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Characterisation by PIXE–RBS of metallic contamination of tissues surrounding a metallic prosthesis on a knee

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Abstract

Implants used as biomaterials have to fulfill conditions of functionality, compatibility and sometimes bioactivity. There are four main families of biomaterials: metals and metal alloys, polymers, bioceramics and natural materials. Because of corrosion and friction in the human body, implants generate debris. This debris may develop toxicity, inflammation and prosthetic unsealing by osseous dissolution. Nature, size, morphology and amount of debris are the parameters influencing the tissue responses.

In this paper, we characterised metallic contamination produced by knee prosthesis, composed with $TiAl_6V_4$ or Co–Cr–Mo alloys, into surrounding capsular tissue by depth migration, *in vivo* behaviour, content, size and nature of debris by PIXE (Particle Induced X-ray Emission) method associated with RBS (Rutherford Backscattering Spectroscopy).

Debris distribution in the whole articulation is very heterogeneous. Debris migrates several thousand micrometers in tissues, with a characteristic decrease. Solid metallic particles of about micrometer size are found in the most polluted samples, in both alloys $TiAl_6V_4$ and Cr–Co–Mo. In the mean volume analysed by PIXE, the concentration mass ratios [Ti]/[V] and [Co]/[Cr] confirm the chemical stability of $TiAl_6V_4$ debris and show the chemical evolution of Cr–Co–Mo debris.

Development of a protocol to prepare thin targets permits us to correlate PIXE and histological analysis in the same zone. The fibrous tissue (collagen fibres, fibroblasts) and macrophage cells are observed with optical microscope in polluted areas. This protocol could locate other pathologies in ppm contamination range, thanks to the great sensitivity of the PIXE method.

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1. Introduction

Pain, malfunction, structural degeneration or diseases sometimes make necessary the uses of biomaterials for replacement of organ or tissue affected. The function of the organ must be restored and the prosthesis component must perform this function as long as the patient is alive.

The biocompatibility describes all the interactions that take place between biomaterial and the tissue: it takes into

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account as well the negative interactions such that the materials should be inert and non-toxic, non-irritant, non-thrombogenic; also it includes as the positive interactions where an interactivity between the material and the host tissue increases the functionality and the retention of the device [1–3]. The performance of implants is described by biofunctionality, biocompatibility and bioactivity properties [4,5]. Many materials are used for implant manufacture: natural materials (coral, bone, collagen, etc.) or synthetic materials (metals and alloys, polymeric, ceramic, etc.). Metal alloys and polyethylene inserts are used as artificial joints.

The three alloys mainly used as implants (orthopaedic implant, dental prosthesis, etc.) are titanium-based alloy, cobalt-based alloy and stainless steel [4,6–8]. Metallic alloys are selected to offer the best corrosion resistance and the best mechanical resistance (under biological and mechanical stress applied by bone). A great toughness and resistance characterise cobalt-based alloys [8]. Stainless steel offers good mechanical properties, corrosion resistance and a low manufacturing cost [7]. Alloying elements are chosen to create a natural oxide layer at the surface [4]. The passive layer (TiO_2) is relatively stable on the device surface and regenerates itself when it is damaged, if oxygen is present [9]. Thanks to these properties, titanium-based alloys show a good corrosion resistance and a better biocompatibility [10]. The two-phase titanium alloys have the mechanical properties (specific strength and lower Young's modulus) for orthopaedic applications and increase the use of this metal as implant [1,9,11].

Many active substances (chemical, biological, physiological environment) and/or mechanical stress can cause wear by dissolution, corrosion, micro-fretting or friction. The passive layer may be also broken in the biological environment by cells [12,13]. Wear of implants is frequently reported and is often associated with release (ions, particles) in the host tissue [14–20]. Several studies, *in vivo* or *in vitro*, have been conducted on metal released from orthopaedic implants to characterise material properties [21–23]. The degradation of the materials can modify the integrity and functionality of the prostheses. Some releases or products, due to degradation, may be dangerous for the tissues. The release of any component from a biomaterial is likely to be associated with increased inflammatory reaction, and other phenomena are possible such as thrombogenicity, carcinogenicity, toxicity and genotoxicity, osteolysis, cutaneous allergic reactions, metal transportation and accumulation in organs, metabolism reaction and implant failure [1,22,24–35]. The pertinent variables associated with osteolysis may include genetic predisposition; number, size, charge, chemical composition, location of the particles, micro-motion, etc. [15,21,25,32,36]. Metallic debris and polyethylene debris play a role in aseptic loosening of the implant and can induce osteolysis (i.e destruction of bony tissues), phenomena leading to failure by unsealing [37].

In vivo metal ions or particles produced by metallic materials have to be understood to improve the material biocompatibility [23].

At the interface tissue (capsular or synovial), in loosened implant, the acidity of the environment may increase and the corrosion is accelerated. As a consequence an excess wear is likely to occur [24]. With Ti-based alloy, the Ti released depends on the media PH. The particles generated are potent stimulators of macrophages and other cells. The number of particles in titanium-based alloys is often very high and a clinical metallosis develops very quickly in the surrounding tissue [14]. A black coloration appears when the metallic debris are very concentrated [14,38]. The corrosive environment of body fluid may dissolve in ionic form a particle. The local tissue reaction may result in aggressive bone lysis for orthopaedic implant or creation of a fibrous tissue at the interface implant. Osteolysis leads to the loss of periprosthetic osseous support. Phenomena associated with cyclic loading lead to the aseptic loosening of the implants [26,27]. Titanium can be found in lymph nodes, in organs, in fluid whereas others studies do not report Ti in tissues surrounding implants. Sometimes Ti is found in high level, locally and/or systemically in the body (blood), except for the organs. Some clinical studies indicate that Ti content increases in serum for patient with titanium implant [21].

Knee prosthesis is a device more recent than hip implant and consequently hip debris characterisation is now well established [15]. It is of major importance to study *in vivo* metal debris released in tissues surrounding knee prosthesis. Knee implants generate more debris and in higher concentration in articulation than hip implants [15,39]. The characterisation of the knee prosthesis wear contributes to understanding the implant behaviour and their properties. Some implant failures are always observed [40].

For several years, research has been directed towards biomaterials characterisation with physical methods [41–44].

In this study, unicompartmental knee implants and biopsies have been retrieved surgically. Implants are made of Cr–Co–Mo alloys and $TiAl_6V_4$ alloys.

We characterise metallic contamination, into surrounding capsular tissue, produced by knee prosthesis: depth migration, *in vivo* behaviour, content, size and nature of debris by the PIXE method. Correlation with histological observations is made too. PIXE method offers a micrometer scale characterisation over an extended area with a great sensitivity and a simultaneous multi-elemental analysis.

2. Materials and methods

2.1. Description of the implants and the biopsies

Unicompartmental prosthesis includes two metal parts. One is inserted in the femur, the other in the tibia; they