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The compression behaviors of titanium/carbamide powder mixtures in the preparation of biomedical titanium scaffolds with the space holder method

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ABSTRACT

Titanium has been widely used as a preferred metallic biomaterial for bone tissue engineering scaffolds. So far, a number of techniques have been developed to produce porous-structured titanium, including the space holder method. However, a number of technological challenges are still present, such as the difficulties in controlling the geometry changes of space-holding particles during the compaction of titanium/space holder powder mixtures. In this research, a series of experiments were performed to investigate the compression behaviors of titanium/carbamide powder mixtures and determine maximum compacting pressures, considering the specific net energy expended during the process, the at-pressure relative density of powder compacts and the yield pressure derived from the load–displacement plots of powder compression cycles. The results showed that the titanium and carbamide powders exhibited dissimilar compression behaviors and titanium/carbamide powder mixtures exhibited intermediate compression behaviors of the monolithic titanium and carbamide powders. The yield pressures of titanium/carbamide powder mixtures, obtained from the Heckel plots and used as a measure for the appropriate compacting pressures, decreased with increasing volume fraction of carbamide in the mixture. The rule of mixtures was found to be applicable to predict the compression behaviors and the appropriate compacting pressures of titanium/carbamide powder mixtures. However, limitations of this model were recognized in the case of compacting mixtures with large volume fractions of carbamide space-holding particles ($x > 50\%$) and at high pressures ($P > 300$ MPa).

1. Introduction

In recent years, bone tissue engineering has been increasingly studied as a preferable approach to the restoration of damaged bone tissue. In this approach, osteogenic cells harvested from the patient are cultured in vitro, seeded onto a graft and then implanted to repair the damaged bone tissue [1]. The graft is of critical importance in this approach, because it acts as a carrier that provides a template and guidance to the bone tissue growth in the defect site. Clinical applications are however quite limited mainly because of disease transmissions and infections from donor to recipient, donor site morbidity, bone defect size, viability of the host bed and availability. Overcoming the limitations associated with the autogenous graft calls for the development of a synthetic material, namely a scaffold [2]. The scaffold with a porous structure is constructed to facilitate the formation of new cells, cellular activities and the transport of nutrients and oxygen required for bone tissue regeneration. In addition, the elastic modulus of the scaffold can be controlled by modifying its porous structure, thereby minimizing stress shielding [3].

Currently, titanium and its alloys are considered preferred materials for bone tissue engineering scaffolds owing to their excellent biocompatibility and corrosion resistance [4]. So far, a number of techniques have been developed to produce porous-structured titanium, such as powder sintering, expansion of pressurized gas bubbles, powder deposition, rapid prototyping and space holder method [5]. Powder sintering leads to porous-structured titanium simply from the neck formation between neighboring powder particles. With this method, however, the maximum porosity of titanium scaffolds is limited to 35% [6]. In addition, the pores in a sintered titanium powder are poorly interconnected and their shapes and sizes depend on the starting powder particles [7]. To overcome these limitations, the space holder method has been introduced. With this method, titanium scaffolds having 55–75% porosity and interconnected pores could be made [8]. Moreover, the control over pore shape and sizes could be exercised by adjusting the geometry and sizes of space-holding particles used in the scaffold fabrication process [8].

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In principle, the fabrication chain of titanium scaffolds with the space holder method is composed of a series of processing steps, i.e., (i) powder mixing, (ii) compaction, (iii) space holder removal and (iv) sintering [9]. At the first step, a titanium matrix powder is mixed with a space-holding powder that serves as a pore former in the scaffold. The mixture is then compacted to produce a scaffold preform prior to the removal of space-holding particles to create new pores in the scaffold. Sintering is performed at the end of the fabrication chain to achieve permanent bonding between titanium powder particles and to form the skeleton of the scaffold. Despite the initial success in using this method, a number of technical challenges are still present [10].

Compaction has been considered a critically important step, as it determines the porous structure of the scaffold product. At this step, a powder mixture composed of titanium and space-holding particles is compacted under a compressive pressure by means of a uniaxial or isostatic powder press. A relatively high compacting pressure is usually applied to produce a scaffold preform in order to obtain a sufficient green strength that keeps the scaffold preform intact at the subsequent processing steps, e.g., during the removal of space-holding particles [11]. However, the geometry of space-holding particles may markedly change under a compacting pressure that exceeds their yield strength or even ultimate strength. As a consequence, the pore shape and sizes of the scaffold will deviate significantly from the initial shape and sizes of space-holding particles [12]. Therefore, a critical compacting pressure that results in geometrical deformation of carbamide space holder needs to be determined to avoid such an undesirable situation.

Recently, a number of ways to determine an appropriate compacting pressure in scaffold fabrication with the space holder method have been reported, for example, by means of visual inspection [13] or evaluation of the mechanical properties [13] and shrinkage percentage [14] of the scaffold. However, there is no information in the open literature on the methods based on powder behaviors under compressive stresses for the determination of appropriate compacting pressures in the preparation of titanium scaffolds with the space holder method.

In this research, a series of powder compaction experiments were carried out to investigate the compression behaviors of titanium/carbamide powder mixtures with various volume fractions of carbamide under compressive stresses. An instrumented uniaxial powder press was devised to acquire load–displacement plots from the powder compaction cycle. By using these data, the compression behaviors of powder mixtures could be analyzed. In addition, possible compression mechanisms responsible for the observed behaviors of titanium/carbamide powder mixtures were established. Critical compacting pressures that result in geometrical changes or fragmentation of carbamide space-holding particles in titanium scaffold preform were determined. An organic carbamide powder $CO(NH_2)_2$ was chosen as the space holder due to its wide applications in the fabrication of titanium scaffolds and its excellent removal capacity from the scaffold preform, in particular through the water leaching process [15].

2. Materials and methods

2.1. Powder mixture preparation

A gas-atomized grade 1 titanium powder (TLS Technik GmbH & Co., Germany) with a spherical particle shape (Fig. 1a) and a cubical carbamide powder (Merck, Germany) (Fig. 1b) were chosen as the matrix and space holder materials, respectively. Particle sizes of both powders were analyzed using a Mastersizer X (Malvern, UK). With this technique, the volume mean diameters D_{av} of titanium and carbamide powder particles were determined to be 72.46 ± 1.81 μm and 417.80 ± 5.64 μm, respectively, and their median diameters D_{50} were 70.32 ± 1.61 μm and 399.23 ± 4.85 μm, respectively.

In order to prevent titanium/carbamide powder mixture from segregating, a binder solution prepared from polyvinyl-alcohol (PVA) powder (Alfa Aesar GmbH & Co KG, Germany) dissolved in tap water was added to the titanium powder and mixed for 1 h, prior to mixing with the carbamide powder. The amount of the PVA binder solution added was 3% of the volume of the titanium powder in the mixing container. Twenty grams of wet titanium powder was then put together with 2.52, 5.88, and 13.72 g dry carbamide powder for 3 h mixing in a cylindrical roller mixer (CAT, Germany) in order to obtain titanium/carbamide powder mixtures with carbamide volume fractions x of 30, 50 and 70%, respectively.

2.2. Powder compaction

Titanium/carbamide powder mixtures were cold-compacted in a die with a diameter of 13 mm. Prior to powder filling, a zinc stearate powder was applied to the die inner surface for lubrication. A powder mixture of 1.3 g was poured into the die cavity, tapped manually and pressed at room temperature by using an instrumented Instron 4505 compression tester (Instron, USA). With this tester, precise loads exerted by the upper punch during a powder compression cycle could be registered. The punch speed applied during powder compression and decompression was set at 2 mm/min. Prior to compaction, the powder mixture was pre-compressed at a load of 50 N to level off the powder bed surface. The load–displacement plots obtained from the powder compression cycles were first corrected for the deformation of the machine and punches by using the plot obtained from a dry run without powder mixture and then used for the analysis of powder compression behaviors. The compaction of each of the powder mixtures was conducted in triplicate.

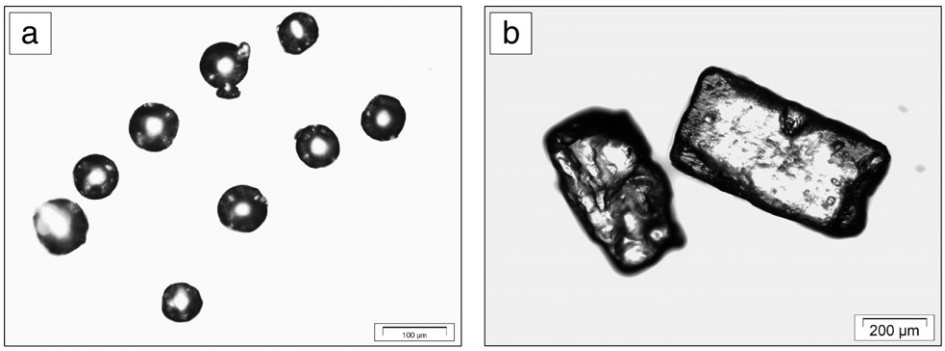

Fig. 1. (a) Titanium powder and (b) carbamide powder.