

**MEASUREMENT OF ENVIRONMENTAL STRESS CRACKING RESISTANCE OF
POLYETHYLENE PIPE: A REVIEW****Yi Zhang¹**Department of Engineering Mechanics
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Qingdao, China**ABSTRACT**

Use of polyethylene (PE) for water and natural gas transportation has increased rapidly due to its good physical and mechanical properties, especially its excellent corrosion resistance property. However, when immersed in adverse environment and subjected to applied stress, PE will suffer from accelerated crack growth in a phenomenon known as environmental stress cracking (ESC). ESC occurs in a brittle manner without little pre-fracture deformation, thus can cause catastrophic, unexpected failure for PE pipe. A number of different test methods have been developed for characterizing ESC resistance (ESCR) of PE materials. Within this paper, a state-of-the-art review is given on the current ESCR characterization methods, including the working principle and limitations of each method.

Keywords: Polyethylene Pipe, Environmental stress cracking resistance (ESCR)

INTRODUCTION

Due to their excellent chemical resistance and reasonable mechanical strength, semi-crystalline polymers (SCPs) such as polyethylene (PE) and polypropylene (PP) are increasingly employed in low-stress applications, e.g. food container, drainage and natural gas pipes, and nuclear power plants. For example, over 90% of the newly installed gas pipeline systems

are now made of polyethylene [1]. Although they have good chemical resistance, SCPs when subjected to relatively low levels of stress and immersed in adverse environment, will suffer from cracking in a phenomenon known as environmental stress cracking (ESC). The presence of aggressive agent such as soaps, wetting agents, oils, or detergents facilitates crack initiation and growth in SCPs, effectively reducing the levels of applied stress necessary for the occurrence of ESC and eventually shortening lifetime of polymer structures. Therefore, ESC resistance (ESCR) is an important performance parameter for different applications of SCPs.

The phenomenon and mechanism of ESC have been extensively studied for the past several decades. In 1959, ESC was first officially defined by Howard [2] as failure in surface-initiated brittle fracture of a polyethylene specimen or part under polyaxial stress in contact with a medium in the absence of which fracture does not occur under the same conditions of stress. Although ESC has been recognized as a critical problem for amorphous polymers such as acrylonitrile-butadiene-styrene (ABS), polycarbonates (PC), poly(methyl methacrylate) (PMMA) and polyvinyl chloride (PVC), it is also a concern for crystalline polymers as has been well-documented for polyethylene in various applications [3]. The ESC mechanisms are complex since they strongly depend on the microstructure of polymers, the level of stress or strain the polymer is subjected to

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and the exposure time and temperature, as well as the compatibility and solubility parameter of the stress-cracking agent into the polymer. The basic mechanism of ESC is similar to slow crack growth (SCG), which is related to the voids initiation and coalescence, and crazes formation and growth. It is believed that the diffusion of molecules from the aggressive

ESCR characterization methods and major factors affecting the ESC behavior.

ESCR MEASUREMENT

Although there is a number of different test methods developed to characterize ESCR of polymeric materials, they can

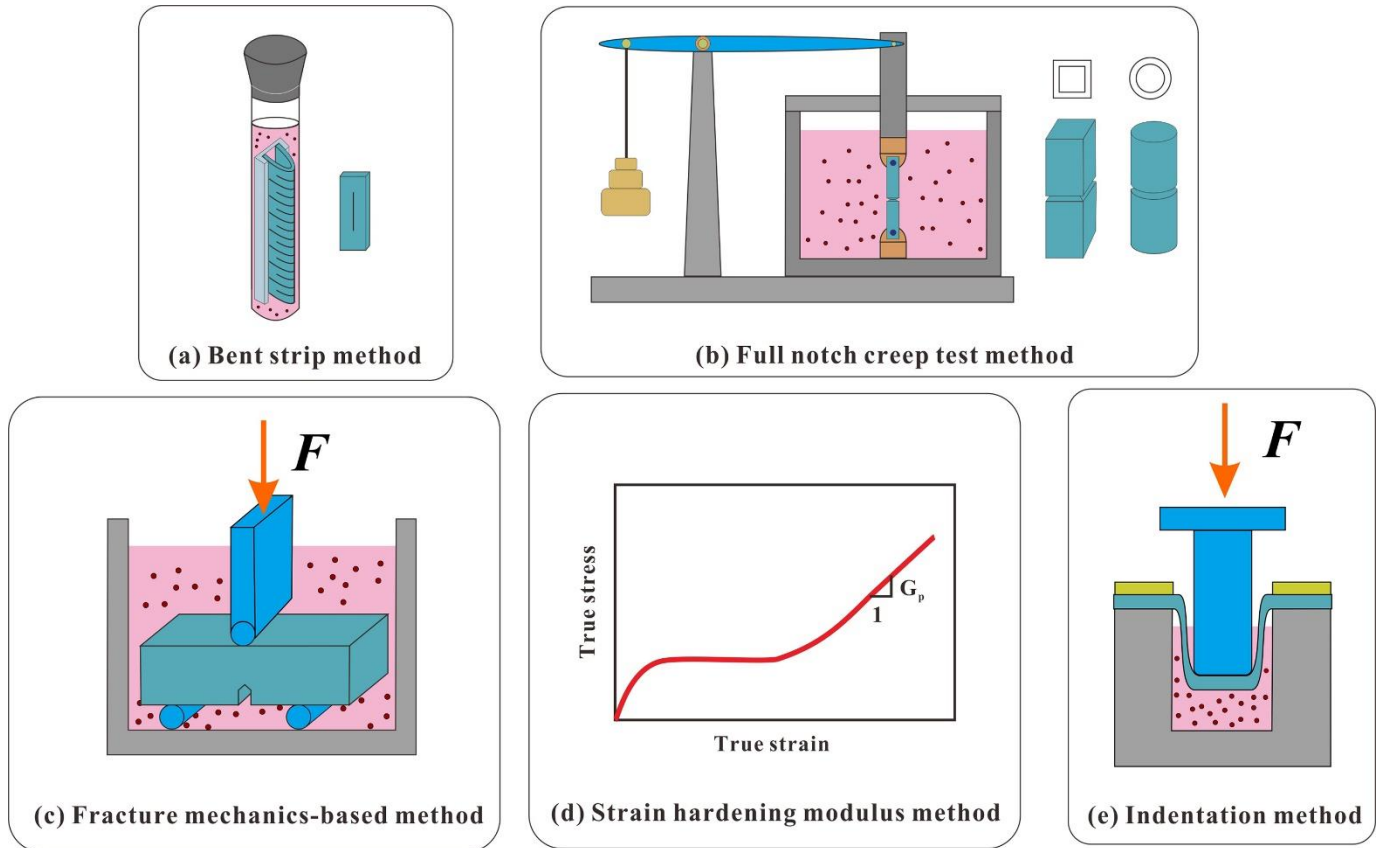


Figure 1 Summary of ESCR characterization methods

agent in to the polymeric materials causes stress-induced swelling and plasticization, which facilitate the craze formation and growth [4].

A variety of testing procedures, as shown in Fig. 1, have been developed and adopted in industry to characterize ESCR of different polymeric materials, such as Bell Telephone Test (ASTM D 1693-15) adopted for ethylene plastics [5] and Bent Strip Test method (ISO 22088-3:2006) for rigid polymers [6], Full Notch Creep Test (FNCT, ISO 16770:2004) [7,8]. However, due to the long test duration (sometimes more than 1000 hours) and critical specimen preparation (i.e. pre-notch procedure) required for such standardized test methods, alternative characterization methods based on the measurement of strain hardening modulus or indentation test have been developed [9]. Furthermore, fracture mechanics-based approaches were proposed to provide information about fracture mechanisms of ESC [10]. This paper summarized the current

be divided into the following categories: constant tensile stress/strain test methods including Bent strip test method (Fig.1(a)), FNCT (Fig.1(b)), fracture mechanics test method (Fig.1(c)), strain hardening modulus test method (Fig.1(d)) and indentation test method (Fig.1(e)).

2.1 Constant strain test methods

Bent strip test (ISO 22088-3:2006 or ASTM D 1693-15), which is the most commonly used method for ESCR evaluation in industry, can be considered as a constant-strain-based test method. In the bent strip test, the pre-notched specimens are slowly bent into “U” shape and placed in a specimen holder. The holder is then placed in a glass tube containing aggressive agent such as Igepal CO-630. The tube is sealed and placed in a constant-temperature (elevated temperature) bath. The number of test specimens that exhibit visible cracks is recorded as a function of time. Constant-strain-based tests are convenient and cheap to perform, however the critical pre-notch procedure for the specimen preparation and visual inspection of the cracks

periodically will lead to significant scattering of the ESCR results. In addition, as improved performance PE pipe materials were developed, the time to failure measured in constant strain tests increased. More importantly, due to stress relaxation the bending stress applied to the specimens will decay to such a low level that it will take extremely long time before the occurrence of cracks.

2.2 Constant stress test methods

FNCT, as shown in Fig. 1(b) belongs to constant stress test method, which applies a constant load on the notched specimen immersed in aggressive chemical agent at elevated temperature until the occurrence of fracture. In North America, Pennsylvania Notch Test (PENT, ASTM F1473) is widely used to measure SCG resistance of PE pipe materials. The main difference between the two test methods is that the FNCT is conducted in aggressive chemical agent on a square section bar with four coplanar notches in the specimen, whereas the PENT is performed in air on rectangular section bar with one main notch and two side notches. Notch Constant Load Test (NCLT, ASTM D5397) is another constant stress test method designed for the ESCR evaluation for Polyolefin geomembranes [11–13]. The failure mechanisms of PE pressurized pipe, represented by the plot of applied hoop stress versus failure time as shown in Fig. 2 can be divided into three characteristic regions: ductile failure, brittle failure and degradation or aging failure. ESC is a brittle-type failure, therefore, it is critical to choose proper stress level applied in constant stress tests to ensure the occurrence of brittle fracture.

2.3 Fracture mechanics methods

In order to provide information about failure mechanisms of ESC and the structural parameters that govern this phenomenon, attempts have been made to develop quantitative methods for the estimation of ESCR of PE materials based on fracture mechanics. The fracture mechanics approach is to conduct three point bending tests on single edge notched (SENB) specimens. The while ESC process including crack initiation and propagation is investigated based on the measurement of key fracture parameters, such as strain energy release rate and crack speed.

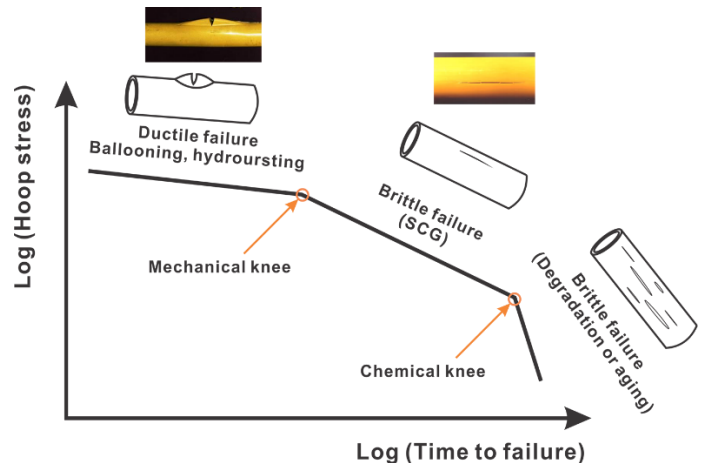


Figure 2 Schematic illustration of the failure behavior of pressurized PE pipes

2.4 Strain hardening modulus method

ESC is believed to be caused by the disentanglement of inter-lamellar links, including tie molecules and entanglements. Since these two are also critical factors for the strain hardening behavior of PE, it follows that ESC and strain hardening behavior of PE can be correlated. As a result, a convenient and reliable test method has been developed to rank the ESCR of PE materials based on the measurement of strain hardening modulus using simple tensile test. Experimental results show that the tensile strain hardening modulus measured at elevated temperature correlates well with the ESCR determined from conventional test methods such as NCLT. A specific concern for this method is the strain hardening modulus is measured from specimens tested in air, not immersed in aggressive chemical agent.

2.5 Indentation test method

Recently, a new approach, which applies indentation tests to generate deflection in PE plates and records time for crack generation under constant displacement during the exposure to aggressive chemical agent, was developed for characterizing ESCR of PE materials [14]. The comparison of ESCR measured from the indentation test and Bent strip test methods confirms the effectiveness of the new approach to quantify ESCR for PE material. The test time required for the new approach has been reduced significantly (less than 2% of the time required for Bent strip test method). In addition, the new method does not require the critical specimen preparation procedure, i.e. pre-notch, thus increasing the reproducibility of the ESCR results. Unlike the Bent strip test method which allows multiple specimens tested at the same time, the new approach relies on the curve of stress decay versus time to detect the crack generation, thus only one specimen can be tested at each time.

INFLUENCING FACTORS

ESCR of semi-crystalline polymers is shown to be highly dependent on molecular parameters, such as molecular weight (MW) [15], crystallinity [16], short chain branch (SCB) contents [17] and lamellar surface area (LSA) [11], testing temperature and the chemical agent [18]. Experimental results suggest the resistance to ESC improves with increasing molecular weight and SCB contents, and with decreasing crystallinity (or density). A larger MW and SCB would result in a higher probability of formation of tie molecule, which is a critical parameter governing the ESC behavior of PE materials. Results presented in indicate that PE with LSA have more interlamellar entanglements, thus better ESCR [19]. Besides, it has been found that the most prominent cracking agents for polyethylene are detergents with Igepal Co-630 solution being a prime example.

DISCUSSION AND CONCLUSIONS

Environmental stress cracking (ESC) is a critical problem that needs to be considered for many polymers including PE in end use applications, especially in natural gas transportation and nuclear power plant. Although a variety of test methods have been developed for the ESCR characterization and ranking, there are limitations for each method. A specific concern for constant stress and constant strain test methods is its inability to isolate the yield stress property as a parameter independent of polymer's mechanical properties. For example, crack in high density polyethylene (HDPE) grows faster than that in low density polyethylene (LDPE) in a constant-strain test whereas the opposite results are observed in a constant stress ESCR testing. This is because the stiffness of HDPE is much higher than that of LDPE, under constant strain condition the higher stress level in HDPE leads to earlier occurrence of cracks. By contrast, LDPE is stressed close to or beyond its yield point more readily than HDPE, and is thus more susceptible to failure. Another limitation for the current ESCR testing method except indentation test method is the quality of the pre-notch introduced to the specimens varies, resulting in poor reproducibility of the test results. While the current ESCR testing methods provide a comparative material ranking, they can not be used for the lifetime prediction of polymer products. Therefore, the development of accurate, reliable and convenient testing method is envisaged.

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REFERENCES

- [1] Kiass, N., Khelif, R., Boulanouar, L., and Chaoui, K., 2005, "Experimental Approach to Mechanical Property Variability through a High-Density Polyethylene Gas Pipe Wall," *J. Appl. Polym. Sci.*, 97(1), pp. 272–281.
- [2] Howard, J. B., 1959, "A Review of Stress-Cracking in Polyethylene," *SPE J.*, 15, p. 397.
- [3] Robeson, L. M., 2013, "Environmental Stress Cracking: A Review," *Polym. Eng. Sci.*, 53(3), pp. 453–467.
- [4] Wee, J.-W., Zhao, Y., and Choi, B.-H., 2015, "Observation and Modeling of Environmental Stress Cracking Behaviors of High Crystalline Polypropylene Due to Scent Oils," *Polym. Test.*, 48, pp. 206–214.
- [5] Chen, Y., 2014, "Investigations of Environmental Stress Cracking Resistance of HDPE/EVA and LDPE/EVA Blends," *J. Appl. Polym. Sci.*, 131(4).
- [6] Hopmann, C., Borchmann, N., Koch, S., and Alperstein, D., 2018, "Influencing the Environmental Stress Cracking Resistance of Amorphous Thermoplastic Parts by the Example of Polycarbonate and Water," *Polym. Eng. Sci.*
- [7] Schilling, M., Niebergall, U., and Böhning, M., 2017, "Full Notch Creep Test (FNCT) of PE-HD—Characterization and Differentiation of Brittle and Ductile Fracture Behavior during Environmental Stress Cracking (ESC)," *Polym. Test.*, 64, pp. 156–166.
- [8] Schilling, M., Niebergall, U., Alig, I., Oehler, H., Lellinger, D., Meinel, D., and Böhning, M., 2018, "Crack Propagation in PE-HD Induced by Environmental Stress Cracking (ESC) Analyzed by Several Imaging Techniques," *Polym. Test.*, 70, pp. 544–555.
- [9] Kurelec, L., Teeuwen, M., Schoffeels, H., and Deblieck, R., 2005, "Strain Hardening Modulus as a Measure of Environmental Stress Crack Resistance of High Density Polyethylene," *Polymer*, 46(17), pp. 6369–6379.
- [10] Kamaludin, M. A., Patel, Y., Williams, J. G., and Blackman, B. R. K., 2017, "A Fracture Mechanics Approach to Characterising the Environmental Stress Cracking Behaviour of Thermoplastics," *Theor. Appl. Fract. Mech.*, 92, pp. 373–380.
- [11] Cheng, J. J., Polak, M. A., and Penlidis, A., 2011, "Influence of Micromolecular Structure on Environmental Stress Cracking Resistance of High Density Polyethylene," *Tunn. Undergr. Space Technol.*, 26(4), pp. 582–593.
- [12] Sardashti, P., Scott, A. J., Tzoganakis, C., Polak, M. A., and Penlidis, A., 2014, "Effect of Temperature on Environmental Stress Cracking Resistance and Crystal Structure of Polyethylene," *J. Macromol. Sci. Part A*, 51(3), pp. 189–202.
- [13] Sardashti, P., Stewart, K. M. E., Polak, M., Tzoganakis, C., and Penlidis, A., 2019, "Operational Maps between Molecular Properties and Environmental Stress Cracking Resistance," *J. Appl. Polym. Sci.*, 136(4), p. 47006.
- [14] Jar, B., and Zhang, Y., 2016, "Evaluation of a New Approach for Characterizing Environmental Stress Cracking Resistance (ESCR) of Polyethylene," *ASME 2016 Pressure Vessels and Piping Conference*, American Society of Mechanical Engineers, p. V06BT06A049 – V06BT06A049.
- [15] Huang, Y.-L., and Brown, N., 1988, "The Effect of Molecular Weight on Slow Crack Growth in Linear

Polyethylene Homopolymers,” *J. Mater. Sci.*, 23(10), pp. 3648–3655.

[16] Soares, J. B., Abbott, R. F., and Kim, J. D., 2000, “Environmental Stress Cracking Resistance of Polyethylene: The Use of CRYSTAF and SEC to Establish Structure–Property Relationships,” *J. Polym. Sci. Part B Polym. Phys.*, 38(10), pp. 1267–1275.

[17] Brown, N., Lu, X., Huang, Y., Harrison, I. P., and Ishikawa, N., 1992, “The Fundamental Material Parameters That Govern

Slow Crack Growth in Linear Polyethylenes,” *Plast. Rubber Compos. Process. Appl.*, 17(4), pp. 255–258.

[18] Brown, H. R., 1978, “A Theory of the Environmental Stress Cracking of Polyethylene,” *Polymer*, 19(10), pp. 1186–1188.

[19] Sardashti, P., Stewart, K. M. E., Polak, M., Tzoganakis, C., and Penlidis, A., 2019, “Operational Maps between Molecular Properties and Environmental Stress Cracking Resistance,” *J. Appl. Polym. Sci.*, 136(4), p. 47006.