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High energy X-ray diffraction study of a dental ceramics–titanium functional gradient material prepared by field assisted sintering technique

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ARTICLE DATA

Article history:
Received 10 March 2014
Received in revised form 17 June 2014
Accepted 20 June 2014
Available online 21 June 2014

Keywords:
Functional gradient material
Dental implants
Titanium
Field assisted sintering
High energy X-ray diffraction

ABSTRACT

A functional gradient material with eleven layers composed of a dental ceramics and titanium was successfully consolidated using field assisted sintering technique in a two-step sintering process. High energy X-ray diffraction studies on the gradient were performed at High Energy Material Science beamline at Desy in Hamburg. Phase composition, crystal unit edges and lattice mismatch along the gradient were determined applying Rietveld refinement procedure. Phase analysis revealed that the main crystalline phase present in the gradient is α-Ti. Crystallinity increases stepwise along the gradient with a decreasing increment between every next layer, following rather the weight fraction of titanium. The crystal unit edge a of titanium remains approximately constant with a value of 2.9686(1) Å, while c is reduced with increasing amount of titanium. In the layer with pure titanium the crystal unit edge c is constant with a value of 4.7174(2) Å. The lattice mismatch leading to an internal stress was calculated over the whole gradient. It was found that the maximal internal stress in titanium embedded in the studied gradient is significantly smaller than its yield strength, which implies that the structure of titanium along the whole gradient is mechanically stable.

1. Introduction

A dental implant is a prosthetic device applied to reconstruct the masticatory function, if the tooth root has to be extracted. Such dental implant is surgically placed in the jaw-bone from the outside and it has to replace both functions of the bone tissue, as well as the tooth itself. Therefore, the function of a dental implant is quite different at different positions i.e. inside, outside and at the boundary of the bone [1].

During the past years research and development of dental implants have been focused on implants composed of only one material, sometimes covered with a coating layer. These implants are essentially uniform in their composition and structure [2–4]. Thus, the concept of a functional gradient material (FGM) may be suitable for designing and obtaining novel dental implants. A FGM is a composite of two or more phases, where the composition and therefore, the mechanical and physical properties, change gradually over the sample [5].

The design of such layered structures has also some disadvantages, such as complexity of scaling up the process to a mass production, difficulty in scaling up the gradient materials to large specimens or the nucleation of stress at the

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http://dx.doi.org/10.1016/j.matchar.2014.06.018
1044-5803/

interfaces which reduces the reliability of the gradient under operating conditions [6]. To overcome these difficulties novel approaches have to be considered. A promising technique for the practical realisation of such gradient materials is the field assisted sintering technique (FAST), also known as spark plasma sintering (SPS) [6–8]. FAST is a rather new powder consolidation technique which applies a pulsed direct current and mechanical load to assist sintering [9–13]. The pulsed current through the die and the sample leads to a rapid heating and to short process times. But the main advantages of FAST over conventional methods like hot pressing or pressureless sintering are lower sintering temperatures, shorter exposure to elevated temperatures, no need for a binder or other additives, as well as important improvements in the properties of the sintered materials [9,10,13]. Another special feature of FAST is the ability to consolidate dissimilar materials like metallic particles, polymers or ceramics into complex nanostructured systems [6–8,10].

Nowadays, titanium and titanium alloys due to their excellent mechanical properties and high corrosion resistance are considered standard materials for dental implants with very well documented high rates of success and survival [2,14]. Potential immunologic and aesthetic drawbacks associated with titanium implants have resulted in the development of alternatives like zirconium dioxide-based dental implants. Zirconium dioxide seems to be a suitable implant material because of its tooth-like colour, mechanical properties, biocompatibility and low plaque affinity [3,4,14].

Previous experiments on binding Ti with a dental ceramics in a traditional firing process revealed that different thermal expansion coefficients of these materials and the formation of oxygen layers lead to the formation of fractures [15,16]. The application of field assisted sintering technique and the gradual change in composition of the dental ceramics–Ti gradient may overcome these problems. Moreover, the structural properties of such gradient materials combining amorphous and crystalline phases are less known and are still open to research. Therefore, it was interesting to study by high energy X-ray diffraction the structural changes i.e. the phase composition, the evolution of the crystallinity and the crystal unit edges, as well as the crystal lattice mismatch of titanium along this functional gradient material.

2. Gradient material and experimental set-up

2.1. Gradient material

As dental ceramics, the zirconium oxide-based, amorphous commercial powder Dentine VitaVM7 (Vita Zahnfabrik Bad Säckingen, Germany) with an average grain size of 18 μm was selected. The titanium powder was a pure α-phase Ti with a purity of 99.5% and an average grain size of 44 μm.

The dental ceramics–Ti functional gradient material was arranged in an eleven layer system with volume fractions of ceramics and Ti varying with 10% step in each layer, as presented in Fig. 1. All layers were designed to have a thickness of approximately 1 mm. The appropriate amounts of powders were weighted using a precise laboratory balance and, in order to ensure homogeneity, blended in grinding jars for 20 min. The obtained mixtures of powders were stacked horizontally layer by layer in a graphite die with an inner diameter of 40 mm, according to the predesigned compositional profile. The stacked material was separated from inner walls of the die and the punches with a graphite foil in order to prevent the material from reacting with the die and to ensure an electrical contact. Furthermore, to reduce radiation heat losses from the outer surface of the die, the graphite die was covered with a porous carbon felt. The application of a carbon felt also reduces possible gradients of temperature in the sample [17,18] and ensures maximal possible in-plane homogeneity of the obtained sample. The field assisted sintering technique was applied in order to consolidate the dental ceramics–Ti gradient material. The sintering procedure was performed in Tycho Sinterlab Rostock using a HP D125 unit from FCT Systeme GmbH Rauenstein, Germany.

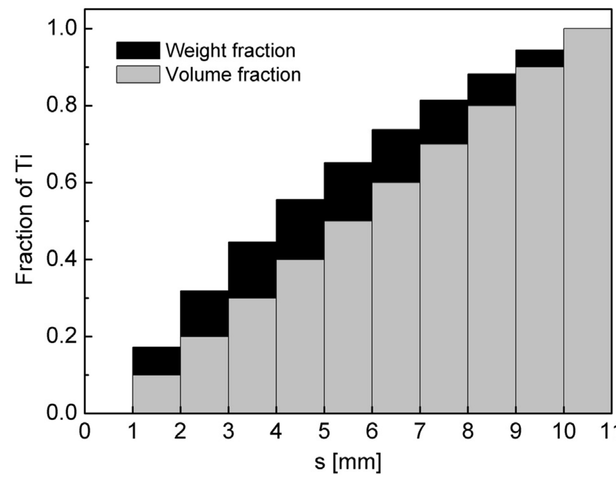

Fig. 1 – Predefined compositional profile of the dental ceramics–Ti functional gradient material: the volume fraction and the corresponding weight fraction of titanium as a function of the position *s* in the sample.

Previous attempts of sintering the dental ceramics–Ti gradient in a one step process at 900 °C with a holding time of 5 min revealed that the dental ceramics tended to darken as a result of carbon diffusion into the sample. Furthermore, a decrease of the sintering temperature to 700 °C with the same process time eliminated the problem of discolouration of the dental ceramics, but in this case Ti was not completely densified. Motivated by the above, a two-step sintering procedure, i.e. pre-sintering at 500 °C and main sintering at 700 °C with a heating rate of 40 K/min and a holding time of 6 min in both cases, was introduced in order to achieve a solid, well densified material and to avoid discolouration of the dental ceramics.

The sintering process was performed in vacuum of approximately 1 mbar. During the sintering a pressure of 28 MPa was applied on the sample. In the last phase of the process, pressure was released and the sample was cooled down with a natural cooling rate of approximately 83 K/min between 700 °C and 400 °C. It is worth to mention that due to technical limitations, the heating process below 400 °C was controlled by a thermocouple mounted in the graphite die and above 400 °C by an optical pyrometer focused on a surface