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Synthesis of biodegradable Mg−Zn alloy by mechanical alloying: Statistical prediction of elastic modulus and mass loss using fractional factorial design

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Abstract: Biodegradable Mg−Zn alloy was synthesized using mechanical alloying where a statistical model was developed using fractional factorial design to predict elastic modulus and mass loss of the bulk alloy. The effects of mechanical alloying parameters (i.e., milling time, milling speed, ball-to-powder mass ratio and Zn content) and their interactions were investigated involving 4 numerical factors with 2 replicates, thus 16 runs of two-level fractional factorial design. Results of analysis of variance (ANOVA), regression analysis and R^2 test indicated good accuracy of the model. The statistical model determined that the elastic modulus of biodegradable Mg−Zn alloy was between 40.18 and 47.88 GPa, which was improved and resembled that of natural bone (30−57 GPa). Corrosion resistance (mass loss of pure Mg, 33.74 mg) was enhanced with addition of 3%−10% Zn (between 9.32 and 15.38 mg). The most significant independent variable was Zn content, and only the interaction of milling time and ball-to-powder mass ratio was significant as *P*-value was less than 0.05. Interestingly, mechanical properties (represented by elastic modulus) and corrosion resistance (represented by mass loss) of biodegradable Mg−Zn alloy can be statistically predicted according to the developed models.

Key words: biodegradable Mg−Zn alloy; mechanical alloying; fractional factorial design; elastic modulus; mass loss

1 Introduction

Recently, magnesium (Mg) and its alloys have attracted increasing attention as innovative biodegradable materials, particularly for their potential use as temporary orthopedic implants and coronary stents. Mg alloys have shown potential for use in biodegradable materials due to their excellent biological performance and biodegradability in the bioenvironment [1]. In terms of mechanical properties, Mg is very compatible with natural bone. Its density (1.74 g/cm^3) and elastic modulus (45−48 GPa) are closer to those of bone (1.8−2.1 g/cm^3 and 30−57 GPa) [2] than in the case of other currently used biomaterials for fixation of fractured bone, like titanium (Ti) alloys, stainless steels or cobalt−chromium (Co−Cr) alloys (approximately 100, 180 and 210 GPa, respectively) [3,4]. For biocompatibility, a large number of Mg ions are present in the human body and are involved in many metabolic reactions and biological mechanisms. The human body usually contains approximately 35 g Mg per 70 kg body, and the daily dietary demand for Mg is about 375 mg [5]. Mg-based biomaterials have been demonstrated to stimulate the formation of new tissue when they are implanted as bone fixtures. It has been accepted that there are no serious concerns regarding the harm that can be caused by Mg ions to the human body [6]. However, Mg-based biomaterials are susceptible to attack in chloride-containing solutions, such as the human body fluid or blood plasma [7].

The most effective way to improve mechanical integrity and degradation behavior of Mg is by alloying with additional elements. Mechanically, zinc (Zn) strengthens Mg alloys and, importantly, Zn could enhance both corrosion potential and Faraday charge transfer resistance of Mg, thus improving corrosion resistance [3]. Clinically, Zn is an essential trace element in the human body, and compared with several other metal ions with similar chemical properties, Zn is relatively harmless [8]. Hence, in this study Zn was chosen as the alloying element and incorporated into the

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Mg matrix with the aim of improving mechanical properties and reducing corrosion rate of Mg.

Recently, the use of powder metallurgy (PM) coupled with mechanical alloying (MA) to synthesize Mg-based alloys is a field of growing interest. This technique is a solid state powder metallurgical process in which elemental powders are alloyed by a repeated deformation mechanism under frequent mechanical impacts [9,10]. MA is one of the simplest and most economical routes for the fabrication of nanocrystalline biometallic materials. In this current work, binary Mg−Zn alloy was produced using mechanical alloying (MA) followed consolidation process by compaction and sintering.

Regularly conventional research methodology via trial and error method is adopted to ascertain the important processing conditions, but certainly, this methodology is both costly and time-consuming for the volume or number of experimental work to be performed [11,12]. Thus, to screen out MA parameters in preparing Mg−Zn alloy we used the design of experiment (DOE) method. This approach helps to understand better how the change in the levels of application of a group of parameters affects the response [13]. When certain high-order interactions are probably negligible, information on the main effects and low-order interactions may be obtained by running only a fraction of complete factorial design [14,15]. Of the available method, a fractional factorial design (FFD) is the most widely used types of design for product and process design and for process improvement. Interestingly, the FFD is a variation of the basic factorial design in which only a subset of the run is used. In developing the regression equation, the test variables were coded according to the following equation:

$$X_j=(Z_j-Z_{0j})/\Delta_j \quad (1)$$

where X_j is the coded value of independent variable, Z_j is the real value of the independent variable, Z_{0j} is the value of independent variable on center point and Δ_j is the step change value. The linear model can be expressed as follows:

$$Y=\beta_0\sum_{j=1}^{n}\beta_j X_j \quad (2)$$

where Y is the predicted response, β_0 is the intercept, β_j is the jth linear coefficient and X_j is the input variable which influences the response.

Referring to the present work, the FFD method enables the establishment of the polynomial functions that describe the effects of MA processing conditions on the final properties of Mg−Zn alloy, knowledge of which is beneficial in further use of fabrication of Mg−Zn alloy. Interestingly, only a fraction of actual experiment number is required to be run without forfeiting the accuracy of the final properties [12]. To sum up, high accuracy of developed mathematical models can quantify the experimental output efficiently and economically using FFD.

2 Experimental

2.1 Materials preparation and characterization

A mixture of elemental Mg (99.00% purity) and Zn (99.70% purity) powders was mechanically milled at room temperature using a high-energy Fritsch Pulveristte P-5 planetary mill under argon atmosphere. The particle size of the powder was measured by particle size analyzer using Helos (H1938) & Rodos equipment. The measurement was carried out in dry condition as the Mg−Zn powder is very reactive to humidity. SEM micrographs of the starting Mg and Zn powders are shown in Fig. 1. Mg powders are irregular-shaped and Zn powders are mostly ellipsoidal, elongated particles.

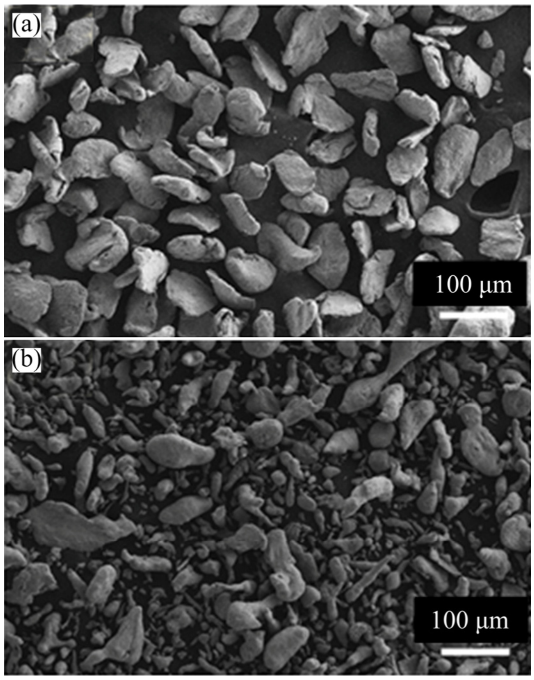

Fig. 1 SEM images of elemental Mg (a) and Zn (b) powders

The particle sizes of elemental Mg powder and Zn powders are up to 227.41 μm and 121.65 μm respectively, as listed in Table 1. 20 mm-diameter stainless steel balls were used during mechanical alloying. 3% n-heptane was added to the powder mixture prior to the milling process to prevent excessive cold welding of the elemental alloy powders.

Then, the milled powders were uniaxially cold pressed under 400 MPa for 2 min at room temperature to produce 10 mm-diameter of green Mg−Zn alloy compacts and sintered at 350 °C under argon flow at