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Wear of crosslinked polyethylene under different tribological conditions

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Abstract Ultra high molecular weight polyethylene (UHMWPE) wear debris has been shown to be a major cause of long term failure of total joint replacements. Recently, crosslinking has been extensively introduced to reduce the wear of UHMWPE. In this study the wear of non-crosslinked and crosslinked UHMWPE were compared under a range of conditions.

The materials examined were UHMWPE GUR 1050, non-crosslinked, moderately crosslinked—5MRad, and highly crosslinked—10MRad. The wear was examined on a multidirectional pin on plate rig. The effect of counterface roughness on wear under different kinematics was examined.

The results from the different counterface conditions showed that highly crosslinked UHMWPE had significantly lower wear against both smooth and scratched counterfaces. However the reduction in wear for crosslinked polyethylene was less for scratched counterfaces. The second part of the study showed that all the UHMWPE's produced lower wear rates under lower multidirectionality because of reduced cross shear frictional forces and work. These findings are relevant to the consideration of the use of crosslinked polyethylene in the knee, where the kinematics have lower levels of cross shear and in the hip and knee against roughened metallic counterfaces.

Introduction

Every year thousands of artificial joints are implanted worldwide. In the UK alone over 50,000 replacement hip joints are implanted annually. Hip replacement operations are one of the most successful surgical procedures of our time, less than 10 percent fail within the first ten years of implantation. However as an increasing number of younger people require joint replacements, the durability and lifetime of the joints needs to be extended. The current limitations are related to the wear of the UHMWPE components. The wear releases polyethylene particles into the surrounding tissue and this can cause aseptic loosening of the femoral or acetabular components. The particles activate macrophages, which release pro-inflammatory cytokines, eventually resulting in bone resorption [1]. The size of the particles produced is important in their biological activity, with phagocytosible particles in the 0.1–1.0 μm size range being most reactive [2]. Larger particles (above 10 μm) stimulate the formation of giant cells, and so are less reactive [3].

Crosslinking has been introduced to reduce the wear of UHMWPE in artificial joints. McKellop *et al.* (4) have shown 5MRad crosslinked UHMWPE had 83% lower wear compared to non-crosslinked material in their hip joint simulator. Muratoglu *et al.* (5) showed an 85% reduction in wear with 10MRad crosslinked material. Both these studies used high concentrations (90–100%) of serum proteins in the lubricant. When lower serum concentrations (25%) were used in a study by Endo *et al.* (6), smaller reductions in wear were found, although only moderate crosslinking was investigated. Additionally, Endo *et al.* (6) used damaged counterfaces and found no reduction in wear rate with moderately crosslinked material. Although there is a general consensus that crosslinking of UHMWPE reduces wear, there remains considerable variation in the percentage reduction in wear reported by different centres, with in some cases zero wear being reported for highly crosslinked polyethylene (7). As many factors such as protein concentration of the lubricant, kinematics, and counterface roughness can control the wear mechanisms of

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polyethylene, it is postulated that they may also play an important role in the relative reduction of wear induced by crosslinking of polyethylene. The aim of this study was to compare the effect of changes in kinematics and counterface condition on the relative wear of polyethylene with three different levels of crosslinking.

Materials and methods

Materials

The materials investigated were ram extruded Ultra High Molecular Weight Polyethylene (UHMWPE) GUR 1050 supplied by DePuy Inc, Warsaw, USA and high nitrogen stainless steel, Ortron 90, supplied by DePuy International, Leeds, UK. The stainless steel had a Vickers hardness (50 g) of 386. The polyethylene represented the material used in an acetabular cup, and the high nitrogen stainless steel represented the femoral head material. The stainless steel plates were 75 mm long, 25 mm wide and 10mm deep.

The UHMWPE was irradiated to two different levels to create different levels of crosslinking. The levels of radiation were 5MRad which gave a medium crosslinked material and 10MRad which resulted in a highly crosslinked material. Crosslinking was achieved using gamma irradiation in nitrogen followed by re-melting at a temperature above 150 °C. The UHMWPE bars were first sealed in pouches under partial vacuum and subsequently exposed to either 5 or 10MRad of gamma irradiation. Following irradiation the bars were remelted at 155 °C for 24 hours in a nitrogen purged oven. This final step was undertaken to extinguish free radicals and to help stabilise against long term oxidation. The bars were then cooled slowly and subsequently machined into pins. The material preparation and irradiation was carried out by DePuy Inc.

The UHMWPE pins were all machined from a single block of material for each radiation level. They were taken from the centre of the block, with the pins oriented along the length of the block, to form a truncated cone with a flat surface of 8mm diameter, which was the contact face (Fig. 1). The wear surfaces of the pins were microtomed to remove any material that was damaged by the machining process. The pins were then put into de-ionised water for a minimum of four weeks in order for them to stabilise their water content (8).

The plates were polished and lapped until they had a smooth surface finish with an average surface roughness Ra $\sim 0.01\ \mu$m. Scratched surfaces were achieved by producing discrete scratches, 5mm apart, perpendicular to the direction of sliding, using a diamond stylus with a spherical tip diameter of 100 μm. High scratches with an average peak height of 1.8 μm and medium scratches with an average peak height of 0.8 μm were generated. A typical scratch profile is shown in Fig. 2. The height of the scratch lips was determined by comparison with the distribution of scratch geometries on previously studied explanted femoral heads (9).

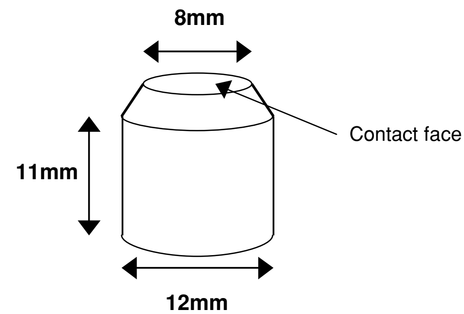


Fig. 1 Dimensions of pin.

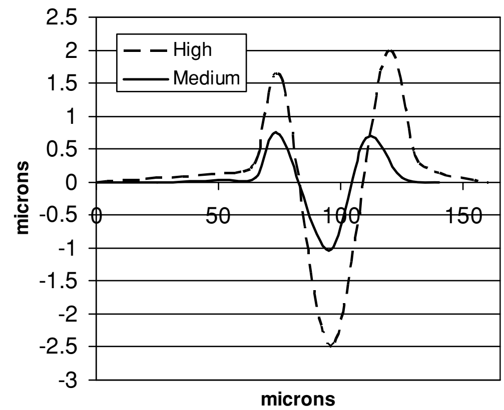


Fig. 2 Typical scratch profile for a high and medium scratch.

Methods

A multidirectional pin on plate wear simulator was used as shown in Fig. 3. The pin on plate machine loaded the pin under a constant force and the plate was reciprocated underneath. Rotation was achieved by a rack and pinion which was attached to the side of the bath and by a cogwheel which was placed on the pin holder (Fig. 4). The pin rotated as the plate oscillated back and forth, subjecting the pin to multidirectional motion (Fig. 5). The pin rotation was directly linked to the plate reciprocation such that the pin was at zero rotation position at the centre of the stroke, and maximum rotation at the end of the stroke. In any one set of test conditions, two pins of UHMWPE at each radiation level (0MRad, 5MRad, and 10MRad) were tested. The test was then repeated to give four replicates for each material under each condition. Control pins were also used and were kept in lubricant for the duration of the test. They were left inside the test rig so that they were subjected to the same conditions of temperature and lubricant as the test pins. Before testing the pins were cleaned and then left for 48hrs in an atmosphere of controlled temperature and humidity to stabilise the polymer

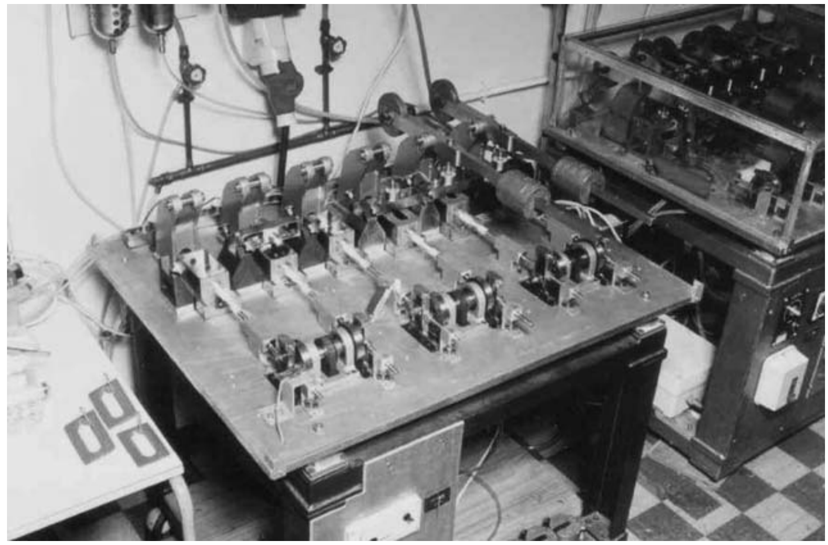


Fig. 3 Six station pin on plate reciprocating rig.

before weighing. The pins were weighed using a Sartorius 2405 balance which was sensitive to 1 μg and accurate to $\pm$ 2 μg. Each pin was weighed until four measurements within a 10 μg range had been obtained.

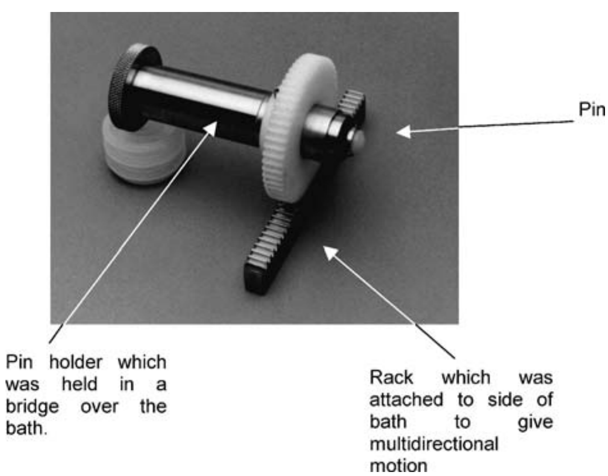


Fig. 4 UHMWPE pin in holder with rack and pinion gear mechanisms to provide rotation.

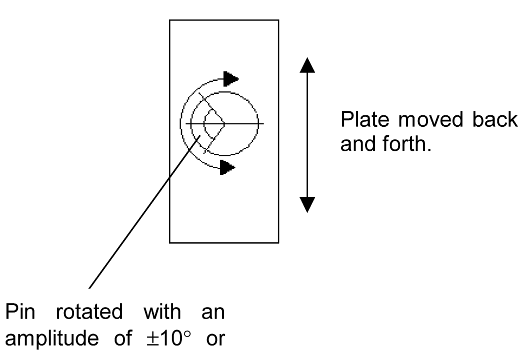


Fig. 5 Plan view of multidirectional pin on plate test.

The wear was determined by measuring the weight of each pin weekly during the test. The mean of the four measurements was calculated and subtracted from the previous results to determine the weight loss. The weight change of the control pins was calculated in the same way and the result was either added or subtracted from the test pins depending on whether they had lost or gained weight. The net weight changes of the test pins were then converted to a volume change using the density of the UHMWPE.

The mass loss was converted to volume loss and the wear factor k was calculated using the following equation (10).

$$k = \frac{V}{PX} \tag{1}$$

where:

k is the wear factor (mm^3/Nm)
V is the volume (mm^3)
P is the applied load (N)
and X is the sliding distance (m).

Two pins of each radiation level were run against smooth counterfaces for three weeks and then the test was repeated for a further three weeks with new pins. The whole method was then repeated with different pins against the medium and higher scratches. During testing the pins were rotated across the plates to reduce the plate variability from affecting the overall wear factors. A 25% (v/v) concentration of bovine serum was used as the lubricant. The serum was diluted using 0.1% (w/v) sodium azide. For the first tests a stroke length of 28mm which gave a total rotation of 60° over the length of the stroke, or ±30° about the centre position and a load of 160N was applied (nominal contact pressure of 3.2 MPa). The tests were then repeated using a stroke length of 10mm which gave a rotation of 20° over the length of the stroke or ± 10° about the centre position.

Variation in frictional force and frictional work acting on the polyethylene with multidirectional motion

During the multidirectional tests the wear pin was continuously rotated. This resulted in a frictional force F that constantly changed direction with respect to the polyethylene as the pin rotated. Furthermore, different points on the polymer pin experienced different relative motion and hence different principal molecular orientation (PMO). The sliding track between a fixed point on the polyethylene pin and the metal plate was firstly determined, as shown schematically in Fig. 6a, with reference to a co-ordinate system fixed to the plate (X, Y) . The PMO direction was then calculated as the average slope of the sliding track, measured from the horizontal axis (X) in the mid stroke in the absence of rotation of the polymer pin (γ) as shown in Fig. 6b. A separate coordinate system (x', y') was fixed to the pin along and perpendicular to the PMO.

For any given instant during the cycle, the relative total velocity (V_{total}) between a fixed point on the polymer pin and the metal plate was determined from the individual rotation and sliding motions of the pin and the plate respectively. The velocity vector thus determined the direction of the frictional force (α), measured relative to the PMO (x') as shown in Fig. 6b. It was therefore possible to resolve both the resultant frictional force (F) and the velocity (V_{total}) along and perpendicular to the PMO taking into account the polymer pin rotation. The corresponding frictional work (E) was calculated for a small time interval (dt) as

$$dE_{x'} = F_{x'} V_{totalx'} dt \tag{2a}$$

$$dE_{y'} = F_{y'} V_{totaly'} dt \tag{2b}$$

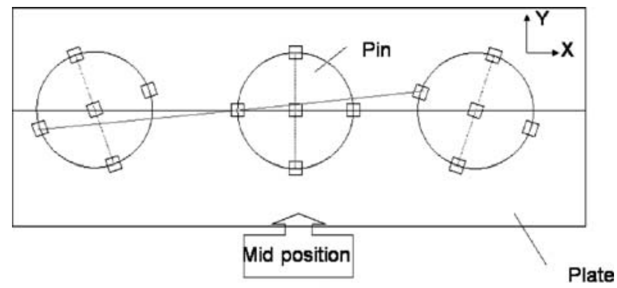
Therefore, the corresponding frictional work in half a cycle $\left(\frac{T}{2}\right)$, corresponding to the pin rotation angle between $-\beta_o$, and β_o was integrated as

$$E_{x'} = \int_0^{\frac{T}{2}} -F \cos\alpha V_{totalx'} dt \tag{3a}$$

released along the PMO direction and

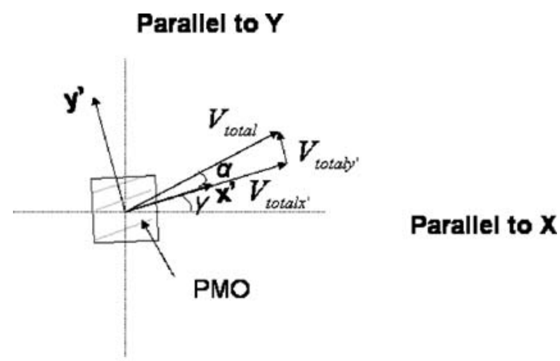
$$E_{y'} = \int_0^{\frac{T}{2}} -F \sin\alpha V_{totaly'} dt \tag{3b}$$

released perpendicular to the PMO direction.



(a)

Fig. 6a Sliding track for a fixed point on the polymer pin.



(b)

Fig. 6b Principal molecular orientation (PMO) relative to the horizontal axis at the mid stroke (γ) for the fixed point on the polymer pin shown in Fig. 6a. Shown are also the coordinates placed along and perpendicular to the PMO (x', y') and the angle between the total resultant velocity and x' (α).

The cross-shear effect was assessed by defining the cross-shear ratio (C), based either on the frictional force or the frictional work released perpendicular to and along the PMO. If the frictional force was assumed to be constant during the cycle,

$$Cf = \frac{\int_{-\beta_0}^{\beta_0} \|\sin\alpha\| \, d\alpha}{\int_{-\beta_0}^{\beta_0} \|\cos\alpha\| \, d\alpha} \tag{4a}$$

$$Cw = \frac{\int_0^{\frac{T}{2}} \sin\alpha V_{totaly'} dt}{\int_0^{\frac{T}{2}} \cos\alpha V_{totalx'} dt} \tag{4b}$$

Equations (4a) and (4b) were evaluated numerically. The numbers of divisions were finally chosen as 41 and 401 for determining the PMO and (C) respectively after mesh sensitivity checks. All the numerical analyses were carried out using Matlab7.0 (The Mathworks, Inc, 1984–2002).

For five typical points chosen on the polymer pin in the mid stroke as shown in Fig. 7, Tables 1, 2 and 3 show the

Table 1 Principal molecular orientation (PMO) for the five points shown in Fig. 7 for an rotation angle of ±30°.

Element	
1	−8.0°
2	0°
3	8.0°
4	0°
5	0°

Table 2 Cross shear ratio based on the frictional force for the five points shown in Fig. 7 for two sets of rotation angles of ±30° and ±10° calculated from different PMO assumptions.

Element	Cross shear ratio (±30°)		Cross shear ratio (±10°)	
	$\gamma \neq 0$	$\gamma = 0$	$\gamma \neq 0$	$\gamma = 0$
1	0.260	0.370	0.086	0.181
2	0.233	0.233	0.077	0.077
3	0.265	0.175	0.086	0.052
4	0.314	0.314	0.102	0.102
5	0.268	0.268	0.087	0.087

Table 3 Cross shear ratio based on the frictional work for the five points shown in Fig. 7 for two sets of rotation angles of ±30° and ±10° calculated from different PMO assumptions.

Element	Cross shear ratio (±30°)		Cross shear ratio (±10°)	
	$\gamma \neq 0$	$\gamma = 0$	$\gamma \neq 0$	$\gamma = 0$
1	0.091	0.115	0.010	0.030
2	0.071	0.071	0.008	0.008
3	0.091	0.115	0.010	0.030
4	0.132	0.132	0.014	0.014
5	0.095	0.095	0.010	0.010

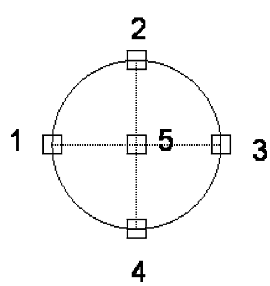


Fig. 7 Five typical elements defined on the polymer pin in the mid stroke.

calculated PMO and the cross-shear ratios for two rotation angles of ±30° and ±10° respectively. The cross-shear ratio, calculated from the assumption that the PMO was along the sliding direction of the plate (*X*), was also included in Tables 2 and 3.

For the conditions considered in the present study, both the PMO and the sliding track were mainly in the sliding direction. Therefore, the angle (α) between the PMO and the resultant velocity direction was approximately the same as the rotation angle of the pin, particularly for the polymer material near the centre of the pin. Under this assumption, equations (4a) and (4b) were simplified as follows:

$$Cf = \frac{\int_{-\beta_0}^{\beta_0} \|\sin\alpha\|\, d\alpha}{\int_{-\beta_0}^{\beta_0} \|\cos\alpha\|\, d\alpha} = \frac{1-\cos\beta_o}{\sin\beta_o} \quad (5a)$$

$$Cw = \frac{\int_0^{\frac{T}{2}} \sin\alpha\, V_{totaly'}\, dt}{\int_0^{\frac{T}{2}} \cos\alpha\, V_{totalx'}\, dt} = \frac{2\beta_o - \sin(2\beta_o)}{2\beta_o + \sin(2\beta_o)} \quad (5b)$$

Equation (5b) was also given by Wang (11), but in a different form. Tables 2 and 3 show the variation of the cross-shear ratio for different pin rotation angles ($\pm\beta_o$). The average cross-shear ratio for the whole polymer pin was calculated by averaging over 17 points evenly distributed along the circumferential and the radial directions. The average cross-shear ratio based on the frictional force was calculated to be 0.268 and 0.087 for the rational angle of ±30 ° and ±10° respectively. The corresponding average cross-shear ratio based on the frictional work was calculated to be 0.096 and 0.010 respectively.

Studies undertaken clinically and using simulators have shown that different levels of cross shear can be observed. In a study by Barbour *et al.* (12) they showed that the shape of the wear path can have a significant effect on the wear of acetabular cups. They tested this using a simulator and by changing the conditions part way through the test to give a different shaped wear path. This change resulted in different levels of cross shear occurring. They showed that greater wear occurred when more cross shear was present. A study by Bennett *et al.* (13) looked at the different wear paths that are found clinically. They found a wide range in the shape and length of wear paths ranging from thin which would give a lower cross shear, up to wide open paths which would have a higher level of cross shear. The work of Bennett *et al.*, (13) defined a range of elliptical wear paths with eccentricities in the range 2.5 to 9.2. McEwen (14) has estimated lower cross shear for fixed bearing knees. These studies show the importance of cross shear on the wear rates produced by UHMWPE and show that clinically a wide range of cross shear can be found.

Results

The wear factors for the three materials against a smooth counterface at 60° rotation and higher cross shear are shown in Fig. 8. The results showed that the highest wear factor was for the 0MRad non-crosslinked material with the wear

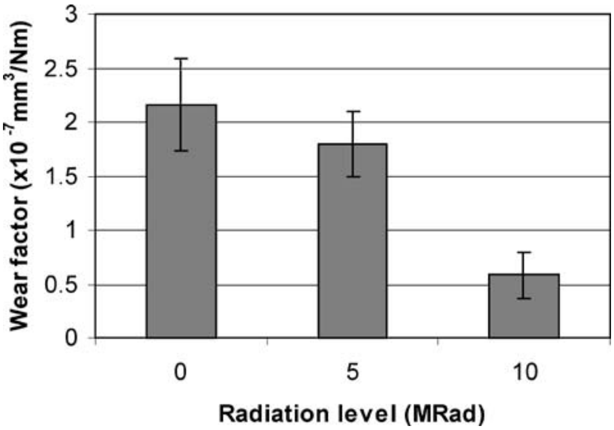


Fig. 8 Comparison of wear factors of different materials against smooth counterfaces at 60° rotation.

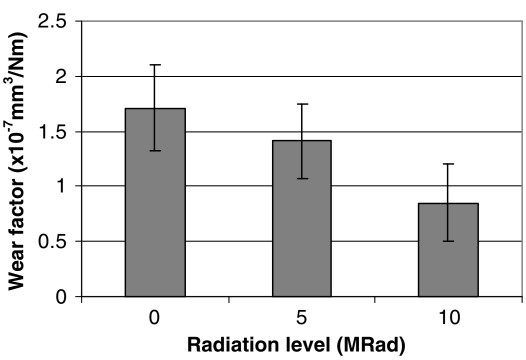


Fig. 9 Comparison of wear factors with of different materials against smooth counterfaces at 20° rotation.

factor decreasing with increased crosslinking. The highly crosslinked material had 73% lower wear compared to the non-crosslinked. Between the 0MRad and 5MRad materials there was a 16% reduction but this was not statistically significant. However the wear factor for the 10MRad material was found to be significantly lower ($p < 0.05$; ANOVA) than for the other materials. These results showed that for a smooth surface which represents an undamaged counterface, the highly crosslinked material had the greatest wear resistance.

The wear factors for the UHMWPE pins against a smooth counterface at 20° rotation are shown in Fig. 9. The results showed that there was a 50% reduction in wear factors from the non-crosslinked to the highly crosslinked material. However this reduction was not statistically significantly different ($p > 0.05$; ANOVA).

The mean wear factors from the three different counterface surfaces at 60° rotation are shown in Fig. 10. These results show that for all the materials the wear factor increased as the scratch height increased. The high levels of crosslinking reduced wear by 64% on the medium scratched plates and by 60% on the highly scratched plates when compared to the results from the non-crosslinked material. The 5MRad material gave a slight but not significant increase in wear factors on the scratched plates when compared to the non-crosslinked material. The results showed that the 10MRad material had a significantly lower wear factor than the other two materials on all of the surfaces ($p < 0.05$; ANOVA).

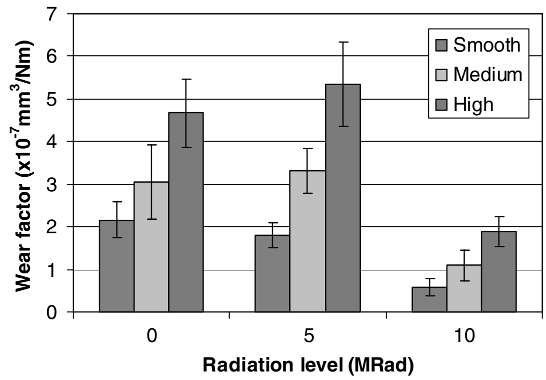


Fig. 10 Comparison of wear factors with all three counterface conditions and radiation level at 60° rotation.

The mean wear factors from all the different counterface surfaces at 20° rotation are shown in Fig. 11. These results show that for all the materials the wear factors increased as the scratch height increased. The high levels of crosslinking reduced wear by 59% on the medium scratched plates and by 67% on the highly scratched plates when compared to the non-crosslinked material. With the medium scratched counterface there was very little difference between the wear factors of the 0MRad and the 5MRad materials. Between these two materials there was no statistically significant difference under any of the counterface conditions. The results showed that the highly crosslinked 10M Rad material gave the lowest wear factors with all of the test conditions, with

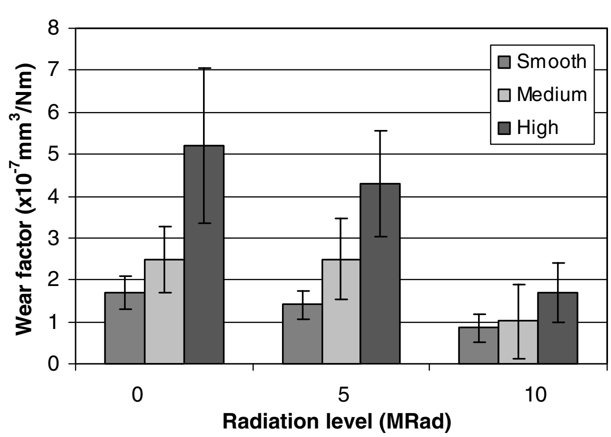


Fig. 11 Comparison of wear factors with all three counterface conditions and radiation level at 20° rotation.

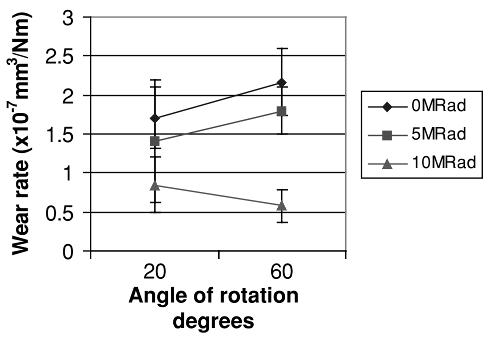


Fig. 12 Comparison of wear rates with different cross shear with smooth counterfaces.

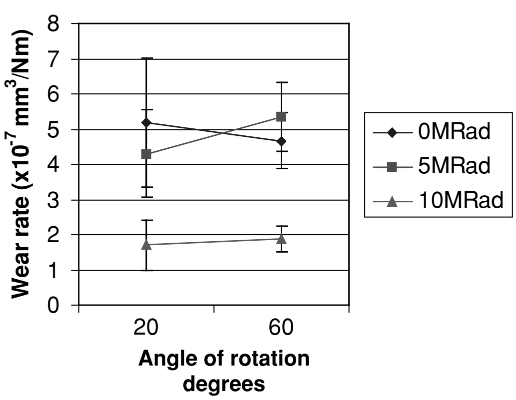


Fig. 13 Comparison of wear rates with different cross shear with medium scratches.

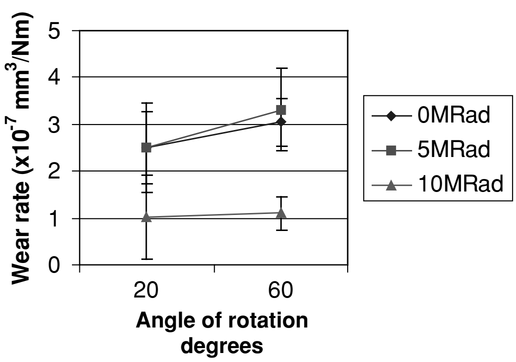


Fig. 14 Comparison of wear rates with different cross shear with high scratches.

the results being statistically significantly different with both of the scratched counterfaces ($p > 0.05$; ANOVA).

Figures 12–14 show the change in wear for the different pin rotations. The non-crosslinked and moderately crosslinked UHMWPE both had lower wear rates at 20° rotation compared to 60° rotation. The cross shear had little effect on the wear of the 10MRad material. An alternative way of considering this is that for the smooth counterface the 10MRad material reduced wear by 73% under high cross shear and by 50% under lower cross shear. The wear rates for the two pin rotation conditions are shown in Figs. 13 and 14 for the medium scratches and the high scratches respectively. For both the counterface conditions the highly crosslinked UHMWPE gave much lower wear factors at each cross shear condition. However for the high scratches there was little effect of cross shear on the wear rates of any of the materials.

Discussion

Smooth pin on plate wear tests

The results from the smooth counterface experiments showed that the wear factor decreased as the crosslinking level increased at both rotations although at 20° rotation the difference was less, and was not statistically significantly different. Previous studies, such as those by McKellop *et al*., (4) and Muratoglu *et al*., (5) with other types of polyethylene have also shown a reduction in wear with crosslinked UHMWPE against smooth counterfaces. However, the reduction seen in this study was not as great as that seen by McKellop *et al*., (4) and Muratoglu *et al*., (5). This study gave a reduction of 73% while the study by Muratoglu et al., (5) gave a reduction of 85% for 10MRad material. For 5MRad material McKellop *et al*., (4) showed a reduction of 83% while this study found a 16% reduction. These differences may be due to the different kinematics used in this study.

The reduction in wear rate is thought to be due to the effect of orientational hardening and softening (11, 15, 16). During unidirectional motion orientational hardening occurs. This means that the molecules orientate themselves with the direction of sliding resulting in a material which is strong in the direction of motion but weaker perpendicular to it. The frictional force remains in an approximately constant direction, and therefore the direction of motion and also the direction in which the molecules are aligned remains roughly constant. In multidirectional motion the frictional force constantly changes direction with respect to the polymer surface. This means that the frictional force is sometimes parallel to the direction of orientational softening and sometimes parallel to orientational hardening. Crosslinking reduces the amount of orientational hardening and softening, as it retards chain mobility and provides resistance to cross shear forces (11, 15, 16). Crosslinking also results in more C-C bonds between molecules which makes it harder to split them. The more crosslinking in the material, the more bonds that are present. This means that there is less orientational hardening and softening, which is why a reduction in wear rate was seen as the crosslinking levels increased.

Medium and high scratched pin on plate wear tests

When worn against both medium and highly scratched counterface conditions there was a significant increase in wear factors for all the materials. However the 10MRad material had a significantly lower wear factor than the other two materials. Between the 0MRad and 5MRad materials there was no significant difference between the wear factors.

The crosslinks gave more protection against the counterface scratches than the non-crosslinked material which was shown by the significantly lower wear factor for the 10MRad material. The links provided resistance to the scratches and prevented particles detaching under multidirectional motion. As the 5MRad material contained fewer crosslinks it did not provide the same protection and this meant that it had a much higher wear factor than the 10MRad crosslinked material.

The effect of cross shear

Against a smooth counterface at 20° rotation with a lower cross shear more orientational hardening was able to occur especially with the non-crosslinked pins which resulted in their lower wear factor compared to the 60° rotation 0MRad pins. This agreed with a hip simulator study by Barbour *et al*., (12) which found an increase in wear rate with increased cross shear. The crosslinking meant that the orientational hardening could not occur as easily and therefore did not give additional wear reductions under low cross shear conditions. However, the crosslinking in the 10MRad material meant it was more resistant to wear under both sets of conditions. Crosslinking gave a reduction in wear for the 10MRad material under both kinematic conditions, while orientational hardening preferentially gave a reduction in wear under the lower cross shear conditions. This may explain the difference in the ratio of the wear rates for the 0MRad and 10MRad materials for the two different conditions.

Figures 13 and 14 show that the effect of the two different levels of cross shear for the three materials under scratched conditions is different to smooth conditions. For the high scratches (Fig. 14) there was no change in wear rate with an increase in cross shear for any of the materials. For the 0MRad material the wear rate did not reduce with reduced cross shear conditions indicating that the strain hardening wear mechanism found on smooth surfaces was not effective under more abrasive conditions. This explains the increase in wear with the 0MRad material with scratched counterfaces under lower cross shear conditions compared to the higher cross shear conditions (Fig. 11 compared to Fig. 12).

For the highly crosslinked material, there was no change in wear with increased cross shear with the scratched counterfaces. This is consistent with that found with smooth surfaces, and confirms that the highly crosslinked material is less sensitive to kinematic conditions. However the wear rate of the highly crosslinked material did increase between 3 to 4 times with the high scratched counterfaces compared to the smooth counterface.

Conclusion

Highly crosslinked 10MRad material had the lowest wear rate under all conditions. Damage to the counterface increased the wear rate for all the materials. This means that it would be an advantage to articulate all materials including crosslinked PE against damage resistant ceramic femoral heads in the hip, (17).

The 73% reduction in wear with highly crosslinked polyethylene compared to non-crosslinked polyethylene under higher cross shear conditions was reduced to 60% under high scratched conditions.

Under lower cross shear kinematic conditions representative of the knee the reduction in wear of highly crosslinked material on smooth counterfaces was 50% compared to non-crosslinked materials.

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