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Investigation of the Hydroxyapatite Crystallites in Bone at the Interface with Implant by Neutron Diffraction.

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Abstract.

The basic principles behind human tissue response to artificial surface implantation may be developed under its biological aspect, it is necessary for a medical use to study the mechanical limits of every biomaterials to predict the tissue and the body's response according to the composition, the structure and the design of a biomedical material. To promote a stable and functional direct connection between bone and implant, titanium implants can be coated with materials based on calcium phosphate ceramics such as hydroxyapatite (HAp)($Ca_{10}(P0_4)_6(OH)_2$). The preferred orientation of HAp crystallites at the interface bone-implant in sheep tibia bones has been measured with the neutron 2-axis diffractometer D20 at the Institut Max von Laue-Paul Langevin (ILL), extracted 60 days after implantation. The implant has two faces, one coated and one non-coated with plasma-sprayed HAp (80 µm). We probed the samples with a spatial resolution of 0.5 mm started from the interface in order to inspect the reorganisation of the HAp crystallite's distribution after implantation.

Introduction

Medicine experiences an accelerated development of new technology, aiming preventive, diagnostic and therapeutic progress. Decision makers of health and the experts have to make choices and to establish strategies according to criterions of safety, of effectiveness and of utility. The world market of biomaterials is very important and in full growth. The European commission estimate this market at 25 G€ with an annual growth rate of 5 to 7%. Europe represents a third of this market. The orthopaedic biomaterials present 8 G€ of this market, with an annual growth rate of 7%. Hip and knee prosthetics alone represent 40% of this market segment, with respectively 750 000 and 500 000 medical interventions per year [1].

Bone is a composite material whose components are primarily collagen and HAp. The c-axes of the apatite crystallites and the collagen fibres are preferentially oriented, e.g., in the long bones in the directions of the stresses that the bones need to withstand [2,3].

Bone occurs in two principal structural forms: cortical, or compact bone, which forms a dense matrix, and spongy bone [4].We used in this work cortical bone.

At the interface with an implant, Ti-6Al-4V, a bone tends to change its properties, affecting the acceptance of the implant. Ti-6Al-4V presents good mechanical proprieties and is biocompatible. HAp has low mechanical strength, but very good osteointegration and biocompatibility. The combination of these two materials gives mechanical strength and good osteointegration proprieties at the interface [5]. Plasma spraying is the most popular method for coating implant parts with HAp. In order to improve these coatings, it is necessary to investigate the texture transformations of the

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bone's properties, as a function of the distance from the implant-interface and the time since implantation. It is necessary to remove substantially the collagen in order to reduce the incoherent scattering of hydrogen.

Materials and methods

We used in this study bone material from one sheep with two implants, Ti-6Al-4V, (20 mm x 10 mm x 1.4 mm) inserted in its left and right tibia bones, the implant having one face coated, the other not. The bone specimens have been prepared in the Pius Branzeu Center of Laparoscopic Surgery and Microsurgery (Romania).

After two months of implantation we extracted the two pieces of the bone with the implants, the samples are preserved in the ethanol in order to keep them under the best conditions. It's very necessary to remove the organic part in order to reduce the intense incoherent scattering of neutrons by hydrogen and that is done in the furnace. The heat treatment does not affect the preferred orientation of the mineral bone crystallites and crystallinity [3].

In this study we used the high-intensity two-axis diffractometer with variable resolution: D20, at Institut Max von Laue-Paul Langevin (ILL), Grenoble. D20 equipped with a large one-dimensional curved microstrip detector.

A piece of sheep tibia (10 × 5 × 5 mm) has been mounted in the Euler cradle. We made an embraced scan with step sizes of 10° for $\Delta\varphi$ and $\Delta\chi$, with φ from 0 to 360° and χ from 0° to 90°, and ω constant at 90°. The size of beam was 9 mm × 0.5 mm and λ = 2.4 Å. It takes 6 hours for each slice of 0.5 mm of the tibia.

Results and discussion

The WIMV method [6] has been used for describing the texture, implemented into the MAUD program [7] (Materiel Analysis Using Diffraction) of L. Lutterotti [8]. We refined first the crystal structure from the sum of all data and afterwards the texture of the 360 whole neutron powder diffraction patterns. The MAUD Rietveld texture analysis program allows to trace the pole figures directly.

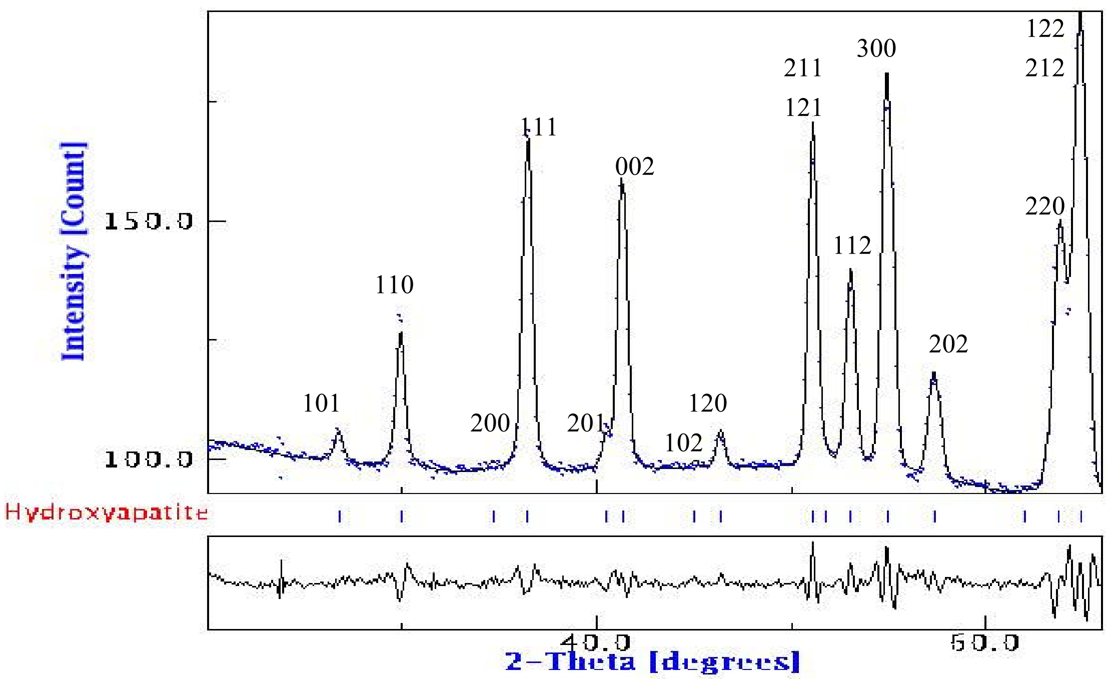

Fig. 1: Reitveld refinement of sum of all data

The texture of the sheep tibia was measured with beam size of 0.5 mm, very far from the interface and the new bone in order to show how the HAp crystallites are oriented in unperturbated parts of the bone.

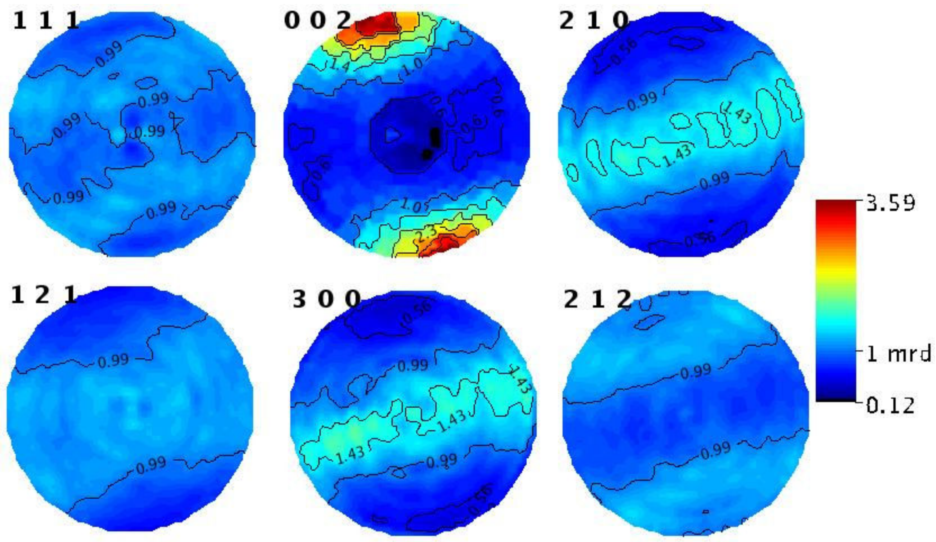

Fig. 2 Reconstructed pole figures from Rietveld texture analysis with MAUD of sheep bone (2 cm × 2 cm × 5 mm) showing the preferred orientation of HAp crystallites in particular when comparing the reflection (002) and the (111) reflection, nearly not affected by texture.

At the interface with the non-coated and coated implant to Hap, we probed the samples in the same conditions with a spatial resolution of 0.5 mm.

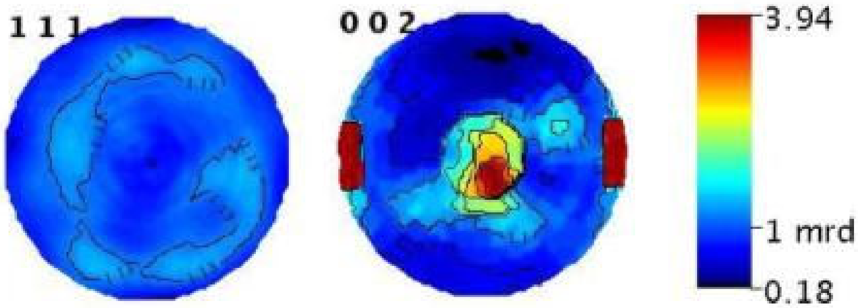

Fig. 3. Pole figures measured at 0 mm from the interface with implant non-coated with HAp

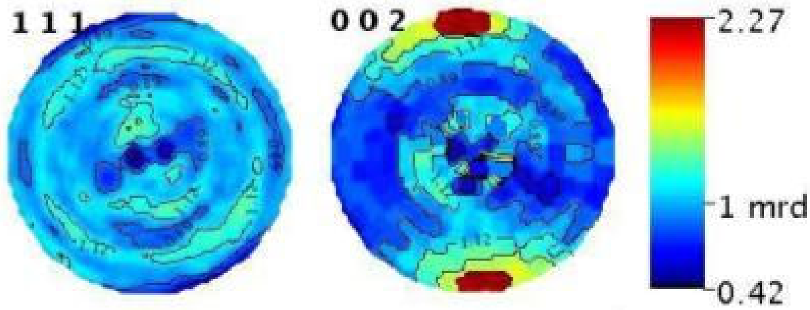

Fig. 4. Pole figures measured at 0 mm from the interface with implant coated with HAp

The figures 3 and 4 show that the HAp crystallites have two various orientations at the interface with the implant. The implant coated with HAp make possible to preserve the preferred orientation of the bone, but with the non-coated implant the HAp crystallites change completely the preferred orientation in an area of 5 mm, after this area we can show the usually preferred orientation of bone.

The maximum of multiples of a random distribution (mrd) of all (002) pole figures at two interfaces revealed that at the interface without HAp the preferred orientation is more intense than the other face, a great part of the crystallites are infected by titanium alloy. The crystallites of HAp grow in another direction, the non-coated face does not allow for the HAp crystallites to have the same initial direction of bone.

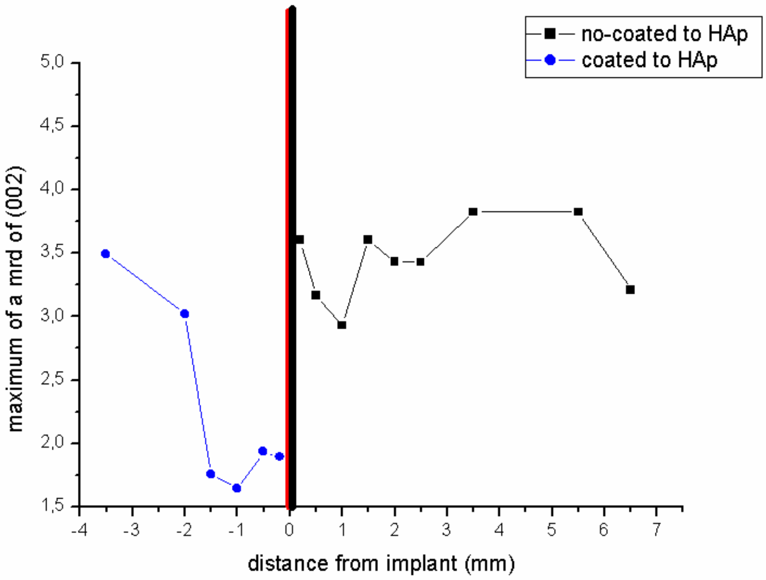

Fig. 4. Maximum of multiples of a random distribution (mrd) of (002) as a function of the distance from the implant interfaces.

Conclusion

The orientation of HAp crystallites at the interface with an implant depend of the coating of the face of the titanium alloy. If the implant is coated with HAp, the HAp crystallites preserve the preferred orientation inside the bone. If the implant is not coated, the crystallites change orientation. The mechanical properties of bone will also depend on this arrangement of HAp crystallites. This change of the preferred orientation creates certainly a perturbation at the interface and generates more probably cracks. The new bone material close to the interface may become unstable. In consequence, this instability increases the rejection rate of prosthesis.

Surface coating can reduce significantly the rejection rate of implants, therefore the HAp coating on titanium alloy should possess a very good combination of biocompability and mechanical properties.

This texture study with neutron diffraction of sheep tibia at the interface with implant revealed that it is necessary to cover implants with HAp.

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