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Tribological behaviour of a 3Y-TZP/Ta ceramic-metal biocomposite against ultrahigh molecular weight polyethylene (UHMWPE)

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ABSTRACT

This study aims to evaluate the tribological behaviour of 3Y-TZP/Ta (20 vol%) ceramic-metal composites and 3Y-TZP monolithic ceramic prepared by spark plasma sintering (SPS) against ultrahigh molecular weight polyethylene (UHMWPE). According to the results of pin (UHMWPE)-on-flat wear test under dry conditions, the UHMWPE – 3Y-TZP/Ta system exhibited lower volume loss and friction coefficient than the UHMWPE – monolithic ceramic combination due to the presence of an autolubricating layer that provides sufficient lubrication for reducing the friction. Owing to the lubrication of the liquid media, under wet conditions obtained using simulated body fluid (SBF), similar behaviour is observed in both cases. Additionally, the ceramic and biocomposite materials were subjected to a low temperature degradation (LTD) process (often referred to as "ageing") to evaluate the changes in the tribological behaviour after this treatment. In this particular case, the wear properties of the UHMWPE-biocomposite system were found to be less influenced by ageing in contrast to the case of the UHMWPE-zirconia monolithic material. In addition to their exceptional mechanical performance, 3Y-TZP/Ta composites also showed high resistance to low temperature degradation and good tribological properties, making them promising candidates for biomedical applications, especially for orthopaedic implants.

1. Introduction

Strategies for decreasing bearing wear in the ball-and-socket joint of artificial hip joints and the sliding-rolling articulation of artificial knee joints have been pursued vigorously by orthopaedic implant manufacturers and material scientists, with variable success. One of the most common biomaterial combinations used for heavy load bearing applications is ultrahigh molecular weight polyethylene (UHMWPE) against ceramics or metals. The cross-linking of polyethylene is a manufacturing strategy designed to reduce the incidence of bearing wear in prosthetic hip and knee joints. However, it is known that UHMWPE wear debris particles generated at the articulating surfaces caused by the tribological interaction with the counterpart of prostheses induced a tissue response that resulted in osteolysis and aseptic loosening, and this effect is recognized as one of the important factors in the failure of joint replacements, especially for long-term failure of UHMWPE in load-bearing implants [1–5].

On the other hand, metals and metallic alloys are the most common materials used for total bone replacement or implant fixations because the mechanical properties of metals meet the requirements for load bearing bone applications [6,7]. However, orthopaedic metallic implants are associated with local and remote adverse tissue responses. Generally, these adverse effects are mediated by the degradation products of implanted materials, which are primarily generated by the wear and corrosion of the metals in body fluids. Corrosion products can accumulate in tissues encapsulating the implant [8]. Corrosion releases metal ions, which enter the bloodstream and become concentrated in the erythrocytes. Thus, metal ions may enter cells and remain in local tissues, or they can be transported throughout the body, possibly leading to cytotoxic, genotoxic and immunological effects, either locally or at a distance from the implant [9].

It has been found that in particular cases, the nickel, chromium and cobalt ions released from joint prostheses lead to hypersensitivity reactions and even to carcinogenesis [10–14]. Therefore, to minimize the polyethylene wear and reduce the negative biological response of metallic implants, ceramic femoral heads have been developed.

The goal was to offer a smooth, low-friction surface that could reduce the wear more than metal-polyethylene bearings. Evidence has been shown for reduced wear when ceramic surfaces are used instead of metals in hip and knee replacement surgery [15,16]. This consideration applies even more acutely to young and active patients who will place greater demands on the prosthetic joint.

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Yttria-stabilized zirconia (Y-TZP) is a potential ceramic material for orthopaedic applications [17]. It shows excellent biocompatibility, the best mechanical properties of oxide ceramics, and high strength and high wear resistance in the ceramic-UHMWPE combination without adverse tissue reactions [18].

Nevertheless, ceramics have some drawbacks. One of the most important drawbacks of ceramic materials is their brittle nature, characterized by low fracture toughness. Load applied to the brittle ceramic can result in very fast catastrophic growth of micro-cracks that are present in virtually any material; such growth then leads to the final unpredictable fracture. This phenomenon is particularly noticeable if a dynamic load is applied such as impacts or strokes.

One of the most effective methods for improving the mechanical properties of ceramic materials, especially the fracture toughness, is to incorporate a ductile metal particle reinforcement [19,20]. Previous works have confirmed that the addition of tantalum lamellar shape metal particles (20 vol%) allows the design of zirconia-metal composites with improved mechanical performance due to the interactions between the transformation toughening of zirconia and the crack bridging mechanisms [21,22].

Despite its enhanced toughness, Y-TZP possesses another critical weakness. It may spontaneously transform to its stable monoclinic form under *in vivo* conditions, giving rise to the degradation of mechanical properties. Moreover, this transformation may produce material surface lifting, increasing its roughness, so the friction coefficient and wear rate will increase as well. This effect has become known as low-temperature hydrothermal degradation (LTD) or ageing [23]. However, previous studies have shown that yttria-stabilized tetragonal zirconia co-doped with tantala improves the LTD of zirconia ceramic [24].

Therefore, tantalum was selected as the second metal phase not only for enhanced toughness of zirconia but also for improved LTD resistance.

In our previous work, it was reported that the friction and wear behaviour of non-aged and aged 3Y-TZP/Nb-UHMWPE couples in dry conditions showed lower friction coefficient and wear rate for the 3Y-TZP/Nb-UHMWPE system than those for the monolithic ceramic-UHMWPE combination [25].

Additionally, SPS-sintered 3Y-TZP/Ta and 3Y-TZP/Nb ceramic-metal composites were evaluated from the point of view of their physico-mechanical behaviour under monotonic and cyclic loading with artificially induced flaws [26]. Compared to 3Y-TZP/Nb composites, 3Y-TZP/Ta composites have demonstrated simultaneous enhancement of damage tolerance and fatigue resistance [26]. Nevertheless, the tribological behaviour of 3Y-TZP/Ta (20 vol%) ceramic-metal composites has not been previously reported in the literature.

Its excellent biocompatibility is an additional advantage of tantalum [27–30]. Our earlier results obtained in an *in vitro* study [31] suggest that a new zirconia-Ta biocomposite displays biocompatibility and, consequently, could be a prospective material for orthopaedic applications.

Therefore, the aim of this work was to evaluate the wear behaviour of UHMWPE against 3Y-TZP ceramic and 3Y-TZP/Ta composites in dry and SBF conditions. The SBF-lubricated wear mechanism can partly simulate the *in vivo* response of the newly developed materials *in vitro*. Additionally, in an attempt to simulate *in vivo* conditions as closely as possible when conducting this type of lubricated test, the specimens were subjected to an accelerated ageing process prior to the wear test in SBF, and the obtained results were compared with those of the non-aged materials under the same testing conditions.

This laboratory wear testing provides an opportunity for the evaluation of materials at an early stage without the expense of simulator tests and allows savings in both time and funds in the selection of new materials. Based on the results from such evaluations, it can be determined whether or not a full series of long-term simulator tests should be carried out.

Moreover, the effect of tantalum addition to the zirconia matrix on the ageing resistance of zirconia-Ta composites was studied as well.

2. Materials and methods

2.1. Starting materials

Commercially available powders were used as raw materials: (1) t-ZrO_2 polycrystals (3Y-TZP, 3 mol% Y_2O_3; TZ-3Y-E, Tosoh Corp., Tokyo, Japan), with an average particle size $d_{50} = 0.26 \pm 0.05$ µm, and (2) tantalum (99.97% purity, Alfa Aesar, Karslruhe, Germany), with an average particle size $d_{50} = 44$ µm.

The starting Ta powder was attrition-milled with zirconia balls in a Teflon container for 4 h using isopropyl alcohol as the fluid. The ball-milled powder consists of flake-like deformed Ta particles with a high aspect ratio of approximately 50:1 and a mean size of 42 µm. To fabricate the zirconia matrix reinforced with lamellar Ta particles, 3Y-TZP powder was wet mixed with 20 vol% of the ball-milled Ta powder. Details of the ceramic/metal slurry processing were reported elsewhere [22].

The obtained powder was compacted by spark plasma sintering (SPS) in vacuum at 1400 °C, applying a heating rate of 200 °C/min and an uniaxial pressure of 80 MPa (FCT Systeme GmbH, HPD25, Germany). The final temperature and pressure were maintained for 3 min. The as-sintered sample disks showed diameters of 20 and 50 mm and a thickness of approximately 3–4 mm.

2.2. Microstructure of sintered specimens

The microstructure of sintered specimens was studied on surfaces polished down to 1 µm by a scanning electron microscope VEGA 3 LMH (SEM Tescan, Brno, Czech Republic) equipped with energy dispersive X-ray spectroscopy (EDS) that was employed for the chemical microanalysis of sintered samples. To obtain a semi-quantitative basis for the tantalum distribution in the ceramic matrix, EDS spectra were collected at one hundred random points. The surfaces of the worn pin after the wear tests under unlubricated conditions were analysed by EDS as well. The error of the EDS measurement was ± 0.4 mol. %. To measure the matrix average grain size, thermal etching was carried out at 100 °C below the sintering temperature in argon atmosphere for 30 min, and the linear intercept method (LIM) was used [32].

2.3. Mechanical characterization

The densities of the sintered samples (ρ) were measured using Archimedes' method in distilled water. The Vickers hardness was measured using a Vickers diamond indenter (Leco 100-A, St. Joseph, MI, USA) on polished surfaces, with an applied load of 196 N and an indentation time of 10 s. The biaxial flexural strength was measured using the piston-on-3-ball method. Disc specimens (diameter 20 mm, thickness 1.2–1.8 mm) obtained from the previously prepared cylinder, with the polished surface as the tension side, were placed on three balls separated by 120° on a 10 mm diameter circle. A piston pushes on the centre of the circle from the other side of the specimen, thus producing a biaxial flexural loading condition. The fracture toughness was measured by using single edge notched beams (SENB, dimension 3 mm × 4 mm × 45 mm). The tests were performed at room temperature using the same testing machine as that used for flexural strength determination at a crosshead speed of 0.5 mm min^{-1} with a span of 40 mm. Notches were introduced by using a diamond blade saw with a thickness of 0.2 mm.

The formulas and calculation procedures used in the measurements of Vickers hardness (Hv), flexural strength (σ_f) and fracture toughness (K_{Ic}) have been reported in previous publications [21,22].