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High-efficiency combinatorial approach as an effective tool for accelerating metallic biomaterials research and discovery

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ABSTRACT

A high-efficiency combinatorial approach has been applied to rapidly build the database of composition-dependent elastic modulus and hardness of the Ti–Ta and Ti–Zr–Ta systems. A diffusion multiple of the Ti–Zr–Ta system was manufactured, then annealed at 1173 K for 1800 h, and water quenched to room temperature. Extensive interdiffusion among Ti, Zr and Ta has taken place. Combining nanoindentation and electron probe micro-analysis (EPMA), the elastic modulus, hardness as well as composition across the diffusion multiple were determined. The composition/elastic modulus/hardness relationship of the Ti–Ta and Ti–Zr–Ta alloys has been obtained. It was found that the elastic modulus and hardness depend strongly on the Ta and Zr content. The result can be used to accelerate the discovery/development of bio-titanium alloys for different components in implant prosthesis.

1. Introduction

There are three categories of metallic biomaterials commonly used for prosthesis implantation: stainless steels, Co-based alloys and titanium alloys [1]. Compared with stainless steels and Co-based alloys, titanium alloys have the following characteristics: low elastic modulus, high corrosion resistance, and superior biocompatibility [2,3]. High elastic modulus of metallic biomaterials, as compared with that of bone tissue (10–30 GPa), can cause so-called stress shielding effect [4]. Consequently it leads to bone resorption and gives rise to failure of implant [5,6]. In the past decade, both near β and β-type titanium alloys comprising non-toxic and non-allergic elements (i.e., niobium, zirconium, tantalum, tin and molybdenum) were widely investigated in order to achieve low Young's modulus and good mechanical properties [7–10]. If alloys have an elastic modulus matching that of bone tissue, 'stress shielding' effect could be mitigated or even eliminated. Song et al. [11] indicated that addition of Zr, Nb, Ta, and Mo elements can effectively increase strength and reduce elastic modulus of titanium alloys by means of electronic structural calculations using discrete variational cluster method (DVM).

Binary Ti–Ta and ternary Ti–Zr–Ta systems, free of toxic elements and with a low elastic modulus, have been studied intensively. Acting as a β-stabilizing element, Ta can significantly reduce the martensitic transformation temperature (Ms) of Ti. With a gradual increase of Ta, it is possible to decrease Ms of Ti alloy to room temperature (i.e., the high-temperature β-phase can be retained to room temperature during rapid cooling). With certain preferred addition of Ta, we can also reduce the elastic modulus of titanium alloys. The relatively neutral element, Zr, can work as a β-stabilizing element to form a homogeneous solid solution in α phase and β phase. Additionally, Zr can increase room temperature tensile strength with little effect on ductility and also improve corrosion resistance and blood compatibility [12].

The binary Ti–Ta system has been investigated extensively [13–16] where Trillo et al. [13] have shown that the corrosion capacity of Ti–40 wt.% Ta and Ti–50 wt.% Ta alloys surpassed that of Ti–6Al–4V alloy in simulated biological solution. Zhou et al. [17] have studied the effects of Ta content on the microstructures, dynamic Young's modulus and tensile properties of quenched Ti–Ta alloys. The results indicate that the microstructures of the quenched binary Ti–Ta alloys are sensitive to Ta content. Ti–30 wt.% Ta, containing needle-like orthorhombic martensite (α″) structure, and Ti–70 wt.% Ta, containing single metastable β phase, both have a good combination of high strength and low modulus among all the studied Ti–Ta alloys. Using first-principles calculation based on the density functional theory, Wu et al. [15] find phase stability, tetragonal shear constant C, bulk modulus, elastic modulus and shear modulus of β type Ti–Ta alloys increasing monotonously

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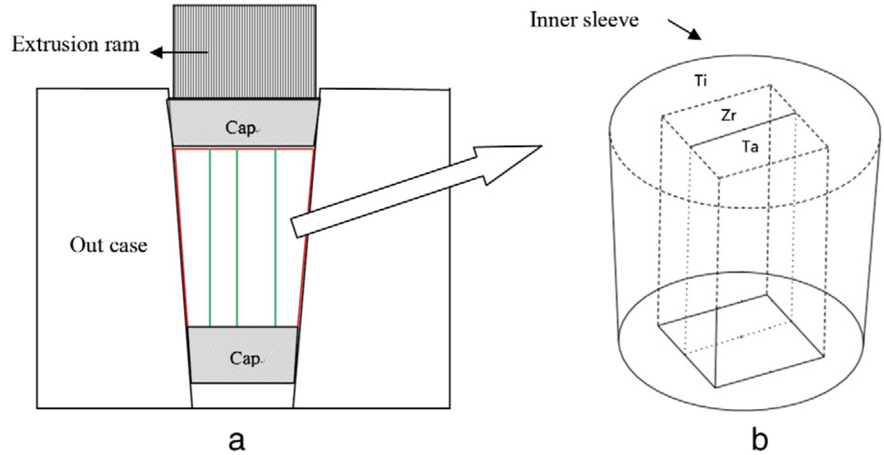

Fig. 1. Method used for the manufacture of the diffusion multiple.

with Ta content. The lowest elastic modulus is reached when valence electron number (e/a) is around 4.25.

As for the binary Ti–Zr system, Equo et al. [18] had studied the hardness and tensile properties. They revealed that hardness of the alloy containing 50% zirconium was approximately 2.5 times higher than that of pure titanium and pure zirconium, and tensile tests showed a similar tendency. Furthermore, Ti–Zr alloys are also reported to have good corrosion resistance and biocompatibility [19,20]. Another attractiveness of alloying zirconium to titanium is that the fusion temperature of titanium (1670 °C) is lowered with an increase in the amount of zirconium, thus facilitating the casting process [21].

Only a restricted subset of alloy compositions that may have a relatively lower elastic modulus in Ti–Zr–Ta system has been experimentally investigated. Thus, the Ti–Zr–Ta system with a large composition space offers tremendous opportunities for optimization of alloy composition and thermo-mechanical treatment. It is currently very time-consuming and expensive to handle one composition at a time for testing of properties. Therefore, combinatorial study is used in our research.

The high-efficiency diffusion multiples, which form continuous composition gradients of alloying elements in one sample, have been developed by Zhao [22]. This kind of combinatorial survey has its advantages compared to the conventional one-alloy-at-a-time practice. It can rapidly analyze rather large compositional space generated by diffusion multiples, and greatly reduces the time spent for each step of making an ingot of individual composition, homogenizing it, preparing a sample, and loading it into different testing systems [23].

In this study, a diffusion multiple of Ti–Zr–Ta was manufactured and annealed at 1173 k for 1800 h to form continuous composition gradient. By combining nanoindentation and electron probe micro-analysis (EPMA), the indentation-based elastic modulus, hardness as well as composition across the diffusion multiple were determined. Based on the experimental data, composition dependent elastic modulus and hardness data of binary Ti–Ta and ternary Ti–Zr–Ta systems were obtained. The results from only one diffusion multiple sample can be used to accelerate the discovery/development of bio-titanium alloys for different components in implant prosthesis. This is a basic research study to investigate the nature of Ti–Ta and Ti–Zr–Ta alloys and to collect useful data for the alloy-design of new biomedical materials.

2. Experimental procedure

2.1. Preparation of the diffusion multiples

The diffusion multiple was manufactured with the method shown in Fig. 1. The outer case was made with titanium. In order to alleviate the influence of the outer case to the internal sample during the annealing process, a tantalum sheet (0.05 mm) was added between the outer case and the Ti inner sleeve. High purity Zr (99.95%) and Ta (99.95%) were machined into the prismatic bars of 5 × 10 × 21 mm by electro-discharge machining (EDM), ground, polished, ultrasonically cleaned in alcohol, and assembled into the Ti inner sleeve. Two Ti caps of 3 mm thickness were put at the ends of the inner sleeve to prevent

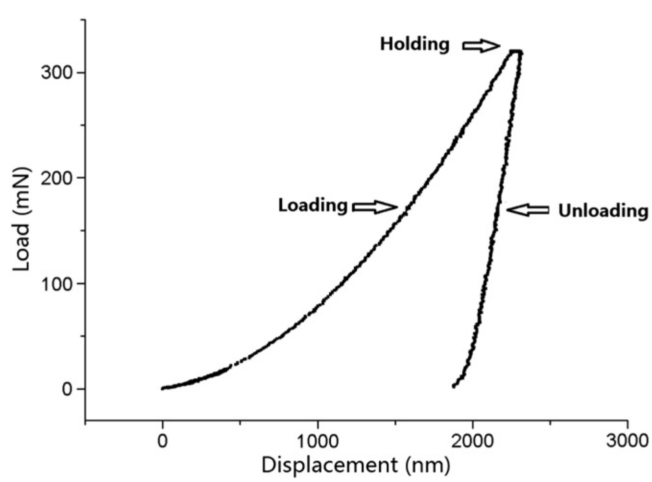

Fig. 2. A schematic representation of displacement *Vs* load.

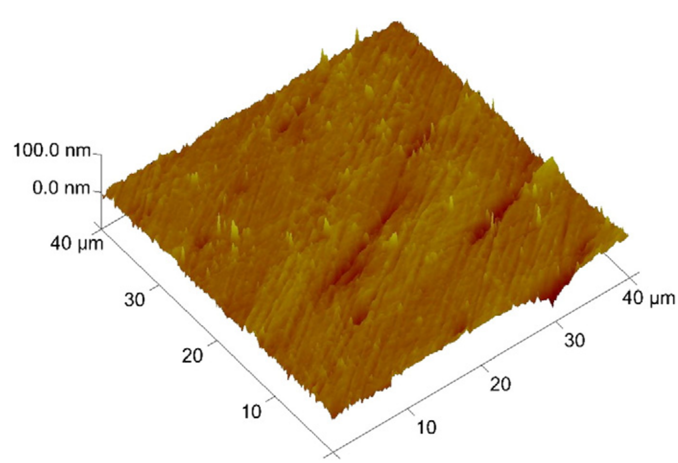

Fig. 3. Typical AFM image of the diffusion multiple surfaces.