Effect of Annealing on the Fracture Behavior of Polyethylene

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ABSTRACT: *The failure behavior of as-molded and annealed high density polyethylene (HDPE) was studied under impact load. The experimental results show that annealing increased the impact fracture energy of HDPE. It was also found that the rise in the propagation energy was responsible for the increased fracture energy of the annealed specimens. Tension tests on as molded and annealed samples show the generation of voids when the material ahead of the notch yielded under tension. This led to the formation of fibrous structures that absorb more energy resulted in increased fracture energy in annealed specimens as compared to that of as-molded specimens.*

INTRODUCTION

Polyethylene (PE) is considered to be a high impact resistant polymer. Heat treatment like annealing influences its mechanical properties. Kitao [1] found that annealing increased the Charpy impact value for both MDPE and HDPE with the effect being more on HDPE. Recent study by Ravi and Takahashi [2] shows that annealing increased the fracture energy of HDPE by more than two times. It was also found that the rise in fracture energy of annealed HDPE was mainly during the propagation phase of the crack growth. To better understand the toughening mechanisms in HDPE, it is necessary to study the morphological changes on the fracture surface due to annealing. The fracture surfaces of quenched HDPE are smooth indicating a fast and brittle like fracture while annealed HDPE shows larger shear lips indicating a ductile failure mode [1]. Studies on notched and unnotched thin films of annealed HDPE subjected to large deformation show the formation of micro fibrils connecting the separated lamellae [3]. These fibrils were composed of highly oriented crystalline material [4]. Similar fibrils were observed on the fracture surfaces of bulk HDPE [5] under constant tension. Depending on the test temperature, these fibbers are either thicker (ductile failure) or finer (brittle failure) [6]. These fibrous structures were formed as a result of crazes generated under load ahead of the notch [6,7]. The purpose of this paper is to investigate cause of increased fracture energy in annealed HDPE and the type of damage zone formed in as molded and annealed polyethylene under dynamic load. Charpy impact test, tension test at four different strain rates and fractographic studies were carried out for this purpose.

EXPERIMENTAL

The material considered for the study was a high density polyethylene with a density of 955 kg/m^3. The specimens were machined from 5 mm thick compression molded plates, which were cooled at room temperature. Annealing was carried out at 100^{o}C for 24 hours and the specimens were cooled to room temperature in the oven itself. The Charpy impact specimens were 80 mm long and 10 mm wide with a span of 40 mm. The total length of tensile impact specimens was 100 mm with 25 mm gauge length and 10mm width. A V-shaped notch with a root radius of 0.25 mm was machined to a depth of 2 mm and then a fresh razor blade was tapped to create a sharp notch of $\cong$ 0.5 mm in depth. The notch was made after annealing treatment.

Charpy impact tests were carried out on an instrumented falling weight impact tested with an impact velocity of 1 m/s. Tension tests at lower strain rate were carried out on conventional UTM while higher strain rate tests were carried out using a dynamic tensile loading machine designed and fabricated in our laboratory. The load was measured using piezoelectric load cell while the displacement was measured using a novel optical extensometer [8] with an optical fiber as position indicator on the specimen. The displacement was measured at 1mm away from the impact point. Fracture energy was determined from the numerical integration of the load-displacement curve. SEM and optical microscope were used to investigate the fracture surface and the damage zone.

RESULTS AND DISCUSSION

Typical load and displacement traces for an as-molded HDPE specimen under Charpy impact are shown in Figure 1. The load rises rapidly to a maximum value and drops suddenly. This drop in load marks the boundary line of two distinct phases i.e., fracture initiation and fracture propagation phase of the total fracture event. The displacement seems to increase monotonically with time till the complete failure. Similar curves were obtained for all the specimens considered for the study.

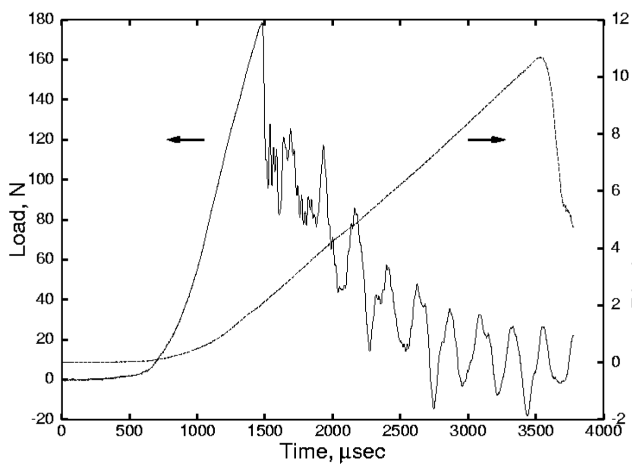


Figure 1: Typical load-displacement curve of an as-molded, HDPE specimen under Charpy impact

The load and displacement data obtained with respect to time were reploted as load versus displacement. Figure 2 shows typical load-displacement curves under Charpy impact for as molded and annealed specimens. After reaching a peak value, the load drops suddenly in as molded specimen. This drop became less steep as the annealing time increased, which indicates that relatively brittle fracture might have occurred in as molded specimens and relatively ductile fracture in annealed specimens. Charpy impact fracture energy for as molded and annealed specimens are shown in Figure 3. It can be seen that annealing for 24 hrs increased the fracture energy of HDPE by about 2.5 times that of as molded specimen. Figure 3 also shows the increase in energy associated with fracture initiation and propagation phases. It can be seen that the raise in fracture energy is mainly due to the propagation energy.

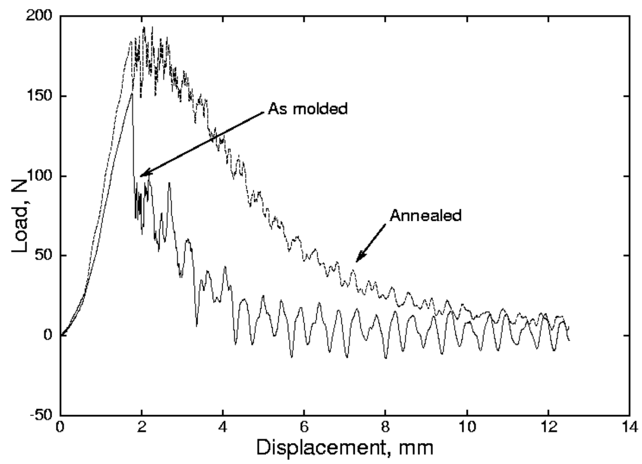


Figure 2: Typical load-displacement curves under Charpy impact

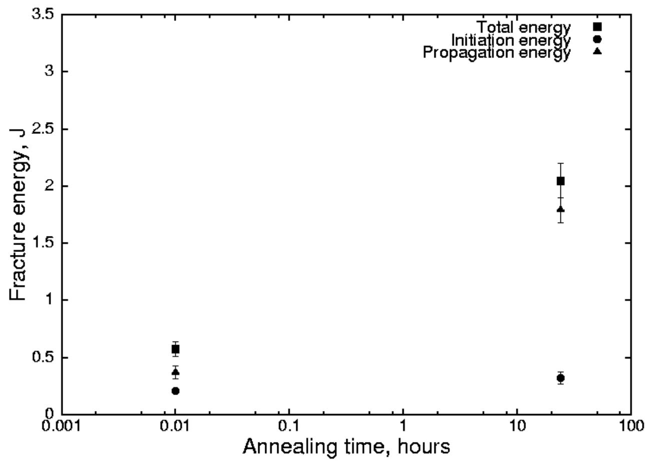


Figure 3: Charpy impact energy, initiation and propagation energies of HDPE specimens

Under uniaxial tension too, the annealed specimens show higher fracture energy. Figure 4 shows the variation of fracture energy with respect to the strain rate. It can be seen that the fracture energy of the annealed samples is higher under all strain rate studied.

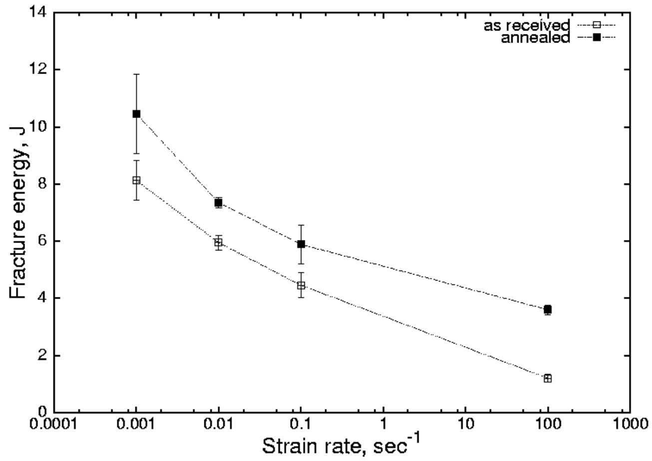
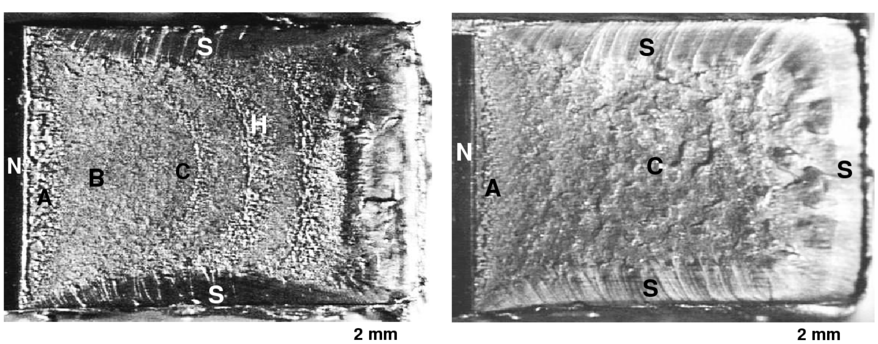


Figure 4: Variation of fracture energy under tension with respect to strain rate

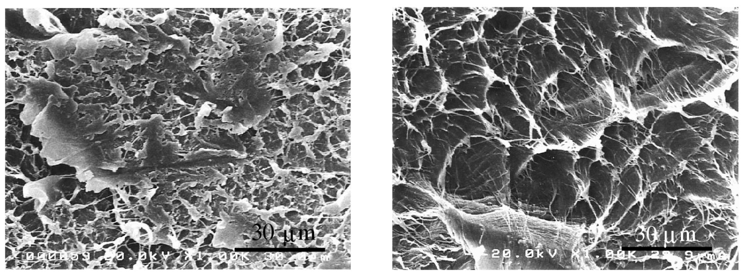
Figure 5 shows the fracture surface of the Charpy impacted specimens. Macroscopically, the fracture surface bears the features of brittle failure. The fracture surface of as molded specimen shows stick-slip lines (Figure 5a). These lines suggest momentary decrease in crack velocity. Annealed specimens did not show any such lines.



(a) (b)

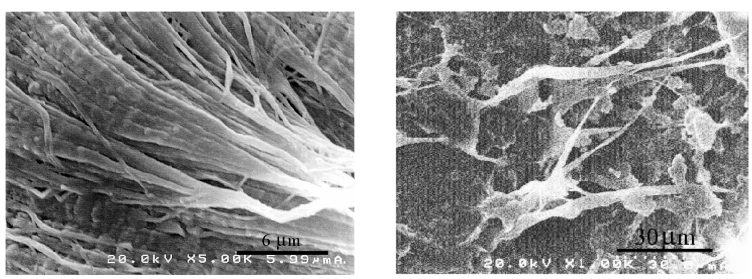
Figure 5: Typical fracture surface of (a) as molded and (b) annealed HDPE (N- notch tip; A-craze zone; B-hackle zone; C-propagation zone; S-shear lip; H-stick-slip line)

The fracture surface in the propagation zone ("C" Figure 5) shows hackle like pattern in as molded specimen while annealed specimen shows dimple pattern suggesting a brittle and ductile fracture respectively in as molded and annealed HDPE (Figure 6). Each dimple pattern is a cluster of dense fibrils as shown in Figure 7a. Formation of such dense fibrils must be responsible for the higher propagation energy. No such dense fibrils were seen in as molded HDPE specimen (Figure 7b).



(a) (b)

Figure 6: SEM photograph showing (a) hackle pattern in as molded and (b) dimple pattern in annealed HDPE specimens



(a) (b)

Figure 7: Propagation zone showing (a) Dense fibrous structures in annealed and (b) few fibrils observed in as molded HDPE specimens

To understand the fracture process, the fracture surfaces of the specimens loaded with tension were investigated. Figure 8 and 9 shows the fracture surface morphology of as molded and annealed HDPE under uniaxial tension. It was observed that the area over which brittle like failure increases with strain rate. Also for a given strain rate, annealed specimens show more ductile like failure compared to as molded one. These changes in fracture surface features between as molded and annealed specimens indicate that the material underwent morphological changes during the annealing process. The fractured surface and yielded region became milky white. It was observed that the occurrence of cavities or voids (Figure 10) causes the whitening as the material yielded under tension. When the load increases further, the material surrounding these voids undergo high stretching before final breakdown leaving fibrous structures. It is also observed the number of voids per unit area of annealed specimens are higher than that of as molded specimens. The generation of increased voids relieves the stresses and therefore higher toughness.

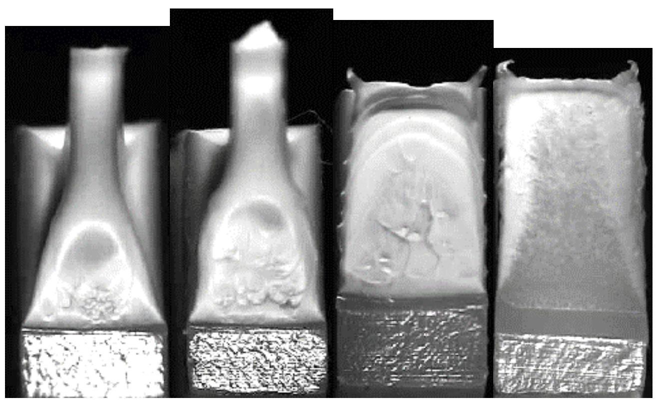


Figure 8: Fracture surface of as molded HDPE specimens under tension

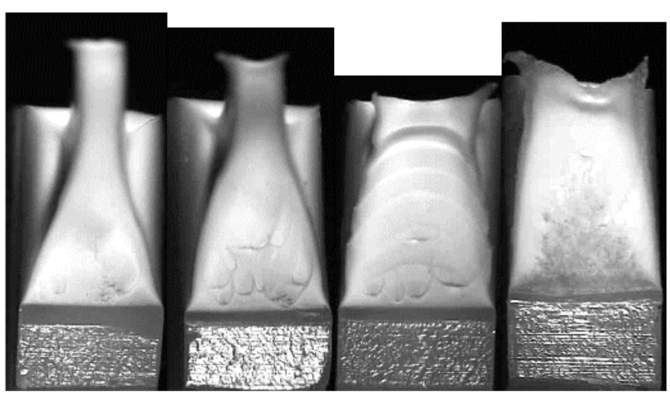


Figure 9: Fracture surface of annealed HDPE specimens under tension

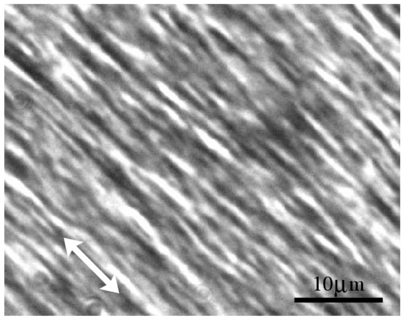


Figure 10: Voids (white zones) and stretched material (dark area) surrounding the voids observed in annealed HDPE

CONCLUSIONS

Annealing of HDPE increased the total fracture energy both under Charpy impact and uniaxial tension. Fractographic studies revealed the formation of dense fibrous structures in annealed specimens. It was also found that the generation of voids led to the formation of fibrous structures. On the basis of these observations, it is proposed that the rise in fracture energy observed in annealed samples is attributed to the void generation and the formation of fibrous structures as the material undergo mode I tensile load.

REFERENCE

1. Kitao, K. (1997) *Polymer Engineering and Science*, **37**, 777.
2. Ravi, S. and Takahashi, K. (in press) *Polymer Engineering and Science*.
3. Adams, W.W., Yang, D. and Thomas, E.L., (1986) *Journal of Material Science*, **21**, 2239.
4. Butler, M.F. and Donald, A.M. (1997) *Journal of Material Science*, **32**, 3675.
5. Lu, X. and Brown, N. (1990) *Journal of Material Science*, **25**, 411.
6. Lu, X. and Brown, N. (1997) *Polymer*, **38**, 5749.
7. Lu, X. and Brown, N. (1990) *Journal of Material Science*, **26**, 612.
8. Mada, T., Takahashi, K., Beguelin, Ph. And Aggag. G. (1997) *Trans. Japan Society for Mechanical Engineers*, **63**, 1431.