A new approach to correlate slow crack growth of HDPE nanocomposites with fracture toughness

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ABSTRACT: The slow crack growth (SCG) behavior of high density polyethylene (HDPE) blends has been conventionally approached by using applied stresses with respect to the failure time. In this paper, the failure behavior of a nonlinear elastic-plastic material such as HDPE is defined by the time-independent plane-strain fracture toughness that is commonly determined by short-term experimental tests. We established the correlation between the time-dependent SCG and the time-independent fracture toughness employing the elastic-plastic fracture mechanics (EPFM). SCG of HDPE blends was evaluated by the notched constant ligament stress (NCLS) test while the plane-strain fracture toughness was estimated by the essential work of fracture (EWF) test using the energy partitioning method. These two different time-domain fracture behaviors can be integrated by a power law relationship under the plane-strain condition. Subsequently, the SCG behavior of various HDPE blends including pristine/recycled HDPE and nanoclay composites was explained in terms of the plane-strain fracture toughness.

Keywords: HDPE, nanoclay composites, slow crack growth, fracture toughness

1 INTRODUCTION

Stress cracking caused by the long-term slow crack growth (SCG) has been a major cause of failure in plastic pipes made of high density polyethylene (HDPE), and the stress cracking resistance (SCR) of HDPE resins and pipes has been extensively studied. The majority of the experimental tests have been performed at elevated temperatures to accelerate crack propagation. The test data are subsequently extrapolated from elevated temperature to a lower ambient temperature condition using either Popelar shift method (PSM) (C. F. Popelar, Popelar, & Kenner, 1990; C. H. Popelar, Kenner, & Wooster, 1991) or the rate process method (RPM) (ASTM D2837, 2013). These tests directly evaluate stress cracking behavior and is expressed by applied stress against the failure time, but they do not provide an insight of failure mechanism that governs SCR.

From the fracture mechanics point of view, fracture failure of a material is defined by the plane-strain fracture toughness (K_c or J_c), which is determined using short-term tests. On the other hand, stress cracking caused by a SCG mechanism is evaluated using time-dependent tests described above, and the fracture toughness is often ignored ironically while analyzing SCR. Although several studies (Beaumont & Young, 1975; Williams & Marshall, 1975; Young & Beaumont, 1976; Zhou, Brown, & Crist, 1995) presented correlations between the time-independent fracture toughness and time-dependent SCG, these correlations were based on linear elastic fracture mechanics (LEFM) which may not be appropriate for nonlinear elastic-plastic materials such as HDPE. This paper, therefore, discusses a new approach to correlate SCG to the fracture toughness based on elastic-plastic fracture mechanics (EPFM). The SCR behavior was evaluated using the notched constant ligament stress (NCLS) test, while the fracture toughness was determined using the essential work of fracture (EWF) concept (Na, Spatari, & Hsuan, 2015, 2016). The correlation was studied in our previous papers (Na, Nguyen, Spatari, & Hsuan, 2016; Na, Spatari, & Hsuan, In Press). In this paper, we verify the correlation by providing statistical test data obtained from various HDPE blended materials such as pristine/recycled HDPE blends and nanoclay composites.

2 EXPERIMENTAL

2.1 *Test materials*

The pristine HDPE resin (ExxonMobil™ HDPE HD 7800P) with density of 0.954 g/cm^3 and melt index (MI) of 0.230 g/10 min was provided by ExxonMobil™. The recycled HDPE pellets with density 0.961 g/cm^3 and MI of 0.642 g/10 min, which were collected and reprocessed from milk jugs and water bottles, were supplied by Envision Plastics. Organo-modified montmorillonite (Nanomer® 1.44P) containing a surface modifier (dehydrogenated tallow dimethyl ammonium) was provided by Nanocor in the form of a master batch consisting of 50% nanoclay and 50% PE. Recycle-blends were prepared by mixing recycled HDPE pellets with pristine HDPE resin with weight fractions of 25, 50, and 75%. Polymer/clay nanocomposites (PCNs) were made from blending the prepared recycle-blends with the appropriate amount of nanoclay master batch to achieve the target concentration of 2, 4 and 6% by weight. The recycle-blends and PCNs were then produced in the form of pellets using a laboratorial twin-screw extruder. Subsequently, the pellets were compression molded into plaques with dimensions of 170 mm X 170 mm and with thickness of 2-mm according to ASTM D 4703-Procedure A (ASTM D4703, 2010). In this study, test materials were coded on the basis of the material composition. For example, P25R75/2% referred to the blend consisting of 25% pristine HDPE and 75% recycled HDPE with 2% nanoclay by weight.

2.2 *Essential work of fracture (EWF) test*

The double edge notched tensile (DENT) specimen with the height of 100 mm, thickness of 8.5 mm and width of 40 mm was prepared for the EWF test to evaluate the plane-strain fracture properties (Na et al., 2015). The pre-notch was made by a fine saw with a thickness of 0.9 mm. Subsequently, a sharp notch was accomplished by pushing a razor blade from the pre-notch tip to a target ligament length. The target ligament lengths were restricted to less than the thickness of the specimen, varying from 0.2t (1.7mm) to 0.8t (6.8mm) to ensure the failure in a plane-strain condition. The EWF test was conducted by applying a tensile displacement at a cross-head speed of 5 mm/min.

2.3 *Notched constant ligament stress (NCLS) test*

The SCR property was evaluated using the Notched Constant Ligament Stress (NCLS) test according to ASTM F 2136 (ASTM F2136, 2008). The dumbbell-shape specimens with dimensions of 3.175 mm wide (W) x 2 mm thick (t) x 63.5 mm long (L), were die out from a 2-mm thick compression molded plaque. Each test specimen was notched to a depth of 20% of the specimen thickness (*i.e.*, 0.4 mm) using a razor blade at a rate of 2.5 mm/min. The notched specimens were then fixed onto the test modules which were then immersed into a bath with distilled water at a temperature of either 30°C or 70°C. Applied stresses from 15 to 22 MPa were tested to achieve ductile failures within measurable times at the test temperature of 30°C. For tests carried out at 70°C, stresses from 1.4 to 9 MPa were applied to capture the brittle failures. The failure time was recorded to the accuracy of ±1.0 seconds for tests performed at 30°C and ±0.1 hours for tests performed at 70°C.

3 RESULTS

3.1 *Essential work of fracture (EWF) test*

The plane-strain specific work of fracture, $w_{e,B}$, of recycle-blended HDPE and PCNs was evaluated on the basis of the EWF concept by employing the energy partitioning method (Kwon & Jar, 2007). The result in Table 1 shows that the $w_{e,B}$ values, which are equivalent to the plane-strain fracture toughness (J_c), decreased linearly as the recycled content increased. For PCNs, adding 2-wt% of nanoclay to the pristine HDPE greatly decreased the $w_{e,B}$ value from 7.51 to 3.04 kJ/m^2, while no significant change was measured in pristine/recycled HDPE blends. Further increase of the nanoclay content to 6-wt%, the $w_{e,B}$ value decreased in both the pristine and recycle blends. Also, adding and increasing nanoclay content led to a significant decrease of $\beta_B w_{p,B}$ value which is an indicator for the ductility of the material. The brittle failure time (t_p) was also calculated from the load-displacement curve of the EWF test based on a strain rate of 5 mm/min (Na, Nguyen, et al., 2016; Na et al., In Press), and the values are included in Table 1.

Table 1: Plane-strain EWF parameters for pristine/recycled HDPE blends and PCNs

Blends	Nanoclay content (%)	$W_{e,B}$ (kJ/m^2)	$\beta_B w_{p,B}$ (MJ/m^3)	t_p *(min)*
P100	0	7.51 ± 5.33	3.84 ± 1.18	0.24
	2	3.04 ± 1.02	1.92 ± 0.24	0.16
	4	2.40 ± 1.55	1.18 ± 0.36	0.12
	6	2.46 ± 1.45	0.53 ± 0.30	0.10
P75R25	0	5.72 ± 3.25	2.25 ± 0.69	0.19
	2	5.75 ± 1.02	0.36 ± 0.20	0.13
	4	3.01 ± 1.74	0.75 ± 0.36	0.11
	6	2.84 ± 1.07	0.37 ± 0.23	0.09
P50R50	0	3.98 ± 1.31	1.43 ± 0.29	0.13
	2	3.56 ± 0.82	0.40 ± 0.19	0.10
	4	2.80 ± 1.13	0.40 ± 0.26	0.09
	6	2.11 ± 0.84	0.49 ± 0.19	0.09
P25R75	0	2.84 ± 1.36	1.11 ± 0.31	0.12
	2	2.88 ± 0.85	0.32 ± 0.21	0.09
	4	1.45 ± 1.14	0.46 ± 0.26	0.07
	6	1.32 ± 0.82	0.27 ± 0.19	0.08

3.2 *Stress Cracking Resistance (SCR) Test*

In analyzing the NCLS test data, the load parameter is conventionally expressed by the stress intensiy factor *K* employing LEFM. In mode 1 fracture, *K* is expressed as

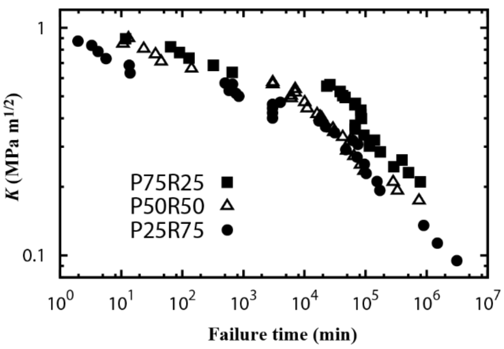
$$K = F(a/b)\sigma_N\sqrt{\pi a} \tag{1}$$

where *F(a/b)* is the specimen geometry factor defined by the notch (*a*)-width (*b*) ratio, and σ_N is the nominal applied stress (Tada, Paris, & Irwin, 2000). Figure 1(a) presents the log-*K* value with respect to the log-failure time for the recycled blends. The results show a bilinear curve consisting of a shallow linear slope followed by a steeper linear slope. The shallow sloped region corresponded to a creep rupture whereas the steeper sloped region represented slow crack growth (SCG) failure (Lu & Brwon, 1990; Grace Hsuan, 2000). Comparing the curves in Fig. 1(a), failure times of SCG decreased as the recycled content increased. Figure 1(b) shows the effect of nanoclay on SCG failure time. Incorporating nanoclay generated a similar bilinear failure curve as the non-reinforced recycled blend, but SCG failure time is noticeable increased.

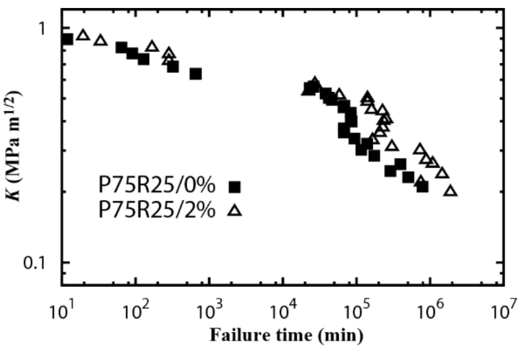
From the fracture mechanics point of view, LEFM is only applicable for brittle materials that deform linear elastically until fracture occurs. However, HDPE and PCN are a nonlinear elastic-plastic material and, therefore, EPFM should be a more appropriate approach than LEFM in the evaluation of overall fracture behavior of PE materials (Bassani, Brown, & Lu, 1988; Lu, Brown, & Bassani, 1989). For an elastic-plastic material, the *J*-integral in EPFM is determined by a combination of the linear elastic component J^e and the fully plastic component J^p, as expressed in Eqn. (2).

$$J = J^e + J^p \tag{2}$$

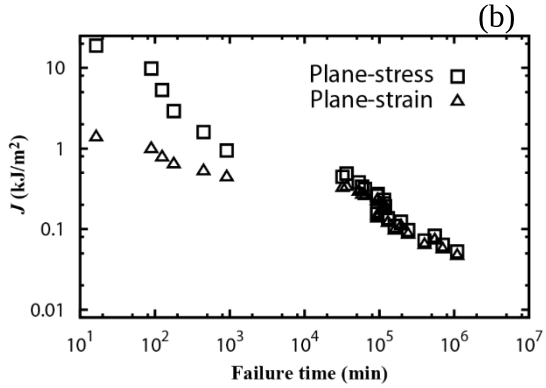
Figure 1(c) repsents the *J*-integral values for P25R75 associated with failure times for both a plane-strain and a plane-stress condition. At high applied stresses (*i.e.*, high *J* condition), the discrepancy between two conditions was significant because the plastic component of *J* (J^P) dominated the total *J*. As the stress decreased, the linear elastic component (J^e) became more dominant in the overall *J*. This result indicates that the conventional method employing *K* is not appropriate to expressing the entire SCR behavior because it does not account for the plastic deformation before failure, particularly for the high stress region.



(a)



(b)



(c)

Figure 1: (a) Log K-log failure time for recycled blends, (b) log J-log failure time for P75R25, and (c) log K-log failure time for PCN

3.3 *Correlation between Plane-strain EWF and SCG*

The failure curve plotting log *J* versus log failure time in a plan strain condition can be separated into four regions based on different slopes as illustrated in Fig. 2. The region I corresponded to the short-term failure where the plastic *J* (J^p) plays a dominant role in the total *J*. Once the test specimen being loaded, a small creep zone, which is comparable to a plastic zone, forms instantaneously at the crack-tip, and the specimen failed shortly afterward. This implies that the crack growth rate was faster than the growth of the small creep zone. Because the creep zone remained small, it is defined as a small scale creep (SSC) condition and the crack-tip condition can be characterized by *J*.

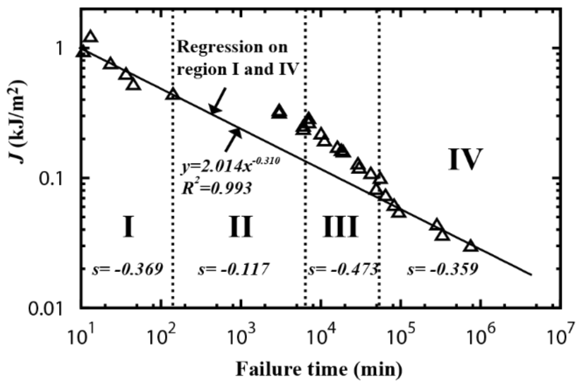


Figure 2: Log J-log failure time plot for P25R75 in the plane-strain condition

In region II, the applied stresses, which are lower than the yield stress at 23°C, enabled the creep zone to grow with time and the creep zone eventually engulfed the entire ligament. In this condition, both the linear-elastic and the elastic-plastic fracture mechanics are unable to characterize the crack-tip condition due to the significant creep strain.

Region III is consistent with the transition from the creep to SCG failure. A large plastic deformation resulting from macroscopic yielding governed the creep failure whereas a craze growing ahead of the crack-tip dominated the SCG failure. These two different mechanisms competed with each other in the transition region (Norman Brown, Donofrio, & Lu, 1987).
Region IV is dominated by the SCG failure which occurred at much lower applied stresses, typically below 40% of the yield stress. At such low stress, the creep strain is very small. As a result, a SSC condition appeared at the crack-tip and can be characterized by the parameter J (Lu et al., 1989).

Because the fracture behaviors in region I and IV can be characterized by the same parameter J under the assumption of a plane-strain condition, it is reasonable to associate these two regions. Also, the slopes of the test data in the regions I and IV were found to be similar and the data in these two regions can be represented by a single regression line with a very high R^2 value as shown in Fig. 2. The linear regression line can be expressed as:

$$J = At_f^{-s} \tag{3}$$

where A is the regression constant and s is the slope of the regression line that is related with time-dependent response of a material. t_f denotes the failure time.

Equation (3) implies that the plane-strain fracture toughness (J_c) can be also calculated once the corresponding failure time is known. The J_c value, which corresponds to $w_{e,B}$, for tested materials was determined using the EWF test, and listed in Table 1. By substituting the brittle failure time (t_p) listed in Table 1 as t_f in Equation (3), the corresponding J value was deduced and is defined as J_{pt} in this study. The comparison between J_c obtained from the EWF test and J_{pt} calculated from Equation (3) is shown in Figure 3. The error bar of each data point represents the 95% confidence interval.

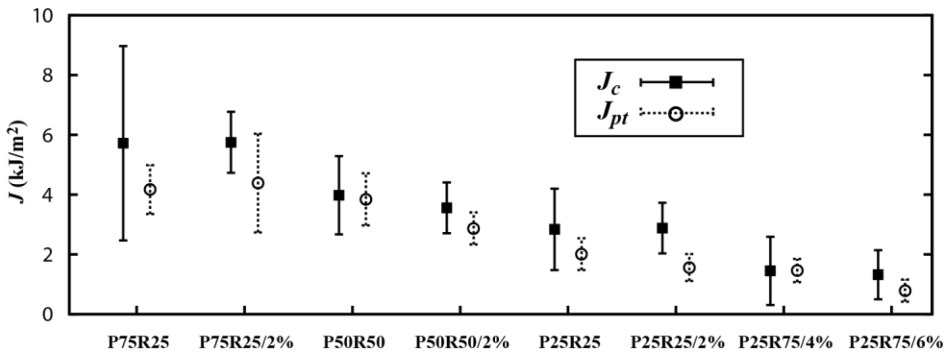


Figure 3: Comparison J_c to J_{pt}

The results indicate that the J_{pt} is statistically similar to the J_c. Considering J_c and J_{pt} were obtained from two methods, a small discrepancy in one sample (*i.e.*, P25R75/2%) is considered to be acceptable. Therefore, the constant A in Eqn. (3) can be replaced by the plane-strain fracture toughness J_c and the failure time t_f was normalized by t_p (*i.e.*, unit time $\tau_f = t_f/t_p$), as shown in Eqn. (4).

$$\tau_f = \left(\frac{J}{J_c}\right)^{-1/s} \tag{4}$$

Equation (4) provides the correlation between fracture toughness and slow crack growth of HDPE blended with recycled HDPE and nanoclay materials. The failure time associated with SCG directly relates to J_c and s. Blending the same type of polymers (i.e., pristine/recycled HDPE blends) had a minimal effect on the slope while it changed J_c. On the other hand, incorporating nanoclay influenced both J_c and s, leading to a change of the failure time.

4 CONCLUSION

This paper introduced a new approach that relates the fracture toughness to the slow crack growth under a plane-strain condition on HDPE blends including pristine/recycled HDPE/nanoclay composites. The calculated J_{pt} values from the NCLS test were comparable to the J_c value obtained from the EWF test within the statistical errors. Therefore, the failure time of time-dependent SCG can be described by the combination of the time-independent parameter J_c and time-dependent response of a material *s*.

The results of the EWF and the NCLS test indicate that replacing pristine HDPE with recycled HDPE reduced the J_c value, and thus decreased failure times of SCG. Incorporating nanoclay up to 6-wt% lowered the J_c value; however, it also decreased the slope (*s*) of the SCG failure curves, resulting in increasing the failure time in the lower *J* values. This paper concluded that a new approach based on the plane-strain *J*-integral analysis is viable to interpret the SCG behavior of HDPE blends in terms of the fracture toughness.

ACKNOWLEDGEMENTS

This study was supported by National Science Foundation (NSF CMMI – 1030783). Any opinions, findings, and conclusions or recommendations expressed in this material are those of the authors and do not necessarily reflect the views of the National Science Foundation.

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