An Investigation into High Temperature Tensile Behavior of Hot-extruded Al-15wt%Mg₂Si Composite with Cu-P Addition

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Abstract The effects of Cu-15%P master alloy addition and hot-extrusion on microstructure and high temperature properties of Al-15wt% Mg₂Si in-situ metal matrix composite have been investigated. For this purpose different concentrations of Phosphorus (0-1 wt%) were added to the remelted composite. P addition changes the morphology of primary Mg₂Si from polyhedral to regular shape and decreases the size of primary Mg₂Si particles. Maximum reduction in size of Mg₂Si particles occurs in 0.05 wt% P content from 26.6 μm to 17.1 μm which is also the most uniform distribution of these particles in the matrix. In order to investigate hot deformation behavior of the hot-extruded composite, a set of isothermal tension tests were conducted in the temperature range of 100–500°C with intervals of 100°C under the initial strain rate of 0.001 s⁻¹. The results indicated that elongation percentage improves by increasing temperature which may be probably because of the morphology change in the eutectic structure leading to easier dislocation movement in the matrix. Fractographic analysis was carried out by using scanning electron microscopy (SEM). Providing the fact that larger number of dimples with more even distribution of them can be observed on the fracture surface of the failed specimens by increasing temperature, ductile mode of fracture is more preferable as the testing temperature rises. These observations coincide with elongation percentage variations.

Keywords Al-15%Mg₂Si Composite, Phosphorus Modification, Tensile Properties, Fractography

1. Introduction

Aluminum and its alloys are capable of substituting steels in aerospace and automotive industries mainly because of their low density and high strength to weight ratio [1]. However, a poor resistance to wear and erosion is of serious concern for prolonged use [2]. Al-Metal matrix composites

(AMMCs), reinforced with particulates of Mg₂Si as a class of advanced engineering materials exhibit excellent castability, good wear resistance properties, higher stiffness and hardness at a lower density as compared to the matrix [3]. The workability of this grade, as a vital aspect of the material processing, is greatly affected by the size, distribution and morphology of Mg₂Si particles [4]. A variety of methods are used for AMMCs processing, each has its advantages and drawbacks. Amongst all, in situ processes have been used widely as a result of good particle wetting, even distribution of the reinforcing phase and thermodynamically stable systems [5].

The Al-Mg₂Si composites have high potential as automobile brake disc material because the intermetallic compound Mg₂Si exhibits a high melting temperature of 1085°C, low density of 1.99×10³ kg m⁻³, high hardness of 4500 MN m⁻², a low coefficient of thermal expansion (CTE) of 7.5×10⁻⁶ K⁻¹ and a reasonably high elastic modulus of 120 GPa [6]. Unfortunately, the primary Mg₂Si particles present in Al-Mg₂Si composites are usually very coarse which leads to poor mechanical properties. Therefore, modification operations should be applied to enhance mechanical strength and ductility. Numerous studies have attempted to refine and modify the Mg₂Si phase by addition of minor elements such as phosphorous [7], Ca or P [8], rare earth elements [9], mischmetal of Ce-30La-17Nd-10Pr [9], sodium salts of NaCl-30MgCl₂-10KCl or NaCl-NaF-KCl [10,11], Strontium [12], Ce [13] and mixture of K₂TiF₆ and KBF₄ [14] to the melt. Among all these, phosphorus has been found to be one of the most effective ones that can change the morphology and size of primary Mg₂Si particles [4]. It has been reported that Cu addition results in formation several intermetallic compounds (primarily Q and θ phases) [15]. These intermetallic compounds segregate at the eutectic cells which increases UTS and decreases elongation value [16].

It is well known that the size, the morphology and the distribution of Mg₂Si phase in the microstructure have a non-negligible effect on tensile properties of cast Al-Mg₂Si composites. Furthermore, casting defects such as porosities

and shrinkages are very disadvantageous to both tensile strength and ductility. Hot extrusion, a conventional thermo-mechanical process widely used in wrought aluminum alloys, is an advantageous method to refine grains and reduce casting defects [17]. Applying extrusion process improves strength and ductility significantly due to: (i) fine grains associated with dynamic recrystallization (DRX); (ii) texture; and (iii) dynamic precipitation [18]. Since extrusion process introduces such suitable characteristics to the structure, it has been carried out for Al- Mg₂Si composites.

While there have been a number of researches to tailor the properties of aluminum using different types and amounts of reinforcements, less attempt has been made to study the effects of hot deformation process on the microstructure and tensile properties of Al-Mg₂Si composites. Also, because of workability limitations at room temperature due to insufficient slip systems [19], deformation is usually performed at elevated temperatures, where additional slip systems would be activated. Toward this end, the high temperature workability is a fundamental aspect of materials processing which is predominantly dictated by the materials ductility through hot tensile deformation [20]. Therefore, Thermo-mechanical processing (TMP) is considered as a more effective microstructural modification method for providing superior mechanical properties. To analyze the thermo-mechanical processes, it is necessary to describe the change in mechanical response under high temperature. On the other hand, material flow behavior during hot deformation processes is complex. The hardening and softening mechanisms are both significantly affected by the temperature and strain rate [21]. The work hardening (WH), and restoration process (i.e. DRX) often occur in metals and alloys. It is well known that DRX can refine the grain during the hot deformation. As an important softening and grain refinement mechanism, DRX controls microstructure significantly and alters mechanical properties. The acceleration of DRX with strain rate is attributed to the increasing rate of dislocation accumulation during high strain rates and high temperatures [22]. When the strain rate is decreased, the dynamic recovery rate increases and the dynamic recovery proceeds adequately or the recrystallization occurs during the hot deformation. However, there is no sufficient deformation energy for the complete recrystallization, and then the recrystallization degree is small [23]. Consequently, the suitable temperature and strain

rate should be taken into account to obtain the best condition of microstructure and mechanical properties. Hence, the effect of thermo-mechanical parameters on the flow behavior of the material, which is mainly dependent on the kinetic of the corresponding restoration phenomena at high temperature, should be enabled.

Several researches have been done to investigate Al-Mg₂Si composites flow behavior at room temperature but less has been carried out to reveal its behavior at elevated temperatures. The aim of this paper is to assess effects of temperature on tensile properties of the hot-extruded Al-Mg₂Si composite.

2. Experimental Procedure

The as-cast Al-15 wt%Mg₂Si composite ingots were prepared using 99.8% Al, 99.9% Mg and 99.2% Si, which were heated in an electrical resistance furnace in a 10 kg SiC crucible [7]. In order to prepare alloy with 0, 0.05, 0.1, 0.3, 0.5, 0.7 and 1 wt% P, the primary ingots were cut into small pieces suitable for being remelted in another electrical resistance furnace. After holding the melt at 750°C for 15 minutes, Cu-15%P master alloy was added. After 15 minutes holding time, a graphite rod was used to stir the molten MMC manually for about 1 min. During the melting period to prevent Mg oxidation, flux powder was added continuously to ensure the melt surface was covered. Degassing was carried out via tablets containing C₂Cl₆ (0.3 wt% of the molten material) for about 3 min. After cleaning the slag, the molten MMCs with different amounts of P were poured into a preheated cast iron mold (200°C), as seen in Fig. 1a.

To investigate the optimum modification, the composite billets were cut crosswise to reveal microstructure of the resultant composite and find the appropriate amount of phosphorus addition which gives the minimum size of Mg₂Si particles. After microstructural measurements, the extrusion billets were prepared by cutting and machining directly from the casted billets with optimum amount of P into cylindrical shapes in the sizes of $\Phi 30$ mm $\times$ H28 mm which fits into the extrusion container. All billets were homogenized in an electric resistance furnace at 500°C for 4 h and subsequently slow cooled in the furnace. Then, the prepared billets were directly extruded at 480°C with an extrusion ratio of 12:1 and ram speed of 0.1mm/min.

Figure 1. schematic drawing of (a) cast iron mould and (b) tensile sample dimensions

High-temperature tensile specimens were machined directly from the as-extruded alloys with tensile axes parallel to the extrusion direction according to ASTM E21 [24] as shown in Fig. 1b. The tests were conducted within the temperature range of 100 to 500°C with intervals of 100°C and under the strain rate of 0.001 s^{-1} . All the tensile test specimens were heated to the required temperature and then held for approximately 10 min prior to loading. The tension tests were conducted at constant cross-head speeds using a computer controlled tension machine, SANTAM STM20, which is equipped with a strain gauge extensometer. The

applied thermo-mechanical cycle is shown in Fig. 2. All thermo-mechanical processes were followed by quenching the specimens in water after failure. The elongation-to-failure was measured from the gauge length of the fractured specimens. Metallographic specimens were prepared through standard routines by polishing and etching in hydrofluoric acid (HF) solution (5%). Microstructural examination was conducted on the as cast, as extruded and tensile tested specimens in a plane containing the extrusion direction or the tensile direction using an optical microscopy equipped with an image analysis system (Clemex Vision.

Pro. Ver. 3.5.025). The relative fracture surfaces were also examined using scanning electron microscopy (SEM).

Figure 2. Thermomechanical cycle applied at different temperatures under strain rate of 0.001 s⁻¹.

3. Results and Discussions

3.1. Microstructural Analysis

The typical microstructures of Al-15%Mg₂Si composite before and after Phosphorus addition in the as-cast condition are depicted in Fig. 3. It is evident that the microstructure of the experimental composite consists of three parts: The dark particles which are known as the primary Mg₂Si surrounded by the bright phase known as α -Al and the Mg₂Si eutectic structure. During eutectic reaction α -Al usually nucleates in primary Mg₂Si particles in order to reduce interfacial energy [25]. This explains why almost all Mg₂Si particles are surrounded by a layer of α -Al. In Fig. 3a, it is clearly seen that the morphology of primary Mg₂Si particles is irregular containing some hollow parts noted by arrows. The average size of particles was found to be $27 \pm 5 \mu\text{m}$ in as-cast condition. Fig. 3b-g shows the microstructure of Al-15%Mg₂Si composite with 0.05, 0.1, 0.3, 0.5, 0.7, 1 wt. %P added respectively. As is observed, the morphology of primary Mg₂Si particles changes to regular polygonal and the hollow parts eliminate. To better understand the effect of Phosphorus on Mg₂Si morphology evolution, the variation in Mg₂Si particle size as a function of P amounts is plotted in Fig. 4. As is observed in Fig. 4, phosphorus addition reduces the size of primary Mg₂Si particles up to 0.05 wt. %P of the order of 5%. It should be noted that the amount of added Cu (from Cu-P) is not too much to influence the mechanical properties of the MMC seriously. In accord with the previous reported results [5] the modification mechanism of alloys can be classified into two types, one is heterogeneous

nucleation in which the modifier elements (or compounds formed by them) act as the heterogeneous nucleus; another is poisoning effect in which the modification elements absorb the forehead of growth and restrict the crystal growth. Since the volume fraction of Mg₂Si particles do not change significantly in the modified composite, there should be an increase in the number of primary Mg₂Si particles. As a result, P can enhance the nucleation of primary Mg₂Si particles in Al melt. Having all the above into consideration, the microstructure of Al-15%Mg₂Si-0.05%P after homogenization and hot-extrusion is depicted in Fig. 5. Extrusion process of Al-Mg₂Si is found to be effective in reduction of Mg₂Si particle size and redistribution of them in the matrix more evenly through applying a proper thermo mechanical cycle. In other words, after the process as it can be seen in Fig. 5, the morphology of eutectic Mg₂Si phase alters from plate-like to rod-like and eutectic network is broken partially. It should be noted that extrusion process, reduces or eliminates porosities in the microstructure considerably. All in all, it is expected that homogenization and hotextrusion have a positive effect on mechanical properties of the discussing composite.

Figure 3. Microstructure of Al-15% Mg₂Si in as-cast condition in 500X magnification with a) No P content, b) 0.05 wt.% P, c) 0.1 wt.% P, d) 0.3 wt.% P, e) 0.5 wt.% P, f) 0.7 wt.% P, g) 1 wt.% P.

Figure 4. The variation of primary Mg₂Si particles size as a function of P amount (%).

Figure 5. The microstructure of experimental Al-15%Mg₂Si-0.05P composite in the as-extruded condition.

3.2. Hot Tensile Deformation Behaviors

The true stress–true strain curves of Al-Mg₂Si composite obtained from corresponding tensile tests in the temperature range of 100-500°C under the initial strain rate of 0.001s⁻¹ are shown in Fig. 6. Obviously, the flow behavior is significantly affected by the deformation temperature. According to the characteristics of flow stress curves, we can realize that the flow stress decreases with the increasing of deformation temperature. The reason for this change is that lower temperatures provide shorter time for energy accumulation and lower grain boundaries mobility which result in the phenomenon that the effect of work hardening is stronger than the effect of dynamic softening.

Figure 6. The typical true stress –true strain curves of Al-Mg₂Si composite obtained from hot tensile tests at various temperatures and initial strain rate of 0.001 s⁻¹

However, the softening behavior is more significant since the rate for cross-slip of screw dislocation and climb of edge dislocations increase with decreasing of flow stress at a higher temperature [27], which is consistent with the variation of the flow stress curves shown in Fig. 6 that the softening behavior at temperature of 500°C is more obvious than it is at other lower temperatures. In general, the flow stress curves can be characterized into two types as: (i) (low temperature) which are the curves showing continues work hardening without decrement of stress level, (ii) (high temperature) where the material undergoes restoration processes. It can be noticed that these curves include a short work hardening region up to UTS followed by a long post-UTS region. They can be further divided into three stages: in the initial deformation stage, the stress increases abruptly because of the influence of work hardening, then the increasing rate of curves decrease with the increase of strain due to the occurrence of restoration phenomena until reaching the flow stress peak, when the softening rate is higher than hardening rate due to the enough energy of restoration process provided by high strain [28]. After all, the stress level decreases slightly with increasing strain followed by a long range of work softening region after UTS, ultimately leading to fracture.

When it comes to the microstructural evolution of the examined composite under hot tension, it is seen that during straining under the initial strain rate of 0.001s⁻¹ in the temperature range of 100-300°C, the microstructure of the eutectic phase in the matrix doesn't change significantly (Fig. 7). Presence of uniform rod-like eutectic structure in the matrix results in high work hardening rate at lower temperatures. For the specimens deformed at higher temperatures (400-500°C), increasing temperature may lead to dissolution and/or coarsening of the eutectic structure in the matrix which can be as a consequence of restoration processes [29]. This behavior is evident in the second type of stress-strain curves.

Figure 7. The optical microstructure (sub-fracture surface) of the tensile specimens elongated to fracture under the strain rate of 0.001 s^{-1} and deformation temperatures of: a) 100°C , b) 200°C , c) 300°C , d) 400°C , e) 500°C .

Figs. 8. (a) and (b) shows the evolution of ultimate tensile strength and fracture elongation of the materials investigated in a wide range of temperature ($100\text{--}500^{\circ}\text{C}$) under the initial strain rate of 0.001 s^{-1} . As it can be seen in Fig. 8. (a), the composite displayed good resistance to high temperature effects and the UTS values of the examined composite have similar amounts in the temperature range of $100\text{--}300^{\circ}\text{C}$. As expected, the materials strength generally decreased with increasing temperature above 300°C . the maximum strength of 220 MPa at 100°C undergoes a degradation of 27% and reduces to 60 MPa at 500°C .

As it is clearly observed in Fig. 8. (b), the tensile ductility of the composite at the lowest testing temperatures (i.e. $100\text{--}200^{\circ}\text{C}$) was relatively constant of the order of 5% . As the test temperature increases the elongation to failure increases to a maximum of 50% at 500°C ; more than ten times the elongation value at 100°C . This phenomenon will be explained in the fractographic analysis of the failed specimens.

Figure 8. Effect of deformation temperature on the (a) ultimate tensile strength, (b) Elongation percentage of the Al-Mg₂Si-0.05P composite.

3.3. Fractography

The fracture surfaces of the specimens tested at temperatures of 100, 200, 300, 400 and 500°C are shown in Fig. 9(a–e). The observed changes in the fracture surface topography coincide with the increase in elongation to failure. As it can be seen in Fig. 9a, at 100°C failure of the composite is resulted from fracture of Mg₂Si particles and void nucleation in the Al matrix. As it is clearly observed, deformation of the Al matrix is localized around Mg₂Si particles seriously because there is a much larger residual plastic strain in this area than the area of the matrix near a plane interface [30]. When an external stress is applied, the flowing matrix will give a compressive stress to the Mg₂Si particles perpendicular to the tensile direction. Then stress concentration around these particles will become more intense. Therefore, they may undergo fracture on a low level of applied strain. Failure of the specimens tested at 100°C is mainly because of this behavior leading to lower elongation percentage. According to Fig. 9b, the elongation behavior at 200°C is also accompanied with fracture of particles and void nucleation similar to 100°C so that they exhibit the same behavior. Figs. 9c shows fracture aspects of the Al-Mg₂Si composite when the specimens are fractured at 300°C. In this case, the fracture shows some particles at the boundaries with high volume of dimples in failure surface. Higher distribution of dimples at 300°C compared to lower temperatures leads to increase in elongation to failure. On the other hand, when Mg₂Si particles were blunted, plastic strain concentration in the matrix around the particles reduces so that elongation to failure of composite is improved. At higher temperatures, corresponding to Fig. 9d, the characteristics of the fracture surface at 400°C contains a non-uniform distribution of both small and large dimples. Furthermore, the ductile dimples with different size and depth are seen without any cleavage facets on the fracture surface which is

an evidence for ductility enhancement at this temperature. As it can be seen in Fig. 9e, the material undergoes restoration phenomena (i.e. DRV or DRX) as the deformation temperature increases up to 500°C. The dynamic softening occurs in the highly strained grain-boundary regions, isolating the microcracks from the boundaries and particle-matrix interfaces, along where they grow, thereby improving the elongation to failure. This has been thoroughly explored elsewhere [31].

Figure 9. The fracture surface of the tensile specimens elongated to fracture at: (a) 100°C, (b) 200°C, (c) 300°C, (d) 400°C and (e) 500°C ($\dot{\epsilon} = 0.001 \text{ s}^{-1}$)

4. Conclusions

In the current study, an analysis of the hot deformation characteristics of an Al-Mg₂Si composite modified by phosphorus has been carried out by performing a set of predetermined hot tension tests. Based on this investigation, the following outcomes are reported:

1. Phosphorus addition (0.05 wt.%P) changed the morphology of primary Mg₂Si phase to regular polygonal and reduced its size considerably.
2. The variation of ultimate tensile strength with temperature showed no significant change at 100-300°C temperature range, but the trend was reversed by increasing deformation temperature. The decrement of UTS value in the temperature range of 400-500°C is attributed to two factors operating simultaneously: (a) dislocation annihilation which is as a result of the effect of restoration processes, (b) dissolution of eutectic phase leading to its morphology change from plate-like to rod-like.

3. Increasing the TMP temperature led to higher ductility values, mainly due to the blunting of the Mg₂Si particles together with the restoration of the deformed microstructure.
4. According to fracture surfaces of the experimental composite, by increasing temperature from 100 to 500°C, the volume fraction of dimples increases. Consequently, ductile mode of fracture becomes dominant.

REFERENCES

- [1] Chong Li, Jing Hou, Degang Zhao, Yuying Wu, Xiangfa Liu, *Mater. Lett.* 68 (2012) 255–257.
- [2] Kaveh Edalati, Maki Ashida, Zenji Horita, Toshiaki Matsui, Hirotaka Kato *Wear* 310 (2014) 83–89.
- [3] C. Li, Y.Y. Wu, H. Li, X.F. Liu, *Acta Materialia*, 59 (2011) 1058–1067.
- [4] Qi Gao, Shusen Wu, Shulin Lü, Xuecheng Duan, Zhiyuan Zhong, *J. Alloys Compd.* 651 (2015) 521e527.
- [5] Q.D. Qin, Y.G. Zhao, W. Zhou, P.J. Cong, *Mater. Sci. Eng. A*, 447 (2007) 186–191.
- [6] M.E. Moussa, M.A. Waly, A.M. El-Sheikh, *J. Magn. Alloys* 2 (2014) 230e238.
- [7] N. Nasiri, M. Emamy, A. Malekan, *Mater. Sci. Eng. A*, 556 (2012) 446–453.
- [8] J. KimJoong, D. KimHyang, K.S. Shin, J.N. Kim, *Scripta Mater.* 41 (1999) 333–340.
- [9] J. Zhang, Z. Fan, Y.Q. Wang, B.L. Zhou, *Mater. Sci. Eng. A* 281 (2000) 104–112.
- [10] J. Zhang, Yu-Q. Wang, Ben-L. Zhou, Xing-Q. Wu, *J. Mater. Sci. Lett.* 17 (1998) 1677–1679.
- [11] J. Zhang, Z. Fan, Y.Q. Wang, B.L. Zhou, *J. Mater. Sci. Lett.* 19 (2000) 1825–1828.
- [12] L. Hengcheng, S. Yu, S. Guoxiong, *Mater. Sci. Eng. A* 358 (2003) 164–170.
- [13] Y.G. Zhao, Q.D. Qin, W. Zhou, Y.H. Liang, *J. Alloys Compd.* 39 (2005) 1–4.
- [14] H.Y. Wang, Q.C. Jiang, B.X. Ma, Y. Wang, J.G. Wang, J.B. Li, *J. Alloys Compd.* 387 (2005) 105–108.
- [15] D.J. Chakrabarti, D.E. Laughlin, *Prog. Mater. Sci.* 49 (2004) 389–410.
- [16] S.E. Vaziri Yeganeh, A. Razaghian, M. Emamy, *Mater. Sci. Eng. A*, A 566 (2013) 1–7.
- [17] D. Ke, L. Hengcheng, J. Qiumin, T. Yun, *Mater. Sci. Eng. A*, 527 (2010) 6887–6892.
- [18] T. Homma, N. Kunito, S. Kamado, *Scripta Materialia*, 61 (2009) 644–647.
- [19] Y. Chino, K. Kimura, M. Mabuchi, *Mater. Sci. Eng. A* 486 (2008) 481–488.
- [20] F.J. Humphreys, M. Hatherly, *Recrystallization and Related Annealing Phenomena*, second ed., Elsevier Ltd., Oxford, 2004, pp. 169–450.
- [21] M. Vedani, F. DeRico, E. Gariboldi, *Composites Science and Technology* 66 (2006) 343–349.
- [22] F. Ma, W. Lu, J. Qin, D. Zhang, *Materials Letters* 60 (2006) 400–405.
- [23] M.R. Rokni, A. Zarei-Hanzaki, Ali A. Roostaei, H.R. Abedi, *Materials and Design* 32 (2011) 2339–2344.
- [24] ASTM E21, *Standard test methods for tension testing of metallic materials*, Annual Book of ASTM Standards, ASTM International, PA, USA, 2010.
- [25] *Standard specification for aluminum-alloy permanent mold castings*. Annual book of ASTM standards, vol. 02.02, ASTM B108, 2003.
- [26] M. Emamy, N. Nemati, A. Heidarzadeh, *Mater. Sci. Eng. A* 527 (2010) 2998–3004.
- [27] C. Wang, K. Wu, M. Zheng, *Mater. Sci. Eng. A* 477 (2008) 179–184.
- [28] R. Hambli, *Int. J. Adv. Manuf. Technol.* 20 (2002) 676–682.
- [29] J. van de Langkruis, W.H. Kool, C.M. Sellars, M.R. van der Winden, S. van der Zwaag, *Materials Science and Engineering A* 299 (2001) 105 – 115.
- [30] N.P. Chenga, B.S.M. Zenga, J.B.Z.Y. Liua, *Journal of Materials Processing Technology* 202 (2008) 27–40.
- [31] B. Mintz, S. Yue, J.J. Jonas, *Int Mater Rev* 36 (1991) 187–217.