

Fracture and Fatigue of UHMWPE: From Microstructure to Mechanical Behavior

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Introduction

For decades, ultra-high molecular weight polyethylene (UHMWPE) has been a popular material of choice for total joint replacements (1). Common UHMWPE failures, such as delamination and following osteolysis, are predominantly caused by wear in areas that often experience cyclic contact stresses or stress concentrations, at the knee or the acetabulum (2). To remedy this problem, most manufacturing processes today implement cross-linking accompanied by thermal treatment. However, these processes negatively impact the fatigue and fracture properties, which are significant elements in biomechanics. Our study reviews the basic fracture and fatigue test methods for UHMWPE. We shall also look into the relationship between the microstructure and the mechanical behavior of UHMWPE.

Fracture Test in UHMWPE

In elastic-plastic fracture mechanics, the J-integral is widely used. J-integral is a path-independent value of a closed contour integral around the crack tip, and it represents how much total energy should be consumed to extend the crack during a monotonic loading condition (3). There are several methods to evaluate J-integral-based fracture toughness, such as multi- and single-specimen methods (4). UHMWPE specimens should be precracked first in order to have the sharpest preexisting crack. The specimens are later loaded statically to a specific displacement to initiate the crack extension. Optical methods and elastic compliance techniques are implemented to measure the crack length for multi- and single-specimen methods, respectively (4). More details are described in ASTM E813, the precursor of ASTM E 1820. However, such methods have obvious drawbacks because they cannot be accurate when materials have viscoelastic properties (4). As a result, the normalization method is proposed, which assumes the load as the product of separable functions of crack length and displacement (4). More details can also be found in ASTM E 1820.

The multi- and single-specimen methods were originally designed for metallic materials, albeit they were also applied to polymers (4). So far there is only one method that is specifically designed for polymers, which is described in ASTM D6068. Compact specimens precracked with three-point bending are prepared first, loaded to a specific displacement and broken to expose the fracture surface. Optical measurements are further taken to obtain the five-point averaged crack lengths. Furthermore, at least seven data points are required to build a J-R curve, which can be applied to different types of polymers including UHMWPE (4).

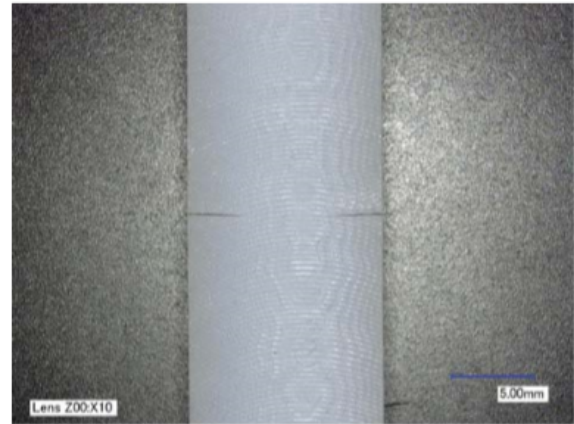


Figure 1. Notched sample prepared for Izod test (8).

There have been a multitude of variants of the J-integral method applied to UHMWPE (4). In 1988, Rimnac et al. evaluated the static fracture behavior of extruded UHMWPE. Details of the test were described in ASTM E813-87. It was found that J_{IC} was 99.5kJ/m² (5,6). Pascaud et al. used the same multi-specimen method to obtain the value of J_{IC} with five analytical methods: 1) power law fit, intersection with 0.2mm offset blunting line; 2) power law fit, intersection with 0.2mm physical crack growth; 3) linear fit, intersection with blunting line; 4) linear fit, intersection with 0.2mm physical crack growth; and 5) linear fit, intersection with $\Delta a = 0$. The conclusion stated that ASTM E813-89 overestimated J_{IC} , and the J-controlled crack growth was found to underestimate J_{IC} (6). With a modified analytical model, the value of J_{IC} was determined to be 66.5kJ/m². Both Rimnac and Pascaud found the specimen thickness had no influence on the value of J_{IC} . So far the roles of strain hardening, rupture etc. during the crack growth process are still unclear (6). As a result, there are still a lot to learn in the field of UHMWPE's failure mechanisms, and many contemporary methods like nonuniform crack initiation appeal promising in solving these problems (7).

Another method to characterize the fracture property of UHMWPE is the Izod test that reflects the dynamic scenario for the fracture resistance, in which deformation and rupture processes happen in a much shorter time scale (4). Here is a brief description per ASTM F648: rectangular prisms are machined first and then samples are notched by a razor blade. Fig. 1 indicates a notched sample (8). The UHMWPE samples should later be impacted by the Izod impact machine. The absorbed energy is measured, corrected for friction and windage, and divided by un-notched cross-sectional area of the sample (8).

Fatigue Test of UHMWPE

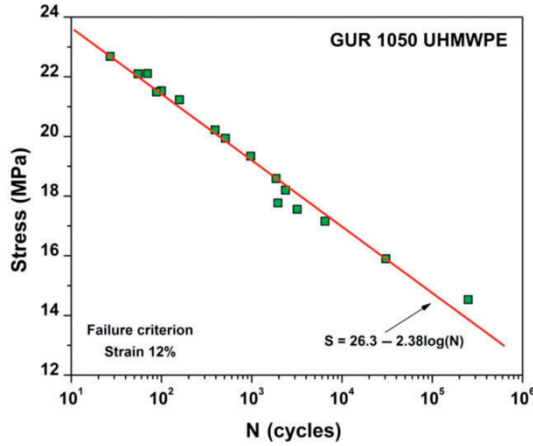


Figure 2. S-N (stress - number of cycles to failure) plot for compression molded GUR 1050 UHMWPE (4).

From a structural point of view, fatigue refers to damage and failure of the material under cyclic loading, where its peak values are considerably smaller than the “safe” loads estimated on the basis of static fracture analyses (9). The fatigue damage process is thought to initiate microstructural changes that lead to the nucleation of microcracks. Furthermore, the microcracks grow and coalesce to form a dominant crack that propagates in a stable manner until the complete structural fracture is reached (4,9). Usually, there are two fatigue evaluation methods: Total-life philosophy and Defect-tolerance philosophy. These approaches could be employed to

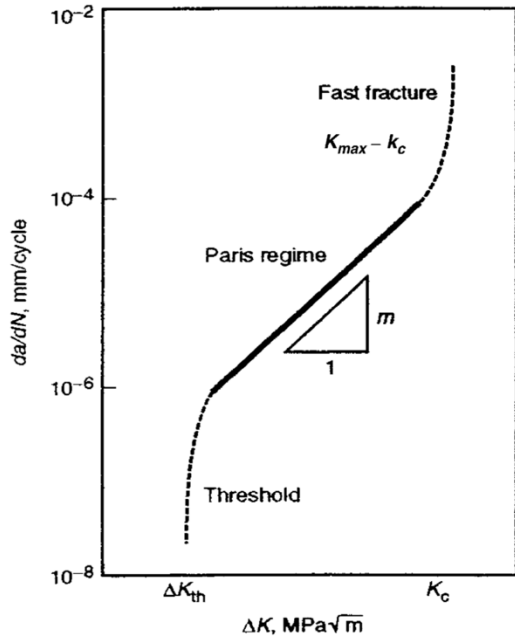


Figure 3. Schematic of fatigue crack propagation plot showing near-threshold regime, Paris regime, and fast-fracture regime (1).

determine the crack nucleation and crack propagation processes, respectively (10).

To predict the total life of a component under fatigue loading, simple experiments are typically run on unnotched, nominally smooth specimens of the component material, where the time to failure (or number of load cycles) is recorded as a function of the applied stress (4). Collected data is typically manifested in a S-N curve (Fig. 2). In total-life philosophy, custom-defined waveform parameters include minimum stress σ_{\min} , maximum stress σ_{\max} , stress range $\Delta\sigma = \sigma_{\max} - \sigma_{\min}$, stress amplitude $\sigma_a = \frac{1}{2}(\sigma_{\max} - \sigma_{\min})$, mean stress $\sigma_m = \frac{1}{2}(\sigma_{\max} + \sigma_{\min})$, stress ratio $R = \frac{\sigma_{\min}}{\sigma_{\max}}$, shape of waveform (sine or square) and frequency (4). For UHMWPE, the total-life philosophy is not widely practiced due to poor understanding of flaw nucleation in the polymer (11,12). In fact, there is only one study that adopts the traditional total-life method (13).

By contrast, the defect-tolerance philosophy is based on the implicit assumption that structural components are intrinsically flawed, and the fatigue life is based on the propagation of an initial flaw to a critical size (1). The plot that describes the defect-tolerance method comprises the velocity of crack propagation $\frac{da}{dN}$, and the stress intensity factor range $\Delta K = K_{\max} - K_{\min}$, where a is the crack length, N is the number of cycles, K_{\max} and K_{\min} are maximum and minimum stress intensity factor based on applied loading, sample geometry and instantaneous crack length (Fig. 3) (1,9). Empirically speaking, three regimes can be distinguished from FCP (fatigue crack propagation): Regime I, where the crack stays inactive ($\Delta K < \Delta K_{TH}$) (ΔK_{TH} refers to atomically sharp pre-crack by fatigue technique); Regime II, where there is a stable crack propagation ($\Delta K > \Delta K_{TH}$, while $K < K_c$); Regime III, where the growth of crack becomes unstable ($K > K_c$) (4).

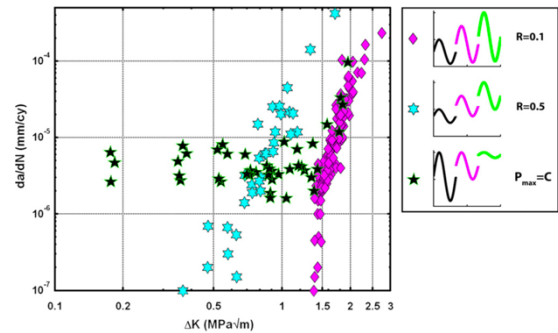


Figure 4. FCP data from $R = 0.1$, $R = 0.5$, P_{\max} constant experiment. Qualitative disagreement between these data over full range on stress intensity factor implies that ΔK is not a sufficient parameter to predict crack propagation behavior (14).

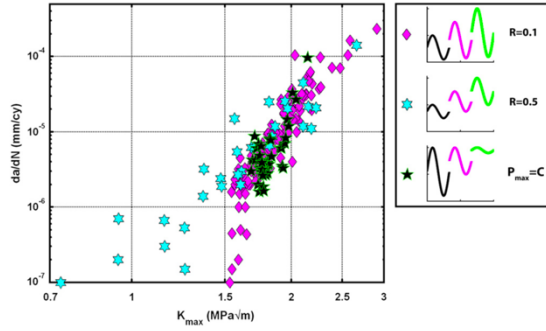


Figure 5. Data in Fig.4 plotted with K_{\max} as the presumed FCP parameter. All data in the stable crack propagation regime collapse to the same curve, confirming K_{\max} as the FCP parameter for this regime of crack growth (14).

For UHMWPE, the rate of stable crack propagation (Regime II) fits a power law relationship with ΔK , also known as the classic Paris equation (Eq. 1) (14).

$$\frac{da}{dN} = C(\Delta K)^m$$

Equation 1. Paris Equation

On the other hand, due to the viscoelastic nature of UHMWPE, the crack growth can also depend on time and K_{\max} (Eq. 2) (14).

$$\frac{da}{dt} = Q(K_{\max})^n$$

Equation 2. Creep Law for UHMWPE

In reality, these two equations can be additive or coupled (Eq. 3 and Eq. 4) (15–17).

$$da = \left[\frac{\partial a}{\partial N} \right]_t dN + \left[\frac{\partial a}{\partial t} \right]_N dt$$

Equation 3. Additive Method

$$\frac{da}{dN} = C(K_{\max})^n(\Delta K)^m$$

Equation 4. Coupled Method

However, Furmanski and Pruitt demonstrated that K_{\max} turned out to be the dominant FCP parameter (14). In that study, loadings with $R = 0.1$, $R = 0.5$ and $K_{\max} = \text{constant}$ were applied, and the rate of crack propagation was distinct against ΔK but collapsed against K_{\max} (Fig. 4 and Fig. 5).

Factors of Fatigue and Fracture in UHMWPE

Prior to the discussion of factors that influence the fatigue and fracture properties of UHMWPE, it is important to explain the purpose of radiation accompanied by thermal treatment, which are the common manufacturing processes in today's UHMWPE applications. Radiation induced cross-linking increases abrasion resistance but creates free radicals that are later

the culprit of oxidation; thermal treatment, such as annealing and remelting, reduces the free radical and prevents the material from oxidation (2). Cross-linking is defined as a process by which doses of gamma or electron beam radiation are applied in order to restrict chain mobility in the amorphous phase of the semicrystalline polymer (18). By reducing the chain mobility, UHMWPE hinders large plastic deformation and prevents the formation of surface fibrils, which is the root cause of wear debris (2). However, radiation also forms carbon free radicals, which over time steal hydrogen atoms from polyethylene chains and become oxidation products (2). To prevent deleterious effects such as chain scission and embrittlement at the crack tip, thermal treatment helps reduce the free radical content (18–20). There are two types of thermal treatments: annealing, which operates below UHMWPE's melting temperature of 135°C, and remelting, which operates above 135 °C (21). Although annealing does not completely annihilate all free radicals and faces the potential of oxidation in the long term, annealing could be preferable when considering its fatigue performance (18,21).

UHMWPE's resistance to fatigue crack is governed by microstructural features such as cross-linking, crystallinity percentage, and lamellae size—all of which are influenced by irradiation and thermal treatment (1,10,19,21). Per ASTM E647, Gencur et al. discovered that an increase in cross-linking density diminished the material's FCP resistance, as portrayed by Fig. 6 (22).

The decrease in FCP resistance was due to the limited chain mobility caused by cross-linking (2,22). With a greater radiation dose, chains in the amorphous region become more restricted, and prevalent sharper crack tips promote brittle crack advancement (10,19,22). In regards to fatigue crack initiation, tested data has

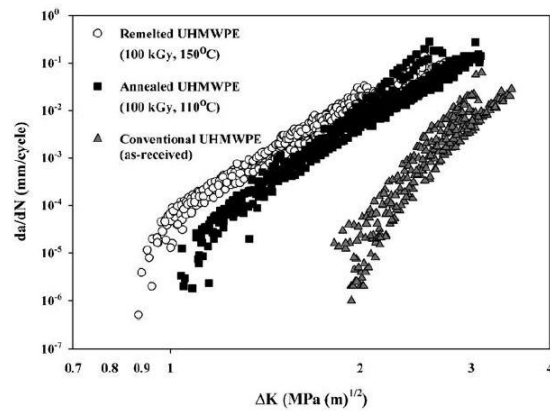


Figure 6. Cyclic stress intensity (ΔK) versus crack growth per cycle (da/dN) for cross-linked remelted, annealed, and untreated UHMWPE. 100 kGy of radiation is considered 'highly cross-linked' (>40 kGy) (22).

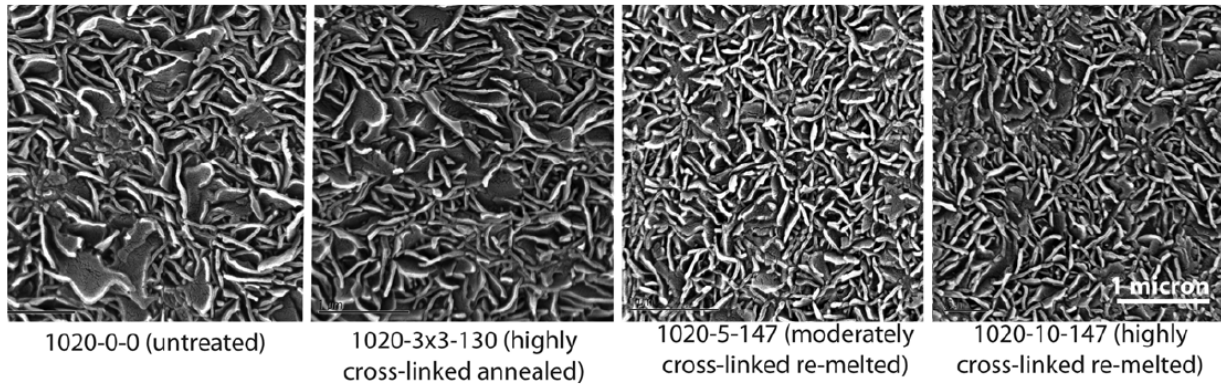


Figure 7. Lamellae size of four material groups. The annealed specimen contains thicker lamellae than the remelted one (21).

shown a 30% - 50% drop in $\Delta K_{\text{inception}}$ (which refers to non-atomically sharp pre-crack by razor blade), which corresponds to the cyclic stress intensity required to produce a crack growth rate of 10^{-6} mm/cycle (22). Comparing cross-linked specimens that later underwent annealing versus remelting, the annealed specimen demonstrated a higher $\Delta K_{\text{inception}}$ than the remelted counterpart (22). This is not to say, however, that cross-linked and annealed UHMWPE is the ideal choice. One must consider the tradeoff between the elements of oxidation and fatigue, which both affect the lifespan of the material *in vivo*.

Crystallinity and lamellae size are properties mainly affected by the type of heat treatment; annealing results in a greater crystallinity percentage and lamellae thickness than remelting (2,19,21). The scanning electron micrographs in Fig. 7 demonstrates this difference: the annealed sample has a bigger lamellar structure than for the remelted case (21). A greater crystallinity percentage improves the FCP resistance, and a thicker lamellae size is capable of blunting and deflecting the crack tip, which reduces the rate of crack advancement and lowers the crack advancement driving force, respectively (19,21).

As for impact toughness, it decreases with an increase of radiation dose because of the large loss in ductility (23).

In summary, it is of importance to take into account all the benefits and costs of radiation and thermal treatment. The use of radiation increases UHMWPE's cross-link density and improves wear resistance. The practice of thermal treatment eliminates free radicals, which are the byproducts of cross-linking, and in result reduces the possibility of oxidation. However, the amount of radiation dosage negatively influences the polymer's fatigue crack resistance, promoting crack-tip sharpening and brittle fracture (19). While annealing yields a greater crystallinity percentage and lamellae thickness, which helps with FCP resistance, it is more susceptible to oxidation than the remelting process. This tradeoff involving fatigue crack resistance, wear

resistance, and oxidation resistance hence represents the shortcomings of UHMWPE applications.

Conclusion

Our study briefly reviewed various methods to test fracture and fatigue properties of the UHMWPE, and also looked into the relationship between manufacture processes (gamma irradiation and heat treatment) and microstructural properties. Furthermore, we summarized the influence of microstructural features on the mechanical behaviors of UHMWPE. It was discovered that radiation and thermal treatment, two essential steps of UHMWPE processing, produce a negative impact on the fatigue performance. The radiation dosage had a negative correlation with FCP resistance. Annealing improved the overall crystallinity and lamellae thickness, whereas remelting did not. On the other hand, remelting possessed a better ability to prevent the oxidation of free radicals. Therefore, the design and manufacturing of UHMWPE should take into account the elements of wear, fatigue, and oxidation.

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