimensions of 250 x 350 mm^2 were used. The microstructure of the sheets was characterized by fully recrystallized grains. The chemical composition of the materials was examined using a Jobin Yvon glow discharge JY 10 000 RF spectrometer. The results are presented in table 1.

Table 1. Chemical composition of the base materials.

Material	Element wt. %									
AA1050	Cu	Mg	Si	Fe	Mn	Zn	Ti	Al		
	0.05	0.05	0.25	0.40	0.05	0.07	0.05	balance		
AZ31	Al	Zn	Mn	Si	Cu	Ca	Ca	Fe	Ni	Mg
	3.00	1.00	0.20	0.10	0.05	0.04	0.04	0.05	0.0005	balance

Then, an 11-layer stack was arranged from alternating Al and Mg sheets according to the scheme shown in figure 1 [23]. The explosive welding process was carried out at ZTW Explomet, Opole. The Saletrol explosive charge (ρ=0.84 g/cm^3), laid in layers was used for the detonation. Two detonation speeds were used: 2200 m/s and 2600 m/s. The detonation velocity was measured using the Kontinitro

Explomet-Fo-2000 electronic device. Some of the composite samples were tested in the as-welded condition, while the remaining samples were annealed (T=673 K, p=3 MPa) in the air. The pressure rate was determined based on the work of Bataev et al. [9] and Vecchio [12]. The one-step heat treatment time ranged from 1 to 10 h, with no protective atmosphere.

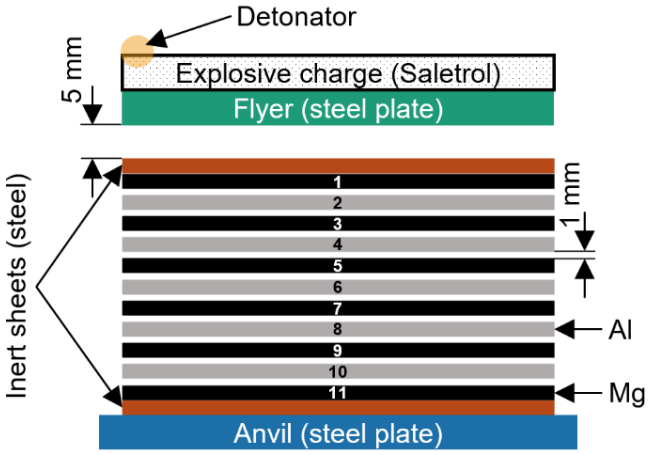


Figure 1. Schematic representation of the stand intended for the EXW process [21].

Samples intended for microstructure studies were cut with a diamond saw along the direction of detonation, both from the as-welded and annealed sheet. The research was performed using Quanta 3D SEM (FEI). The samples after welding and after annealing were ground and then polished. For this, the Vibrometer with Al_2O_3 and colloidal silica were used.

The study of the reaction products formed along the interface in the as-welded condition was carried out using the FEI Tecnai G2 TEM. Thin foils intended for TEM observation were cut-off with a focused ion beam (FIB) method using an FEI Quanta 3D 200 Dual Beam microscope from the central regions of the clads. The samples were installed to a grid (Cu) using an OmniProbe lifting instrument and a tungsten gas supply system. In the next step, the samples were thinned to <100 nm and polished using Ga+ current beam adjustment. TEM analysis was performed for the interfaces between 1/2, 5/6, and 10/11 sheets. Vickers microhardness tests were performed using LECO AMH 2000 hardness tester (100 G, 15 s).

3. Results

Figure 2 presents the structure of the as-welded Al/Mg layered material in the junction area. No micro/macro-cracks in Mg sheets were found in the composites produced with a detonation velocity of 2200 m/s, whereas in the case of the composites produced with a detonation velocity equal to 2600 m/s, micro-cracks were found, in upper sheets of the AZ31 alloy. However, independently of applied detonation velocity macro-scale analysis shows no traces of delamination in the area of the interface. For this reason, in the further part of the paper, the results of tests for the Al/Mg metallic composite produced with the detonation velocity of 2600 m/s are presented.

Based on the observations of the SEM/BSE microstructure, a differentiation of the interfacial morphology was found in the Al/Mg composite, in particular in the case of the first interface compared to the others. The differences occurred regardless of the detonation speed used. The significant kinetic energy of the upper sheet occurring over the explosive welding led to the development of a wavy interface (figure 2) with a large period and amplitude in the first and second layers (respectively Al and Mg).

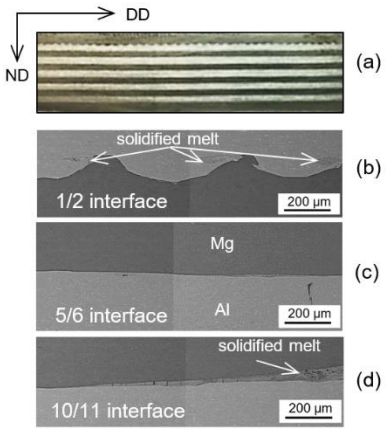


Figure 2. The microstructure of a multilayer composite Mg/Al after EXW, (a) the macrostructure of a multilayer composite Mg/Al (section ND/DD), (b) Al/Mg composite microstructure with visible solidified melt in 1/2 wavy interface, (c) Al/Mg composite microstructure in 5/6 interface, (d) Al/Mg composite microstructure with solidified melt in 10/11 interface.

In the area of interfaces, the solidified melt regions (SMR) were revealed (figure 3a-b). The interfacial boundaries near the intermetallic compounds and Mg/Al were characterized by an irregular shape. This proves the large fluctuations of the chemical composition at the interface. It was found that the largest SMR closed in the vortex of the waves revealed in Al in the first interface. Similar observations were made by Hokamoto et al. [24] during tests of explosively welded materials with different melting points. They found that the irregular internal structure is only noticed at the interface of the intermetallic inclusion and in the less melting material. This particular interface morphology is slighter visible in the case of the following interfaces, where SMR has the shape of a semi-continuous or continuous layer (figure 3b).

Chemical composition mapping indicates a nonidentical concentration of magnesium and aluminium inside the solidified melt area (figure 3c-d). Changes in the chemical composition inside the narrow regions of the solidified melt were insignificant.

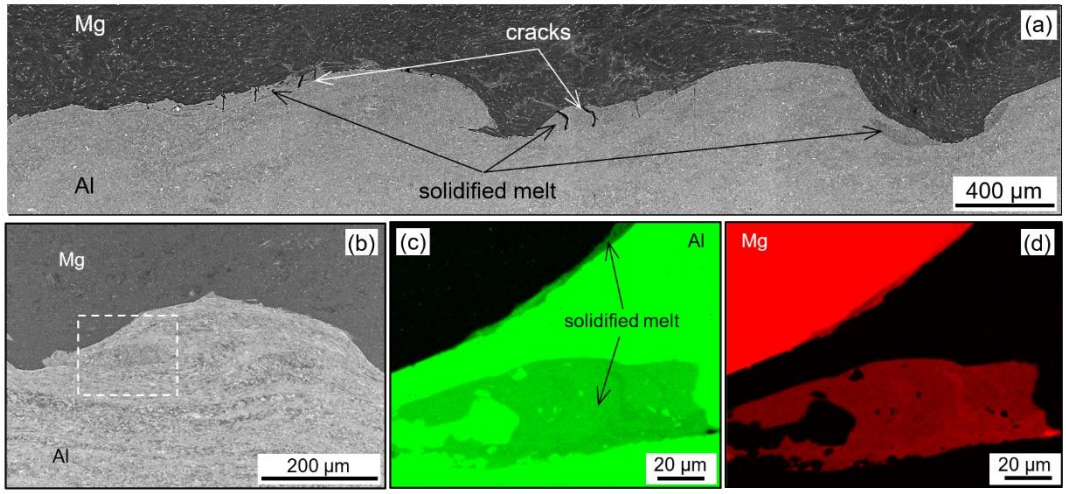


Figure 3. Morphology of the interfacial layers and elements content (section ND/DD). SMR at the 1/2 interface. There are noticeable cracks in the structure (a), an interrupted layer of the SMR at the 10/11 interface (b), and maps of the chemical composition presenting the content of Al (c), Mg (d).

The diversified phase formation and microstructure of the SMR were confirmed by TEM analyses. Figure 4a shows the area of solidified melt, which is located on the top of the vortex at the 1/2 interface. The SMR consisted mainly of amorphous or ultra-fine-grained phases. Nucleation of γ-$Mg_{17}Al_{12}$ grains was observed close to the edges of the SMR and magnesium plate. The chemical composition fluctuated in the reaction region. Fluctuations in chemical composition were dependent on structural changes. The predominantly (near) non-crystalline phases were characterized by a content proportion of about 50/50,

whereas the greater grains had a composition similar to one of the β-Mg_2Al_3 or γ-$Mg_{17}Al_{12}$ equilibrium phases (figure 4b-e).

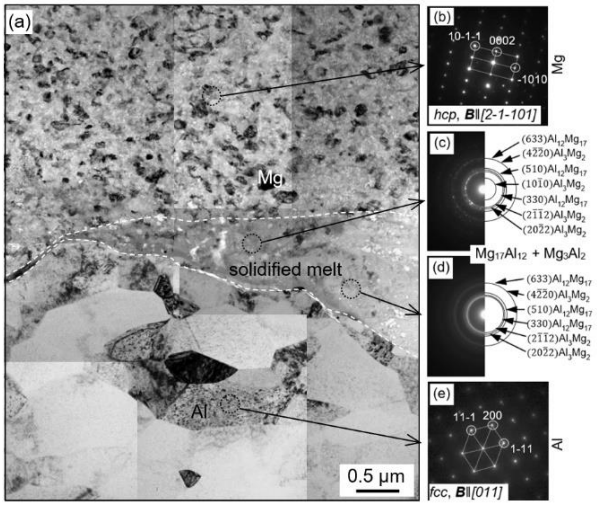


Figure 4. SMR microstructure formed on the top of the vortex (1/2 interface) (a), selected area diffraction of the base plates, and SMR (b-e).

As part of the research work, analyses of the chemical composition and microstructure of the regions close to joined area were carried out too. The location of the solidified melt regions was determined based on the EBSD and EDS analysis (figure 5). Figure 5a presents an orientation map showing changes in chemical composition (figure 5b) and structure in the vortex region (1st interface). In the case of the magnesium plate, mapping exposed darker areas with poorer diffraction patterns, representing shear band formation. In the case of the aluminium sheet, the creation of a flat grain microstructure was noticed. Their shape shows the rotational character of the material flow during vortex formation.

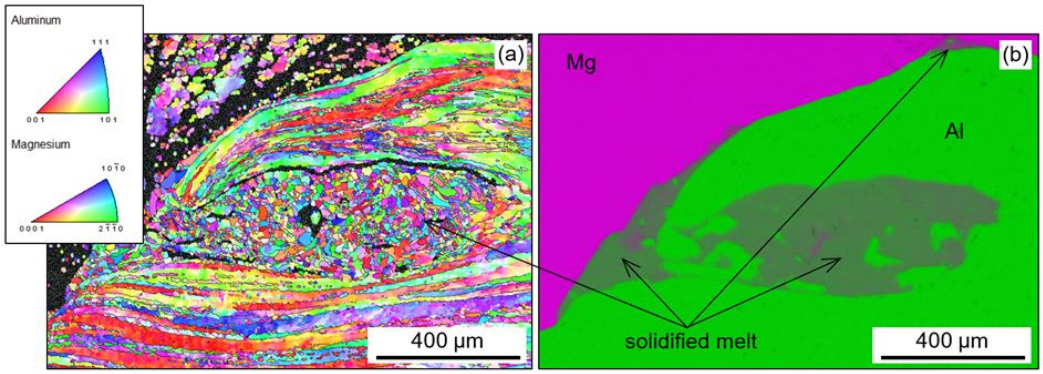


Figure 5. Orientation maps near the 1/2 interface (a), mapping revealing the chemical composition (b).

Heat treatment (673 K) for 60 s caused the growth of the intermetallic layers on the aluminium/magnesium boundary in all analysed cases and the transformation of pre-existing SMR. Figure 6a shows the microstructure of the Al/Mg interfacial layers presented using TEM bright field contrast. The intermetallic layer is made of two sublayers of different chemical compositions and structures, i.e. β-Mg_2Al_3 and γ-$Mg_{17}Al_{12}$ (figure 6b-c), the width of which increased with the annealing time.

The most intense structural changes were noticed during short times of heat treatment of below 30 min, where two processes were preferred. Firstly, the severely deformed layers of the composite were recrystallized. Secondly, the migration of atoms through the interfaces caused the creation and quick growth of equilibrium phases. As diffusion has a decisive role in this transformation, the regions of reaction among aluminium and magnesium may show increased nucleation of the equilibrium phases. During the short-term heat treatment (< 30 min), phases situated in the SMR transformed into the

equilibrium phase of β-Mg_2Al_3. Structural observations and measurements of the chemical composition reveal that right after heat treatment (673 K, 30 min), SMR rich in aluminium transformed into fine, non-regular areas a bit larger ($Mg_{0.30}Al_{0.70}$) and smaller ($Mg_{0.16}Al_{0.84}$) of the Mg content (figure 7a). The variability of the chemical content achieves about 10 at. % (figure 7b). Extending the time of the heat treatment had consequences in chemical composition variability, with a concurrent escalation in the Mg volume. The changes that were observed show an analogous character to those occurring in the Ti/Cu system. These are spinodal transformations, during which phases of different chemical compositions pass into the $TiCu_4$ phase [25]. In addition, another phenomenon was observed during the short heat treatment time, namely, a magnesium-poor layer formed around the SMRs at the aluminium/molten interface. This was due to an increase in the magnesium content inside the SMR. This transformation was completed after heat treatment lasting 30 min. at 673 K when the phases located in the regions of the solidified alloy transformed into the stable β-Mg_2Al_3 phase.

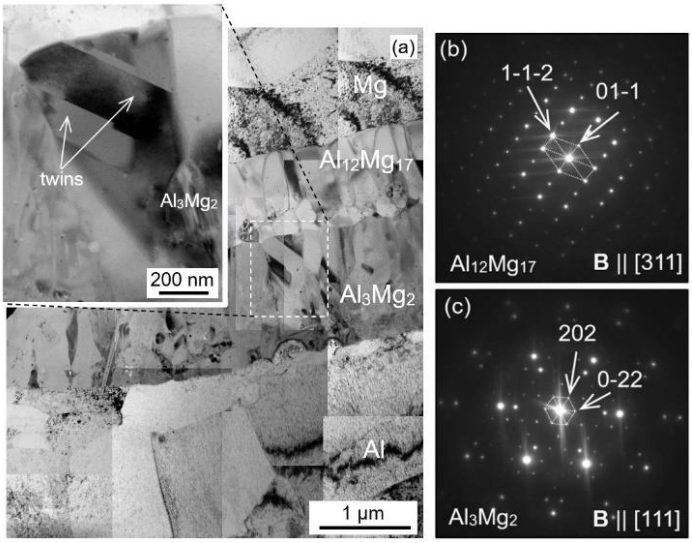


Figure 6. Phase composition and structure of the diffusion layers (a) and corresponding diffraction patterns of γ-$Mg_{17}Al_{12}$ (b) and β-Mg_2Al_3 (c) phases.

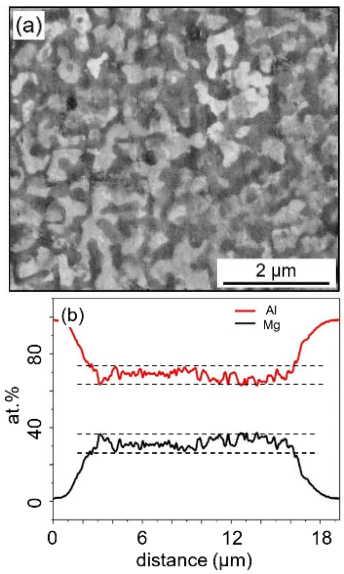


Figure 7. Solidified melt region near the 1/2 interface after annealing (a) with point chemical composition measurements (b).

Vickers microhardness measurements of the samples after heat treatment showed that the intermetallic compounds were characterized by higher hardness than that of the parent materials (figure 8). The β-Mg_2Al_3 phase had a hardness of 310–321 μHV, while the γ-$Mg_{17}Al_{12}$ phase had a hardness of 345–355 μHV. These values were higher than the hardness values for the parent materials (for Al~35 μHV, for Mg~65 μHV).

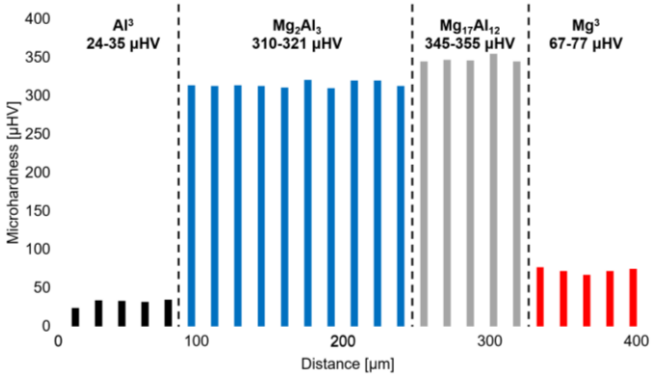


Figure 8. Microhardness of the samples after heat treatment on 5/6 interface.

4. Conclusions

Rapid solidification and local melting occurred in the entire range of tested interfaces due to the dynamic bonding process (EXW). These processes led to the formation of reaction regions consisting of non-equilibrium phases with an amorphous or ultrafine-grain structure with some quantity of equilibrium phases β-Mg_2Al_3 and γ-$Mg_{17}Al_{12}$. The interfacial layers of the base plates showed signs of large plastic deformation, which resulted in strong structure refinement.

Annealing of the Al/Mg composite for just very short times resulted in rapid nucleation and growth of the γ-$Mg_{17}Al_{12}$ and β-Mg_2Al_3 phases. As a result of heat treatment, the areas of different chemical compositions revealed in the reaction regions systematically transformed into β-Mg_2Al_3.

Acknowledgments

This research was funded in part by the National Science Centre (NCN) of Poland, within project no.: 2018/31/B/ST8/00942.

References

[1] Heidarzadeh A, Mironov S, Kaibyshev R, Çam G, Simar A, Gerlich A, Khodabakhshi F, Mostafaei A, Field DP, Robson JD, Deschamps A and Withers PJ 2021 *Prog. Mater. Sci.* **117** 100752.
[2] Rakshith M and Seenuvasaperumal P 2021 *JMA* **9(5)** 1692–714.
[3] Chen ZJ, Liu Q, Wang GJ and Wang DM 2015 *Mater. Res. Innov.* **19(4)** S147–51.
[4] Trykov YP, Shmorgun VG, Rogozin VD, Dolgii YG. 2003 *Weld. Int.* **17(8)** 661–4.
[5] Çam G 2013 *Int. Mater. Rev.* **56 (1)** 1–48.
[6] Findik F 2011 *Materials & Design* **32(3)**1081–93.
[7] Paul H, Petrzak P, Chulist R, Maj Ł, Mania I and Prażmowski M 2022 *Mater. Des.* **214** 110411.
[8] Lazurenko DV, Bataev IA, Mali VI, Bataev AA, Maliutina IuN, Lozhkin VS, Esikov MA and Jorge AMJ 2016 *Mater. Des.* **102** 122–30.
[9] Bataev IA, Bataev AA, Mali VI, Pavliukova DV 2012 *Mater. Des.* **35** 225–34.
[10] Bataev IA, Ogneva TS, Bataev AA, Mali VI, Esikov MA, Lazurenko DV, et al. 2015 *Materials & Design* **88** 1082–7.
[11] Paul H, Maj Ł, Prażmowski M, Gałka A, Miszczyk M and Petrzak P. 2018 *Procedia Manuf.* **15** 1391–8.
[12] Vecchio KS 2005 *JOM* **57(3)** 25–31.
[13] Ghaderi SH, Mori A and Hokamoto K 2008 *Mater. Trans.* **49(5)** 1142–7.
[14] Ben-Artzy A, Stern A, Frage N and Shribman V 2008 *Sci. Technol. Weld. Join.* **13(4)** 402–8.
[15] Yan YB, Zhang ZW, Shen W, Wang JH, Zhang LK and Chin BA 2010 *Mater. Sci. Eng. A* **527(9)** 2241–5.
[16] Zhang T, Wang W, Yan Z and Zhang J. 2021 *Chin. J. Mech. Eng*. **34(1)** 8.
[17] Chen P, Feng J, Zhou Q, An E, Li J, Yuan Y and Ou S 2016 *J. Mater. Eng. Perform.* **25(7)** 2635–41.
[18] Balga D, Ostroushko D, Saksl K, Mazancová E and Milković O 2014 *Arch. Metall. Mater.* **59(4)** 1593–7.
[19] Arisova VN, Trykov YuP, Slautin OV, Ponomareva IA and Kondakov AE 2015 *Met. Sci. Heat. Treat.* **57(5–6)** 291–4.
[20] Lee KS, Yoon DH, Kim HK, Kwon YN and Lee YS. 2012 *Mater. Sci. Eng. A* **556** 319–30.
[21] Macwan A, Jiang XQ, Li C and Chen DL 2013 *Mater. Sci. Eng. A* **587** 344–51.
[22] Zhang N, Wang W, Cao X and Wu J. 2015 *Mater. Des.* **65** 1100–9.
[23] Wright ES and Levitt AP 1974 *Metal Matrix Composite* (K.G. Krieder, Academic Press, New York) **4** 37-99.
[24] Hokamoto K, Chiba A, Fujita M and Izuma T 1995 *Composites Engineering* **5(8)** 1069–79.
[25] Paul H, Chulist R, Miszczyk M, Lityńska-Dobrzyńska L, Cios G, Gałka A, Petrzak P and Szlezynger M 2020 *Mater. Sci. Eng. A* **784** 139285.