

Plasticity-Induced Damage Layer Is a Precursor to Wear in Radiation-Cross-Linked UHMWPE Acetabular Components for Total Hip Replacement

Avram A. Edidin, PhD,* Lisa Pruitt, PhD,† Charles W. Jewett, MS,‡
Deborah J. Crane, MS,† Daniel Roberts, PhD,‡ and Steven M. Kurtz, PhD§

Abstract: The mechanism for the improved wear resistance of cross-linked ultra-high-molecular-weight polyethylene (UHMWPE) remains unclear. This study investigated the effect of cross-linking achieved by gamma irradiation in nitrogen on the tribologic, mechanical, and morphologic properties of UHMWPE. The goal of this study was to relate UHMWPE properties to the wear mechanism in acetabular-bearing inserts. Wear simulation of acetabular liners was followed by detailed characterization of the mechanical behavior and crystalline morphology at the articulating surface. The wear rate was determined to be directly related to the ductility, toughness, and strain-hardening behavior of the UHMWPE. The concept of a plasticity-induced damage layer is introduced to explain the near-surface orientation of the crystalline lamellae observed in the wear-tested acetabular liners. Cross-linking reduces abrasive wear of acetabular components by substantially reducing—but not eliminating—the plasticity-induced damage layer that precedes abrasive wear. **Key words:** ultra-high-molecular-weight polyethylene, wear, cross-linking, hip simulator, small punch test, transmission electron microscopy, ultimate properties, morphology, fracture, plasticity-induced damage layer.

Researchers have hypothesized that wear at the articulating surface of total hip replacements is related to the mechanical response of ultra-high-molecular-weight polyethylene (UHMWPE) at large strains under multidirectional loading [1,2]. Direct correlations between the wear behavior of UHMWPE

and its mechanical behavior at large strains have been limited, however, primarily because bulk material tests are not performed on specimens scaled for the articulating surface. Advances in miniature specimen testing techniques have greatly facilitated direct mechanical characterization of UHMWPE implants [3–7]. The small punch test, originally developed to characterize the tensile and fracture properties of large metallic components using a miniature specimen [8], is particularly well suited for the mechanical characterization of retrieved components. Small punch testing technology was applied to and validated for virgin UHMWPE using disk-shaped specimens measuring 0.5 mm in thickness and 6.4 mm in diameter [3]. A subsequent investigation demonstrated that the small punch test can be used successfully to characterize as irradiated and

*From *Stryker Howmedica Osteonics, Allendale, New Jersey; †Department of Mechanical Engineering, University of California at Berkeley, Berkeley, California; ‡Exponent Failure Analysis Associates, Menlo Park, California; and §Exponent Failure Analysis Associates, Philadelphia, Pennsylvania.*

Funds were received from a research grant from Stryker Howmedica Osteonics in support of the research material described in this article.

Submitted March 15, 1998; accepted October 26, 1998.

Reprint requests: Avram A. Edidin, PhD, Stryker Howmedica Osteonics, 59 Route 17, Allendale, NJ 07401.

Copyright © 1999 by Churchill Livingstone®

0883-5403/99/1405-0016\$10.00/0

oxidatively degraded UHMWPE that had been gamma irradiated in air or nitrogen [4]. Thus, miniature specimen testing currently provides a reliable and effective means to characterize the mechanical properties of UHMWPE using specimens scaled to the articulating surface.

As a semicrystalline polymer, the mechanical behavior of polyethylene is strongly related to its molecular weight and crystalline morphology. For UHMWPE with an average molecular weight of 6 million g/mol, the molecular backbone consists of approximately 400,000 carbon atoms. Visualizing the polymer chain as an entangled strand of string, its unentangled length would be greater than 1 km were the string to have the same aspect ratio as a UHMWPE molecule. Despite the length of the polymer chain, UHMWPE in the solid state is a two-phase material, consisting of a crystalline phase embedded within an amorphous phase. The crystalline phase in UHMWPE consists of folded rows of carbon molecules packed into lamellae, typically 10 to 50 nm in thickness and on the order of 10 to 50 μm in length. The amorphous phase consists of randomly oriented and entangled polymer chains from neighboring molecules. The amorphous phase is also traversed by tie molecules, which interconnect remote crystalline domains and provide additional resistance to mechanical deformation. Thus, at a microstructural level, UHMWPE can be considered to be a complex composite material.

Although the crystalline phase within UHMWPE is typically randomly oriented within the amorphous phase, the size, shape, and orientation of the lamellae are sensitive to mechanical loading. For example, when deformed in uniaxial tension to strains greater than 0.17, the lamellae within UHMWPE begin to show irreversible changes, which persist even after unloading [9]. The phenomenon of irrecoverable, permanent strain resulting from mechanical loading is termed *plasticity*, and the study of lamellar plasticity, in particular, holds clues to the mechanical loading history of UHMWPE. Because the liberation of particulate debris from UHMWPE components is expected, at a microscopic level, to involve locally large deformations, from the examination of worn articulating surfaces one should theoretically be able to detect lamellar plasticity, or mechanical damage to the original crystalline morphology. This damage would thus be the precursor to wear and wear debris. Only a few detailed studies are currently available in the literature regarding the crystalline morphology of UHMWPE after processing into extruded rod or compression-molded sheet [10,11]. Goldman et al. [12] previously evaluated the crystalline structure and morphology of

gamma-irradiated UHMWPE from tibial components. The near-surface morphology in UHMWPE implants and its likely connection to wear mechanisms, however, has yet to be explicitly investigated.

Previous research has suggested that the mechanical loading conditions at the articulating surface of acetabular components may be conducive to the development of plasticity-induced damage near the surface [1,2,13–18]. While investigating the abrasion resistance of polyethylene under dry (unlubricated) conditions, Pooley and Tabor [13] concluded that molecular orientation was responsible for differences between static and kinetic friction at the articulating surface. More recently, wear surface and particle morphology characterization has been performed on retrieved UHMWPE implants as well as components that have been tested in conventional hip joint simulators [1,2,17,19,20]. Although wear testing machines do not precisely mimic the complex loading and kinematics of the human hip joint, conventional biaxial rocking hip joint simulators have been shown to reproduce clinically relevant wear rates and wear debris for conventional UHMWPE when operated under the appropriate conditions [21]. Detailed investigations of the wear surfaces and debris from acetabular components that were previously implanted in patients or exercised in joint simulators have suggested that the mechanisms of wear in conventional UHMWPE are related to orientation of long-chain molecules at the articulating surface [1,2]. The accumulation of plastic strains is believed to result in localized anisotropy and *strain softening* in a direction transverse to the oriented molecular chains [1]. Because of the crossing wear paths and multidirectional motion prevalent at the surface of total hip replacements, the shearing of the oriented UHMWPE molecules is hypothesized to be the primary mechanism of abrasive/adhesive wear in acetabular components [1,2].

Modifications to the structure of UHMWPE, such as cross-linking of the molecular chains, has previously been shown to reduce dramatically abrasive/adhesive wear in several *in vitro* hip and knee joint simulator studies [22–24]. It has been suggested that cross-linking of UHMWPE reduces abrasive wear by improving the resistance of the polymer molecules to plastic deformation at the articulating surface. The role of cross-linking on the local mechanical properties and crystalline morphology at the articulating surface, however, has yet to be studied in detail. Consequently, the purpose of the present study was to investigate the effect of cross-linking, as induced by gamma radiation in nitrogen, on the mechanisms of wear in UHMWPE acetabular

components for total hip arthroplasty. This study tested the hypothesis that the wear behavior of UHMWPE in a biaxial rocking hip simulator is directly related to the mechanical behavior measured at the articulating surface. The small punch test was used to provide quantitative measures of the large deformation mechanical behavior of UHMWPE, as characterized by ultimate strength, ductility, and toughness of the polymer. In addition, using transmission electron microscopy (TEM), we investigated the hypothesis that cross-linking increases the abrasive wear resistance of UHMWPE by reducing local plastic deformation at the articulating surface.

Materials and Methods

Twelve UHMWPE acetabular liners of 28-mm inner diameter were machined from a ram-extruded rod of GUR 4150 HP (PolyHi Solidur, Fort Wayne, IN). The reported molecular weight for the resin was 4.0 to 6.0 million g/mol. Implants were packaged in nitrogen and submitted to a commercial irradiation facility for repeated doses of gamma radiation (Table 1). Sets of 2 implants each were subjected to 0 to 5 repeated doses of gamma radiation in nitrogen without violation of the original packaging. Inserts were then subjected to wear testing in a biaxial rocking hip simulator. Wear testing was followed by detailed characterization of the resulting mechanical, physical, and morphologic properties of the UHMWPE.

Wear Testing

Wear testing was performed by using an MTS Multi-Station Hip Simulator fitted with commercially available cobalt-chromium femoral heads (MTS Systems Corp., Eden Prairie, MN). Before testing, specimens were presoaked in distilled water for 30 days. The acetabular liners were then mounted in an anatomic position in polyurethane molds

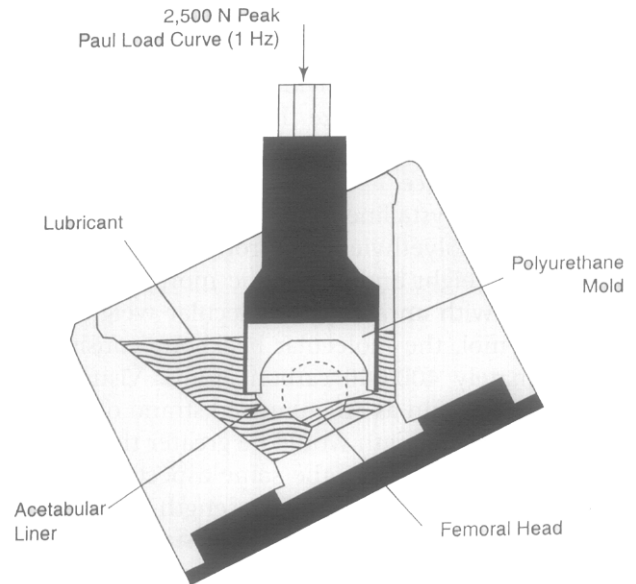


Fig. 1. Schematic of the MTS hip simulator used to test acetabular liners after gamma irradiation in nitrogen.

within the testing machine (Fig. 1). The Paul load curve was employed (2500 N maximal load) at a rate of 1 Hz for 3 million cycles.

During testing, triple-filtered bovine calf serum with 0.1% sodium azide, 20 mM ethylenediaminetetra-acetic acid (EDTA), and 30% distilled water was used as the lubricating medium. Serum was continually replenished with distilled water to account for evaporation. The lubricant was replaced every 2.6×10^5 cycles. The temperature of the lubricant was not controlled during the experiment but typically ranged between 30° and 35°C during the test because of frictional heating.

Every 5.7×10^5 cycles, the acetabular liners were removed from their individual test chambers and cleaned. The mass of each liner was determined using an analytical balance with a precision of 0.01 mg. Loaded and soaked controls were not used to account for lubricant absorption during the current experiment. Previous research at the first author's institution using loaded and soaked controls has shown that fluid absorption accounts for less than a few percent of the total change in mass of the specimen. Thus, the change in mass for each liner during the present study was not corrected for fluid absorption, which was judged to be negligible. The wear rate was calculated using standard linear regression from the steady-state slopes of mass loss as a function of cumulative loading cycles over the duration of the experiment.

Table 1. Summary of Irradiation Cross-Linking and Test Conditions

Repeat Gamma Doses in N ₂ (No.)	Lot Dosage (kGy)	Cumulative Dosage (kGy)	Average Density (g/mL)
0	0	0	0.934
1	27.1	27.1	0.938
2	26.3	53.4	0.940
3	26.7	80.1	0.941
4	26.5	107	0.941
5	26.8	133	0.942

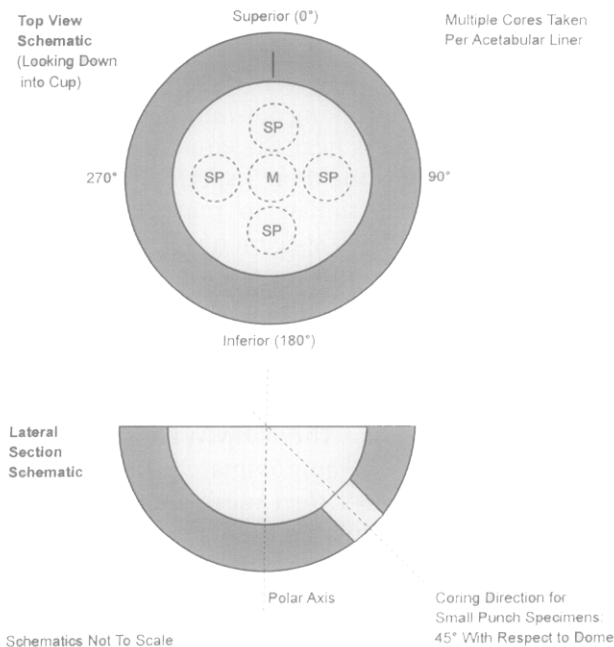


Fig. 2. Schematic of coring locations on the wear-tested acetabular liners. Four cores were taken for the preparation of small punch specimens (SP), and one core was taken for density and morphologic characterization (M).

Small Punch Testing

At the conclusion of wear testing, 4 6.4-mm-diameter cores were taken perpendicular to the worn articulating surface of 1 acetabular component per irradiation condition for the preparation of small punch specimens (Fig. 2). The cores were all taken at 45° with respect to the polar axis of the cup, and all of the cores were located within the worn area of the articulating surface. To test the hypothesis that local variations in loading might influence the measured mechanical properties of the UHMWPE, the location of each core (ie, at 0°, 90°, 180°, and 270° with respect to the superior-inferior reference plane) was carefully noted for consideration in the subsequent statistical analysis (Fig. 2).

The surface of each core was carefully machined to prepare a 500- μ m-thick, disk-shaped specimen starting within 25 μ m of the articulating surface; a second specimen from the core was prepared at a depth of 1.5 to 2.0 mm from the surface. Because of the difficulty in preparing specimens near the concave surface of the core, however, it was not always possible to manufacture all specimens within the required thickness tolerances [3]. As a result, 2 to 5 miniature disk-shaped specimens per cup had acceptably tight tolerances for subsequent small punch testing. A total of 25 small punch tests were per-

formed on the 6 multiply irradiated liners; 19 of the tests were performed on specimens starting within 25 μ m of the articulating surface, whereas the remaining 6 tests were performed on specimens taken in the depth range of 1.5 to 2.0 mm.

The small punch testing method used in this study has been previously described in detail [3,4,8], so only a brief overview of the procedure is provided here. The small punch specimens were placed in a custom-built apparatus and deformed against a hemispherical-head punch moving at a constant displacement rate of 0.5 mm/min (Fig. 3). The resulting load-displacement curve was characterized by an initial peak load, an ultimate load, and ultimate displacement. The work to failure, calculated as the area under the load-displacement curve, provided a measure of toughness. Finally, the shape of the load-displacement curve, which is sensitive to resin, irradiation, and oxidative degradation [3,4,8], was interpreted to provide insight into the large-strain plastic deformation response of the UHMWPE.

The effects of cross-linking by multiple irradiation doses in nitrogen were statistically evaluated using standard analysis of variance (ANOVA) procedures using SAS Version 6.12 (SAS Institute, Inc., Cary, NC). The effects of secondary experimental design variables, such as specimen location and depth, were also investigated. A *P* value of .05 was taken to indicate statistical significance.

Density and Morphology

For 1 component per irradiation condition, an additional core was taken for the characterization of average density and crystalline morphology through the thickness. The core was obtained parallel to the

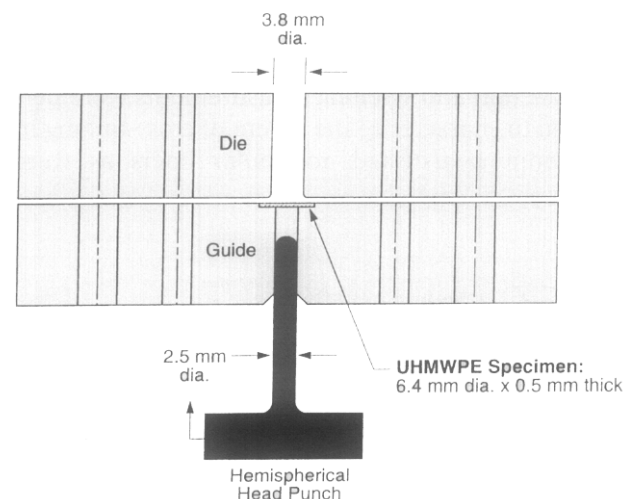


Fig. 3. Small punch test apparatus.

polar axis (Fig. 2), near the center of the worn region at the articulating surface. From each test case, 2 specimens prepared from each core were evaluated in the density gradient column prepared with isopropanol and distilled water according to ASTM D1505-85.

The crystalline morphology and lamellar alignment of the UHMWPE was determined as a function of depth below the articulating surface using TEM. Using a diamond knife, representative cores were sectioned into 65-nm slices and placed on grids containing 10 sequential sections spanning a range of 650 nm (0.65 μm) in depth (Fig. 4). The morphologic characterization was performed for each of the 6 dosages by performing TEM on successively ultramicrotomed slices of UHMWPE, starting at the articulating surface and continuing to a maximal subsurface depth of 9 μm . Specimens from each test group were prepared for TEM by staining the UHMWPE in chlorosulfonic acid (99% concentration) for 6 hours at 60°C. Chlorosulfonation of the UHMWPE stains, cross-links, and stabilizes the amorphous regions of the material [25]. After acid stain, the samples (allowed to reach ambient temperature) were rinsed with acetone (0°C) then with distilled water. The samples were dried at 60°C for 1 hour and embedded in epoxy resin with a curing time of 24 hours at 60°C. Each specimen was ultramicrotomed using a diamond knife into 65-nm-thick sections and collected onto Formvar-supported, carbon-rubidium slot grids. Slot grids allow for unobstructed viewing over larger areas of the sample. Before TEM evaluation, the specimens were poststained with 0.2% aqueous solution uranyl acetate for 3 hours to intensify contrast. Polymer morphology was characterized using a JEOL 100CX operating at 80 kV. Lamellar alignment was identified in TEM as domains of stacked crystalline lamellae.

Extraction and Swelling Measurements

Extraction and swelling measurements were performed to characterize the extent of cross-linking in the same wear-tested acetabular liners as were

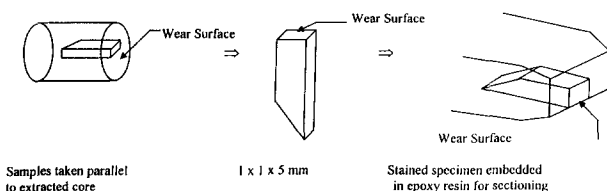


Fig. 4. Schematic of the transmission electron microscopy sample preparation and slicing orientations taken from the cores from acetabular liners.

subjected to small punch testing, density measurement, and morphologic characterization. Specimens of reproducible geometry were obtained from the 6 wear-tested liners by taking 6.4-mm-diameter cores through the thickness near the equator of the cup. Extraction was performed on 3 specimens per liner using an apparatus similar to that described by ASTM D 2765-95. Each specimen was placed in an individual 500-mL round bottom flask attached to a water-cooled condenser. Into each flask was added p-xylene and Irganox 1010 (0.5% by weight, Ciba-Geigy Corp., Tarrytown, NY), an antioxidant, to reduce degradation of the UHMWPE during the prolonged extraction. The UHMWPE specimens, each with a mass of approximately 0.3 g, were enclosed in a stainless steel mesh cage (U.S. 120 gage) and suspended within the solvent, which was heated to a rolling boil for 48 hours. Specimens were weighed before and after assembly in the mesh cage using a calibrated analytical balance with a precision of 0.1 mg.

After extraction, specimens were allowed to equilibrate for 2 hours in a fresh bath of p-xylene (also loaded with 0.5% antioxidant) that was maintained at a controlled temperature of 130°C. Specimens were then removed from the bath; carefully wiped to remove excess solvent; quickly transferred to the adjacent balance; and then weighed in the hot, swollen, and extracted state. Finally, specimens were dried in a vacuum oven at 60°C for 48 hours, which was previously determined to be sufficient time for the specimens to reach constant weight. Specimens were weighed once again in the dried and extracted state. Gel content, defined as percentage of polymer mass remaining after extraction, was calculated per ASTM D 2765. Swell ratio, defined as the volume ratio of the swollen extracted polymer to the dried extracted polymer, was also calculated in accordance with ASTM D 2765.

Results

Wear Testing

After an initial wear-in period, all of the cups exhibited linear wear behavior for the duration of the wear testing (Fig. 5). Five repeated doses of gamma irradiation in nitrogen (133 kGy cumulative dose) resulted in an 88% reduction in average wear rate when compared with the rate after a single irradiation dose (27.1 kGy cumulative dose). Based on ANOVA, the cumulative dose of gamma radiation was associated with statistically significant reductions in the wear rate ($P < .0001$). Multiple irradiation cycles of gamma radiation in nitrogen

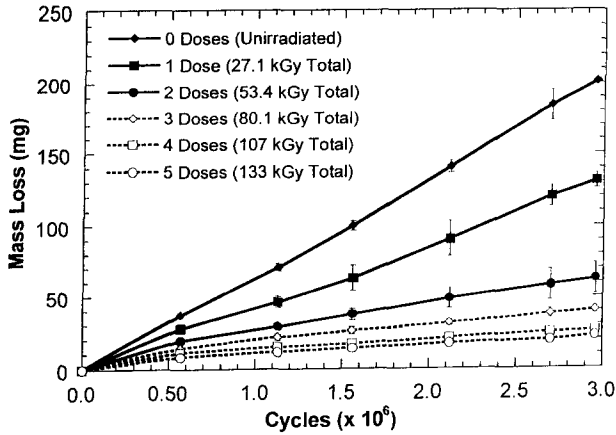


Fig. 5. Wear behavior (mass loss) of UHMWPE as a function of loading cycles and cumulative irradiation dose in nitrogen.

resulted in progressive and systematic decreases in the rate of wear up to 107 kGy (Fig. 6). Beyond 107 kGy, increased irradiation did not appear to provide further reduction in the wear rate.

Small Punch Testing

Mechanical characterization of the wear-tested acetabular liners using the small punch test also revealed systematic changes to the large-deformation mechanical behavior of the cross-linked UHMWPE at the articulating surface (Fig. 7). The load-displacement behavior of the irradiation-cross-linked UHMWPE specimens obtained within 25 μm of the articulating surface exhibited an initial peak load, followed by a membrane drawing phase dur-

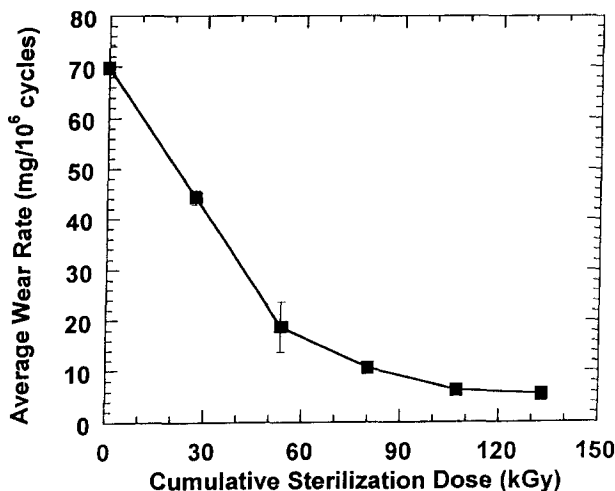


Fig. 6. Effect of cumulative irradiation dose in nitrogen on the average steady-state wear rate of UHMWPE.

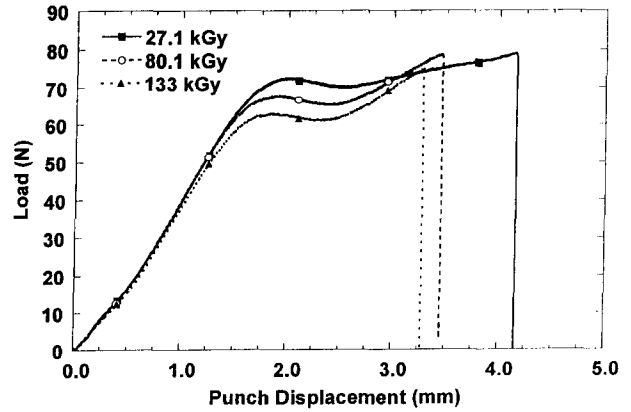


Fig. 7. Effect of cumulative irradiation dose on load-displacement behavior of UHMWPE during the small punch test.

ing which the UHMWPE deformed in equibiaxial tension around the head of the punch. The irradiation-cross-linked UHMWPE exhibited strain-hardening behavior after the initial peak load. The strain hardening of the cross-linked UHMWPE was especially notable after 5 cumulative doses of gamma irradiation in nitrogen (Fig. 7).

The small punch test reproducibly characterized the large-deformation mechanical response of the UHMWPE at the articulating surface for the multiply irradiated and wear-tested acetabular liners (Table 2). For example, after 1 irradiation dose in nitrogen, the relative experimental uncertainties in initial peak load, ultimate load, ultimate displacement, and work to failure were each less than 3% (Fig. 8). As indicated by Fig. 8, the small punch test results did not vary considerably as a function of location near the articulating surface.

Cumulative radiation doses of greater than 107 kGy were required to induce any measurable differences in the mechanical behavior of UHMWPE near the articulating surface as opposed to the depth of the acetabular liners. Up to a cumulative dose of 80 kGy, the load-displacement behavior of the UHMWPE at a depth of 1.5 to 2.0 mm was qualitatively similar to the near-surface behavior (Fig. 9A). At cumulative doses of 107 and 133 kGy, however, the subsurface mechanical behavior no longer displayed an initial peak load, whereas the mechanical behavior evaluated at the articulating surface, in contrast, still showed evidence of an initial maximum in the load-displacement curve (Fig. 9B). The ultimate load, ultimate displacement, and work to failure did not vary significantly as a function of depth into the liner ($P > .05$).

Using ANOVA, the wear rate of the acetabular liners was directly correlated to the metrics of the

Table 2. Summary of Hip Simulator and Small Punch Test Results

Cumulative Dosage (kGy)	Wear Rate (mg/10 ⁶ cycles)	Initial Peak Load (N)	Ultimate Load (N)	Ultimate Displacement (mm)	Work to Failure (mJ)
0	73.0	75.8	69.0	4.34	256
27.1	45.2	73.7 ± 1.0	76.9 ± 1.7	3.96 ± 0.08	228 ± 8
53.4	15.1	68.5 ± 3.6	73.2 ± 2.9	3.44 ± 0.09	176 ± 6
80.1	11.1	65.6 ± 1.6	74.0 ± 2.9	3.33 ± 0.04	165 ± 3
107	5.85	64.1 ± 1.8	73.3 ± 1.8	3.15 ± 0.07	152 ± 5
133	5.40	64.8 ± 2.4	69.7 ± 8.1	3.04 ± 0.13	144 ± 11

load-displacement curve obtained from the small punch tests performed at the articulating surface. The initial peak load was linearly correlated to the wear rate ($r^2 = .78, P < .0001$) (Fig. 10A). The ultimate load, in contrast, was not significantly affected by the wear rate ($P > .05$) (Fig. 10B). Ultimate displacement and work to failure were both found to increase linearly with increasing wear rate ($r^2 = .93, P < .0001$) (Fig. 10C, D). Thus, improved wear performance of radiation-cross-linked acetabular liners in the hip simulator was associated with statistically significant reductions in the initial peak load, ultimate displacement, and work to failure of the UHMWPE determined using the small punch test.

Density and Crystalline Morphology

The average density of the UHMWPE increased nonlinearly with cumulative radiation dose (Fig. 11). The greatest changes in density were observed during the first 27.1 kGy of gamma radiation dosage, for which density was observed to increase

from 0.934 g/mL to 0.939 g/mL. For additional cumulative doses, the rate of density change progressively decreased. The maximal average density was found to be 0.942 g/mL and occurred at a cumulative dosage of 133 kGy.

The TEM analysis showed evidence of lamellar alignment in all of the wear-tested UHMWPE, regardless of cumulative exposure to gamma radiation. Previous work [12] and TEM analysis of the

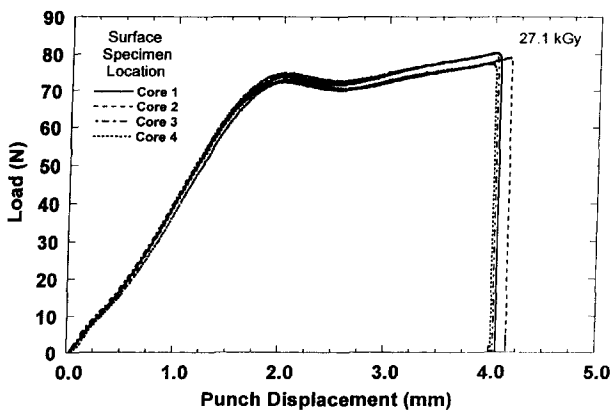


Fig. 8. Reproducibility of the small punch test performed on 4 specimens prepared from the articulating surface of the same wear-tested acetabular liner. Before wear testing, the liner was gamma irradiated in nitrogen with a dose of 27.1 kGy.

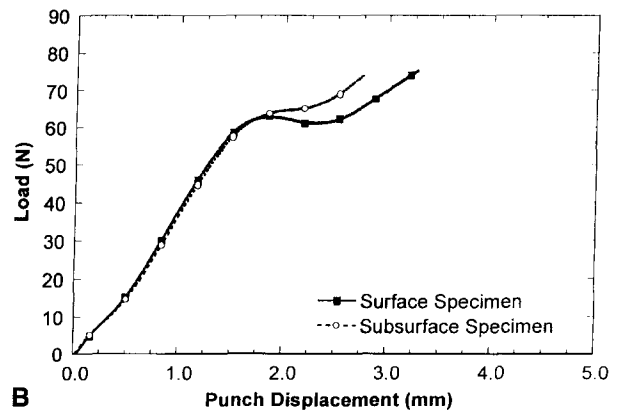
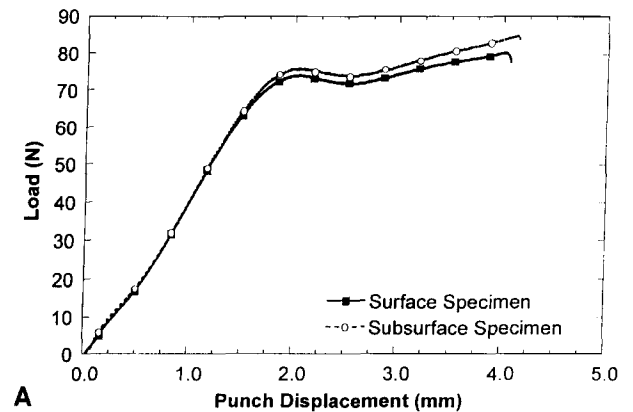
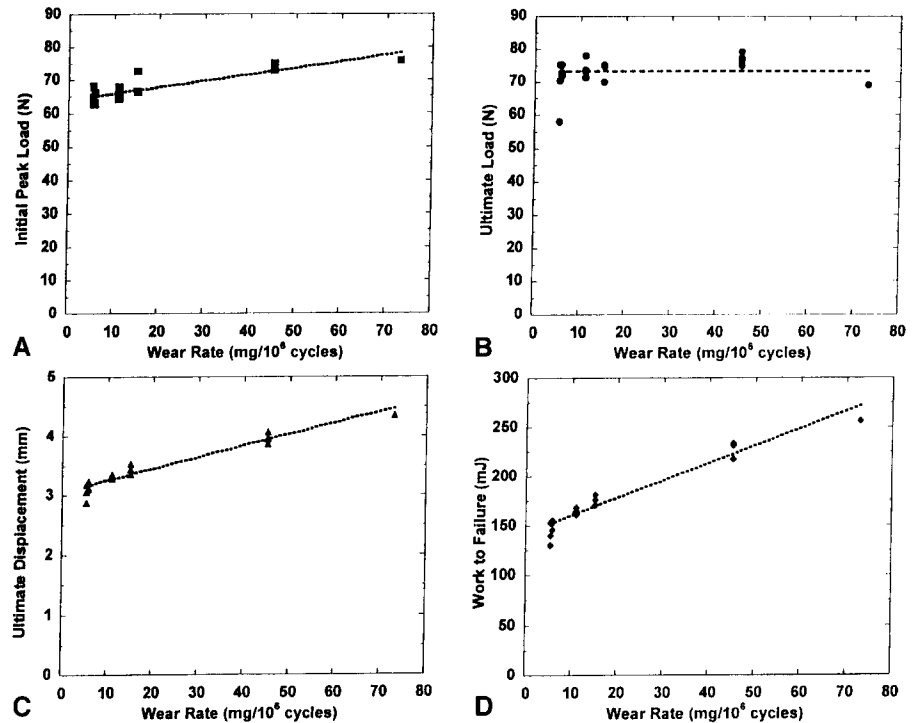


Fig. 9. Comparison of load-displacement behavior for specimens prepared from the articulating surface versus subsurface specimens prepared at a depth of 1.5 to 2 mm. (A) Cumulative irradiation dose of 27.1 kGy. (B) Cumulative dose of 133 kGy.

Fig. 10. Correlation of abrasive wear rate from the hip simulator with (A) initial peak load, (B) ultimate load, (C) ultimate displacement, and (D) work to failure as determined by the small punch test. Statistically significant correlations were observed between the wear rate and the initial peak load, ultimate displacement, and work to failure ($P < .0001$).



bulk material provided evidence that the microtoming process did not induce lamellar alignment, and hence the texture was judged not to be an artifact of the specimen preparation technique. For the virgin, non-cross-linked UHMWPE, lamellar alignment was evident to depths less than 9 μm below the articulating surface (Fig. 12A, B). At a depth of 9 μm , the morphology of the virgin UHMWPE was random and isotropic (Fig. 12C). The cross-linked UHMWPE also showed evidence of near-surface lamellar alignment after wear testing (Fig. 12D, E). Although there was still substantial texture development in the cross-linked UHMWPE, lamellar orientation

was observed only up to a depth of 4 μm below the articulating surface. This region of lamellar alignment in the wear-tested virgin and cross-linked UHMWPE was termed the *plasticity-induced damage layer*.

Extraction and Swelling Measurements

As expected, the unirradiated UHMWPE almost completely dissolved after the prolonged extraction (gel content of 1%). Although the extraction and swelling measurements clearly discriminated between irradiated and unirradiated UHMWPE, gel content and swell ratio were found to reach a nearly constant value after a single irradiation dose of 27 kGy in nitrogen (Fig. 13). The average gel content of the nitrogen-irradiated and wear-tested UHMWPE was found to be $99\% \pm 1\%$, whereas the average swell ratio was found to be 2.87 ± 0.13 .

Discussion

The results of this study support the hypothesis that the wear behavior of radiation-cross-linked UHMWPE acetabular components is directly related to the large-deformation mechanical behavior determined at the articulating surface. The small punch test has been previously shown to characterize reproducibly UHMWPE in equibiaxial tension to failure. This is the first study, however, in which the

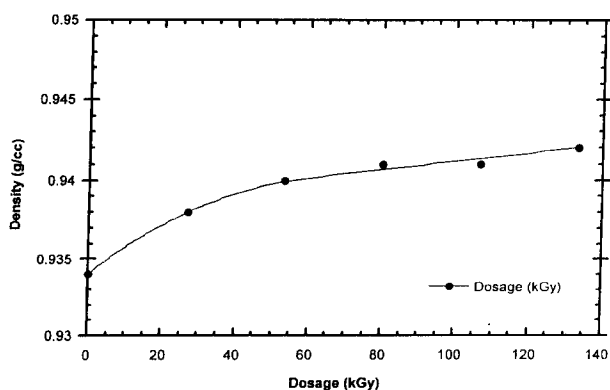


Fig. 11. Average density of the UHMWPE as a function of cumulative radiation dose.

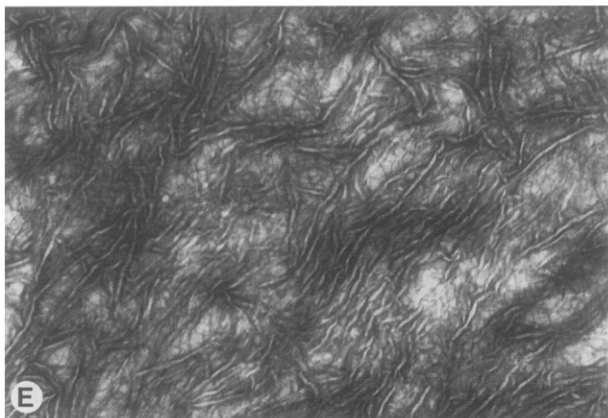
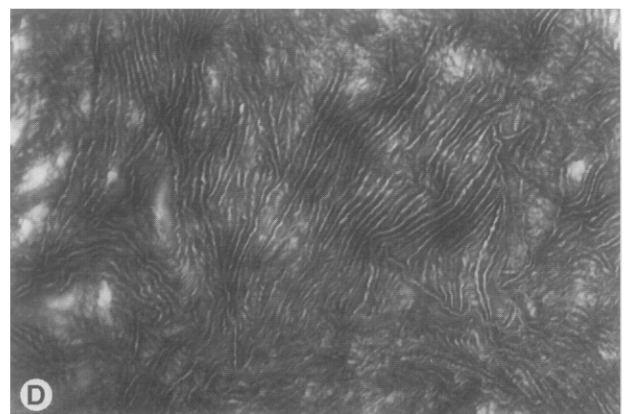
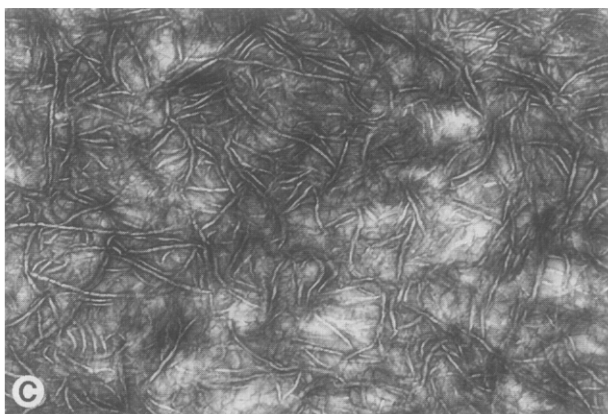
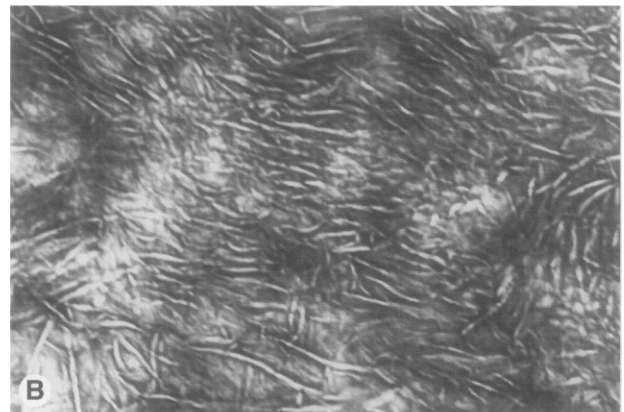
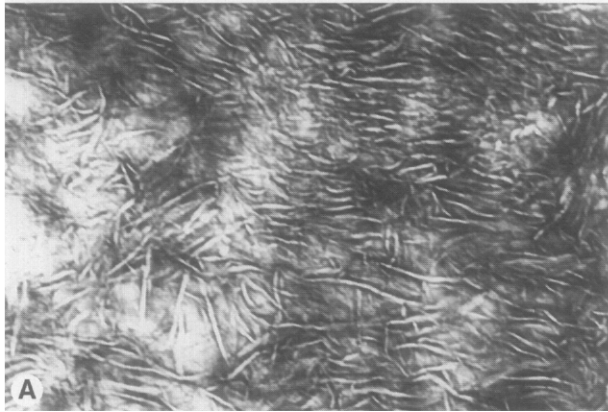


Fig. 12. After wear testing, the virgin, non-cross-linked (0 kGy) UHMWPE showed evidence of lamellar alignment at depths of (A) 1.3 to 2.0 μm and (B) 3.3 to 3.9 μm from the articulating surface. (C) At a depth of 9 μm , the morphology of the virgin UHMWPE was random and isotropic. The cross-linked UHMWPE also showed substantial lamellar alignment after wear testing but only to a maximal depth of 4 μm . (D) Depth of 1.3 to 2.0 μm for the cup irradiated with 80 kGy. (E) Depth of 3.3 to 3.9 μm for the cup irradiated with 133 kGy. The region of lamellar alignment in the wear-tested UHMWPE was termed the *plasticity-induced damage layer*. (Original magnification $\times 16,000$)

miniature specimen testing technique has been applied at the articulating surface of wear-tested orthopedic implants. In contrast with virgin UHMWPE, which exhibits strain softening after the initial peak load, the radiation-cross-linked materials in this study were observed to strain harden.

Previous researchers have shown that radiation-cross-linked UHMWPE components exhibit improved wear resistance in conventional hip simulators [22–24]. It has been proposed that cross-linking

of UHMWPE reduces wear *in vitro* by improving the resistance of the polymer molecules to lamellar alignment and plastic deformation at the articulating surface. The TEM performed in this study provides evidence that cross-linking does not eliminate texture development at the articulating surface but rather reduces the thickness of the plasticity-induced damage layer (Fig. 12). Cross-linking reduced the plasticity-induced damage layer 2-fold over the non-cross-linked material (as schematically

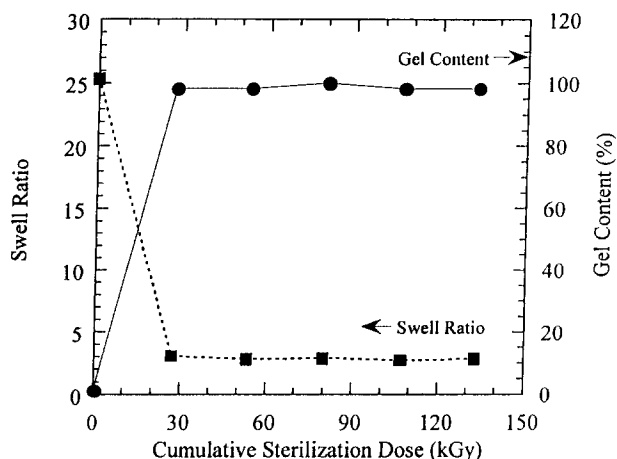


Fig. 13. Effect of cumulative dose on gel content and swell ratio for the UHMWPE.

depicted in Fig. 14). The results of this study suggest that the thickness of this damage layer plays a critical role in the wear mechanism of UHMWPE in acetabular components.

Exposure to high-energy radiation in an inert environment has been shown to promote cross-linking in UHMWPE [26,27]. In the present investigation, acetabular liners were subjected to repeated irradiation cycles to reach the desired cumulative dosage. Thus, the results of the present study are not directly comparable to prior work because each repeated irradiation cycle can be expected to result in both degradation (ie, chain scission) and cross-linking. The average density of the UHMWPE was observed to increase with repeated irradiation cycles, consistent with chain scission and degradative changes to the polyethylene [28–31]. Despite evidence of degradative changes to the UHMWPE during repeated cycles of irradiation, the wear performance improved significantly as a function of cumulative dosage. Work by Greer et al. [32] has shown that the improved wear performance of

radiation-cross-linked acetabular liners in a conventional hip simulator was unaffected by oxidative degradation. Thus, prior research and the present study suggest that the beneficial effects of cross-linking may predominate over the undesirable effects of concomitant degradation.

A noteworthy finding of this study was that the miniature specimen mechanical testing, density, and morphologic investigations provided more discriminating information about the wear-tested UHMWPE than did the swelling or extraction experiments. Our findings in this regard are consistent with the conclusions of previous researchers [33], who have also noted the lack of discriminatory power of standard swelling techniques to characterize cross-linking in UHMWPE. Novel and nonstandard techniques have been proposed by Muratoglu et al. [33] for direct measurement of swell ratio, which may provide the additional sensitivity that is apparently necessary to distinguish between UHMWPE of subtly varying cross-link densities.

The complex plasticity mechanisms in polyethylene can be categorized as intralamellar or interlamellar [9,34–37]. The intralamellar mechanisms occur within the crystalline lamellae and include twinning, martensitic transformation, dislocation chain slip, and unraveling. The interlamellar mechanisms are accommodated through the amorphous region and include interlamellar shear, separation of lamellae, tilting or rotation, and cavitation. The findings from the present study suggest that cross-linking affects the depth of crystalline plasticity and suppresses strain softening in the UHMWPE. Taken together, these observations suggest that radiation cross-linking hinders the interlamellar mechanisms of plastic deformation in the amorphous regions but does not prevent intralamellar plasticity from occurring within the crystalline regions of UHMWPE. Thus, the cross-linked UHMWPE investigated in the present study was not immune to abrasive wear but exhibited a significantly lower wear rate because of its smaller plasticity-induced damage layer.

Some of the findings in this study should be interpreted with caution. In addition to improved abrasive wear resistance, radiation-induced cross-linking was accompanied by significant decreases in the ductility and toughness of the UHMWPE. The reduction in ductility and toughness for cross-linked UHMWPE is of potential clinical concern, primarily because of their potentially negative impact on the fatigue and fracture resistance of the polymer [14,38]. Based on analyses of retrieved acetabular components, it appears that although abrasion and adhesion are the primary wear mechanisms in the hip, accelerated wear may infrequently be caused

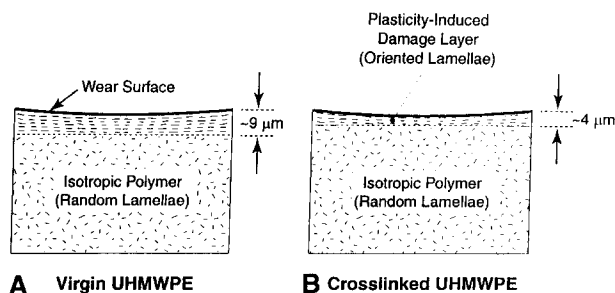


Fig. 14. Schematic of the plasticity-induced damage layer and its relative thickness for (A) the virgin UHMWPE and the (B) radiation cross-linked UHMWPE.

by scratches in the femoral head or by the introduction of third bodies, such as poly(methylmethacrylate) PMMA debris [2,17,21]. Historically, no alterations of UHMWPE have been shown to improve conclusively the clinical, *in vivo* performance of a hip arthroplasty. Therefore, from both a biomaterials and a biomechanics perspective, any modification of UHMWPE for total hip replacements must necessarily strike an optimal balance between wear performance, ductility, ultimate strength, fatigue endurance, and fracture resistance.

Conclusions

In this study, the mechanical and morphologic properties were determined for UHMWPE acetabular liners that were previously subjected to 3 million cycles of hip simulator testing. This work is novel in that miniature specimen testing methods were used to determine the mechanical behavior near the articulating surface of the UHMWPE liners. Further, the mechanical behavior of the cross-linked UHMWPE was correlated with the radiation dosage, *in vitro* wear rate, and polymer morphology.

The following conclusions can be drawn from this study:

- Exposure to high-energy radiation in an inert environment promotes cross-linking and significantly enhances abrasive wear resistance of UHMWPE in a conventional hip simulator.
- The *in vitro* wear behavior of radiation-cross-linked UHMWPE is directly related to the large-deformation mechanical properties at the articulating surface.
- Cross-linking does not eliminate plasticity-induced texture in the UHMWPE but rather limits the thickness of the plasticity-induced damage layer that precedes abrasive wear.

Acknowledgments

Special thanks to S. Taylor, R. Windmiller, and A. Kalinowski for technical assistance and for many helpful discussions.

References

1. Wang A, Sun DC, Yau S-S, et al: Orientation softening in the deformation and wear of ultra-high molecular weight polyethylene. *Wear* 203:230, 1997
2. Jasty M, Goetz DD, Bragdon CR, et al: Wear of polyethylene acetabular components in total hip arthroplasty: an analysis of one hundred and twenty-eight components retrieved at autopsy or revision operations. *J Bone Joint Surg Am* 79:349, 1997
3. Kurtz SM, Foulds JR, Jewett CW, et al: Validation of a small punch testing technique to characterize the mechanical behavior of ultra-high molecular weight polyethylene. *Biomaterials* 18:1659, 1997
4. Kurtz SM, Jewett CW, Foulds JR, Edidin AA: A miniature-specimen mechanical testing technique scaled to the articulating surface of polyethylene components for total joint arthroplasty. *J Biomed Mater Res (Appl Biomater)* 1998 48:75, 1999
5. White SE, Paxson RD, Tanner MG, Whiteside LA: Effects of sterilization on wear in total knee arthroplasty. *Clin Orthop* 331:164, 1996
6. Sutula LC, Collier JP, Saum KA, et al: Impact of gamma sterilization on clinical performance of polyethylene in the hip. *Clin Orthop* 319:28, 1995
7. Collier JP, Sperling DK, Currier JH, et al: Impact of gamma sterilization on clinical performance of polyethylene in the knee. *J Arthroplasty* 11:377, 1996
8. Foulds JR, Woytowicz PJ, Parnell TK, Jewett CW: Fracture toughness by small punch testing. *J Testing Eval* 23:3, 1995
9. Butler MF, Donald AM, Ryan AJ: Time resolved simultaneous small- and wide-angle X-ray scattering during polyethylene deformation: II. Cold drawing of linear polyethylene. *Polymer* 39:39, 1998
10. Bellare A, Cohen RE: Morphology of rod stock and compression-moulded sheets of ultra-high-molecular-weight polyethylene used in orthopaedic implants. *Biomaterials* 17:2325, 1996
11. Pruitt L, Bailey L: Factors affecting the near-threshold fatigue behavior of surgical grade ultra high molecular weight polyethylene. *Polymer* 39:1545, 1998
12. Goldman M, Gronsky R, Ranganathan R, Pruitt L: The effects of gamma radiation sterilization and ageing on the structure and morphology of medical grade ultra-high molecular weight polyethylene. *Polymer* 37:2909, 1996
13. Pooley CM, Tabor D: Friction and molecular structure: the behavior of some thermoplastics. *Proc Roy Soc Lond* 329:251, 1972
14. Wang A, Stark C, Dumbleton JH: Role of cyclic plastic deformation in the wear of UHMWPE acetabular cups. *J Biomed Mater Res* 29:619, 1995
15. Wang A, Essner A, Stark C, Dumbleton JH: Comparison of the size and morphology of UHMWPE wear debris produced by a hip joint simulator under serum and water lubricated conditions. *Biomaterials* 17:865, 1996
16. Wang A, Essner A, Polineni VK, et al: Wear mechanisms and wear testing of ultra-high molecular weight polyethylene in total joint replacements. Presented at the Polyethylene Wear in Orthopaedic Implants Workshop, Society for Biomaterials, New Orleans, April 30-May 5, 1997
17. McKellop HA, Campbell P, Park SH, et al: The origin of submicron polyethylene wear debris in total hip arthroplasty. *Clin Orthop* 311:3, 1995

18. Cooper JR, Dowson D, Fisher J: Macroscopic and microscopic wear mechanisms in ultra-high molecular weight polyethylene. *Wear* 162-164:378, 1993
19. Wang A, Stark C, Dumbleton JH: Mechanistic and morphological origins of ultra-high molecular weight polyethylene wear debris in total joint prostheses. *Proc Inst Mech Eng* 210:141, 1996
20. Bragdon CR, O'Connor DO, Lowenstein JD, et al: The importance of multidirectional motion on the wear of polyethylene. *Proc Inst Mech Eng* 210:157, 1996
21. Sauer WL, Anthony ME: Predicting the clinical wear performance of orthopaedic bearing surfaces. In Jacobs JJ, Craig TL (eds): *Alternative bearing surfaces in total joint replacement*. American Society for Testing and Materials; West Conshohocken, PA, 1998
22. McKellop HA, Shen FW, Yu YJ, et al: Effect of sterilization method and other modifications on the wear resistance of UHMWPE acetabular cups. Presented at the Polyethylene Wear in Orthopaedic Implants Workshop, Society for Biomaterials, New Orleans, April 30-May 5, 1997
23. Jasty M, Bragdon CR, O'Connor DO, et al: Marked improvement in the wear resistance of a new form of UHMWPE in a physiologic hip simulator. Presented at the 43rd Annual Meeting of the Orthopaedic Research Society, San Francisco, February 9-13, 1997
24. Wang A, Polineni VK, Essner A, et al: Effect of radiation dosage on the wear of stabilized UHMWPE evaluated by hip and knee joint simulators. Presented at the 23rd Annual Meeting of the Society for Biomaterials, San Francisco, March 18-22, 1997
25. Sawyer LC, Grubb DT: *Polymer microscopy*. Chapman & Hall, New York, 1987
26. Streicher RM: Ionizing irradiation for sterilization and modification of high molecular weight polyethylenes. *Plast Rubber Proc Appl* 10:221, 1988
27. Rose RM, Goldfarb EV, Ellis E, Crugnola AN: Radiation sterilization and the wear rate of polyethylene. *J Orthop Res* 2:393, 1984
28. Rimnac CM, Klein RW, Betts F, Wright TM: Post-irradiation aging of ultra-high molecular weight polyethylene. *J Bone Joint Surg Am* 76:1052, 1994
29. Kurtz SM, Rimnac CM, Bartel DL: Degradation rate of ultra-high molecular weight polyethylene. *J Orthop Res* 15:57, 1997
30. Bostrom MP, Bennett AP, Rimnac CM, Wright TM: The natural history of ultra high molecular weight polyethylene. *Clin Orthop* 309:20, 1994
31. Kurtz SM, Rimnac CM, Santner TJ, Bartel DL: Exponential model for the tensile true stress-strain behavior of as-irradiated and oxidatively degraded ultra high molecular weight polyethylene. *J Orthop Res* 14:755, 1996
32. Greer KW, Schmidt MB, Hamilton JV: The hip simulator wear of gamma-vacuum, gamma-air, and ethylene oxide sterilized UHMWPE following a severe oxidative challenge. Presented at the 44th Annual Meeting of the Orthopaedic Research Society, New Orleans, February 21-24, 1998
33. Muratoglu OK, Cook JL, Jasty M, Harris WH: A novel technique to measure the cross-link density of irradiated UHMWPE. Presented at the 44th Annual Meeting of the Orthopaedic Research Society, New Orleans, February 21-24, 1998
34. Lin L, Argon AS: Review: structure and plastic deformation of polyethylene. *J Mater Sci* 29:294, 1994
35. Bowden PB, Young RJ: Review: deformation mechanisms in crystalline polymers. *J Mater Sci* 9:2034, 1974
36. Keller A, Pope DP: Identification of structural processes in deformation of oriented polyethylene. *J Mater Sci* 6:453, 1971
37. G'Sell C, Dahoun A: Evolution of microstructure in semi-crystalline polymers under large plastic deformations. *Mater Sci Eng* 175:183, 1994
38. Gul R: Improved UHMWPE for use in total joint replacement. Dissertation, Massachusetts Institute of Technology, Boston, September 1997