Microstructure and Corrosion Rate of the Hot Rolled Mg-1.6Gd Alloys as Prospective Degradable Implant Materials

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Abstract. At present, degradable materials implants with the capability of high loads for the temporary replacement of bones and biocompatible are not been available much more. Mg-Gd alloys have been intensively observed as appropriate degradable materials, since they combine a good strength and corrosion rate. Thus, the aim of this study is development of Mg-Gd alloys as materials implant with biomechanical tailored properties. Mg-1.6Gd alloys of hot rolled at severe reduction of 80% at 40 % per stage with a speed of 10 mm/min. The hot rolled was employed by cross rolling with 0° rolling direction first and followed by changing the rolling direction to 90° each pass (for two time). The hot rolled of the alloys have been investigated in the temperature range recrystallization (400-550 °C). Meanwhile, the corrosion experiments also were research after 3, 7 and 14 days of immersion in Ringers solution. The phenomena of corrosion was observed using optical and weight of the samples before and after immersion. The Results show that the hot rolling techniques reveals refine microstructure with the grain size range of 50-60 µm. The number of precipitates have been founded on the cross rolling, particularly at the higher temperature and the highest corrosion rate occurs on the short immersion time for 3 days.

Keywords: microstructure, corrosion, bones, biocompatible, implant

1. Introduction
The small addition of Gd in Mg have been significantly improved microstructure and mechanical properties by thermo-mechanical process. Susanti et al showed that the Mg-1.6Gd alloys have been exhibited values of its mechanical properties through the hot extrusion. One of ways is the micro-alloying addition of Gd (1.6 wt%) in magnesium. The strength can increase due to precipitates hardening. The hot extrusion of the Mg-1.6Gd alloying also showed the recrystallization grain size and the other excellent mechanical properties. This discussion already explained in the paper before [1-4]. Before, it well known that the elastic modulus of Mg is close to that of human bone properties which it is suitable and is used as the requirement of implant materials. Therefore, magnesium alloys have been great attention as a degradable implant. However, many efforts must be employed to improve mechanical properties and degradation characteristic of the alloy. In the term of mechanical properties that can be related to the microstructure control to purpose for applications such as strength, hardening, ductility and elastic modulus etc [5].

Another paper, Susanti et al clearly reported that the effect of the micro alloying on microstructures by the hot rolling with a total reduction of 95 % [6]. This study explained that the microstructures of the Mg-1.6Gd alloys reveals nearly similar grain morphology with tendency fine equated grain on each temperature for the both of the methods. Especially for cross rolling (CR) which has larger grain size compared to the unidirectional rolling (UR). It has visually more pretending homogenized with the size
of about 60-100 µm. The number of precipitates found at the higher temperature of the rolling. The grain size and shape of the alloy that were caused by the mechanical processing to that related the mechanical process. This study is to prove the previously research which Luo et al. showed that the addition of Gd in to Mg can improved the microstructure and the mechanical properties with the ASTM standard, Mg-xGd (x= 0.8, 1.5, and 2.5) [7].

The effect of rolling on microstructure can change the grain size of the MgGd which is correlated on the corrosion rate. The grain size increases with the increasing the temperature of rolling. The grain size can be also changed by the addition of rare earth elements (REE), such as Ce, La, Nd Mo or Gd as showed on the papers [8, 9]. Moreover, it can be be summarized that the addition of REE can from corrosion resistance by reducing the grain size and the formation of surface layer as protection. The other way is to give the use of REE in order to form surface layer by oxide as reported by Susanti et al [10]. This study explained that the hot rolled of Mg-1.6Gd alloys can be improved the grain size and can be also founded Mg(OH)₂ surface layer as corrosion resistance for the MgGd alloys after the longer immersion time.

2. Experimental

Mg-1.6Gd alloys is prepared the number of specimen for hot rolling which were machined by the initial size of 50 mm in length, 25 mm in width and 10 mm in thicknesss. Specification of specimens are the same with the paper before [6]. The Mg-1.6Gd alloys was employed cross rolling with reduction of 80% which is 0° rolling direction first and followed by changing the rolling direction to 90° each pass (for two times). This cross rolling was carried at a temperature range recrystallization (400- 550°). All the samples were observed by standard metallography procedure and then followed the corrosion process which all the procedures nearly the same explanation with the before paper [10]. The samples were weighed and the corrosion rate was also calculated with using equation (1):

\[ CR = \frac{W K}{\rho A t} \]

where, \( CR \) refers to corrosion rate, \( W \) is the weight loss from sample, \( \rho \) is density of the sample, \( A \) is area of sample and \( t \) is time in Ringers solution.

3. Result and discussion

The microstructures of cross rolling of Mg-1.6Gd alloys result nearly the same the grain size for all the samples with tendency fine equiaxed as shown in Figure1. A lot of twin grow spread on each the microstructure and also found the formation of precipitates. The highest temperature exhibits the number of precipitates much more compared the others. As well known that these precipitates in grain are Mg-Gd as explained by Susanti et al [10] and then the formation of precipitates grow with the increasing temperature of rolling.
Figure 1. Microstructure of the hot rolled Mg-1.6Gd Alloys at various temperature with reduction of 80%; a) 400°C, b) 450°C, c) 500°C, d) 550°C of cross rolling.

The strength of the Mg-1.6Gd alloys is very influenced by the grain size and shape of the as-rolled. At the early temperature of the rolled Mg-1.6Gd alloys (400°C) has about the grain size of 65 µm. The grain size decrease to be about 48 µm at a temperature of 450 °C and then drop to be about 46 µm at a temperature of 400°C and the increasing to be 52 µm at a temperature of 550 °C as shown in Figure 2.

Figure 2. The grain size of the cross rolling Mg-1.6Gd at various temperature.
The degradation of the Mg-1.6Gd alloys are tested with use of immersion test with Ringer solution. The immersion test with using Ringer solution for 3 days, 7 days and 14 days to get corrosion rate on the each sample as shown in Figure 3. It can be seen that the early immersion (3 days) occurs pitting corrosion on around the surface of samples and result the white colour as the corrosion product. And then the number of degradation spread and reaches a depth of samples for 7 days and the degradations of sample reach the peak on the immersion of 14 days.

![Image](a) ![Image](b) ![Image](c) ![Image](d)

**Figure 3.** Typical corrosion morphologies of the Mg-1.6Gd alloys sample: (a) sample before immersion; (b). immersion for 3 days; (c) immersion for 7 days; (d) immersion for 14 days.

Generally, the highest corrosion rate occurs at the short immersion duration. In this case, the initial corrosion stage no occur the protection on samples yet. The formation of surface layer as a protection is obtained by MgO in water to form Mg(OH)$_2$ layer after the longer immersion time as explained by Song et al [11]. Moreover, the protection can reduce due to attack Cl$^-$ from the water and then gradually the thick protection layer can be loss and the samples can be finish. Meanwhile, the corrosion rate occurs higher at the low temperature compared the others which is very correlation to grain size and precipitate. At the low temperature of rolling result the corrosion rate of about 28 mmpy and then drop to about 22 mmpy at a temperature of 550 °C. On the each temperature always find the highest corrosion rate after the immersion time for 3 days as shown on Figure 4.

![Image](Figure 4. Corrosion rate of cross rolled Mg-1.6Gd alloys at different temperature.)

**Figure 4.** Corrosion rate of cross rolled Mg-1.6Gd alloys at different temperature.

4. Conclusions
In this study, the microstructure and corrosion rate observation of the hot rolled Mg-1.6Gd with reduction of 80 % at temperature range of 400-550 °C. It is resulted that the The number of precipitation are found on microstructure and with the increasing temperature of rolling has more precipitation which
spread in the grain. A lot of twin also found in the grain on every temperature of rolling which it can improve toughness of the alloys. Following that the corrosion rate occur higher on the immersion time and then the corrosion rate is slowly at the longer immersion time. This is caused by the formation of Mg(OH)\textsubscript{2} as protection surface layer. The corrosion rate is influenced by the number of grain boundaries.

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