A Study of the Morphology and Mechanical Properties of Crosslinked UHMWPE

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Introduction

Ultra-high molecular weight polyethylene (UHMWPE) is a widely used bearing material in orthopedic implants. A significant factor affecting the longevity of in vivo use of implant components is the formation of billions of wear particles generated by articulation against metallic or ceramic prosthesis components. Particle debris can elicit a foreign body response leading to bone resorption (osteolysis) and ultimately component loosening necessitating complicated revision surgery.[1]

In vitro joint simulator studies have demonstrated that crosslinking greatly enhances the wear resistance of UHMWPE.[2] However, in accompaniment to improved wear-resistance, there is a reduction in mechanical properties including: fracture toughness, modulus of elasticity, tensile strength, yield strength, hardness, and fatigue crack propagation resistance.[3] Clinically, the degree of crosslinking is of particular concern in high-stress applications, such as in knee joint prostheses where mechanical properties are critical to performance. This study investigated the effects of various degrees of radiation crosslinking on both the morphology of UHMWPE as well as the resistance to crack propagation and crack initiation using the model of Rice and Sorensen.[4]

Materials and Methods

Commercially available, ram extruded GUR 1050 (Ticona, Bayport, TX) rod-stock (PolyHi Solidur, Ft. Wayne, IN) was used for all experiments. Rod-stock, which was 8 cm in diameter, was machined into 30 cm segments and gamma-irradiated at doses of 25, 50, 100, and 200 kGy (Isomedix, Northboro, MA). Following radiation, the material was heated to 170˚C (well above the melting temperature of 138˚C measured by DSC), held at 170˚C for 4 h, subsequently annealed at 125˚C for 48 h, and finally slowly cooled to ambient temperatures. As a control, unirradiated rod-stock was subjected to an identical thermal history. USAXS was performed on 2-mm-thick specimens at the UNI-CAT beamline of the Advanced Photon Source (APS) using 10 keV x-rays. The beam had a cross sectional area of 2 mm × 0.6 mm. The degree of crystallinity was determined by differential scanning calorimetry (DSC) on a Perkin Elmer Pyris 1 instrument. Percent crystallinity was calculated by normalizing the heat of fusion of each sample to the heat of fusion of polyethylene crystal (293 J/g). Resistance to crack propagation (J_{ss}) and resistance to crack initiation (J_{1c}) were measured using compact tension (CT) and tensile tests.

Results

The long period, or inter-lamellar spacing, for all UHMWPE samples was determined using paired distance distribution functions (PDDF) or p(r) which were identical to the one dimensional correlation function for lamellar systems. The scattering functions were converted to PDDFs using the computer program ITP developed by Glatter.[5] PDDF are related to the scattering function I(q) by the equation:

$$p(r) = (1/2\pi^2 A)\int_0^\infty q^2 I(q)\cos(qr)dq \qquad (1)$$

The USAXS long period was measured from the first maximum of the PDDF (Fig. 1).

Interlamellar spacing and DSC crystallinity values were used

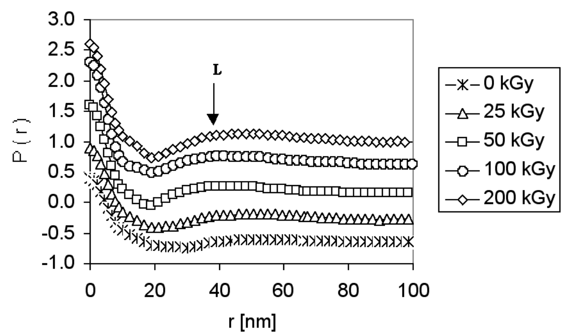

FIG. 1. Paired distance distribution functions p(r). Values of p(r) have been offset for clarity.

to determine the lamellar thickness using the following equation:

$$D = X_c L \qquad (2)$$

where D is the lamellar thickness, X_c is the degree of crystallinity (%) measured by DSC, and L is the inter-lamellar spacing. The thickness of the amorphous region (A) was calculated by taking the difference between the inter-lamellar spacing (L) and the lamellar thickness (D). Table 1 shows the morphological features revealed using USAXS and DSC.

Table 1. Characterization of the crystalline morphology of crosslinked UHMWPE.

Dose [kGy]	X_c [%] (DSC)	L [nm] (USAXS)	D [nm]	A [nm]
0	52.7	49.8	26.2	23.6
25	42.2	49.8	21.0	28.8
50	46.3	40.0	18.5	21.5
100	46.2	40.6	18.8	21.8
200	42.9	48.6	20.8	27.8

The resistance to crack propagation (J_{ss}), was calculated from CT fracture tests using the Rice and Sorensen model[4] and the following equation:

$$J = \alpha\varepsilon_0\sigma_0 c h_1(a/b, n)\,(P/P_0)^m \qquad (3)$$

where a = crack length measured from pin-line
b = pin-line to free end of specimen distance
c = b-a
$P_0 = 1.455\eta c\sigma_0$
$\eta = [(2a/c)^2 + 2\,(2a/c) + 2]^{1/2} - (2a/c + 1)$
$\varepsilon_e = \alpha\varepsilon_0(\sigma_e a/\sigma_0)^n$

where ε_e is the power-law fit, strain-hardening expression that relates the stress σ_e and ε_e with the fitting constants $\alpha\varepsilon_0$ and σ_0 (tensile yield stress). The expression $h_1(a/b,\mathrm{n})$ is a tabulated function of a/b and n, where n is the strain-hardening exponent (1/m). P is the maximum load per unit specimen thickness.

Resistance to crack initiation (J_{1c}) was calculated using the following equation:

$$\frac{J_{ss}}{J_{1c}} = \frac{\alpha}{\lambda e}\frac{\sigma_0}{E}\exp\left(\frac{E}{\sigma_0}\frac{1}{\beta}\theta^{p}\right) \qquad (4)$$

where E = Tensile Modulus
σ_0 = Tensile Yield Stress
$\alpha = 0.50$
$\beta = 3.9$
$\lambda = 0.160$

The values of α, β, and λ are theoretical constants [4], e is the basis of the natural logarithm, and θ^p is the crack opening angle at steady state.

Dose [kGy]	J_{ss} [kJm^{-2}]	J_{1c} [kJm^{-2}]
0	116.9 ± 0.1	2.1
25	101.2 ± 0.1	23.8
50	98.5 ± 0.2	76.2
100	87.6 ± 0.1	= J_{ss}
200	79.3 ±1 .9	= J_{ss}

Table 2. Resistance to crack propagation (J_{ss}) and resistance to crack initiation (J_{1c}).

Discussion

The data showed no apparent relationship between the interlamellar spacing (long period) and the dose level of gamma radiation of the crosslinked samples. While the reduction in the long period was not consistent with increasing degree of crosslinking, the smaller long period of the 50 and 100 kGy UHMWPE samples as compared to other doses indicated a morphology comprised of smaller lamellae. The post gamma-irradiation isothermal crystalization produced smaller, less perfect crystals.

While the resistance to crack propagation of UHMWPE decreased upon crosslinking, this property was accompanied by a larger increase in the resistance to crack initiation. Resistance to crack initiation and particulate wear are important properties for knee prostheses. A low degree of crosslinking may be beneficial in the fabrication of knee prostheses since it greatly increases the resistance to crack initiation with only a small reduction in resistance to crack propagation. In conclusion, higher dosages of crosslinked UHMWPE may prove to be more appropriate in applications where applied stresses are lower, such as in the hip joint prosthesis.

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