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Comparative wear under four different tribological conditions of acetylene enhanced cross-linked ultra high molecular weight polyethylene

H. MARRS, D. C. BARTON, R. A. JONES*, I. M. WARD*, J. FISHER
*School of Mechanical Engineering, and *IRC in Polymer Science & Technology, Department of Physics and Astronomy, University of Leeds, Leeds, LS2 9JT, UK*

C. DOYLE
Howmedica Worldwide Research & Development, Staines, Middlesex, UK

In this study, the wear of ultra high molecular weight polyethylene (UHMWPE) (Grade RCH 1000) crosslinked by gamma irradiation in acetylene was compared to virgin (non-irradiated) UHMWPE using four different wear configurations: (i) unidirectional motion with a smooth counterface, (ii) multidirectional motion with a smooth counterface, (iii) unidirectional motion with a rough counterface and (iv) multidirectional motion with a rough counterface.

Nomenclature

ε_n = engineering/nominal strain
σ_n = engineering/nominal stress
ε_t = true strain
σ_t = true stress
A_o = original area
F = translational friction force
F_x = friction force along x-axis
F_y = friction force along y-axis
Gel_i = measured gel content of the irradiated sample
Gel_{true} = true gel content of the irradiated sample
Gel_v = measured gel content of the virgin sample
K = wear factor
l = final length
L = load
l_o = original length
$M_{n,0}$ = initial number of average molecular weight prior to irradiation
p = probability determined from Students' *t*-test
R_a = arithmetic mean of departure of the profile from the mean line
UTS = ultimate tensile strength
V = volume loss
x = axis of element on pin wear face perpendicular to direction of translational motion
X = sliding distance
y = axis of element on pin wear face parallel to direction of translational motion

It was found that the wear rates of the two types of UHMWPE were dependent on the test conditions. The unidirectional smooth test produced less wear than the multidirectional smooth test for the virgin material. Both rough tests produced more wear than the corresponding smooth tests with the unidirectional rough test producing less wear than the multidirectional rough test. The wear rates for the two materials were not significantly different in the unidirectional smooth and the multidirectional rough tests. In the unidirectional rough test, the polymer crosslinked using gamma irradiation in the presence of acetylene wore 1.5 times more rapidly than the virgin material. In contrast, for the multidirectional smooth test, the wear rate of the virgin material was 9.3 times greater than that of the acetylene enhanced crosslinked material.

This study confirms previous work that a multi-directional friction force can accelerate wear in a non-crosslinked material and that, in the presence of multidirectional motion such as in a hip joint, the crosslinked material has considerable advantage. However, with roughened counterfaces and a more abrasive wear mode, this advantage was negated. Thus crosslinking of UHMWPE using gamma irradiation in the presence of acetylene has the potential to reduce wear and therefore osteolysis in hip prostheses as long as femoral heads remain smooth and undamaged.

1. Introduction

During the last 25 years, acetabular cups manufactured from ultra high molecular weight polyethylene (UHMWPE), have been mainly sterilized in air using γ-irradiation. It has been shown conclusively by Besong *et al.* [1] that the wear rate of UHMWPE Grade GUR 412 sterilized via this method when tested *in vitro* against counterfaces of varying roughness increased significantly with increasing shelf-life. By collecting the wear debris from these tests, it was also shown that the total number of particles produced per unit load, per unit sliding distance was increased by 34 times when the gamma irradiation sterilization process and 10-year aging was carried out on UHMWPE [2]. For a particle size of less than 0.5 μm (i.e. submicron), the total number of particles per unit volume of debris for the aged irradiated material was greater than for the non-irradiated material. This result was statistically significant.

The reason for this reduction in wear properties lies in the chemical reactions taking place following sterilization. During γ-irradiation, free radicals are formed within the polymer and, in the presence of air, some of these free radicals combine with oxygen molecules, causing oxidation and thus degradation of the material [3]. This has been extensively documented through measurement of the degradation of material properties such as toughness [4]. The reduction of the mechanical and wear properties with time after sterilization is a result of the remaining available free radicals combining with more oxygen molecules. However, the rate of degradation may be less in the body than on the shelf.

In the human body, the production of UHMWPE wear particles can lead to bone resorption. The particles are engulfed by macrophages, stimulating the release of cytokines, which produce osteoclastic bone resorption. This wear debris induced osteolysis is dependent on the rate of generation of submicron wear particles, and is therefore accelerated by the degradation and increased wear due to oxidation following irradiation.

Widespread concern about oxidative degradation has led to the development of alternative sterilization methods such as electron-beam irradiation [3], varying the γ-ray dose [5] and using inert atmospheres during irradiation such as nitrogen [6] or a vacuum to prevent oxidation of the material. By irradiating UHMWPE in an inert atmosphere, the free radicals combine to form crosslinks. The number of crosslinks can be further increased by irradiating in the presence of acetylene [7]. The elimination of free radicals reduces the chance of oxidation and aging and the presence of crosslinks alters the mechanical properties of the material along with the wear properties. Meng Deng and Shalaby [5] compared the tensile properties of UHMWPE Grade GUR 415 irradiated using gamma rays in four different environments: air, nitrogen, acetylene and vacuum. UTS was greatest and ultimate elongation the least when irradiated in acetylene. It was also demonstrated that the temperature at which oxidation occurred, decreased for materials irradiated in all the different environments except acetylene.

Sun *et al.* [8] compared the wear of unirradiated, air-irradiated and ''stabilized'' (i.e. crosslinked) UHMWPE using a hip simulator (the gel contents for these materials were 0.10, 0.46 and 0.75 respectively). The ''stabilized'' material was irradiated in an inert atmosphere to prevent oxidation, and then was held at an elevated temperature for a period of time to help crosslink any remaining free radicals. The unirradiated material gave the greatest wear rate and the stabilized or crosslinked material gave the least. The wear tests were carried out in a hip joint simulator utilizing multidirectional friction forces, on materials that had not been aged.

Due to the benefits of crosslinking UHMWPE, alternative methods of producing the crosslinks have been pursued. Chemical crosslinking is possible using either chlorosulphonation or dicumyl peroxide treatment followed by u.v. irradiation [9]. It has been suggested by Penning *et al.* [9] that crosslinking of UHMWPE by means of organic peroxides can occur without scission of the main chain. This may provide an improved route for crosslinking high strength fibers when compared with γ-irradiation in the absence of acetylene and chlorosulphonation where crosslinking is accompanied by a reduction of tensile strength [9]. However, there remains much debate about the relative benefits of the different types of crosslinking.

The present research stems partly from an extensive program of research at the IRC in Polymer Science and Technology at the University of Leeds directed at the use of irradiation crosslinking for the improved creep performance of melt-spun high modulus polyethylene fibers. Following the classic work of Charlesby and Pinner [10], it was recognized that X-ray or γ-irradiation of polyethylene can produce both crosslinking and chain scission, the latter potentially leading to loss of strength. A very useful discovery by Woods *et al.* [11] in 1984 was that irradiation in an atmosphere of acetylene followed by high temperature annealing gave a major enhancement of crosslinking. This discovery led to subsequent research and the establishing of practical protocols for crosslinking fibers. Woods *et al.* [7] document procedures that have been successfully applied to bulk UHMWPE. The fundamental issues are that irradiation in the presence of acetylene leads to chain reactions and that annealing after irradiation ensures that all the free radicals have reacted, enabling an optimal degree of crosslinking to be obtained rather than chain scission. The mechanical behavior and the chemistry of the crosslinked material have been discussed in a number of publications [12–15].

The aim of the present study was to investigate the properties and wear of acetylene enhanced crosslinked UHMWPE for use in acetabular cups in hip prostheses and compare its performance with virgin UHMWPE that had not been irradiated. This was achieved by carrying out tensile tests and wear tests of different configurations and using both rough and smooth counterfaces on samples from acetylene enhanced crosslinked UHMWPE as well as from the virgin non-irradiated material.

2. Materials

2.1. Method of crosslinking

A compression-molded UHMWPE RCH 1000 (now marketed as GUR 120) block (approx. $70 \times 70 \times 70$ mm) was placed in a stainless steel cylinder and maintained at 100 °C. The cylinder was evacuated of air gases, maintained under vacuum for 16 h, and then filled with acetylene at atmospheric pressure for 31 h. These timings were calculated to be sufficient to remove dissolved oxygen and equilibrate dissolution of acetylene into the UHMWPE at 100 °C [14, 15]. The UHMWPE sample was then cooled to room temperature (ca. 18 °C) and γ-irradiated, at Atomic Energy Authority, Harewell, UK, with a dose of

2.5×10^4 Gy (2.5×10^6 Rad) whilst still in the acetylene atmosphere of the cylinder. Following irradiation, the UHMWPE was annealed at 100 °C for 5 h again still in the acetylene. This latter annealing step ensured chain-reactions of almost all the free radicals generated in the polymer with either the dissolved acetylene or the polymer itself in the absence of oxygen. The cylinder seals were then broken and the UHMWPE sample removed for wear and tensile tests and gel-fraction analysis [13, 14].

2.2. Pins and plates

The tensile specimens and wear pins were machined from both the acetylene irradiated and virgin UHMWPE (RCH 1000) blocks whilst the counterface plates and disc were made from cobalt chrome alloy (low carbon content). The surfaces of the smooth metal counterfaces were polished and lapped, giving the desired low surface roughness (R_a). The rough counterfaces were first polished and lapped and then roughened by a grinder to the desired R_a value. This initial polishing and lapping removed any background periodicity of the surface. The lubricant used in all the wear tests was a solution of 25% bovine calf serum and 75% of a deionized water solution containing 0.1% sodium azide.

3. Methods

3.1. Gel fraction analysis

Samples of UHMWPE were placed into weighed stainless steel gauze containers (120 gage). Gel-fraction determinations were carried out as per the method of Kang *et al.* [16], with the exception that decahydronaphthalene (dekalin, b.pt. 192 °C) containing 2,6-di-*t*-butyl-*p*-cresol antioxidant (1% w/v) was used as the solvent. Since the samples were UHMWPE, they were solvent extracted in the dekalin for 96 h. Following solvent extraction, the samples were rinse-washed in boiling acetone (61 °C) and then dried in an oven at 100 °C for 12 h.

3.2. Tensile tests

The material was tested in the form of flat dumb-bells of 1.6 mm thickness cut out from strips of UHMWPE using a die-cutter. The dimensions of these specimens are shown in Fig. 1, for which there is no standard.

This dumb-bell shape was chosen as it was the smallest available and the size of the crosslinked polyethylene sample limited the use of larger specimens. (Indeed, the change in width of the dumb-bells at the grips was severe, and most of the specimens failed at the end of the narrow gage section rather than in the gage section itself.) The width and thickness of the gage section of each dumbbell was measured using a micrometer, so that the initial cross-sectional area could be calculated. Tensile tests were carried out on a Howden universal testing machine at a separation rate of 180 mm/min. Each dumb-bell was pulled to failure, and the loads and crosshead displacements were recorded at set intervals of time (usually every 20 ms). At least seven specimens were tested for each material type. The true stresses and strains were calculated from the following equations

$$\text{True strain} \quad \varepsilon_t = \ln(1 + \varepsilon_n) \tag{1}$$

$$\text{True stress} \quad \sigma_t = \sigma_n(1 + \varepsilon_n) \tag{2}$$

where σ_t = true stress, σ_n = nominal stress, ε_t = true strain and ε_n = nominal strain.

The yield stress, 5% proof stress, UTS, strain to failure, and energy to failure were calculated from plots of true stress vs true strain for each material.

3.3. Wear tests

The wear properties of crosslinked and virgin RCH 1000 UHMWPE were studied using four different test configurations: (i) unidirectional motion pin-on-plate reciprocator (smooth counterface); (ii) multidirectional motion pin-on-plate reciprocator (smooth counterface); (iii) unidirectional pin-on-disc reciprocator (rough counterface) and (iv) multidirectional motion pin-on-plate reciprocator (rough counterface).

Table I lists the test parameters for each wear configuration. Conical-ended wear pins were used and all had the same dimensions (see Fig. 2).

For tests (i), (ii) and (iv), a six station reciprocating pin-on-plate wear machine was used. Three stations were used to test the pins made from the crosslinked RCH 1000 UHMWPE whilst the other three tested the virgin RCH 1000. After each interval of testing, the pins were swapped so that they were wearing against different plates in an attempt to eliminate the effect of different surface roughness of individual plates as a variable.

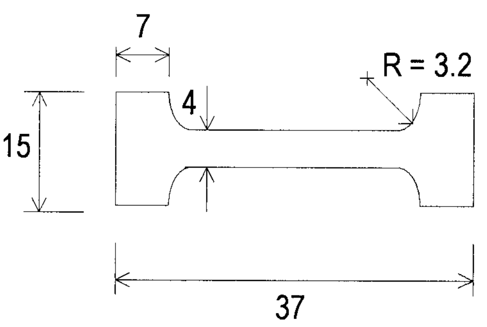


Figure 1 Tensile specimen. Thickness = 1.6 (dimensions in mm).

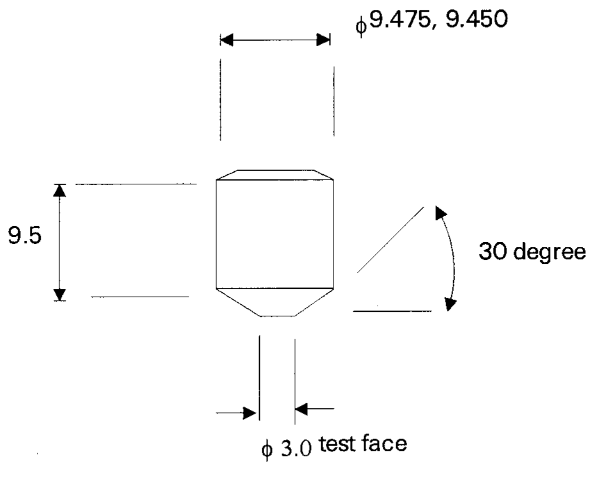


Figure 2 Wear pin dimensions (in mm).

TABLE I Test parameters

Test type	Load (N)	Frequency (Hz)	Average counterface Roughness, R_a (μm)	Minimum no. of wear data points for each material
Unidirectional (smooth)	160	2	0.01	11
Unidirectional (rough)	80	1	0.09	9
Multidirectional (smooth)	160	1	0.01	10
Multidirectional (rough)	80	1	0.09	12

Control pins from each material were placed in the same lubricant as the wear pins in order to monitor the moisture uptake during the test. These control pins were prepared in the same manner as the test pins and both sets were allowed to soak in deionized water for a minimum of 2 weeks prior to testing in order to reduce the moisture uptake during the test. The counterface and test pins were all contained in a stainless steel bath into which the lubricant was added until it completely submerged the counterface surface. The test pins were loaded axially in compression via the pin holder and a cantilever mechanism which enabled the load on the pin to be varied by simply moving the load along the cantilever arm (see Fig. 3).

3.3.1. Unidirectional motion pin-on-plate test (smooth)

This test was run using smooth plates with $R_a \sim 0.01$ μm. To generate measurable wear with highly polished surfaces, a load of 160 N was used which gave a nominal contact stress on the pins of 22.6 MPa. The frequency of the reciprocating cycle was 2 Hz, the sliding distance being 52 mm. The duration of time between wear measurements was 4 days. The motion is illustrated in Fig. 4.

3.3.2. Multidirectional motion pin-on-plate test (smooth)

In this multidirectional motion test, the pin was rotated as the plate oscillated back and forth by means of a simple rack and pinion gear mechanism (see Fig. 4b), subjecting

Figure 3 Reciprocating pin-on-plate wear machine.

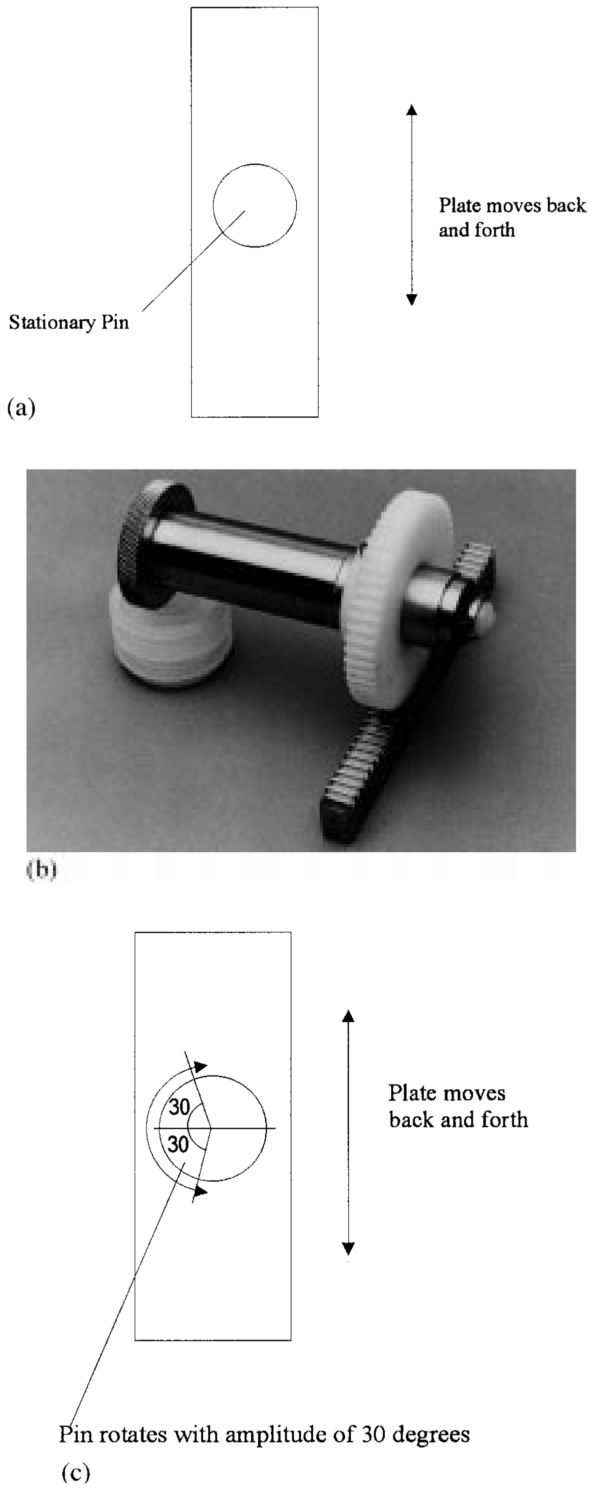


Figure 4 (a) Unidirectional motion pin-on-plate wear test. (b) UHMWPE pin in pin holder with rack and pinion gear mechanism to provide rotation. (c) Plan view of multidirectional pin-on-plate test.

the wear face to multidirectional motion and varying the direction of the resultant frictional force (see Fig. 4c). The load used was again 160 N and because of the highly polished plates, the duration of each test was again 4 d. The sliding distance was 64 mm and the total pin rotation was 120° per cycle. A test cycle frequency of 1 Hz was used to prevent the lubricant from spilling out of the containers.

3.3.3. Unidirectional motion pin-on-disc test (rough)

By using a tri-pin-on-disc machine (see Fig. 5a), the three pins were worn simultaneously against the same rough counterface disc which removed the effect of plate to plate variability seen in the pin-on-plate tests. The test pins were fixed in a cylindrical holder and loaded axially in compression via the pin holder and a cantilever mechanism onto the disc as it moved beneath them with a rotating motion.

The CoCr disc was rotated against either three crosslinked RCH 1000 pins or three virgin RCH 1000 pins. In order to subject the pin to abrasive wear, the surface of the CoCr plate was lapped smooth initially to remove the periodicity of the surface and then roughened to $R_a \sim 0.09$ μm. This value of R_a was the same for all the pins in this test. A frequency of 1 Hz, and a load of 80 N per pin (giving a nominal stress of 11.3 MPa) were sufficient to create measurable wear over a 2-day testing period. The total sliding distance was 80 mm per cycle. Between the wear tests, the rough CoCr counterface was measured using a Rank Taylor Hobson Talysurf 6 surface measuring machine to ensure that the surface roughness had not changed substantially. The motion of this rig is illustrated in Fig. 5b.

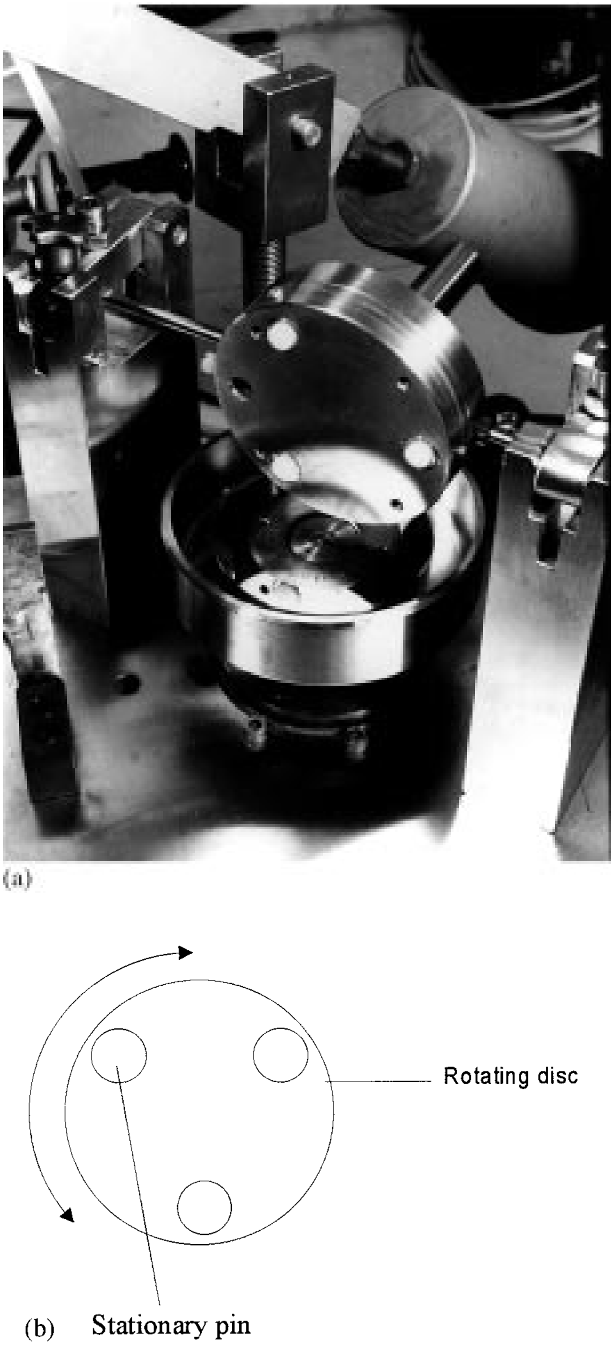


Figure 5 (a) Tri-pin on disc wear machine. (b) Plan view of pin-on disc test.

3.3.4. Multidirectional motion pin-on-plate test (rough)

This test had the same set-up as the smooth multidirectional motion test, except that rough plates with an average R_a of 0.09 μm were used. The test was run at 1 Hz, for a duration of 4 d. between measurement intervals. Because of the high wear being produced by the rough counterface, a load of only 80 N per pin was needed, giving a nominal stress of 11.3 MPa. The sliding distance per cycle was 64 mm. Once more, the rotation of the pin was 120° per cycle.

3.3.5. Calculation of wear factor

After the desired sliding distance had been reached, each set of test apparatus was dismantled and cleaned. The test pins were removed from the holders and cleaned ultrasonically along with the control pins to remove all traces of debris and lubricant. The pins were then placed in a controlled environment for 2 d after which they were carefully weighed using a Gallenkamp balance accurate to 1 μm. The weight changes of the unworn control pins were then either added or subtracted from the weight changes of the test pins to enable the weight loss due to wear of the test pins to be calculated. This was then converted to a volume loss and the corresponding wear factors were calculated using the following equation

$$\text{Wear factor, } K(\text{mm}^3/\text{Nm}) = \frac{\text{Volume loss, } V}{\text{Load, } L \times \text{Sliding distance, } X} \quad (3)$$

3.3.6. Statistical analysis

A Student's t-test was used to analyze the wear data as per the method documented by Mould [17]. Confidence limits of 95% were also calculated by multiplying the standard error by the Students' t-value found for a set of data from one material [17]. These confidence limits have been presented in the form of error bars on the wear factor histograms. Statistical significance between the mean wear factors of the two materials was determined using a Students' t-test for each test condition. The probability p is taken as the probability that the difference between the means occurred purely by chance. Significance is taken for $p < 0.05$.

4. Results

4.1. Gel fraction analysis

Ordinarily virgin, or non-crosslinked, polyethylenes do show a very low or zero gel fraction following solvent extraction. Usually a degree of crosslinking is required before a gel fraction can be achieved. The dose required to produce a gel fraction is known as the dose to incipient gelation. This dose depends largely on the molecular weight of the unirradiated polymer. The diffusion time to extract a non-crosslinked non-branched entangled chain into the solvent is proportional to the square of the number of rotatable bonds. Hence, for UHMWPE, the number of reptations (bond rotations) required to do this is excessively high and cannot be reached in normal time scales. The virgin non-crosslinked UHMWPE in this study still had a gel fraction of 0.146 even after 96 h of extraction. This gel fraction is not likely to be improved upon until the extraction time is increased by several orders of magnitude. This was therefore an impractical proposition, rendering it impossible to directly calculate the degree of crosslinking in the acetylene treated UHMWPE RCH 1000, which had a measured gel fraction of 0.937. However, it is possible to estimate the true gel fraction ($\mathrm{Gel}_{\mathrm{true}}$) of the irradiated sample of gel fraction ($\mathrm{Gel}_{\mathrm{i}}$), by simply subtracting the gel fraction of the virgin material ($\mathrm{Gel}_{\mathrm{v}}$)

$$\mathrm{Gel}_{\mathrm{true}} \approx \mathrm{Gel}_{\mathrm{i}} - \mathrm{Gel}_{\mathrm{v}} \tag{4}$$

This yields a $\mathrm{Gel}_{\mathrm{true}}$ value of 0.791 for the irradiated UHMWPE RCH 1000, from which the degree of crosslinking can be estimated. The true gel fraction versus number of crosslinks per initial preirradiated number average molecule ($M_{n,0}$) is not expected to change significantly from one PE to another. Hence, using the work of Jones *et al.* [15] and particularly Fig. 9 therein, the 'gel-effective' and 'total' numbers of crosslinks in the UHMWPE RCH 1000 of the present study were estimated to be 0.88 and 2.01 per $M_{n,0}$ respectively. 'Gel-effective' crosslinks are those which contribute to increasing the average molecular weight of each chain and the number is low, but structurally, in total, there are about two crosslinks per initial number average molecule of the treated UHMWPE RCH 1000, meaning that on average each molecule is connected to four others. These results indicate that an extensive network has developed in the UHMWPE RCH 1000 following γ-irradiation and annealing in acetylene with an absorbed dose of only 2.5×10^4 Gy (2.5×10^6 Rad).

4.2. Tensile tests

From Table II, it can be seen that in comparison to the virgin material crosslinking UHMWPE RCH 1000 increased the yield stress and 5% proof stress, whereas the strain to failure was decreased, resulting in a similar energy to failure to that of the virgin RCH 1000. This can also be seen in the average true stress-strain plots for the two materials shown in Fig. 6.

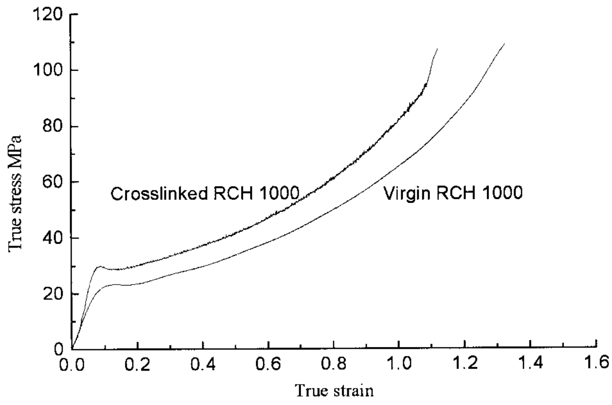


Figure 6 Mean tensile results for crosslinked RCH 1000 and virgin RCH 1000. Specimens were pulled to failure.

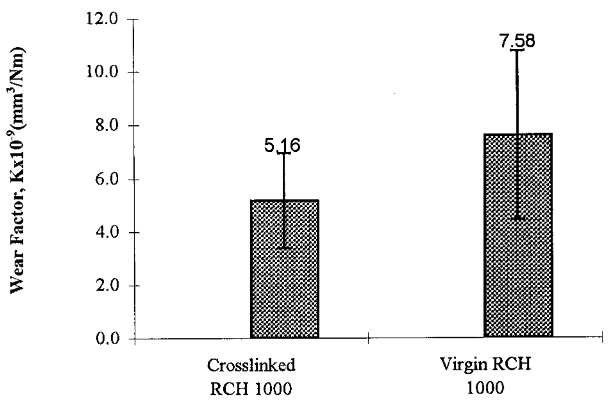


Figure 7 Mean wear factors for unidirectional motion pin-on-plate test (smooth counterface) $\pm$ 95% confidence limits.

4.3. Wear tests

4.3.1. Unidirectional motion pin-on-plate test (smooth)

The average results shown in Fig. 7 indicated that the virgin material had a slightly higher ($\times 1.5$) wear factor than the crosslinked material. Statistical analysis using a Students' t-test gave a probability (P) of 0.14 that these mean results were different due to chance, i.e. that both sets of results came from the same populations. Thus there was no significant difference in the wear factors for the two materials under these test conditions.

4.3.2. Multidirectional pin-on-plate test (smooth)

It can be seen from Fig. 8 that the wear factor for the virgin material was nine times greater than for the

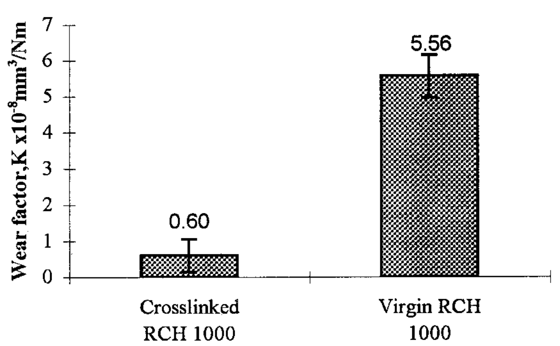


Figure 8 Mean wear factors for multidirectional motion wear test (smooth counterface) $\pm$ 95% confidence limits.

TABLE II Mean tensile test results $\pm$ 95% confidence limits

Materials	Yield stress (MPa)	5% Proof stress (MPa)	UTS (MPa)	Strain to failure	Energy to failure ($MJ\,m^{-3}$)
Virgin RCH 1000	20.0 ± 0.4	22.3 ± 0.2	108.9 ± 6.6	1.32 ± 0.02	60.5 ± 1.8
Crosslinked RCH 1000	25.8 ± 1.1	29.2 ± 0.3	107.2 ± 3.2	1.10 ± 0.00	60.3 ± 1.6

crosslinked material in this smooth multidirectional test. In fact the average wear factor for the crosslinked material in the multidirectional smooth wear test was roughly the same as in the corresponding unidirectional test. However, there was seven times more wear in the virgin material in the multidirectional test compared to the unidirectional smooth test.

The Students' *t*-test also showed that the difference between the mean wear factors for the two materials in this test configuration was significant with a probability, $P < 0.001$.

4.3.3. Unidirectional motion pin-on-disc test (rough)

This rough test produced much greater wear factors than for the multi- and unidirectional smooth tests (see Fig. 9). The results showed less difference between the virgin and crosslinked materials than for the multidirectional smooth test. However, the crosslinked material gave a 1.5 times greater wear factor than the virgin material and this difference between the means was statistically significant ($P = 0.05$).

4.3.4. Multidirectional motion pin-on-plate test (rough)

This test produced the greatest wear factors of all (see Fig. 11b). Compared with the unidirectional smooth test, the biaxial motion coupled with the rough counterface increased the wear factor by a factor of 410 for the crosslinked material and by 360 for the virgin material. As for the unidirectional tests, the two materials in this particular test had similar wear factors, with the isotropic material having the slightly higher mean value. However, the difference was not statistically significant. Fig. 10a shows that there was considerable scatter in the wear factors for each material. This was probably due to small differences in the topographies of the individual rough counterfaces. Fig. 10b shows the wear factors for the two materials for the three counterfaces which had distinctively different R_a values. This plot shows that the counterface roughness was the dominant variable controlling the wear rate.

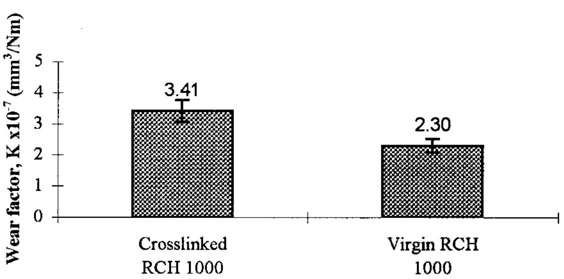


Figure 9 Mean wear factors for unidirectional motion pin-on-disc wear test (rough counterface) $\pm$ 95% confidence limits.

5. Discussion

5.1. Effect of surface roughness

The four different wear tests produced very different wear rates as shown in Fig. 11(a) and (b) and were thought to promote different wear mechanisms in the UHMWPE. The smooth tests are likely to subject the polymer pin primarily to adhesive and deformation wear and the rough tests to primarily a more abrasive wear mechanism as defined below.

(i). Adhesive and deformation fatigue wear involves the removal of polymer by the harder counterface asperities after many interactions. In unidirectional tests the adhesive frictional force associated with the multiple asperity interactions may cause molecular orientation in the surface layers of the UHMWPE [18].

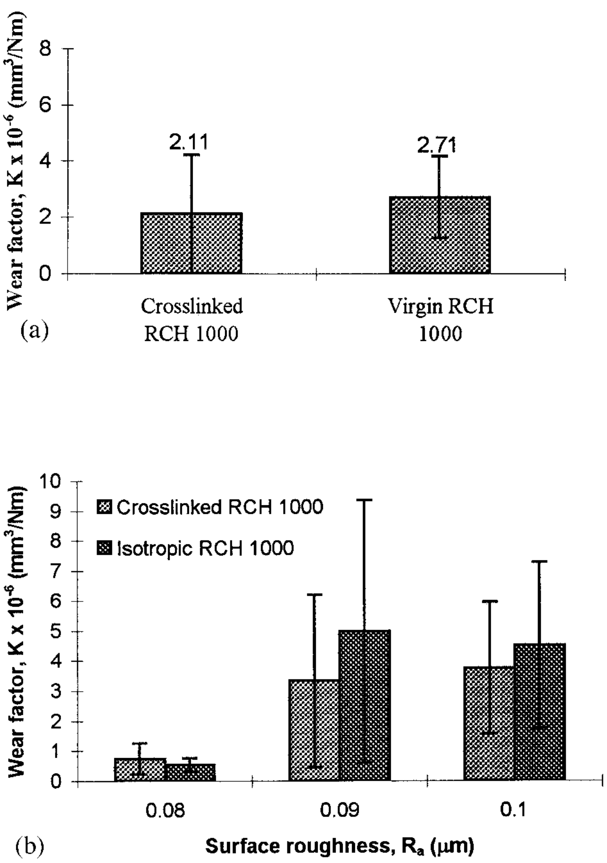
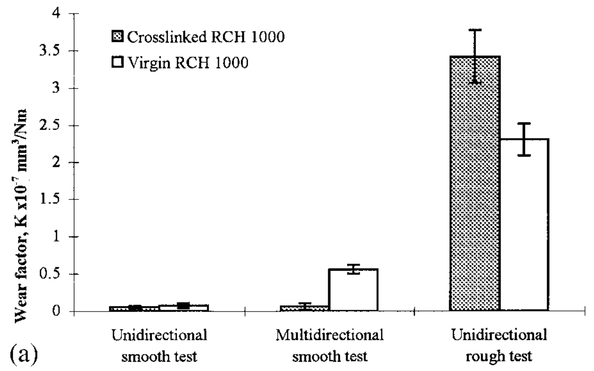


Figure 10 (a) Mean wear factors for multidirectional motion pin-on-plate wear test (rough counterface): $\pm$ 95% confidence limits. (b) Mean wear factors for individual plates of rough multidirectional test $\pm$ 95% confidence limits.



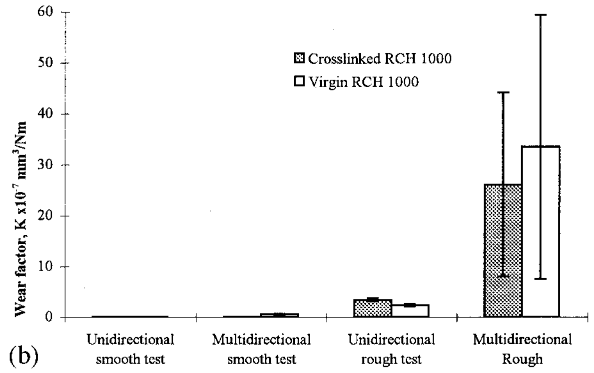


Figure 11 (a) Mean wear factors for three different tests $\pm$ 95% confidence limits (excluding multidirectional rough test). (b) Mean wear factors for all four wear tests $\pm$ 95% confidence limits.

(ii). Abrasive wear involves the ploughing of a soft material by the large surface asperities of a harder material, and is associated with rough surfaces. In this case, wear debris may be generated by a single rough asperity interaction. The counterface used in the pin-on-disc test had an $R_a \sim 0.09\ \mu m$, causing the asperities of the hard metal counterface to cut into the softer polyethylene pin, creating a greater level of stress concentration and removing the polymer by a low cycle fatigue or abrasive action.

The rough tests were used to indicate the behavior of the clinical material interface after third body wear. Third body wear occurs, *in vivo*, as a result of cement and bone particles that free themselves and work their way into the cavity between the articulating surfaces causing scratches and other surface damage [19]. Isaac *et al.* [20], have showed that damage to stainless steel prostheses is predominantly caused by the X-ray contrast medium in acrylic bone cement. Wear tests using simulated scratches of similar R_a to those produced by these particles *in vivo* have shown up to a 70-fold increase in wear factor [21]. In this study, the rougher counterfaces increased the wear rate of both materials between 30 and 410 times.

Although the crosslinked material showed substantial advantage over the virgin material in the smooth multidirectional test, this was not realized in the corresponding rough tests. The multidirectional rough test showed the highest wear factors and no difference between the materials. In the unidirectional rough test, the crosslinked material had a significantly higher wear rate. It is likely that, under the more abrasive wear of the rough counterface, less molecular orientation is produced as the surface polymer is removed with a single asperity interaction. Under these abrasive conditions, other factors such as strain energy to failure may be more important [22].

5.2. Effect of multidirectional motion

In the unidirectional tests, the friction force acting on the wear face of the pin was always in the direction of sliding motion. However, in the multidirectional tests, due to the rotation of the pin against the reciprocating plate, the wear face of the pin was subjected to a frictional force that constantly changed direction, i.e. with components acting both parallel and perpendicular to the sliding direction. If the pin wear face is split up into elements (see Fig. 12a) and the forces are shown on an element they would resemble those shown in Fig. 12(b) and (c) as the pin rotates. F is the force acting on the pin due to friction and this can be assumed not to significantly change direction with respect to the plate although it will change direction with respect to the pin. If we resolve the translational friction force F along the fixed x and y axes of the element and plot the variation in the components of the friction force against rotation of the pin, a sinusoidal relationship is apparent (see Fig. 12d). It can be seen that there is a great deal of variation in the frictional forces along the principal axes, x and y, of the element.

It was in this multidirectional test that the crosslinked RCH 1000 showed considerably less wear than the virgin RCH 1000, by almost an order of magnitude, whereas in the unidirectional motion tests there was little difference between the two materials. Indeed, in both uni- and multidirectional smooth tests, the crosslinked material had similar wear factors, whereas the virgin material gave a seven-fold increase. It has been proposed by Wang *et al.* [18], that in unidirectional motion tests, the molecular chains at the surface of the virgin RCH 1000 become oriented parallel to the sliding direction giving the surface of the material some resistance, i.e. ''orientational hardening''. In effect the UHMWPE becomes stronger in the sliding direction, resulting in a high wear resistance and low wear rate. Wang *et al.* [18] also suggest that the virgin material becomes strain softened in the direction perpendicular to the sliding direction causing ''orientational softening''.

Assuming that orientational hardening and softening occurs in both uni- and multidirectional smooth tests, the reason for the great difference in wear results will lie with the constant direction of the frictional force in the unidirectional test and the varying direction in frictional force in the multidirectional test. As the frictional force acts in the direction of sliding in the unidirectional test and the orientation of the molecules also lie in this direction, the materials show low wear. For the multidirectional test, though, the frictional force is constantly changing direction with respect to the polymer surface and is sometimes parallel and sometimes perpendicular to the orientated molecules (i.e. parallel to the direction of orientational softening); this latter may account for the increase in wear factor of the virgin material in multidirectional tests. In the case of the crosslinked material, the network of crosslinks may prevent the same degree of orientational softening and hardening that

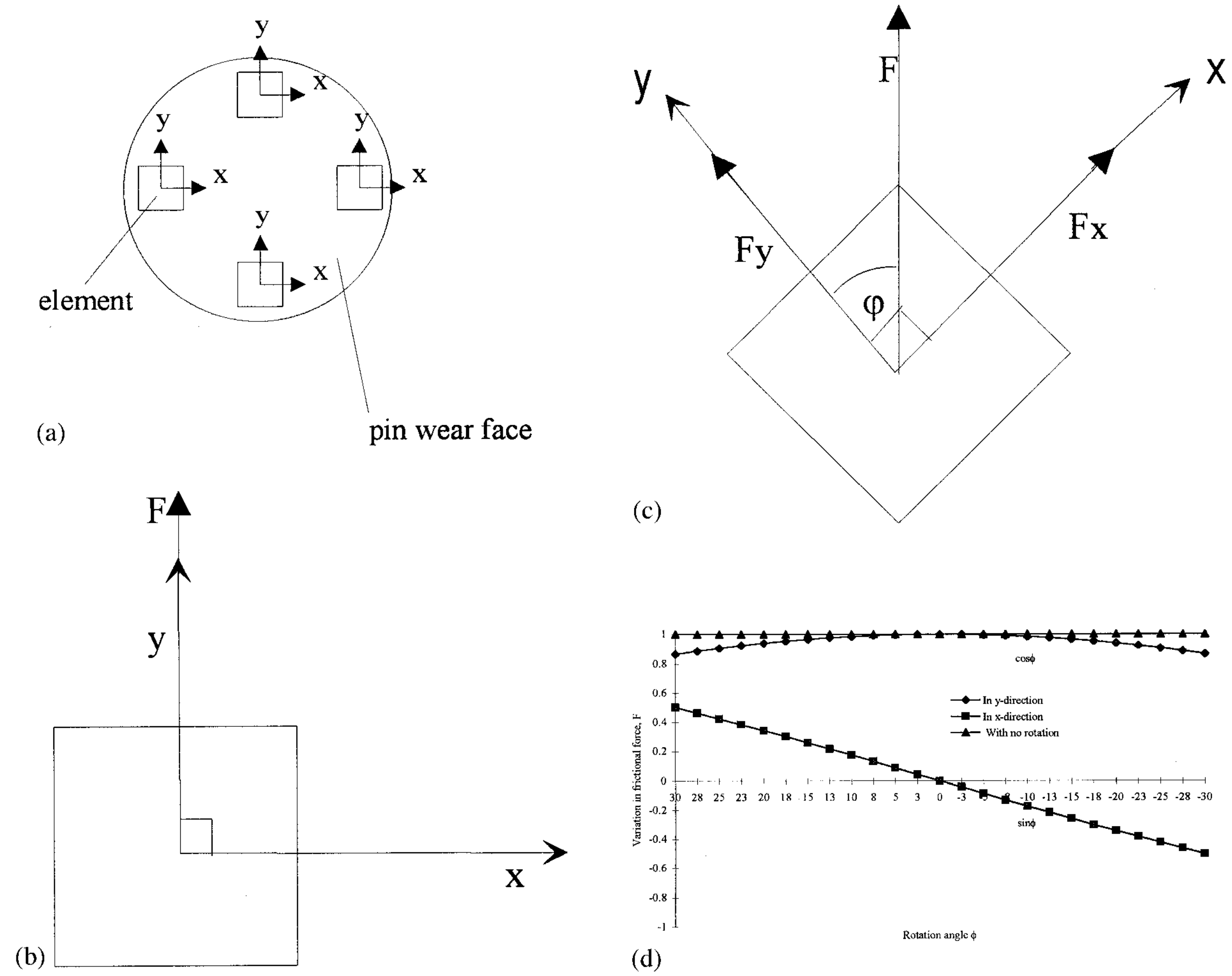


Figure 12 (a) Elements on wear surface of pin. (b) Translational frictional force F along the principal axes. x and y, of an element on the pin wear face for zero pin rotation F is in the direction of sliding motion. (c) Translational frictional force components F_x, F_y along the principal axes, x and y, after the pin has rotated through an angle ϕ. (d) Variation in the frictional force components F_x and F_y along the principal axes (x and y) of an element on a pin wear face per half cycle.

occurs in the virgin material. Hence, the crosslinked material did not wear as rapidly as the virgin material in the multidirectional smooth test.

The difference in the wear rates between the unidirectional and multidirectional motions may also be explained by an alternative but similar approach. Regarding the virgin material in the multidirectional motion tests, the degree of strain or orientational hardening will be less due to the direction of the resultant friction force constantly changing and causing the wear rate to increase. Whereas, with the crosslinked material, the additional molecular crosslinks may produce lower wear rates for both uni- and multi-directional smooth tests, as they provide resistance to both deformation and adhesive fatigue wear processes. This may be associated with the elevation in the yield stress of the crosslinked material.

It has been commented [23] that linear wear machines are not suitable for testing polyethylene bearing materials for hip prostheses. Wear factors generated by linear wear machines are always considerably lower than those found from clinical results. This is considered to be a result of the different motions encountered in these two situations. A multidirectional test such as a hip simulator or a rotating pin mechanism such as described in this paper yield wear factors closer to those found in clinical trials because, as illustrated by Wang *et al.* [18] with the aid of a computer model, the shear stresses on the articulating surface of the natural hip joint due to friction constantly change direction, and most certainly are multidirectional.

5.3. Effect of crosslinking

By crosslinking UHMWPE RCH 1000, the yield stress of the material was increased and the strain to failure decreased without significantly altering the energy to failure. Due to the network of crosslinks, the material required more force to allow the molecular chains to untangle and slide. Moreover, the crosslinks limited the relative movement of the molecular chains causing a reduction in the strain to failure. The tensile results from the crosslinked material, supported the wear results in the multidirectional smooth test showing a greater resistance to wear.

There is an increasing body of evidence of reduced wear with crosslinked materials for multidirectional tests and smooth counterfaces (as shown by Wang *et al.* [24] and Shen *et al.* [25]), and this study supports these findings. There has already been clinical evidence of low wear with crosslinked materials. Oonishi *et al.* [26] suggest that the best total hip prosthesis is one with an alumina head and a UHMWPE socket irradiated with a higher dose of 10^6 Gy (10^8 Rad) of γ-radiation, whilst

Wroblewski *et al.* [27] have also shown low wear with crosslinked polyethylene and ceramic heads. However, this paper has shown that these advantages are negated with roughened counterfaces. In interpreting the results of the crosslinked material sliding against roughened counterfaces, in the context of a metallic femoral head that may become roughened *in vitro*, it is important to consider the degree of roughness. Although the R_a values in this study were similar to that of 0.07 μm found in damaged heads [20, 21], it is important to recognize that this is a harsh test condition in the context that *in vivo* typically only a small proportion of the area of the head is damaged in comparison to the whole counterface in the pin-on-plate or pin-on-disc test. *In vivo*, not all the wear surface of the polyethylene is exposed to the roughened portion of the head, but nevertheless, there remains considerable advantages in using ceramic femoral heads with crosslinked polyethylene.

6. Conclusion

This study demonstrates that the wear factor of a material depends greatly on the type of motion created in a wear test as well as on the surface roughness of the counterface. It is evident that UHMWPE RCH 1000 crosslinked using γ-irradiation in the presence of acetylene has greatly reduced wear in a smooth multidirectional motion test compared with the virgin material. These benefits are not present in a simple unidirectional motion test where the virgin material is more resistant in the sliding direction due to the effect of surface strain induced orientation. This is of great significance as the multidirectional motion test is closer to the relative motion in the natural hip than is the standard unidirectional test. However, in both uni- and multidirectional tests using a rough counterface, there was little difference in wear rates between the virgin and the crosslinked material. The results indicate considerable benefits will be gained by using the acetylene enhanced crosslinked polyethylene in artificial hip joints but only if the femoral head remains smooth.

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