Acta Biomaterialia 8 (2012) 1948–1956

Contents lists available at SciVerse ScienceDirect

Acta Biomaterialia

journal homepage: www.elsevier.com/locate/actabiomat

Osteoclast resorption of thermal spray hydoxyapatite coatings is influenced by surface topography

Karlis A. Gross [a,b,*], Dirk Muller [b], Helen Lucas [c], David R. Haynes [c]

[a] Institute of Biomaterials and Biomechanics, Riga Technical University, Latvia
[b] Department of Materials Engineering, Monash University, Victoria 3168, Australia
[c] Department of Anatomy and Pathology, School of Medical Sciences, University of Adelaide, Australia

ARTICLE INFO

Article history:
Received 27 December 2010
Received in revised form 10 January 2012
Accepted 16 January 2012
Available online 25 January 2012

Keywords:
Topography
Implants
Hydroxyapatite
Coating design
Osteoclast resorption

ABSTRACT

Coating characteristics such as composition, crystallite features and topography collectively impact the cell response. The influence from splats has not yet been assessed for hydroxyapatite (HAp) thermal spray coatings. The objective of this work is to (a) survey the topography on commercial implants, (b) ascertain topography formation from single splats, and (c) determine the osteoclast resorption pattern on a topographically refined coating compared to dentine. Coatings on dental implants, an orthopedic screw, a femoral stem and a knee implant were studied for reference. The effects of substrate pre-heat, roughness, spray distance and particle size on the coating roughness and topography were studied. Human-derived osteoclasts were placed on a coating with refined topography and compared to dentine, a polished coating and polished sintered HAp. A pre-heat of at least 200 °C on titanium was required to form rounded splats. The greatest influence on coating roughness and topography arose from particle size. A 2-fold increase in the mean particle size from 30 to 72 μm produced a significant difference ($P < 0.001$) in roughness from 4.8 and 9.7 μm. A model is shown to illustrate topography formation, nanostructure evolution on single splats, and the topography as seen in commercial implants. Osteoclasts showed a clear preference for activity on coatings with refined topography. A one-way ANOVA test revealed a significantly greater pit depth ($P = 0.022$) for dentine (14 μm) compared to the as-sprayed and polished coating (5 μm). Coatings with topography display a similar number of resorption pits with dentine, but a 10-fold greater number than polished coatings, emphasizing the importance of flattened droplet topography on implant surfaces.

1. Introduction

Orthopedic implants are exposed to osteoclasts, bone-resorbing cells, and osteoblasts, bone-forming cells. The individual contribution of both cell types dictates bone regeneration through the processes of resorption, and bone formation. Biomaterial surface characteristics impact the chain of biological events, leading to a successful clinical outcome. To date, there has been little or no attention on the topography of thermal spray hydroxyapatite (HAp) coatings. This work will survey the surface topography of implants and prostheses, investigate topography control from thermal spraying, and show the human osteoclast resorption of topographically improved HAp coatings.

In the past, attention has been focussed on providing a stable and strong interface with bone. The direct bonding capability of HAp was supplemented by mechanical interlocking of bone with porous surfaces made from sintered beads, wire-mesh or a titanium plasma coating bond-coat (Fig. 1). Pores larger than 100 μm allowed bone ingrowth or abutment into surface cavities. A study on thermally sprayed fluorhydroxyapatite with large changes in surface roughness (R_a of 6 μm vs. 21 μm) provided openings for interdigitation with bone, but did not show any difference in the osteoblast response [1], posing the question of whether more organized topography at the micro- and nano-scale could be important for cell response. The work developed here is motivated by the question, "Is it possible to control the surface topography with thermal spraying, and will this have any effect on the cell response?" This research is an extension of unpublished data from a Masters thesis that showed textured HAp coatings with a topography from flattened molten droplets [2].

Significant effort has been directed at improving the coating mechanical stability with approaches such as strengthening with a secondary phase or an increase in the HAp crystallinity. Inclusion of a second phase, such as zirconia, increases the fracture

* Corresponding author at: Biomaterials research laboratory, Institute of Biomaterials and Biomechanics, Riga Technical University, Riga LV-1048, Latvia.
E-mail addresses: kgross@rtu.lv, karlis.gross@monash.edu (K.A. Gross).

1742-7061/$ - see front matter Crown Copyright © 2012 Published by Elsevier Ltd. on behalf of Acta Materialia Inc. All rights reserved.
doi:10.1016/j.actbio.2012.01.023

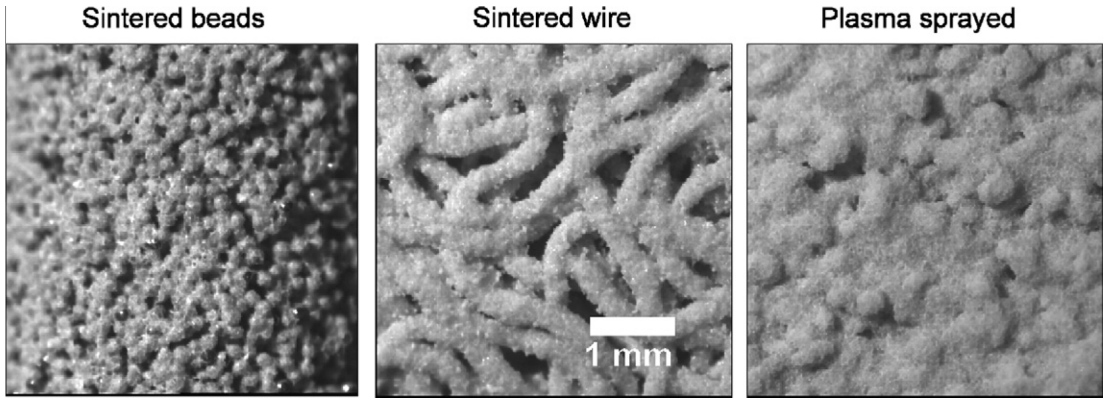

Fig. 1. A hydroxyapatite coating on a sintered bead, a sintered wire and a plasma-sprayed titanium bondcoat shows the macrotopography to facilitate bone ingrowth.

toughness [3], but zirconia should be retained within the coating to provide the sole purpose of reinforcing HAp. Use of a strengthening phase, however, requires a homogeneous crystalline coating. A crystalline coating has been obtained with (a) the use of a ceramic bond-coat under the HAp coating [4,5], (b) a change from air plasma spraying to radio-frequency plasma spraying [6] or vacuum plasma spraying [7], or (c) post treatments such as hydrothermal processing [8]. The alternative has been to spray a thermally stable fluorapatite instead of the more thermally sensitive HAp. While the main focus has been the production of a uniform stable coating, these modifications influence the coating roughness and topography. This work will use increased heat input from the spray gun to encourage crystallization and columnar growth within the coating. Crystal orientation and a designed roughness offer new surface characteristics.

It is difficult to review the details on the coating surface due to the range of terms used in the scientific literature. ‘‘Surface characteristics’’ is used as a general term. ‘‘Roughness’’ implies a measured value from profilometry. ‘‘Morphology’’ describes the shape of features on a surface. ‘‘Topology’’ places attention to the height of features, but ‘‘topography’’ describes the shape and height of surface features. Further discussion will use ‘‘roughness’’ as a quantitative measure and ‘‘topography’’ to refer to the shape and height of surface features.

There has been little research on the effect of thermal spraying on the resulting surface topography. Chemistry has been separated from topography with a thin titanium film to reveal that 80% of the maximum bone-forming response is attributed to topography [9]. Since the coating surface contains numerous features, the most important topographical characteristic could not be determined. Improved cell adhesion and proliferation occur on sintered HAp ground with different SiC papers [10]. A recent literature review on surface topography the need to establish routine measurement methods and a report on specific surface features to allow a comparison with surfaces in journal articles and clearly show the cell response to topographical features to understand the effect of topography on cell response [11]. This work will generate a surface with reproducible features from molten droplets, a control of droplet spreading and crystallization within the splat.

The thermal spray process is an additive process that can control the shape and thickness of each flattened solidified droplet, herein referred to as a splat. This building block offers a new surface feature for which the cell response is presently not known. This work will (a) characterize the surface of commercial coatings, (b) investigate the conditions for producing a rounded splat, (c) determine coating parameters that influence the coating topography, and (d) determine the human osteoclast (OC) resorption of an as-sprayed coating compared to dentine, a polished coating and sintered HAp.

2. Materials and methods

2.1. Assessment of commercial coatings

HAp coatings from three dental implants (Interpore, LifeCore, Biovent), an orthopedic screw (Osseotite®, Orthofix), a hip prosthesis (Margron) and a knee prosthesis (ASDM) were examined in a scanning electron microscope. Joint prostheses presented large areas that allowed the measurement of the coating roughness. The orthopedic screw and the dental implants did not present suitable surfaces for roughness measurement. Conditions for scanning electron microscopy (SEM) and roughness measurement are given in Section 2.4.

2.2. Production of single splats

Commercially pure titanium substrates ($15 \times 15 \times 2.5$ mm) were ground on silicon carbide paper (#400 and #800) for 1 min at a speed of 300 rpm, followed by polishing on an MD-Largo surface with a 3 μm diamond suspension and final polishing with a 0.05 μm colloidal silica on an MD-Chem cloth. Titanium substrates were mounted in an aperture of a muffle furnace to set the titanium surface temperature to 25, 100, 200, 400 and 600 °C to determine the temperature for disc-shape splat formation. Single splats were produced with a Metco 5P torch (Sulzer Metco, Wohlen, Switzerland) used for producing flame-sprayed coatings. A total of 100 splats were counted for each surface temperature condition. The degree of splashing was calculated by dividing the number of circular splats by the number of rounded and splashed splats.

2.3. Coating production

Coatings were flame-sprayed using spray-dried HAp powder (CAM Implants, The Netherlands). Powder was classified by wet sieving to three different particle sizes (20–40, 40–63 and 63–80 μm), dried, re-sieved and then thermally sprayed with an oxygen and acetylene gas. Since all coatings were to be made from rounded splats, the effect of substrate temperature (25 and 400 °C) on the coating roughness was initially assessed for a 20–40 μm powder sprayed at 9 cm onto a polished surface. Then a pre-heat of 400 °C was constant for the remaining experiments. The effects of spray distance (9 and 15 cm) and substrate roughness (R_a of 0.2, 2.2 and 3.6 μm) were then determined. Based on conditions for producing well-molten and spread droplets, three types of coatings were produced on pre-heated (400 °C) polished surfaces with a spray distance of 10, 12 and 15 cm for the 20–40, 40–63 and 63–80 μm powder, respectively. Usually coating adhesion requires spraying onto a grit-blasted surface for a mechanical