Aging effect on the tribological behavior of a Novel 3Y-TZP/Nb Biocomposite against Ultra High Molecular Weight Polyethylene (UHMWPE)

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Abstract

The aim of this study was to evaluate the friction and wear behavior of ultra high molecular weight polyethylene (UHMWPE) against 3Y-TZP and a novel 3Y-TZP/niobium (Nb) biocomposite. The ceramic and the biocomposite materials were subjected to an accelerated aging process to evaluate the changes in the tribological behavior after this treatment. To carry out the tribological tests, a reciprocating pin-on-flat contact configuration was used. The results show a lower friction coefficient and wear rate for

biocomposite/UHMWPE couples than for 3Y-TZP/UHMWPE couples. The present study demonstrates that the excellent wear properties of this new biocomposite should never be influenced by aging. Based on these data, the potential of zirconia-Nb as an alternative bearing biomaterial is discussed.

1. Introduction

The wear of ultra high molecular weight polyethylene (UHMWPE) in acetabular cups is one of the main causes of long-term loosening and failure of artificial hip joints, due to the wear debris generated at the articulating surface of the cup[1]. Wear debris promotes osteoclastogenesis, which interferes with osteogenesis, causing periprosthetic inflammatory bone loss and resorption[2]. In addition, for 22 mm diameter head prostheses, wear and penetration of the femoral head into the UHMWPE cup may lead to impingement of the neck of the femoral component on the rim of the cup, causing its loosening[3].

Yttria-stabilized tetragonal zirconia polycrystal (Y-TZP) is a very attractive material for hip joint prosthesis bearings because of the high wear resistance of the ceramic-UHMWPE combination without adverse tissue reactions[4]. Compared with alumina, Y-TZP ceramics have better fracture toughness, higher flexural strength[5; 6] (which allows the design of more extreme shapes), and lower wear rates against polyethylene *in vitro*[7]. The excellent wear and mechanical properties of zirconia are considered very promising; however, problems still remain concerning the low-temperature aging degradation (LTD) caused by phase transformation, which may cause deterioration on the surface quality of zirconia bearing components[8]. This deterioration is believed to cause increased wear rates and has been observed in many clinical reports[9; 10; 11].

The degradation of Y-TZP is caused by a tetragonal to monoclinic (t-m) phase transformation accompanied by microcracking. This degradation is time dependent and enhanced by water or water vapor. Increasing the Y_2O_3 content and decreasing the average grain size of the zirconia may reduce the transformation rate; however, it does not make sense to reduce the grain size too much, because the grains can lose their metastability[12]. In addition, increasing the concentration of stabilizing oxide above 3.5 mol% can allow the nucleation of significant amounts of the stable cubic phase. Consequently, these solutions lead to a decrease in fracture toughness as a result of lack of the stress-induced t-m phase transformation. Doping Y-TZP with pentavalent oxides such as Nb_2O_5, is a promising way to improve resistance to LTD without any consequential loss in toughness[13]. Niobium ions reside as substitutional defects in the zirconium lattice, annihilating the oxygen vacancies generated by yttria doping[14]. As described in a previous article, a new synergic 3Y-TZP/Nb composite with both high flexural strength and extremely high fracture toughness was engineered by the interactions between transformation toughening and crack bridging[15]. Niobium was chosen because of its high melting temperature (~2477 °C), the stability and adhesion of ZrO_2–Nb interfaces[16], its ductile constitutive behavior and proven biocompatibility[17; 18], which keeps the use of these composites as biomaterials. In addition, it was shown in a previous publication that this composite is unaffected by low-temperature degradation due to the presence of a solid solution of Nb_2O_5 in the 3Y-TZP matrix, which may eliminate the deterioration of the surface quality of zirconia-based components[19].

The aim of this work was to evaluate the wear behavior of UHMWPE against zirconia and zirconia/Niobium systems when exposed to an accelerated aging process and compare the results with those for the non-aged materials.

2. Experimental Procedure

The following commercially available powders have been used as raw materials: (1) Tetragonal zirconia polycrystals (3Y-TZP, 3 mol% Y_2O_3; TZ-3YE, Tosoh Corp., Tokyo, Japan), with an average particle size of d_{50} = 0.26 µm. (2) Niobium (Goodfellow, Huntingdon, U.K., 99.85% purity) with an average particle size of d_{50} = 35 µm.

2.1. Materials Processing

Niobium powder was attrition milled with zirconia balls in a Teflon container for 4 h using isopropyl alcohol as the liquid medium. Due to this process, niobium particles with a lamellar-flaky shape (LFS) with a high aspect ratio and a d_{50} = 41 µm were obtained.

Zirconia/milled niobium suspension of 80 wt% solid content was prepared using distilled water as liquid media and a 3 wt% addition of an alkali-free organic polyelectrolyte as surfactant. The relative proportion of Nb was 20 vol%. The mixture was homogenized by milling with zirconia balls in polyethylene containers at 150 rpm during 24 h and then dried at 90^{o}C during 12 h. The resulting powder was ground in an agate mortar and subsequently passed through a 75 µm sieve and hot pressed for 1h at 1400 °C and 45 MPa. For comparison purposes, monolithic 3Y-TZP material was prepared under the same conditions. The produced materials were machined carefully in order to obtain prismatic bars with 15 x 3 x 4 mm^3 dimensions.

2.2. Aging Experiments

Aging experiments were performed in a steam autoclave (Microclave 4001404, J.P. Selecta, Barcelona, Spain) at 134 °C and 2 bar. Polished bars were placed into the autoclave and left in a steam atmosphere for 10 h (ISO standard 6474-2). The experimental procedure is explained in a previous work[19].

X-ray diffraction method, using a Philips X'Pert (Eindhoven, Netherlands) equipment with a Ge (111) incident beam monochromator (Johansson type) and the CuKα1 (λ = 1.5405981 Å) radiation, was used to evaluate the crystallographic composition. The scan conditions and calculations for estimating the ratio of monoclinic to tetragonal zirconia are explained elsewhere[19].

2.3. Wear Experiments

Wear experiments were carried out in a tribometer (Model MT/60/NI, Microtest, Spain) with a reciprocating motion, in which the stationary pin is loaded with a dead weight on the horizontal UHMWPE reciprocating plate. The rectangular pin contact surface (3 x 4 mm^2) was polished with diamond paste down to 1 µm and then with a colloidal silica suspension to avoid geometrical discontinuities in the contact region, providing a relatively constant contact pressure between the pin and the UHMWPE plate (40 x 30 mm) during the test. Previous to the wear test, the samples were washed ultrasonically in an acetone bath for 5 min and then dried at 90 °C for 30 min.

The tangential friction force between the pin and the plate was measured by the strain gauges fixed on leaf springs attached to the transverse bar holding the wear pin. The strain-gauge output voltage was passed through an amplifier and plotted on a chart recorder. Subsequently, by converting this voltage to friction force, friction coefficient data were collected. A constant force of 30 N was applied, resulting in an apparent contact pressure of about 2.5 MPa, a value similar to those borne by the femoral heads[20].

A sliding velocity of 0.06 m/s was applied. The test duration was associated with a traveling distance (*S*) of 10 Km. After each sliding test, the worn surfaces were cleared by blowing pressurized air before post-mortem observations. All tests were performed under the same conditions.

The wear rate (W) was calculated by using Eq. (1):

$$W = \frac{\Delta V}{F_N S} \qquad (1)$$

ΔV being the volume loss after the tests (mm^3), F_N the applied load (N), and S the sliding distance (m). In order to estimate the volume losses correctly, the track profiles were measured with a surface profilometer (Talysurf CLI 500, Taylor Hobson, Leicester, UK) that maps the surface morphology by putting a stylus in mechanical contact with the sample, with a step of 0.01 µm and a scanning speed of 0.1 mm/s.

The profilometer was used to determine the three-dimensional UHMWPE surface topographic map. The average surface roughness (R_a) of the surfaces of the different materials before and after aging was obtained using the same apparatus. The UHMWPE worn surfaces were observed with a field emission – scanning electron microscopy (FE-SEM) (Model DSM-950, Carl Zeiss, Germany).

3. Results and Discussion

Figure 1 represents the friction coefficient as a function of the sliding distance registered during the wear test for the ceramic and ceramic-metal composite before and after aging, when worn against UHMWPE. As can be observed, the friction coefficient has different values before and after aging, but no significant differences were found in the case of the ceramic-metal composite. The friction coefficient increases rapidly throughout the first meters of sliding and then stabilizes. This behavior can be attributed to a wear process of the UHMWPE during the first 100 m. As the wear proceeds further, the wear track becomes smoother and the friction coefficient settles to a steady level.

The 3D UHMWPE wear track surface topographies, after sliding against both aged ceramic and ceramic-metal composite, are presented in Figure 2(A) and 2(C) respectively. From the 3D surface topographies, the corresponding wear track dimensions (i.e., depth and width), as well as the volume of wear scars were extracted and summarized in Table1. Under identical test conditions, sliding speed and contact load, the smallest depth of the wear tracks was measured for the 3Y-TZP/Nb composite before aging. According to the equation 1, the UHMWPE wear rate (W) against 3Y-TZP rises from 5.45 x 10^{-5} to 7.25 x 10^{-5} mm^3/Nm when the ceramic pin is subjected to the aging process. In the case of UHMWPE against 3Y-TZP/Nb, there is no such increment on the wear rate (from 3.62 x 10^{-5} to 4.08 x 10^{-5} mm^3/Nm).

In Table 1, the R_a values for UHMWPE before and after wear tests are compared for all the sliding combinations. After the wear tests, it was observed that average surface roughness of UHMWPE against aged 3Y-TZP increased notably, while the R_a values measured on UHMWPE against aged and non-aged 3Y-TZP/Nb materials were similar to the pre-test ones. In order to study the effect of roughness changes on the UHMWPE plates, SEM observations were performed on these worn surfaces. Figure 2 shows the results. It can be observed that the worn surface (B) shows multiple scratches parallel to the slide direction when compared to (D).

X-ray measurements performed on the surface of the pins before the aging experiment showed a similar concentration of monoclinic zirconia in both samples (0.4 % for the 3Y-TZP and 1.7 % for the 3Y-TZP-Nb). After an accelerated aging period of 10 hours, these values rose to 17 % for the 3Y-TZP and 2.5 % for the 3Y-TZP-Nb, showing the important role that the Nb_2O_5 solid solution plays in zirconia aging behavior, which has been discussed elsewhere[19]. The zirconia martensitic transformation is associated with a volume expansion of about 4%, which may lead to a substantial roughness increase. This fact is responsible for the larger number of protuberances produced in the 3Y-TZP pin surface as compared with the 3Y-TZP/Nb pin. The Ra values of the pins, measured with the profilometer, were 11.8 and 11.2 nm for immediate readings and 20.4 and 12 nm after 10 h aging for 3Y-TZP and 3Y-TZP/Nb, respectively. The Ra values of the 3Y-TZP/Nb pins before and after aging are similar. Consequently, roughness increase on the surface of this composite is almost negligible, explaining the low increment in the UHMWPE wear rate after the aging process, which remains almost equal to that of the non-aged material. On the other hand, the scratches observed on the UHMWPE against aged 3Y-TZP and therefore the higher wear rate, are a consequence of the increase of pin surface roughness after aging because of the phase transformation of zirconia. Moreover, these microcraks produce pull out of the material during the wear test, caused for the large stress concentration towards the edge of the contact region, and generate a pin with chamfered edges. This is the reason for the different morphology of the wear tracks of figure 2(A) and 2(C).

In summary, the present study suggests that the 3Y-TZP/Nb composite has much greater phase stability than 3Y-TZP, and that its wear properties are not influenced by aging.

4. Conclusions

The influence of phase stability on wear properties of UHMWPE after accelerated aging of 3Y-TZP and a novel 3Y-TZP/niobium biocomposite was investigated. According to the results of pin on flat wear test using ceramic pins with or without autoclave aging, the wear rate of UHMWPE was almost the same against 3Y-TZP/niobium samples (3.62×10^{-5} mm^3/Nm, 4.08×10^{-5} mm^3/Nm, respectively). In contrast, the effect of 3Y-TZP aging on the wear behavior of UHMWPE was clear (50% greater wear than non-aged 3Y-TZP). The results indicate that the surface roughness of the ceramic-metal composite remain undamaged during the aging test; the wear rates of the UHMWPE counterface are therefore likely to be similar before and after aging of the biocermet pin. Consequently, this new biocermet is a more reliable bearing material against UHMWPE than 3Y-TZP because of its resistance to accelerated aging.

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TABLES

	R_a (µm)		Friction coefficient (µ)	Width (mm)	Depth (µm)	Volume of wear scar (mm^3)	W (mm^3/Nm)
S (km)	0	10					
3Y-TZP	0.67	3.62	0.35	3.8	301	16.35	5.45×10^{-5}
aged 3Y-TZP	0.65	5.53	0.42	3.8	403	21.75	7.25×10^{-5}
3Y-TZP/Nb	0.66	1.23	0.27	3.8	200	10.86	3.62×10^{-5}
aged 3Y-TZP/Nb	0.67	1.89	0.30	3.8	225	12.24	4.08×10^{-5}

Table 1. Friction coefficient, average wear track dimensions, volume of wear scars, wear rates (W) and average surface roughness *(R_a) of UHWMPE* against ceramic and ceramic-metal composite before and after aging.

FIGURE CAPTIONS

Fig.1. Friction coefficient as a function of sliding distance corresponding to UHMWPE plate against ceramic and ceramic-metal composite before and after aging

Figure 2. (A) Three-dimensional topographic map of the wear track made by the aged 3Y-TZP pin and the corresponding SEM observation (B). (C) and (D) are the equivalent images for the wear track made by the aged 3Y-TZP/Nb pin on the UHMWPE counterpart.