Assessment of Damage and Long-Term Strength of Polyethylene

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Abstract Tensile strength of polyethylene is examined at various crosshead speeds using cylindrical specimens of 20 mm in gauge length. Through monotonic stretch with intermittent halt for load relaxation, the study shows that long-term tensile strength of polyethylene should be around half of the short-term strength determined in the standard test conditions. The study discovers a transition in the localized deformation (commonly known as necking), of which the appearance changes from opaque white to translucent when the crosshead speed is reduced from 0.05 to 0.01 mm/min, possibly due to the suppression of cavitation damage at the low crosshead speed. At a sufficiently high crosshead speed, damage generation can occur at a threshold strain as low as 4% which is way below the yield strain of the material. It is therefore concluded that determining long-term tensile strength of polyethylene needs to consider accumulation of the damage. Since this type of damage is not quantified in any of current standard methods, the study suggests that results from those tests need to be analyzed carefully to ascertain their validity for characterizing long-term performance of polyethylene products.

Keywords Long-term strength, polyethylene, damage, neck

1. Introduction

Polyethylene (PE) is known for its excellent ductility, superb corrosion resistance and strong strain hardening ability [1]. Its reliable performance has expanded the applications to products such as pipes for natural gas transportation that requires service lifetime for more than 50 years. Although standard methods, e.g. ASTM D1598 and D2837, are available to assess serviceability of PE pipes for this purpose, accuracy of the assessment is hindered by uncertainty in the resistance to a fracture behaviour known as slow crack growth (SCG) [2].

SCG development was discovered from early tests on full-sized pipe specimens subjected to internal hydrostatic pressure [3-5]. Results show a transition on the trend line of hoop stress versus time to failure, which yields a much steeper change of stress drop with increase of time to failure after the transition than before, with the former showing crack growth in much more brittle manner than the latter. Therefore, evaluating performance of PE pipes in service requires testing that takes at least one year for each specimen to ensure that the failure behaviour after the transition does not occur in service. In view of the lengthy time required for such tests, many studies have turned to accelerating tests that use small coupon specimens with sharp notches to shorten the time to initiate the crack growth [6, 7]. Although the use of sharp notches prevents the tests from determining the critical stress level for the transition to the SCG development, the results show a trend line that bears some similarity to the transition observed in full-sized pipe tests. As a result, the accelerating test becomes very popular for materials comparison, and is standardized as ASTM F1473.

Fracture generated in the notched specimens is known to be governed by shear deformation, and the transition is due to the change of shear deformation from the macroscopic scale (i.e. in a ductile manner) to the microscopic scale (in a brittle manner) [8, 9]. The latter is also known as craze that contains fibrils in a constrained space in front of the notch tip [10]. Based on results from the accelerating tests, formation of crazes is suggested to be the dominant mechanism for the SCG development [11]. An extensive testing program has shown that the test method can generate a transition of the trend line for a wide range of PE materials [6].

Results from the above accelerating test are always analyzed as a function of the applied load, in terms of stress without the notch or stress intensity factor [11]. Since the applied load remains constant during the test, it is perceived that loading rate does not play any role on the deformation and fracture behaviour. However, the applied load needs to increase to the targeted level at the beginning of the test, for which the duration is relatively short. At the notch tip, rate of the initial stress increase should be pretty high. If the high loading rate generates damage that leads to different fracture behaviour from that occurs during the SCG development, results from the accelerating test should not be used to evaluate the SCG resistance. To my knowledge, such a scenario has never been seriously considered for the development of the accelerating test.

Work by O'Connell et al. [12] shows a transition from ductile to brittle fracture by testing notched specimens at an extreme combination of low loading rate and high temperature, that is, at a crosshead speed of 0.005 mm/min and temperature of 110^{o}C. The transition is opposite to the effect of loading rate and temperature commonly known for deformation and fracture behaviour of polymers. Fracture surfaces generated in such an extreme condition are featureless and without any indication of whitening. Note that this type of fracture behaviour is different from that shown in the work by Lu et al. [13] using a similar specimen design but subjected to a higher initial loading rate at a lower temperature. The latter always contains a thin but distinct whitening zone adjacent to the fracture surface. Unfortunately, due to the presence of sharp notches, difference of the true loading rates at the notch tips of those specimens cannot be quantified.

Some studies on the accelerating test have adopted notch-free specimens so that the loading rate (in terms of rate of strain increase) and its effect on the deformation behaviour can be quantified. Most of this type of studies applied a strain rate above 10^{-3} s^{-1} [e.g. 14-16], which is equivalent to a test speed about 1.2 mm/min for a gauge length of 20 mm. At the moment when this manuscript is prepared, the lowest strain rate that can be found in the literature is in the order of 10^{-5} s^{-1} [17].

A strain rate of lower than 10^{-5} s^{-1} is used in the work described in this paper to introduce a deformation transition that changes the neck appearance from opaque white to translucent. Based on the observed deformation transition, this paper investigates effects of loading rate on the necking phenomenon of notch-free PE specimens. In addition to the reduction of the crosshead speed, a special loading scheme is used to determine the stress-elongation curve of PE, in order to determine the long-term strength and its change caused by different initial loading rate.

2. Experimental

2.1. Materials

Specimens used for the mechanical testing are prepared from PE plates of 10 mm thick, provided by NOVA Chemicals, of which molecular weight, molecular weight distribution, and density are given in Table 1. The plates were compression-molded from pellets to ensure isotropy of the mechanical properties. Cylindrical specimens with geometry same as those used in ref. [18] were machined from those plates, with dimensions shown in Figure 1. To ensure that necking was always initiated in the middle of the gauge section where an extensometer was placed to monitor the change of cross-sectional dimensions, a small imperfection was introduced there to reduce the diameter by less than 2%. Note that despite the presence of such an imperfection, all specimens could show full neck development in the whole gauge section prior to the final fracture.

Table 1. Material characteristics of PE used in this study [19].

Weight-average molecular weight (M_w) (g/mol)	Number-average molecular weight (M_n) (g/mol)	Branches per 1000 C	Density, ρ (g/cm^{-3})	Crystallinity (%)	Tensile yield strength (MPa)
73,074	30,391	3.4–4.2	0.941	63.6	20.2

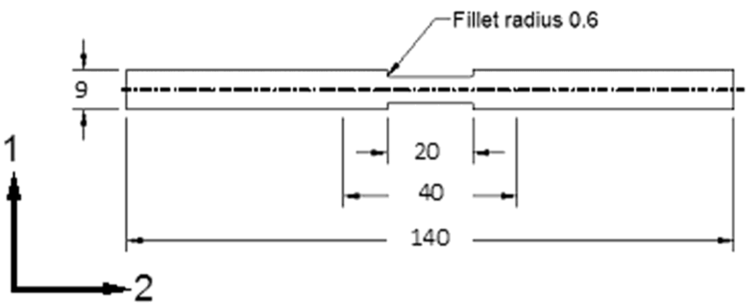


Figure 1. Dimensions of cylindrical specimen used in the study.

2.2. Mechanical Testing

All tests were conducted using a universal testing machine (QUASAR 100) at room temperature. Eight crosshead speeds between 0.001 and 5mm/min were selected for the testing, corresponding to the initial straining rate between $8x10^{-7}$ and $4x10^{-3}$ sec^{-1}. Two types of tensile tests were used. One, named simple tensile test here, applies a constant crosshead speed in the range between 5 and 0.001 mm/min until the stroke reaches a pre-set value. The other, named relaxation test, applies a constant crosshead speed of 1 or 0.001 mm/min, to reach a preset stroke and then, holds the stroke for 100,000 seconds (about 28 hours) while change of the load is recorded. For both types of tests, stress and area strain were calculated. The latter was defined as *ln* (A_o/A) where A_o is the original cross-sectional area and A the cross-sectional area at the moment of measurement.

3. Results and Discussion

3.1. Simple Tensile Test

Figure 1 shows the summary of engineering stress versus stroke from simple tensile tests. As expected, peak stress decreases with the decrease of crosshead speed. In addition, profile around the peak load changes with the crosshead speed. In particular, the curve section around the maximum stress changes from single-peak to double-peak, with the two peaks separating further away with the decrease of the crosshead speed. The post-tested specimens also show a transition with the decrease of the crosshead speed. As shown in Figure 3, by decreasing the crosshead speed, noticeably from 0.05 to 0.01 mm/min, the neck appearance changes from opaque white to translucent. The latter has a similar color of the original specimen.

Figure 4 summaries maximum engineering stress as a function of crosshead speed, in logarithmic scale in Figure 4(a) and linear scale in Figure 4(b). Insert in Figure 4(b) depicts change of the maximum engineering stress at crosshead speeds below 0.01 mm/min. Both figures suggest that around the lowest crosshead speed used in the study, 0.001 mm/min, the maximum engineering stress does not show any tendency of reaching a plateau value, but prediction from the two figures for the maximum engineering stress at zero crosshead speed is very different. Figure 4(a) suggests that the value could be way below 10 MPa; while Figure 4(b) based on specimens with translucent neck appearance (for crosshead speeds at and below 0.01 mm/min), the value should be about 10.5 MPa.

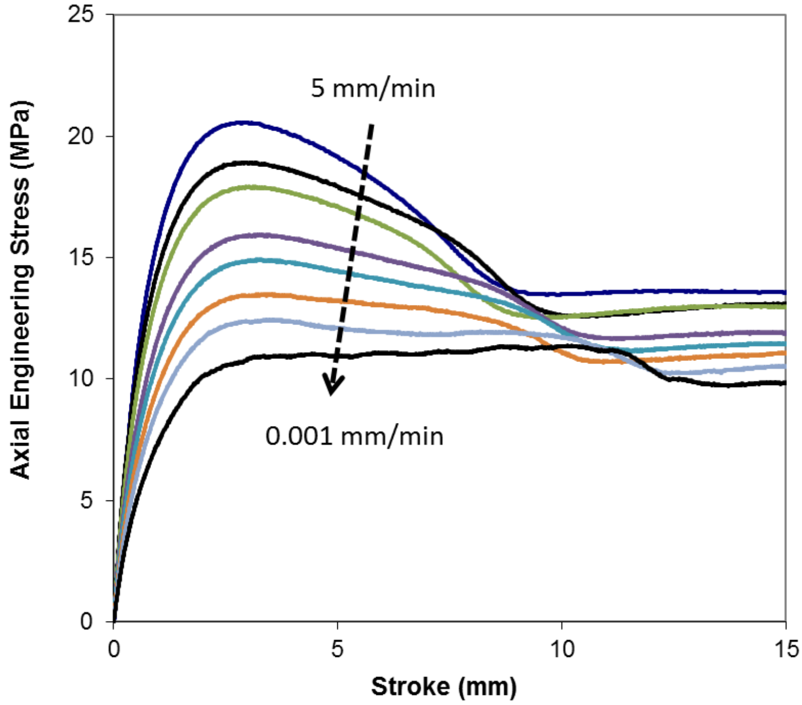


Figure 2. Engineering stress-stroke curves for crosshead speeds from 5 to 0.001 mm/min.

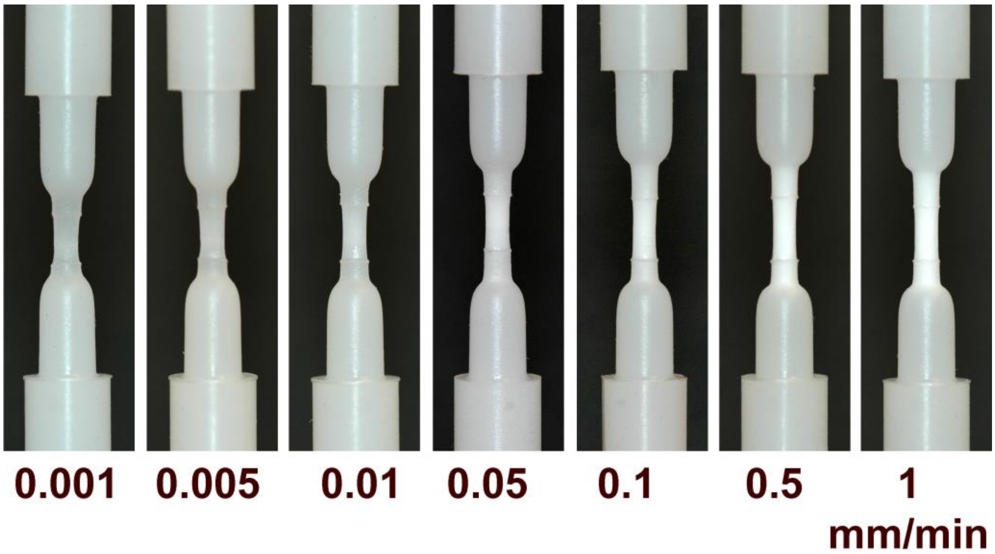
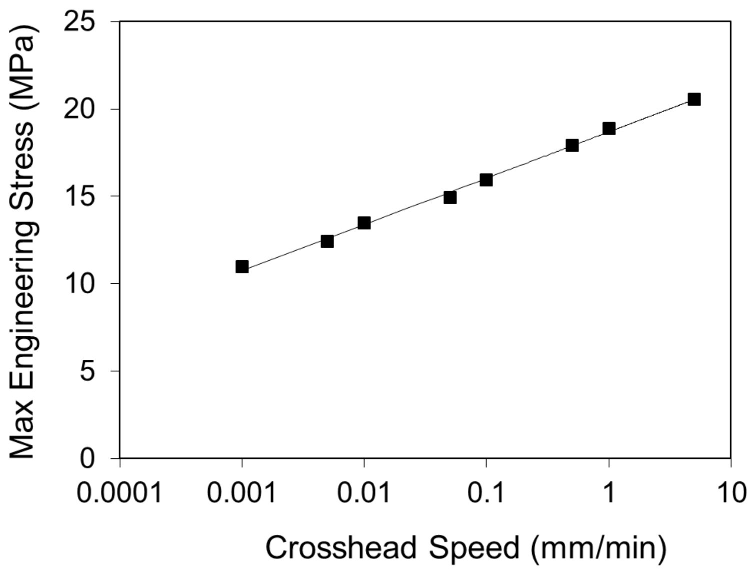
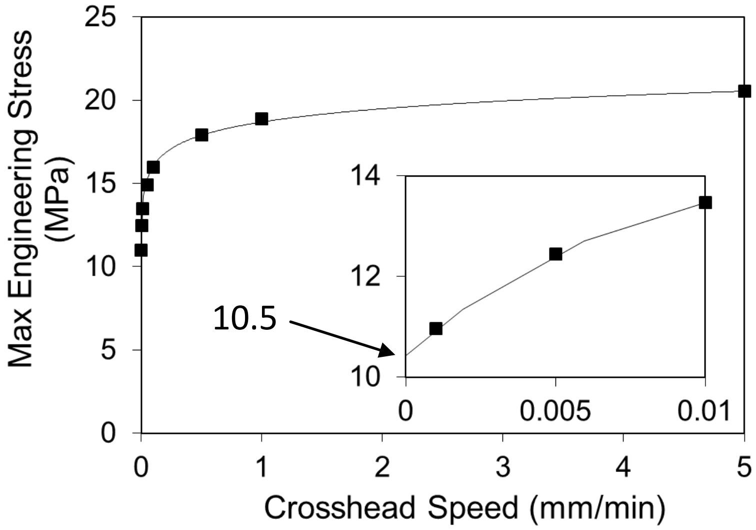


Figure 3. Transition of specimen deformation by changing the crosshead speed from 1 to 0.001 mm/min.



(a)



(b)

Figure 4. Maximum engineering stress versus crosshead speed in (a) logarithmic scale and (b) linear scale.

In view of the uncertainty in predicting the strength at zero crosshead speed (regarded as the long-term strength here), another test (named intermittent tensile test) was conducted, at the lowest crosshead speed that is provided by the test machine, 0.001 mm/min. Instead of applying a monotonic increase of stroke, stroke for the intermittent tensile test was programed to reach a pre-set value at 0.001 mm/min and then, the load was allowed to relax for 100,000 seconds. This time period was chosen based on some preliminary tests which suggest that the load maintained at a fairly constant level after 100,000 seconds, after which the further load decrease within the same period is within two times of the load cell resolution (± 0.5N). An example of the engineering stress-stroke curve from the intermittent tensile test is shown in Figure 5, in which the pre-set stroke values range from 0.8 mm to 6 mm. The corresponding long-term stress values at those strokes are then connected to form the long-term stress-stroke curve, also shown in Figure 5. Peak value of the long-term stress-stroke curve is 9.9 MPa, which is very close to that predicted from Figure 4(b), using values from three lowest crosshead speeds, from specimens with translucent neck appearance.

Note that the above pre-set stroke values cannot be applied to intermittent tensile test at a crosshead speed of 1 mm/min. This is because after the relaxation, the stroke required to resume the tensile loading is much longer than the pre-set increment of the stroke values. As a result, individual relaxation tests, as presented below, are used to compare the long-term stress-stroke curves between two crosshead speeds, 1 and 0.001 mm/min.

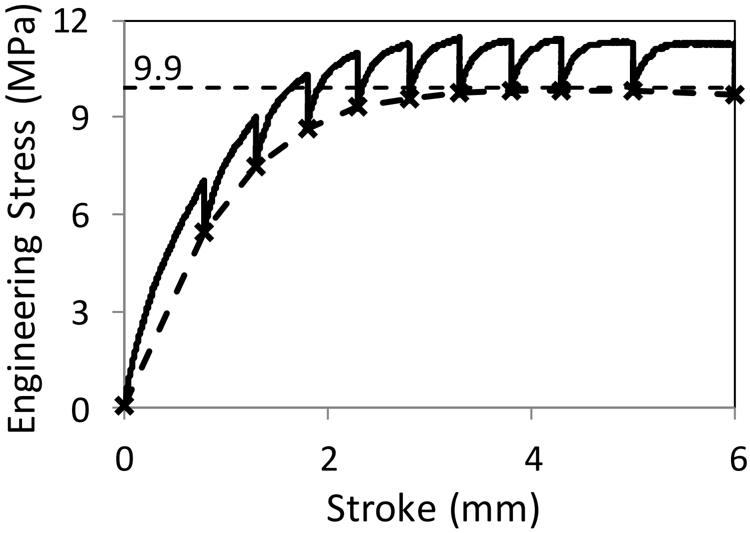


Figure 5. Engineering stress-stroke curves from an intermittent tensile test at crosshead speed of 0.001 mm/min, showing a long-term strength of 9.9 MPa, at a stroke between 3.8 and 4.3 mm.

3.2. Relaxation Test

Relaxation tests for the same period of 100,000 seconds were conducted at pre-selected strokes to establish long-term stress-stroke curves at crosshead speeds of 1 and 0.001 mm/min. Note that for the relaxation tests, each specimen is used only for one pre-set stroke.

Figure 6 summarizes results from the relaxation tests. The overall curve profile at 0.001 mm/min is similar to that shown in Figure 5 with peak stress at 9.9 MPa, and is distinctively above the curve from 1 mm/min. Peak stress for the latter is 8.9 MPa, suggesting that by increasing the crosshead speed from 0.001 to 1 mm/min, the long-term tensile strength of PE is reduced by about 10%. Figure 6 also suggests that with the increase of the stroke, difference of the long-term stress between curves at the two crosshead speeds increases, and the difference exists even at the smallest stroke of 0.8 mm which corresponds to an area strain of 3.6%. The stress decrease by increasing the crosshead speed is an indication of damage introduced by the deformation. Therefore, Figure 6 suggests that damage must have been generated at a very early stage of the deformation process.

Figure 7 depicts the post-test specimens with different strokes at the crosshead speed of 1 mm/min. For strokes below 6 mm, there is no clear neck formation. Therefore, only 1 photograph, taken from a specimen with stroke of 1.3 mm, is shown in the figure. Similar behaviour was found from specimens tested at 0.001 mm/min, except that the neck appears to be translucent, instead of opaque white.

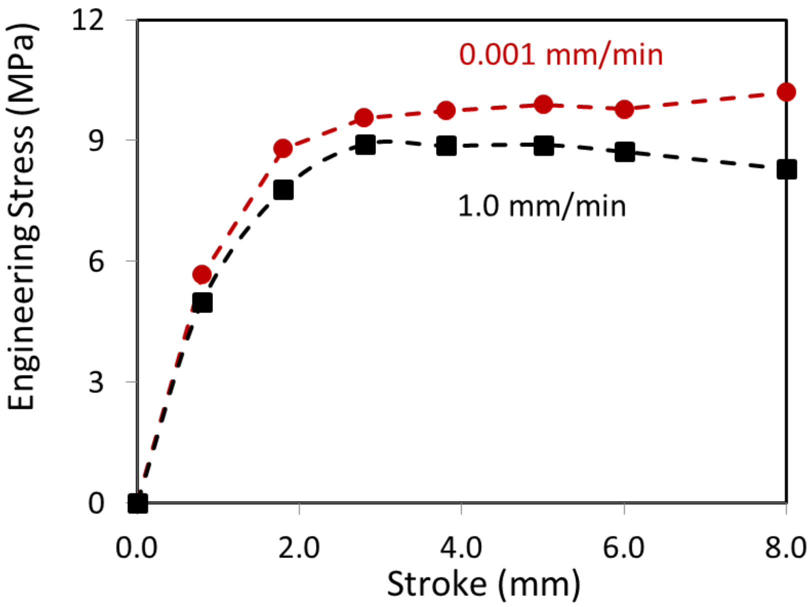


Figure 6. Engineering stress versus stroke from relaxation tests at 2 different crosshead speeds.

Further evidence for the damage generated by a high crosshead speed of 1 mm/min is shown in Figure 8 which contains three engineering stress-stroke curves, all from simple tensile test to a preset stroke of 1.8 mm at a crosshead speed of 0.001 mm/min. The three curves are from a virgin specimen, labeled "As is," and after stretch to 1.8 mm at two crosshead speeds, as labeled by "After 1 mm/min" and "After 0.001 mm/min," respectively. The figure suggests that the prior stretch at 0.001 mm/min does not change much of the stress increase with time, but the prior stretch at 1 mm/min has decreased the stress response. Therefore, even at a crosshead speed of 1 mm/min, corresponding to an initial strain rate of around $8x10^{-4}$ s^{-1}, damage is introduced way before the yield point. This phenomenon has also been observed by similar tests at other pre-set stroke values of less than 5 mm.

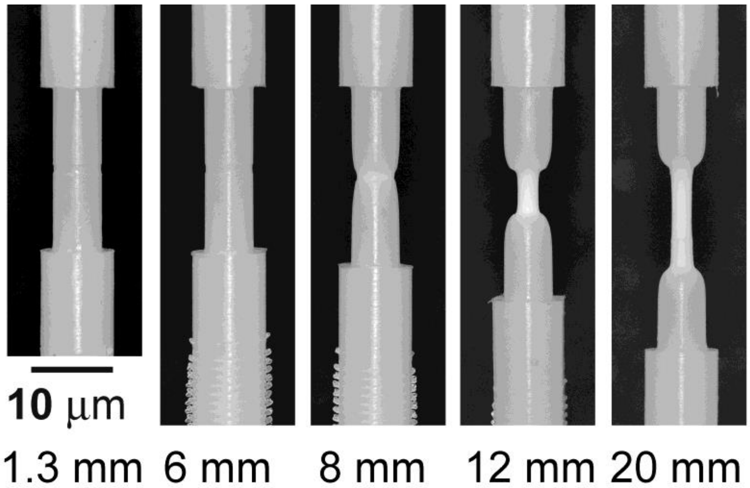


Figure 7. Specimens after being tested at 1 mm/min for different pre-set strokes (given under each photograph). Specimens tested at 0.001 mm/min have similar dimensional changes but do not show any whitening.

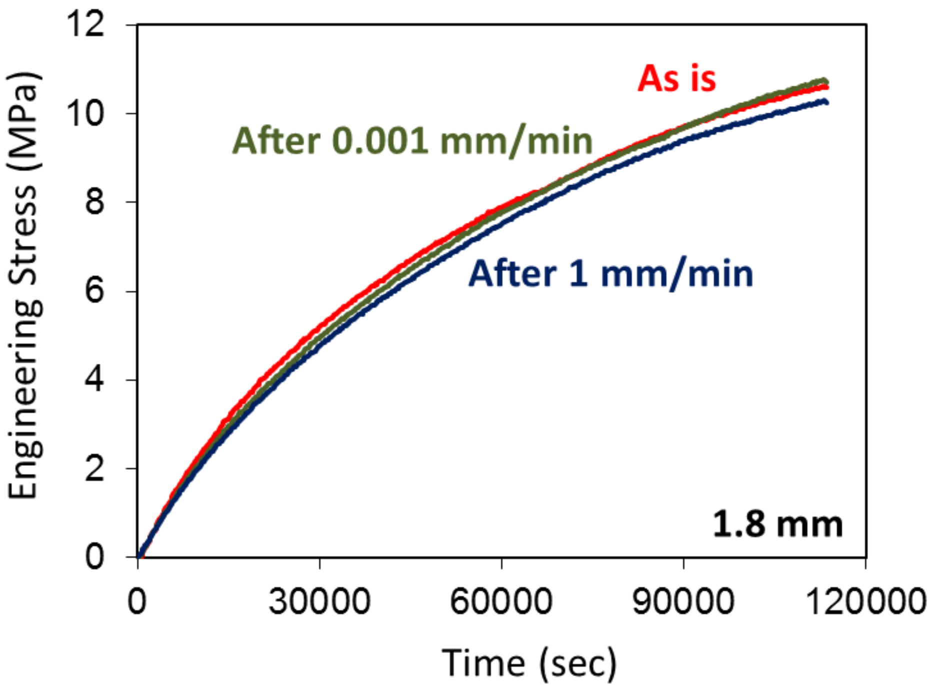


Figure 8. Comparison of engineering stress response with different loading history: as is, after stretch of 1.8 mm at 1 and 0.001 mm/min, respectively.

4. Conclusions

This paper presents an experimental study to assess long-term strength of PE and influence of loading rate on the damage generation. The study is based on a newly discovered "deformation transition" in notch-free specimens which generates translucent neck instead of the common opaque white. The discovery leads to development of a short-term test that uses coupon specimen to quantify the long-term strength of virgin PE. The results suggest that long-term strength of virgin PE is about 50% of its strength determined from the standard test.

The study also shows that long-term strength of virgin PE only represents the upper limit of the strength for PE pipe, as damage generated during the pipe transportation and installation may further reduce the strength. The results indicate that even under a loading rate of 1 mm/min, corresponding to a strain rate around $8x10^{-4}$ s^{-1}, damage can be generated in polyethylene at a strain level less than 4%. Therefore, long-term damage resistance of polyethylene pipes can be very different from the short-term test results that are often determined from virgin material.

The results suggest that assessment of long-term performance of PE pipe should include the damage assessment which may be generated during installation and service. Since such damage assessment is not considered at present, a new methodology is needed to quantify and monitor the damage evolvement in a plastic pipe and to quantify its effect on PE's long-term performance.

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