Fatigue characteristics of ultra high molecular weight polyethylene with different molecular weight for implant material

MITSUO NIINOMI[1], LEI WANG[2], TAKUMI ENJITSU[3]*, KEI-ICHI FUKUNAGA[1]
[1]*Department of Production Systems Engineering, Toyohashi University of Technology, 1-1, Hibarigaoka, Tempaku-cho, Toyohashi 441-8580, Japan*
[2]*Department of Mechanical Engineering, Tottori University, Koyama 4-101, Tottori 680-8552, Japan*
[3]*Graduate Student of Toyohashi University of Technology*
E-mail lwang@mech.tottori.u.ac.jp

In order to investigate the effect of molecular weight on fatigue characteristics in the ultra high molecular weight polyethylene (UHMWPE), tension–tension fatigue tests of notched specimens were carried out in the present study. The effects of frequency and stress ratio on the fatigue characteristics were also investigated and fractography was discussed.

The fatigue strength does not increase with increasing molecular weight. The fatigue strength might be influenced by the high degree of crystallinity in spite of the decreased tie molecule density in this study. Almost no effect of frequency on the number of cycles to failure can be observed. However, the higher the frequency, the higher the crack tip temperature. The effects of heat and strain rate on the fatigue strength must be considered in polymer materials. At a high stress ratio, the stress–number of cycles to failure (S–N) curves shift to high number cycles to failure side. Both stress amplitude and mean stress influence the fatigue life of UHMWPE.
© 2001 Kluwer Academic Publishers

1. Introduction

Ultra high molecular weight polyethylene (UHMWPE) has been used as a biomaterial for replacing failed hard tissue because of its greater impact resistance, greater wear resistance and superior biocompatibility. For example, in the most advanced structure of artificial hip joint, UHMWPE is used for the socket contacted with the ceramic bone head composed of alumina or zirconia attached to the stem made of titanium alloys. Those parts are usually operated under the cyclic loading due to walking for a long time. Understanding fatigue characteristics of UHMWPE is, therefore, very important to prolong the life of the instrumentations like artificial hip joints used in the body. Since polyethylene is composed of crystal and amorphous parts, its fatigue characteristics are influenced by various factors, and then the fatigue life prediction is very difficult. Moreover, the fatigue problems are more complex because of the visco-elasticity.

Many reports on the fatigue characteristics of the polyethylene have been appeared up to now. According to those reports [1–5], molecular weight, the degree of crystallization or tie molecule concentration has been found to influence on the fatigue characteristics of polyethylene by those reports [1–5]. These findings are limited to polyethylene. There are just a few reports on the fatigue characteristics of UHMWPE.

Fatigue tests were performed on UHMWPE with various molecular weights in order to investigate the fatigue characteristics relating to molecular weight in this study. The fatigue characteristics of UHMWPE were also investigated when the loading frequency and stress ratio were varied.

2. Experimental procedures
2.1. Materials
The materials used in the present study are UHMWPE plates with a thickness of 10 mm fabricated by press forming. The molecule weight, density and average particle size of powder before press forming are listed in Table I. Each material will be described as PE575, PE360, PE230, PE210 and PE200, respectively in order of the largeness of molecular weight in this study. The number in each one indicates the molecular weight.

Among the testing materials, PE210 was fabricated using a slightly smaller powder with an average diameter of 130 μm and was a relatively narrow particle size distribution. PE200 was fabricated using much smaller powders with an average diameter of 30 μm.

Sample	Average molecular weight (g/mol)	Density (g/cm³)	Average-particle size (μm)
PE575	575×10^4	0.929	160
PE360	360×10^4	0.934	150
PE230	230×10^4	0.935	160
PE210	210×10^4	0.937	130
PE200	200×10^4	0.940	30

T A B L E I I Tensile properties, crystallinity and melting temperature of samples

Sample	Yield strength (σ_y/MPa)	Tensile strength (σ_F/MPa)	Elongation (E_l/%)	Degree of crystallinity (W_c/%)	Melting temperature (T_m/K)
PE575	22.7	43.3	374	48.5	410.0
PE360	23.0	41.1	431	50.3	410.6
PE230	21.2	34.0	511	51.1	398.5
PE210	24.0	40.4	504	52.9	410.7
PE200	23.1	40.6	506	53.2	411.1

2.2. Measurement of degree of crystallinity

The degree of crystallinity of the testing materials was measured using a differential scanning calorimeter (DSC). The sample was cut from the plate with a weight of 5 mg, and measured within Ar gas atmosphere up to 453 K at a heating rate of 10 K/min. The fusion heat (Δ Hm) was calculated from the endothermic curve. Then the degree of the crystallinity, Wc (%), was calculated using the following equation:

$$\text{Wc } (\%) = 100 \times \Delta Hm/\Delta H \qquad (1)$$

where ΔH is the fusion heat needed for complete crystallization of the polyethylene. 293 J/g [2] was adopted as ΔH in the present study.

2.3. Tensile tests

Tensile specimens without a notch whose geometry is shown in Fig. 1, were machined from the plate. The tensile tests were carried out using an Instron type testing machine at a cross head speed of 8.3×10^{-4} m/s.

2.4. Fatigue tests

The specimens with a razor notch whose depth is 1 mm were used for fatigue tests. The geometry of the specimen for fatigue tests is same as that for tensile tests as shown in Fig. 1. Fatigue tests were performed in air at a temperature of 296 ± 1 K with a sine wave under a constant loading condition using a electro-servo-hydraulic testing system. The stress ratio (R) and the frequency (f) were in general 0.1 and 1 Hz, respectively.

In order to investigate the effects of the stress ratio and the frequency on the fatigue behavior, the tests were also done with $R = 0.5$, and $f = 3$ and 5 Hz.
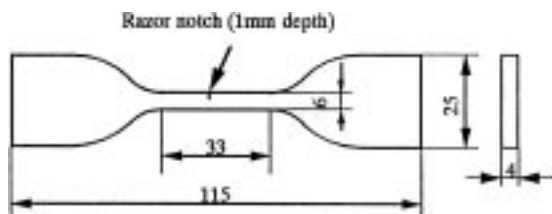
2.5. Fracture surface observation

The fracture surfaces were coated with Platinum, and were then characterized using a scanning electron microscope (SEM).

3. Experimental results and discussions
3.1. Tensile characteristics

The tensile properties, degree of crystallinity and melting temperature of the materials are shown in Table II. The tensile strength and melting temperature of PE320 are a little lower than those of the other materials. The other materials except PE320 have nearly the same tensile strength and melting temperature. However, the elongation of the materials decreases with increasing the molecular weight. Meanwhile, the degree of crystallinity of the materials also decreases with increasing the molecular weight.

The typical load-deflection curve obtained in a tensile test on each material is shown in Fig. 2. There are some differences in the shape of load-deflection curves. The slope of the curve after the yield point increases with increasing molecular weight. In other words, the work hardening ratio of the material increases with increasing the molecular weight. This may be caused by increasing the tangling of molecular chains with increasing molecular weight of the material.

3.2. Fatigue strength

The S–N curve of each material is shown in Fig. 3. Among the tested materials, PE200 and PE210 are superior to the other materials in fatigue strength. In general, the larger the molecular weight, the greater the fatigue strength [1]. However, it does not hold in the present materials. For instance the degree of crystallinity or, the size or size distribution of powder at press forming
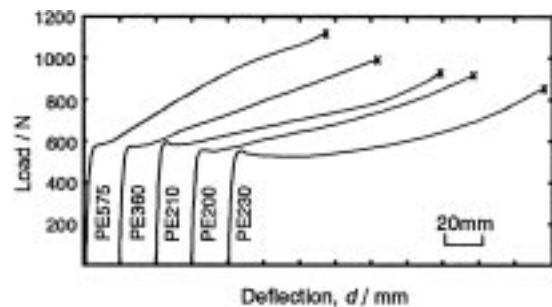

Figure 1 Geometry of tensile (without notch) and fatigue (with notch) specimen.

Figure 2 Typical load–deflection curve of testing polyethylene materials.
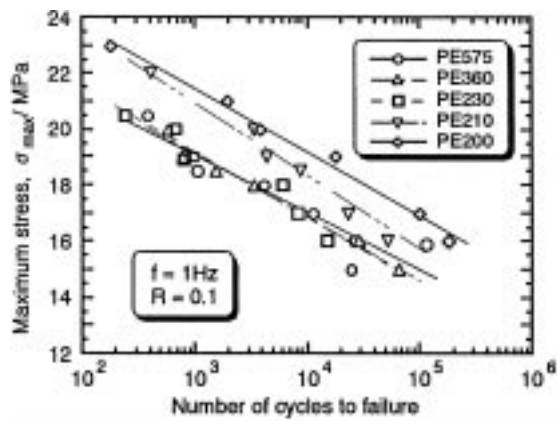
Figure 3 Relationship between maximum stress, σ_{max}, and number of cycles to failure.

will influence the fatigue behavior of the present materials.

There are many reports on the relationship between the degree of crystallinity and fatigue strength of high molecular polyethylene [2, 4]. However, some have reported that the greater degree of crystallinity brings greater fatigue strength [4] while others have reported that the smaller degree of crystallinity brings smaller fatigue strength [2, 3].

Yen *et al*. [6] have reported that the fatigue strength is related to tie molecule chains. The schematic drawing of tie molecule chains is shown in Fig. 4. The tie molecule chains form in the non-crystalline part, and connect the crystalline parts, which are related to both the fracture of crystalline part and the strength of non-crystalline parts. However, it is very difficult to measure the tie molecule density, Runt [2] and Strebel [3] have reported that the fatigue strength increased with the increase of the number of tie molecule chain. Brown and Ward [7] have been, however, successfully calculated the tie molecule chain density (f_T) with the brittle fracture stress (σ_B) at low temperature by the following equation:

$$f_T = (C\sigma_B - \beta E_{ISO})/\beta(E_T - E_{ISO}) \qquad (2)$$

where E_T is the Young's modulus of the tie molecular, E_{ISO} is the Young's modulus in van der Waals bonding, β is the constant, and C is the stress concentration factor. For the present materials, $E_T = 300$ MPa, $E_{ISO} = 8$ GPa, $\beta = 0.1$, and $C = 20$ [7]. In order to calculate the tie molecule chain density, tensile tests were also carried out in liquid N_2 (77 K) in the present study. The brittle fracture stress, σ_B, obtained from the tensile tests in liquid N_2 is shown in Table III. The tie molecule chain density, (f_T) calculated by the Equation (2) is also shown
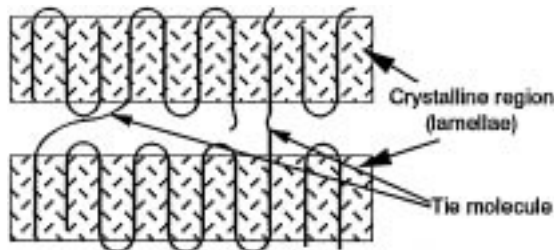

Figure 4 Schematic drawing of tie molecule.

T A B L E   I I I Brittle fracture stress σ_B and tie molecule density f_T of the testing materials

Sample	PE575	PE360	PE230	PE210	PE200
σ_B/MPa	157	157	160	143	146
f_T	0.080	0.080	0.082	0.071	0.073

in the same table. The brittle fracture strength (σ_B) is 3.5–4.7 times greater than the tensile strength, (σ_F) at room temperature. σ_B of PE200 and PE210 are a little smaller than those of others. The tie molecule chain densities of PE200 and PE210 are a little smaller than those of others.

The relationship between the molecular weight and the degree of crystallinity or the tie molecule chain density is shown in Fig. 5. The degree of the crystallinity decreases with increasing the molecular weight. The degree of the crystallinity of PE200 and PE210 is, in particular, much greater than those of others. However, the tie molecular chain densities of PE200 and PE210 are much smaller than those of others. The fatigue strength of the present materials is, therefore, strongly influenced by increasing the degree of the crystallinity rather than by decreasing the tie molecular chain density.

3.3. Effect of frequency on fatigue strength

The *S–N* curves of the present materials obtained at $f = 3$ and $f = 5$ Hz are shown in Fig. 6(a) and (b), respectively. The *S–N* curves obtained at $f = 1$ Hz are also shown in each figure for the comparison. The fatigue strength obtained at $f = 5$ Hz looks a little greater than that obtained at $f = 3$ Hz. However, the difference between them is very small.

The fatigue strength of the polymer, in general, is influenced by the frequency because the temperature of the specimen is raised by the cyclic deformation due to its lower heat conductivity [8], and the fatigue behavior of polymer is also influenced very much by the strain rate due to its visco-elasticity [7]. Bucknall and Dumpleton
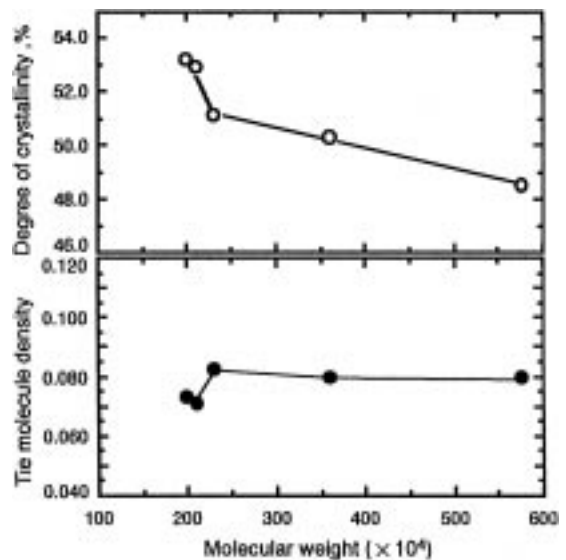

Figure 5 Degree of crystallinity and tie molecule density as a function of molecular weight.
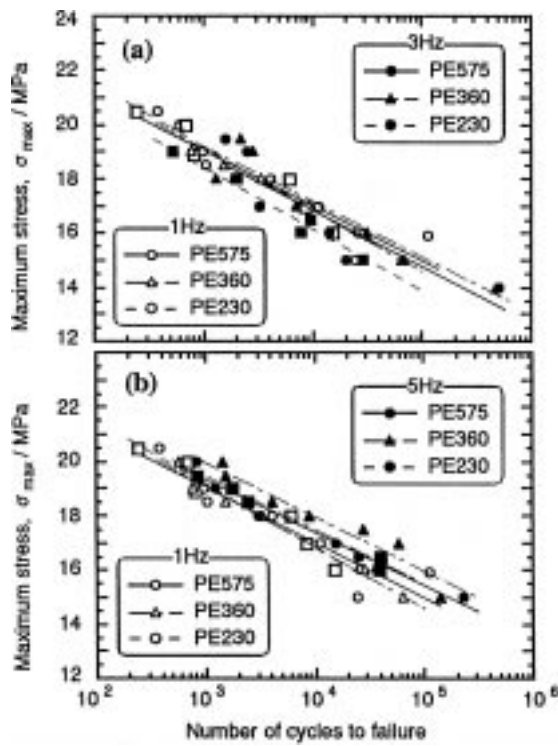
269

Figure 6 Effect of frequency on *S–N* curve. (a) $f = 3$ and $1\,Hz$, $R = 0.1$; (b) $f = 5$ and $1\,Hz$, $R = 0.1$.

[9] have reported that the number of cycles to failure increases with the frequency (f) between 0.5 and 2 Hz in the fatigue tests of the high-density polyethylene. The frequency dependency on the number of cycles to failure is not recognized in the present materials as stated above.

3.4. Effect of increase in temperature

Sauer *et al*. [10] have pointed out that the effect of temperature on the mechanical properties of the polyethylene is strong. Therefore, the temperature change in the specimen during fatigue tests was also investigated in the present study. One thermoelectric-couple was put near the notch of the specimen surface of PE360, and the temperature was recorded continuously. At the same time, the testing environment temperature was controled to be constant at $293 \pm 0.5\,K$. Typical results of the measurement of temperature change near
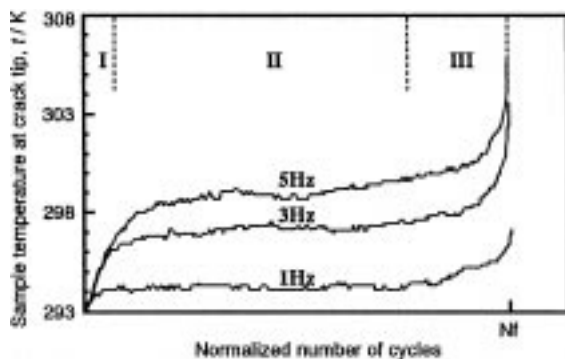
the crack tip of the specimen surface versus normalized number of cycles obtained in PE360 at a constant maximum stress of 18 MPa are shown in Fig. 7. The temperature near the crack tip of the specimen surface clearly increases during the fatigue tests, and also clearly increases with increasing frequency. The temperature change can be divided into three stages as marked in the figure with I, II and III. The temperature increases rapidly at the first stage, I, increases steadily at the second stage, II, and increases rapidly again at the third stage, III. The temperature rises, for example, about $1\,K$ at $f = 1\,Hz$, about $4\,K$ at $f = 4\,Hz$, and about $6\,K$ at $f = 5\,Hz$. In fact, the temperature at the crack tip of the specimen interior will rise much greater than that on the specimen surface. From this result, it can be suggested that the number of cycles to failure will increase with increasing strain rate due to the increase in frequency while the number of cycles to failure will be decreased by heating due to the heat accumulation with increasing frequency. These two effects will cancel each other, and the effect of frequency on fatigue strength will not be clearly recognized in this study. Anyhow, the fatigue tests at high frequency are necessary to be carried out carefully because the temperature rising of the specimen is remarkable. The effects of stress state at the artificially introduced pre-crack tip and the visco-elasticity on the crack tip blunting is necessary to be taken into account in analyzing the results of fatigue tests with the effect of frequency and temperature. However, further study is needed to solve these effects.

3.5. Effects of stress ratio on the fatigue behavior

The relationship between maximum stress and number of cycles to failure with $f = 3\,Hz$ at $R = -0.5$ and $R = 0.1$ is shown in Fig. 8. And the relationship between stress amplitude and the number of cycles to failure is shown in Fig. 9. The fatigue life of each material is longer at $R = 0.5$ than at $R = 0.1$. This trend is clearly seen in PE575 and PE360. The difference in the fatigue life among PE575, PE360 and PE230 can be also defined at $R = 0.5$ although that can not be defined at $R = 0.1$. The fatigue life of each material may be considerably influenced by the average stress because the fatigue life of each material is shorter at $R = 0.5$ where the stress
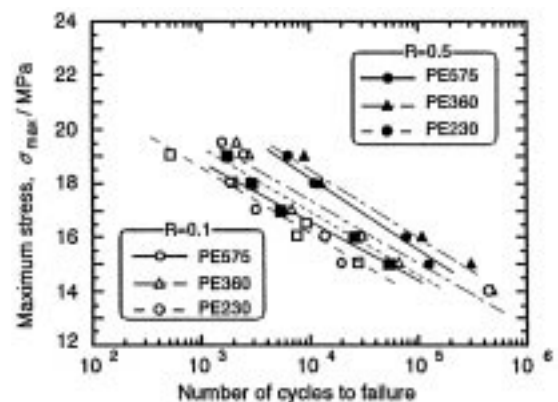

Figure 7 Effect of frequency on crack tip temperature as a function of normalized number of cycles at a maximum stress of 18 MPa.

Figure 8 Relationship between maximum stress, σ_{max}, and number of cycles to failure with the $f = 3\,Hz$ at $R = 0.5$ and $R = 0.1$.
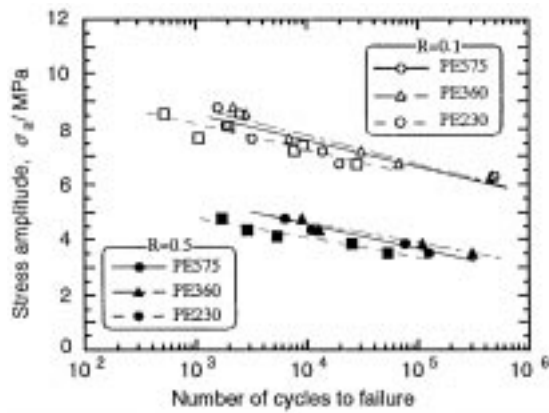
Figure 9 Relationship between stress amplitude, σ_a, and number of cycles to failure with the $f = 3$ Hz at $R = 0.1$ and $R = 0.5$.

Figure 11 Tensile fracture mechanism of ultra high molecular weight polythylene.

amplitude is relatively smaller, than at $R = 0.1$. The effect of tensile strength shown in Table II will appear on the fatigue life of each material when the R ratio is relatively greater. The effect of heating will be decreased when the R ratio is greater, which coincide to smaller stress amplitude. Assuming linear visco-elasticity, the amount of heat generation per unit time due to the visco-elastic loss energy, W, is described by the following equation [11]:

$$W = \pi f C'(f, T)\sigma_a^2 \qquad (3)$$

where C' is the loss compliance, T is the testing temperature, f is the frequency, and σ_a is the average stress amplitude. It is clear from the above equation that W is increases proportionally to the testing frequency (f) and the square root of average stress amplitude, σ_a. The heat generation, that is, temperature rising of the specimen can be understood to have great possibility to affect greatly on the fatigue life of the UHMWPE.

3.6. Fracture behavior

The typical SEM fractograph of PE210 after tensile test is shown in Fig. 10. It can be seen from the SEM fractograph (a) taken at low magnification that the crack initiates at specimen surface. The crack propagates from the initiation site with creating striped pattern in the stable crack propagation state as can be seen in the SEM
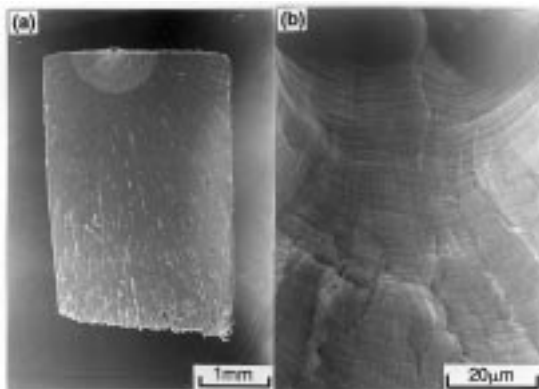
fractograph (b) shown in Fig. 11. The fatigue crack initiated from sub-surface of the some specimens, but in almost of the specimen, the crack initiated from the specimen surface as shown in Fig. 11.

Based on the fracture surface observation stated above, the tensile fracture surface morphology of the UHMWPE can be schematically shown in Fig. 11. The tensile fracture process of the UHMWPE can be divided into three stages. They are the crack initiation, stable crack propagation and unstable crack propagation stages. This fracture morphology is similar to that of the polymer fiber, that is, the nylon-filament, which has been reported by Ogata and Yoshida et al. [12].

The light micrographs of the regions near the crack tips of PE210 as a representative UHMPH and usual polyethylene with a molecular of 2.5×10^5 and a density of 0.956 as a reference material are shown in Fig. 12(a) and (b), respectively.

The craze, which is intrinsic in polymer materials [13], can be observed near the crack tip of usual polyethylene as shown in Fig. 12(b). The large failed region such as craze can not be observed near the crack tip of present UHMPH as shown in Fig. 12(a). This trend was confirmed by the SEM observation of fracture surface.
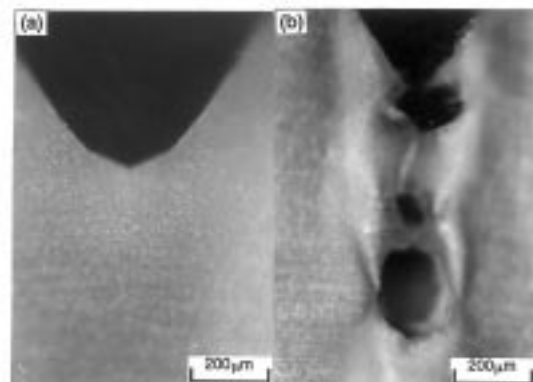
SEM fractograph of PE340 as a representative

Figure 10 SEM micrographs of typical tensile fracture surface (PE210).

Figure 12 Optical micrographs of crack tip regions. (a) Molecular weight: 210×10^4; (b) Molecular weight: 25×10^4.
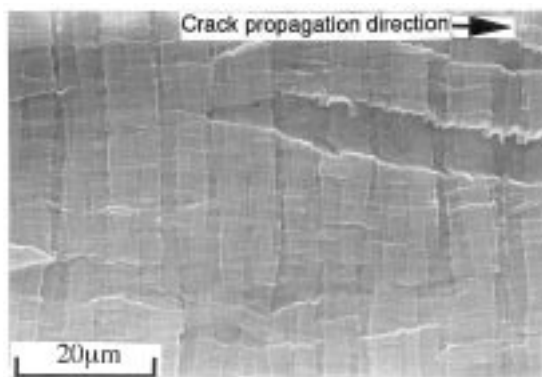
271

Figure 13 SEM micrograph of fatigue fracture surface (PE340).

observation result of fatigue fracture surface of present UHMWPE is shown in Fig. 13. The striation like cyclic patterns can be seen on the fracture surface perpendicular to the crack propagation direction. The striation like pattern seems to be different from that observed on the fatigue fracture surface of the metallic materials [14]. In the present materials, the fracture surface was firstly stretched by tear stress, then shrank and rolled in to form the striation like pattern as the crack propagates.

4. Conclusions

The fatigue tests were carried out on UHMWPE with different molecular weights in order to investigate the effect of molecular weight on the fatigue strength of UHMWPE. The fatigue tests at various frequencies and stress ratios were also conducted with UHMWPE. The effects of frequency and stress ratio on the fatigue strength of UHMWPE were investigated. The following conclusions were obtained:

1. In the present materials, the fatigue strength does not increase with increasing molecular weight. Fatigue strength of the present materials will be influenced more strongly by the crystallinity than by molecular weight.

2. In the present materials, the fatigue life is not clearly changed with change of the frequency. However, the specimen temperature during fatigue tests increases with increasing frequency.

3. The fatigue life of the present materials increases with increasing stress ratio when the maximum stress is plotted versus the number of cycles to failure. On the other hand, when the stress amplitude is plotted versus the number of cycles to failure, the fatigue life of the materials decreases with increasing stress ratio because the average stress is greater with increasing stress ratio.

References
1. H. NISHIMURA and I. NARISAWA, *Polym. Eng. Sci.* **31**(6) (1991) 399.
2. J. RUNT and M. JACQ, *J. Mater. Sci.* **24** (1989) 1421.
3. J. J. STREBEL and A. MOET, *J. Polym. Sci. Part B. Polym. Phys.* **33** (1995) 1969.
4. M. IWAMOTO and S. JINE ZIRYO, *J. Soc. Mater. Sci. Japan* **37**(418) (1987) 807.
5. X. LU, R. QIAN and N. BROWN, *Polymer* **36**(22) (1995) 4239.
6. J. T. YEN and J. RUNT, *J. Mater. Sci.* **24** (1989) 2637.
7. N. BROWN and I. M. WARD, *J. Mater. Sci.* **18** (1983) 1405.
8. S. NAGAI and S. ISHIKAWA, *Proceedings of Polymer* **52**(3) (1995) 125.
9. C. B. BUCKNALL and P. DUMPLETON, *Polym. Eng. Sci.* **25**(6) (1985) 313.
10. W. L. SAUER, K. D. WEAVER and N. B. BEALS, *Biomaterials* **17**(20) (1996) 1929.
11. R. W. HERTZBERG and J. A. MONSON, in Proceedings of the 27th Sagamore Army Materials Research Conference, 1983, p. 231.
12. N. OGATA and Y. YOSHIDA, *J. Mater. Sci. Soc. Japan* **18**(5) (1982) 240.
13. J. J. STREBEL and A. MOET, *J. Mater. Sci.* **26** (1991) 5671.
14. R. ODERAZAWA ''Fractograph and its Applications'' (Nikan Industry News Press, Tokyo, 1983) p. 34.