Microstructure Transformation of Mg-1.6Gd during Hot Rolled at High Deformation Ratio

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Abstract. Mg-1.6Gd alloy was prepared by hot rolling to produce a potentially degradable implant material. Microstructural transformation during hot rolling was observed and correlated to mechanical properties. The objective of this study was to examine the constituents of the microstructure such as grain size, phases which are contributed to mechanical properties. Hot rolled Mg-1.6Gd alloy was performed at a temperature range of recrystallization (400-550 °C) with a speed of 10 mm/min and reduction of 95%. The hot rolled was employed by two methods: unidirectional rolling (UR) and cross rolling (CR). The results show that both of the hot rolling techniques reveal refine microstructure with the grain size range of 50-130 µm. The number of lamellar in every grain were shown on the UR, while for the CR resulted in many precipitates, especially at the highest temperature hot rolling. The hot rolled sample of 95% reduction had the relatively smaller of the grain size than the hot rolled sample of 30% reduction (200 µm).

Keywords: Mg-rare earth element, microstructure, lamellar, precipitate, biodegradable, prospective implant materials

1. Introduction
Binary Mg-Gd alloys have been received much attention as degradable implant materials [1, 2]. The alloys are well known for their low density, high strength, creep resistance, and inherent biocompatibility. The elastic modulus is about 45 GPa, which is close to that of human bone properties. It is suitable and is used as the requirements of implant materials[3-6]. However, magnesium alloys have been limited in application due to their low ductility[7, 8]. Many efforts have been performed to improve mechanical properties and/or degradable characteristic of the alloy, such as the addition of other elements and/another mechanical process. In terms of mechanical properties, that can be attributed to the microstructure control that is responsible for the properties relevant to applications, such as strength, hardening, ductility, and elastic modulus[9-18]. Therefore, Mg-Gd alloys have a great potential as candidates for the future design of degradable material.

Recently, Peng et al. reported that the melt-spun Mg-20Gd alloy are found in a few metastable (amorphous) phases, and they transform into a stable phase (Mg2Gd) after at the high temperature (350-400 °C)[10]. The tensile strength of the melt spun is higher than that of as-cast alloy, while the elongation of the two samples is less than 2%. The fracture of the alloy sample is a combination of brittle fracture and ductile fracture. Other study showed that hot extrusion is one of technique to refine the microstructure morphology and to improve the mechanical properties of Mg-Gd alloys. Mg-1.6 Gd
(wt%) alloys exhibit the tensile strength of 232 MPa, the yield strength of 142 MPa and elongation to failure of 72%. These alloys have been fabricated by direct extrusion at the temperature of recrystallization range of 400-550 °C [12]. Rolling process is also an effective way to refine the microstructure which will affect the mechanical properties. Cizek et al. studied the properties of cold rolled Mg-15Gd alloy with a reduction ratio of 8%, 21%, and 29%. Precipitation was found from the rolled samples which cause strong hardening[11]. Luo et al. also reported that the microstructure and the mechanical properties could be improved by the addition of Mg-xGd (x = 0.8, 1.5, and 2.5) [16]. Further study is still needed to understand the microstructure transformation of Mg-1.6Gd alloys that performs the thermo-mechanical process. In this paper, severe hot rolled processing, in terms of unidirectional (only 0° rolling direction) and cross rolling (simultaneously rolling in 0° and 90° direction) with a total reduction of 95% is conducted to Mg-1.6Gd (wt%) alloys. Microstructure morphology and their transformation of the alloy sheet will be observed and discussed.

![Image](http://example.com/image1.png)

**Figure 1.** Microstructures of hot rolled Mg-1.6Gd alloy at various temperatures with reduction of 95%: (a) 400 °C, (b) 452 °C, (c) 491 °C, and (d) 560 °C unidirectional rolling (UR)

2. **Experimental Method**

Mg-1.6 Gd (wt%) alloy ingot were machined for hot rolling specimens to the initial size of 50 mm in length, 25 mm in width and 10 mm in thickness. The alloy was homogenized at 500 °C for 5 hours and then followed by normalizing at room temperature (about 37 °C). The process was employed by two methods: unidirectional rolling (UR) and cross rolling (CR) at a total reduction of 95%. The UR has employed one direction each pass, while the CR was employed by 0° rolling direction first and followed by changing the rolling direction to 90° each pass (for four times). The hot rolling, both UR and CR was carried out at a temperature range of recrystallization (400-550 °C). The specimens were
rolled by multi-pass to the final thickness (h = 3 mm) with speed of 10 mm/min. Each the rolling samples were reheated to 400 - 550 °C for 15 min before per pass rolling. The sheets rolling were examined by standard metallography procedures. Samples were ground by using up to 2000 grade silicon carbide paper and then polished with alumina paste. An etching solution was prepared containing 20 mL acetic acid + 1 mL nitric acid + 60 mL ethylene glycol + 20 mL aquades for about 15 seconds. Then the microstructure of the specimen was observed using optical microstructure after cooled to room temperature after hot rolling.

3. Results and Discussion

The microstructures of Mg-1.6Gd alloys reveal nearly similar grain morphology with tendency fine equiaxed for each temperature for both of UR and CR as shown in Figure 1 and 2. Figure 1 shows the hot rolled of UR for different temperatures. Generally, the grain size of UR is smaller than CR. The average of grain size for UR is about 70 µm. At a temperature of 400 °C the grain size is about 69 µm. The grain size increases as temperature increases where the grain size growth occurs at a temperature of 452 and 560 °C are 72 and 89 µm, respectively. The grain size drops significantly to 49 µm at a temperature of 491 °C. In this temperature, the hot rolled process can influence clearly to the grain size refinement. All of the hot rolled samples show lamellar phases inside the grain which grows in the different orientation. A sharp form of Mg-rich phase (α-Mg) is also observed in the grain of the samples which hot rolled unidirectional at a temperature of 491 °C.

![Figure 2](image)

**Figure 2.** Microstructures of hot rolled Mg-1.6Gd alloy at various temperatures with reduction of 95%: (a) 410 °C, (b) 455 °C, (c) 521 °C, and (d) 561 °C of Cross rolling (CR)

Figure 2 shows the microstructure of the hot rolled for CR which has larger grain size compared to the UR which is visually more pretending homogenized. At a temperature of 410 °C has the biggest grain size about 127 µm, and the grain size goes down to 64 µm at the highest temperature of 561 °C. The smallest grain size is found at a temperature of 521 °C with the size of 56 µm. The microstructure also exhibited the number of lamellar in every the grain. The lots of lamellar are found in the hot rolled sample of 410 °C. The formation of precipitates occurs at higher temperature. It can be predicted
that these precipitates particles in grain are Mg₅Gd as explained by Hort et al. in phase diagram [9]. The Mg-1.6Gd alloy in the different conditions at a temperature below 500 °C are composed mainly of α-Mg solid solution and second phase or precipitates of Mg₅Gd. However, the precipitation during the hot rolled is hardly detected because of the limited particles size at a temperature range of 400-550 °C. Moreover, the grain size and shape of the as-rolled plays an important role in the high strength of the Mg-1.6Gd alloy. It should be noted that the formation of precipitates only occurs in cross rolled samples. The formation of precipitation increases at higher temperature.

![Figure 3](image_url)

**Figure 3.** The grain size of Mg-1.6Gd alloys after unidirectional hot rolling (UR) and cross rolling (CR) at various temperatures.

The temperature effect on the grain size of both rolling methods, *i.e.* UR and CR is shown in Figure 3. The grain size of UR samples is relatively similar at the range of rolling temperature. However, for CR samples, at a temperature below 500 °C give higher grain size than that of UR samples at the same range of temperature. The grain size refinement of CR samples occurs clearly at a temperature higher than 500 °C.

4. Conclusion

In this study, the microstructure observation of the hot rolled Mg-1.6Gd had been conducted by two methods i.e. UR and CR at a temperature range of 400-550 °C. It is obtained that the lamellar phase which is a Mg-rich phase (α-Mg), are found in both of UR and CR samples at all range of the temperatures. It is also found that CR samples which were rolled at a temperature higher than 500 °C, there is precipitation of Mg₅Gd that spread in the grain. The microstructure of the samples of UR has smaller grain size compared to CR samples at a temperature below 500 °C. However, refinement of grain size of CR samples occurs at rolling temperature higher than 500 °C.
5. References

[1] Hort N, Huang Y, Fechner D, Störmer M, Blawert C, Witte F et al. 2010 Magnesium alloys as implant materials – Principles of property design for Mg–RE alloys Acta Biomaterialia 6 pp. 1714-1725

[2] Grillo C A, Alvarez F and de Mele M A F L 2014 Cellular response to rare earth mixtures (La and Gd) as components of degradable Mg alloys for medical applications Colloids and Surfaces B: Biointerfaces 117 pp. 312-321

[3] Zheng W-w, Li X-g, Zhang K, Li Y-j, Ma M-l and Shi G-12015 Reactive diffusion in Mg–Gd binary system at 773 K Transactions of Nonferrous Metals Society of China 25 pp. 3904-3908

[4] Fronzi M, Kimizuka H and Ogata S 2015 Atomistic investigation of vacancy assisted diffusion mechanism in Mg ternary (Mg–RE–M) alloys Computational Materials Science 98 pp. 76-82

[5] Qiu Z, Xiaofeng D, Yanping L and Xiaojia W 2017 Analysis on micro-structure and mechanical properties of Mg-Gd-Y-Nd-Zr alloy and its reinforcement mechanism Journal of Alloys and Compounds 690 pp. 961-965

[6] Liu X, Zhang Z, Le Q, and Bao L 2016 Effects of Nd/Gd value on the microstructures and mechanical properties of Mg–Gd–Y–Nd–Zr alloys Journal of Magnesium and Alloys 4 pp. 214-219

[7] Drynda A, Deinet N, Braun N and Peuster M 2009 Rare earth metals used in biodegradable magnesium-based stents do not interfere with proliferation of smooth muscle cells but do induce the upregulation of inflammatory genes Journal of Biomedical Materials Research Part A 91 pp. 360-369

[8] Mueller W D, Fernández Lorenzo de Mele M, Nascimento M L and Zeddies M 2009 Degradation of magnesium and its alloys: dependence on the composition of the synthetic biomedical journal of biomedical materials research Part A 90 pp. 487-495

[9] Hort N, Huang Y, Fechner D, Störmer M, Blawert C, Witte F et al. 2010 Magnesium alloys as implant materials–principles of property design for Mg–RE alloys Acta biomaterialia 6 pp. 1714-1725

[10] Peng Q, Wu Y, Fang D, Meng J and Wang L 2006 Microstructures and properties of melt-spun and as-cast Mg-20Gd binary alloy Journal of Rare Earths 24 pp. 466-470

[11] Čižek J, Prochazka I, Smola B, Stulíková I and Očenášek V Influence of deformation on precipitation process in Mg–15wt.% Gd alloy Journal of alloys and compounds 430 pp. 92-96

[12] Susanti O, Harjanto S and Mochtar M 2015 Microstructure and Mechanical Properties of Extruded Mg–1.6Gd as Prospective Degradable Implant Materials Advanced Material Research 1112 p. 592

[13] Kulyasova O, Islamgaliev R, Kil’mametov A and Valiev R Superplastic behavior of magnesium-based Mg-10 wt% Gd alloy after severe plastic deformation by torsion The Physics of Metals and Metallurgy 101 pp. 585-590

[14] Nie J, Gao X and Zhu S 2005 Enhanced age hardening response and creep resistance of Mg–Gd alloys containing Zn Scripta Materialia 53 pp. 1049-1053

[15] Peng Q, Wu Y, Fang D, Meng J and Wang L 2007 Microstructures and properties of Mg–7Gd alloy containing Y Journal of Alloys and Compounds 430 pp. 252-256

[16] Luo S, Yang G, Liu S, Wang J, Li J and Jie W 2016 Microstructure evolution and mechanical properties of directionally solidified Mg-xGd (x = 0.8, 1.5, and 2.5) alloys Materials Science and Engineering: A, 662 pp. 241-250

[17] Cao G, Zhang D, Zhang W and Qiu C 2015 Microstructure evolution and mechanical properties of Mg–Nd–Y alloy in different friction stir processing conditions Journal of Alloys and Compounds 636 pp. 12-19

[18] Li X, Qi W, Zheng K and Zhou N 2013 Enhanced strength and ductility of Mg–Gd–Y–Zr alloys by secondary extrusion Journal of Magnesium and Alloys 1 pp. 54-63, 3/2013.
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