Investigation of Physical Properties on HA/nGO Coating Magnesium Alloy

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Abstract. AZ91D magnesium alloys have been referred to as a revolutionary material for biomedical applications, particularly in orthopaedics as a bone implant material. This is attributable to the good strength and strong biocompatibility of magnesium and its alloys with bone tissue relative to other metallic alloys. The rapid degradation of pure magnesium has, sadly, reduced its therapeutic use. As a consequence, the efficacy of hydroxyapatite (HA) and graphene oxide (GO) in corrosion resistance has demonstrated tremendous potential to improve its defensive properties in order to address this vulnerability. AZ91D alloy has been coated with HA/GO by micro-arc oxidation (MAO) at a specific concentration and voltage, with varying treatment times. Surface roughness, elemental composition analysis and surface morphology analysis were tested at different treatment time. The sample enduring 15 min of treatment time was found to have the greatest resistance to corrosion due to its surface morphology and the importance of surface roughness was within the range of recommended roughness for a biomedical implant.

1. Introduction

Biomaterial with biodegradable activity is desirable for biomedical implants [1]. Biodegradable materials like magnesium, iron and zinc have a fast advancement in the biomedical application of temporary implants due to their intrinsic ability to degrade and disappear in the body within a certain amount of time, which prohibits the need for another surgery to remove the implant after end of the healing process [1][2]. Among these biodegradable implant materials, only the magnesium-based alloy in this paper is involved.

Magnesium (Mg) and its alloy are very important in different industries due to their excellent properties such as high specific intensity, high processing capabilities, low metal density, high thermal conductivity and electromagnetic wave shield characteristics. As a result, this glossy grey solid is known as having a significant effect on the pharmaceutical, agriculture and construction industries. However, the biggest challenge posed by magnesium-based substance implants is the high corrosion rate, which could harm the body if the implants failed before the healing process was completed [3]. A decent surface modification is required to encounter this problem.

Surface improvement is intended to improve the mechanical properties of the material [4]. The biocompatibility of a surface-modified Mg implant is determined by both the surface coating and the alloy substrate [5]. In this analysis, hydroxyapatite (HA) and graphene oxide (GO) are involved in the surface improvement of magnesium alloys. HA is the most common substance used in biomedical
implants, owing to the composition of HA nearly the same as human bone tissue, and the excellent biocompatibility that occurs within the physiological environment [6][7]. Whereas GO attracted considerable interest to biomedical application due to its excellent result in resistance to corrosion [8], besides, GO also has exceptional mechanical properties, high stability and excellent biocompatibility [9]. Recent investigations conducted by Wen [10] indicated that GO could facilitate adhesion and accelerate bone regeneration, as well as induce biominalization and accelerate bone regeneration.

In this study, the increase in corrosion efficiency that can be associated with the creation of an external HA/GO coating has been seen to be more consistent and adherent. The effect of the state of the coating on the properties of the MAO film was also studied.

2. Methodology
The process of micro-arc oxidation was used to form an oxide layer on the 1 mm × 1 mm × 1 mm dimension AZ91D magnesium alloy. There was four sample were prepared and three procedures have been completed for the creation of a coating that is a method of mounting, polishing and MAO coating. Epoxy resins were used to mount the magnesium until it was polished with emery paper #1000. A constant applied to the samples with various time treatment (5, 10, 15 and 20 min) was kept at ambient temperature by DC power supply MODEL QRP-60H15D in 230 voltages. The MAO procedure involved positioning the sample as an anode and the stainless-steel plate as a cathode. The electrolyte is prepared by using, 54 g/L sodium phosphate, 2 g/L sodium hydroxide, 5 g/L calcium carbonate, 3 g/L hydroxyapatite and 2g/L graphene oxide. The coating samples were then put in the desiccator for 24 hours for further processing. MITUTOYO CS-3100 machine was used for testing the surface roughness of the sample after the coating process. Each analysis was conducted three times at various horizontal direction. Ra was used to calculate the magnitude of the arithmetic average roughness. The overall surface roughness average was determined on the basis of the equation (1).

\[ Ra = \frac{Ra_{(1)} + Ra_{(2)} + Ra_{(3)}}{3} \]  

(1)

Scanning electron microscopy was also observed to analyze the surface morphology on the coated sample and Energy dispersion x-ray spectrometry (EDS) were used to characterized the elemental composition of the coated sample microstructure.

3. Result
3.1. Morphology surface analysis
The microstructure of the samples after the HA/GO coating is shown in figure 1. Samples composed of many circular pores with a pattern of inhomogeneous waves during the MAO rising stage, molten oxide trapping and gas bubbles. Figure 1 (a) with a period of 5 minutes during MAO coating had a rougher surface than (b) with a duration of 15 minutes, and several cracks were observed on the MAO substrate because the moulded oxide cooled rapidly in the comparatively cooling electrolyte at a temperature of thousands degrees [11]. Due to the fact that the oxide coating interface to the air is trapped by the MAO coating, the total thermal force in the accelerated solidification of the metal will easily form these cracked coatings [12].

In comparison, the HA/GO coating shown in figure 1(b) showed a smooth surface with a small pore size but a rougher surface in figure 1(a) and a large crack. Most small pores on the surface can lead to hydroxyapatite and graphene additives, which are excreted on the surface and the small pores sealed on the surface with the leading force produced by the electrical field during the MAO process (figure 1 (b)) [10]. It can be concluded the longer the time of the MAO coating, the fewer cracks form.
3.2. Elemental composition analysis

The study of the elemental composition was shown in figure 2 by the composition and percentage of the coated samples which were treated for 5 and 15 minutes at the time of treatment. These samples were coated with Magnesium (red) and some areas with other components. Carbon (dark blue) and phosphate (green) elements were found on the surface coating (a) and (b) suggesting that HA/GO had penetrated the coating. Conversely, the formation of micro cracks on the sample (a) and (b) is packed with oxygen from the GO solution [13]. The homogeneous HA/GO factor on the coating (b) improves the resistance of the sample to corrosion [14][15]. The greater the existence of GO factor, the better the improvement of corrosion resistance for that sample was in close agreement with Han, which stated the effect of corrosion resistance on AZ91D [16]. Besides, the presents of Phosphate in the sample helped for modifying the mechanical and corrosion properties of Mg alloys [17]. A part of it, the percentage of elements for those samples showed the O was still the highest followed by P, C, and lastly Mg. The presence of Carbon element at 15.6 % for 15 minutes (b) and 12.4 % for 5 minutes (a) exhibited that the samples were successfully coated by the coating process.

**Figure 1.** SEM morphology of sample (a) with 5 minutes and (b) 15 minutes of time treatment.

**Figure 2.** Distribution of composition by colours and percentage time treatment of (a) 5 minutes (b) 15 minutes.
3.3. Surface roughness

The surface roughness was measured using the average arithmetic roughness, Ra. The surface roughness was measured using the average arithmetic roughness, Ra. From the study, the value of surface roughness was increased as the duration of treatment increased, as seen in figure 3. The highest surface roughness was 18.907 μm at 20 min length of treatment and the lowest surface roughness was 9.582 μm at 15 min. In this case, the optimal value for roughness was 9.582 μm. The corrosion rate will also be decreased as the roughness of the sample reduces [18]. This is because the contact area between the solution and the specimen was reduced in an alkaline solution and the corrosion reactions decreased indirectly [19]. In acidic solution (NaCl), as the roughness reduces, the collection sites of Hydrogen Chloride, HCL have been decreased and the chloride reactive has become low [19]. However, Guangling Song [20] suggested that the deeper and rougher the surface morphology, the more hydrogen gas is emitted. Excessive gas evolution can inhibit bone consolidation and change the bone transformation mechanism [21]. In conclusion, Ra=9.582μm was accepted due to the corrosion rate decrease with decreasing surface roughness [22].

![Figure 3. Surface roughness value with different time treatment in minutes.](image)

3.4. Conclusion

The findings and a series of studies have been shown to explore the physical properties of hydroxyapatite and graphene-coated magnesium alloys relevant to morphology and structural phases. New hydroxyapatite and graphene coatings on Mg substrates is used in a convenient MAO method for biodegradable implants. Hydroxyapatite and graphene elements have been successfully incorporated into the composite layer. The inclusion of hydroxyapatite and graphene electrolyte minimizes the porosity of the coated samples. Working with variable parameters that are time during the coating process has affected the surface morphology that can be used in the study of the SEM and EDS effects.

In fact, the surface texture of the powder coating in relation to the roughness measure, in which the average surface roughness values are determined, was approved at 15 minutes by 9,582 μm due to a substantial decrease in bone-implant integrity. This is because the final result of SEM and EDS confirms that hydroxyapatite and graphene oxide are well coated after 15 minutes of coating time compared to other samples. As a final conclusion, it should be noted that, these experiments have focused on one or two coating properties and the maximum range of functionality and coating properties. However, full characterization is necessary for use as an implant, including all essential factors such as surface chemistry, adhesion and morphology of the coating. This study has demonstrated the possible safety of AZ91D magnesium alloy, which is intended to help biomedical approaches.
4. References

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