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Procedia Structural Integrity 2 (2016) 227–234

Structural Integrity Procedia

www.elsevier.com/locate/procedia

21st European Conference on Fracture, ECF21, 20-24 June 2016, Catania, Italy

Fracture mechanics testing for environmental stress cracking in thermoplastics

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Abstract

Under the combined influence of an aggressive environment and applied stress, engineering thermoplastics may undergo a phenomenon known as environmental stress cracking (ESC). This can result in adverse effects such as embrittlement and premature failure in service, due to the growth of environmentally-induced cracks to critical sizes, with little to no fluid absorption in the bulk material. Fracture mechanics is proposed as a suitable scheme to study and quantify ESC, with the aim being to obtain characterising data for different polymer-fluid combinations of interest, as well as to develop a reliable fracture mechanics test protocol. In the proposed method, slow crack growth is monitored to assess the effect of a range of applied crack driving forces (K, or alternatively G) on observed crack speeds, as opposed to simply measuring time-to-failure. This paper presents the results of experiments performed on the following materials: linear low density polyethylene (LLDPE) in Igepal solution and high impact polystyrene (HIPS) in sunflower oil. A discussion of the various issues surrounding the data analysis for these long-term tests is also included, as the attainment of consistent and repeatable results is critical for a method to be internationally standardised, which is a goal of the European Structural Integrity Society (ESIS) Technical Committee 4 from whose interest this work is drawn.

Peer-review under responsibility of the Scientific Committee of ECF21.

Keywords: environmental; stress cracking; thermoplastics; fracture mechanics; compliance

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Peer review under responsibility of the Scientific Committee of ECF21.
10.1016/j.prostr.2016.06.030

Nomenclature

a	crack length
B	specimen thickness
C	specimen compliance
E	Young's modulus
G	strain energy release rate
K	stress intensity factor
P	applied load
S	span
W	specimen width
x	normalised crack length
Y	shape factor
δ	central deflection of specimen
ε	applied strain
ρ	notch tip radius

1. Introduction

Environmental stress cracking (ESC) occurs in certain thermoplastics when immersed in particular environments. In such cases, it is acknowledged that ESC is not due to bulk absorption of the environment, but rather is the result of the environment acting on an existing, loaded defect in the material, as noted by Williams (1984). ESC causes the incubation, initiation and resulting propagation of cracks leading to fast fracture of components, at applied stress levels significantly lower than normally required for the fracture of the material in air at quasi-static speeds. Incubation is defined here as the time between initial loading and the first instance of an increase in the crack length, i.e. crack initiation.

ESC has been investigated using several methods including chemical compatibility, time-to-failure, and strain hardening. The former for example, as in Hansen and Just (2001), has enabled the identification of polymer-liquid pairs having a higher tendency to stress crack as judged from the proximity of their solubility numbers. This allows the determination of polymer-liquid pairs which are inert and thus safe for mutual contact. The downside of these methods, however, is that they do not provide sufficient information for use as design criteria in the development of load-bearing components. Existing standards in usage, such as the Bell telephone (ASTM (2015a)), full-notch creep (ISO (2004)) and bent strip tests (ISO (2006)) tend only to provide a relative indicator of stress-cracking resistance; furthermore, they do not provide any information as to the mechanisms affecting the fracture rate of the material.

A fracture mechanics framework is thus considered suitable to study ESC phenomena, by loading notched specimens in environment and subsequently monitoring the rate of crack growth, i.e. crack speed under different levels of crack driving force (K, G). It was shown previously by Williams (1978) that unique relations exist between the crack driving force and crack speed, for any given polymeric material and temperature. An advantage of such tests is that they take shorter times than those measuring time-to-failure; in addition, the tests are able to distinguish between the different phases of crack growth, namely incubation, initiation and propagation.

2. Development of test method

The method adopted draws upon existing standards to measure K and G at initiation, such as for quasi-static plane strain fracture toughness (ISO (2000)). The specimen proposed is the single edge notched bend (SENB) configuration in three-point bend, as shown in Fig. 1. As with quasi-static fracture tests, the specimens are notched by razor sliding, taking care not to press the blade into the notch to avoid introducing residual stresses which may affect the test results.

The constant load (increasing G) set-up is a robust arrangement which allows for ESC testing across a wide range of applied crack driving forces, increasing from a fraction of the quasi-static fracture toughness (depending on the

initial crack value). Alternative fracture mechanics test configurations, such as the buckled plate and double cantilever beam specimens as in Andena et al. (2013), while valid, only provide limited ranges of G and K in comparison to SENB.

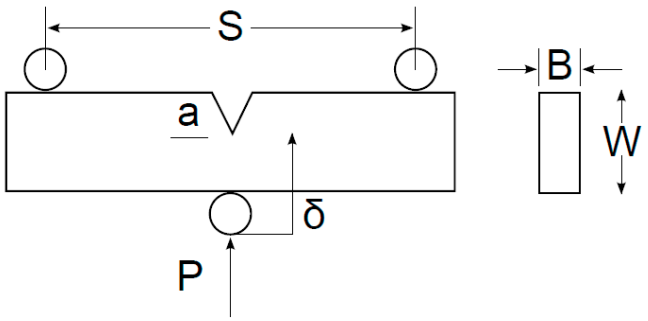

Fig. 1. Single edge notched bend (SENB) specimen. B is the specimen thickness, W its width, a is the crack length, P is the applied load, δ is the central deflection of the specimen, and S is the span.

2.1. Data capture

After preliminary testing, it was decided that crack growth measurements via the compliance method were preferred over optical measurements. An advantage of the latter is that it allows simultaneous measurement of specimen creep during testing; thus, the time-dependent modulus of the material may be obtained. The optical method can only be used with environments that are relatively clear, and as such is not suited for environments that obscure the specimen from view, for example Igepal solution. Stress whitening may also appear on the specimen sides ahead of the notch tip, complicating the optical measurement. Fig. 2 shows typical output from the compliance method:

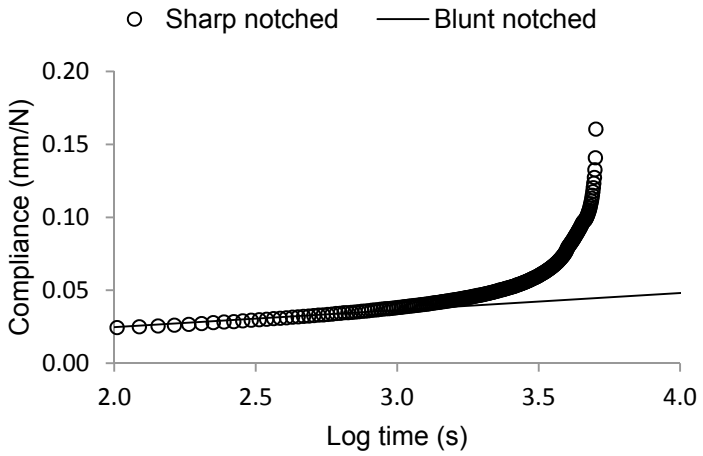

Fig. 2. Typical example of compliance vs. time curves for LLDPE/Igepal.

Eqn. 1, adapted from Bakker (1990), relates the (normalised) crack length, x to P and δ, as measured throughout the test using a linear variable differential transformer (LVDT) as a displacement transducer:

$$C = \frac{\delta}{P} = \frac{16}{EB}\left[1 + \frac{9}{2}\int_0^x Y^2 x dx\right] = \frac{16}{EB}\phi(x) \tag{1}$$

where C is the specimen compliance, E is the Young's modulus of the specimen material, Y is the shape factor and φ is the bracketed term, the latter two both depending on x. For an equivalent blunt notched specimen, the compliance expression is similar, however in this case the bracketed term is fixed as the crack does not extend. The compliance thus increases only due to creep:

$$C_b = \frac{\delta}{P} = \frac{16}{EB}\left[1 + \frac{9}{2}\int_0^{x_b} Y^2 x dx\right] = \frac{16}{EB}\phi_b(x_b) \tag{2}$$

where C_b is the compliance for a blunt notched specimen (notch tip radius $\rho \approx 1000$ μm), x_b is its crack length, and φ_b is a constant depending on x_b. By monitoring C(t) and C_b(t), φ(x) can be determined, which is used to find x and a. Crack speeds can then be calculated by differentiating successive values of a. Numerical values of C_b/C as related to x are provided in Table 1; the case with $x_b = 0.35$ is provided.

Table 1. Numerical and fitted values of C_b/C for $x_b = 0.35$.

x=a/W	C_b/C (numerical)	C_b/C (fitted)	Error *(%)*
0.25	1.326	1.338	0.9
0.30	1.161	1.161	0.0
0.35	1.000	0.995	0.5
0.40	0.847	0.842	0.6
0.45	0.706	0.701	0.6
0.50	0.576	0.573	0.6
0.55	0.459	0.456	0.7
0.60	0.357	0.352	1.4

For x > 0.25, the plot of Cb/C against the square of the (normalised) ligament (1-x) tends to a straight line, for which a fit may be utilised:

$$\frac{C_b}{C} = p(1-x)^2 + q \Rightarrow x = 1 - \left[\left(\frac{C_b}{C} - q\right)\left(\frac{1}{p}\right)\right]^{\frac{1}{2}} = \frac{a}{W} \tag{3}$$

where p and q are constants. The relevant values of p and q, together with φ_b are provided for different values of x_b in Table 2:

Table 2.Values of φ_b, p and q as a function of x_b.

x_b	Φ_b	p	q
0.00 (un-notched)	1.00	1.30	0.02
0.25	1.43	1.85	0.03
0.30	1.63	2.11	0.03
0.35	1.90	2.45	0.04
0.40	2.24	2.90	0.04
0.45	2.69	3.48	0.05
0.50	3.29	4.26	0.06

2.2. Data processing

ESC tests bear some resemblance to fatigue tests in that both are slow crack growth processes involving large amounts of collected data. In the case of fatigue tests, it was noted by De Vries et al. (2004) that the recommended data processing procedures as per existing standards are not fully adequate, and result in large amounts of discarded data causing deviations in practice from the published protocols.

Different processing schemes were evaluated on sample output data produced using a MATLAB script to ascertain their effects and to establish an optimum processing scheme to be followed. The desired objective is to recommend a smoothing procedure which identifies trend-lines while reducing scatter to a minimum. The script in question generates data about a base curve with a specified amount of scatter (Fig. 3), while keeping the crack length constant or increasing, i.e. not permitting any negative crack growth, as in physical ESC experiments.

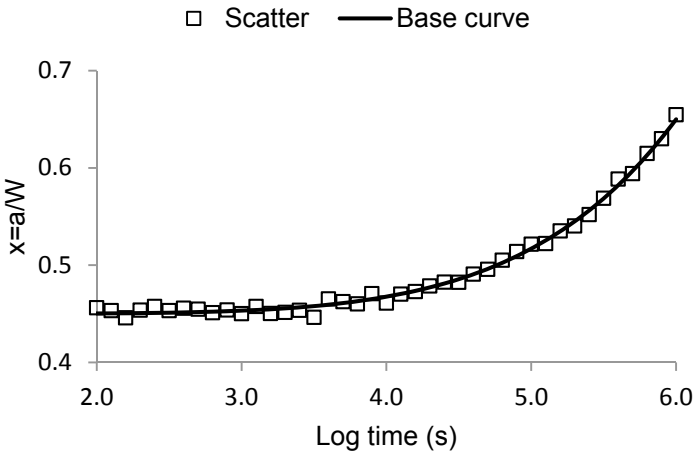

Fig. 3. Simulated crack data using with x^6 polynomial as a base curve.

The data should be sufficiently discrete to facilitate further processing; there should be adequate crack extension between subsequent measurements, amounting to at least 10 times the measurement accuracy as recommended by ASTM (2015b). While the LVDT output can be highly precise, a resolution value of the order of 10 μm is considered suitable (similar to optical measurements); thus an interval of 0.1 mm (100 μm) is suggested between data points.

It is recommended to smooth both the raw crack values and the resulting crack speeds. Five-point simple averaging and secant differentiation are adequate schemes for smoothing and differentiating, respectively. In most

cases, no further processing or data reduction need subsequently be done. Polynomials should not normally be fitted to the raw data, so as not to pre-define the shape of the compliance curve, which may mask important features in the results.

3. Materials

The material was received in the form of compression-moulded plates, which were then machined down, annealed and conditioned prior to testing. There were 4 materials in total: 2 grades of linear low density polyethylene (LLDPE or PE), and 2 grades of high impact polystyrene (HIPS). The specimen dimensions were B = 6 mm, W = 25 mm, and x ≈ 0.35. A span, S of 100 mm was used in all the tests. Side-grooving was not performed on the test specimens.

Table 3. Material and environment properties.

Material	Environment	Temperature	ESC test method	ESC resistance	
LLDPE A	10% Igepal CO-630	50 °C	Bell telephone test	1000 h	Andena et al. (2012)
LLDPE B				20 h	
HIPS A	Sunflower oil	Room temperature	Bent strip method	94 $\varepsilon_r/\varepsilon_0$%	Andena et al. (2013)
HIPS B				26 $\varepsilon_r/\varepsilon_0$%	

where $\varepsilon_r/\varepsilon_0$ is the ratio of applied strain. Both "A" grades were quoted as having a higher ESC resistance compared to the "B" grades. The fluid chosen for each test material was either one which has been known to affect the material in usage (sunflower oil), or alternatively one which has historically been used in ESC tests for that material (Igepal solution).

4. Results and discussion

Figs. 4 and 5 show the results of tests performed; three different stages of crack growth are noticeable for each material: two stages of similar slope separated by a transition stage, as in Williams (1984):

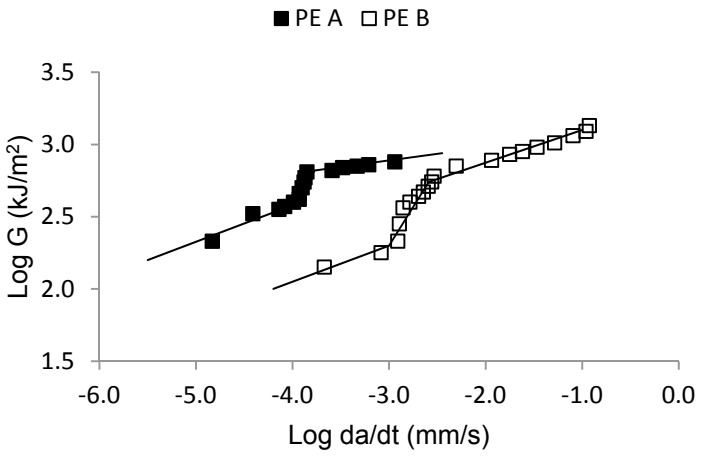

Fig. 4. G vs. crack speed data for for.LLDPE/Igepal. The lines are of slope 0.25 and the transitions slope 1, except for the upper line in PE A which is of slope 0.1.

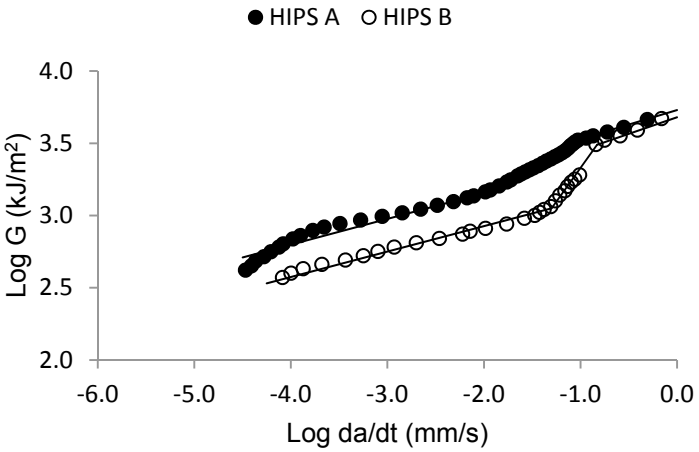

Fig. 5. G vs. crack speed data for.HIPS/sunflower oil. The lines are of slope 0.2, except for the transitions, which are of slope 0.5 (HIPS A) and 1 (HIPS B).

In each case, data for the material with lower ESC resistance as judged by conventional ESC tests falls to the right of and below the material with higher ESC resistance. This means that for any value of applied crack driving force (G), the material with the lower ESC resistance cracks faster than that with the higher ESC resistance. One way to quantify this is to assign a "critical crack speed" criterion corresponding to the right-most "knee" where the transition stage changes to the upper slope: as such, the critical crack speeds were 0.2 μm/s and 3 μm/s respectively for PE A and PE B, and 90 μm/s and 150 μm/s respectively for HIPS A and HIPS B. All other things being constant, a material with a lower critical crack speed indicates a higher ESC resistance.

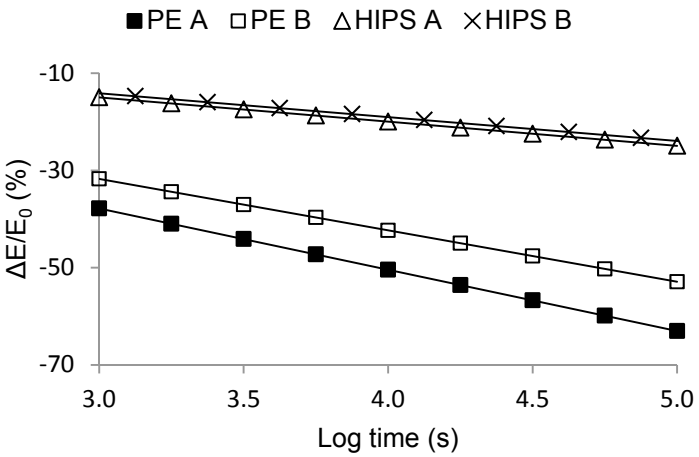

Fig. 6. Relative modulus change vs. time. $E_0 = E(t=0)$; $\Delta E = E(t) - E_0$.

An alternative method to quantify ESC from the tests conducted is by assigning a "critical time" criterion corresponding to the time taken for the crack to initiate, i.e. the incubation time. Taken as such, the critical times

were 1000 s and 300 s for PE A and PE B, and 1800 s and 900 s for HIPS A and HIPS B, on average, respectively. Again, all other things being constant, a material with a higher critical time indicates a higher ESC resistance.

Fig. 6 shows the relative modulus change with time for the materials tested. For the time-scales involved, it is shown that creep is not negligible, particularly at elevated temperatures, and thus should be included in the compliance calculations.

5. Conclusions

Constant load tests were performed on SENB specimens in environment, with crack growth monitored via the compliance method using an LVDT. It was found in both the LLDPE/Igepal and HIPS/sunflower oil combinations that the less ESC-resistant grade cracked sooner, and at a higher rate for the same values of crack driving force as compared to the more ESC-resistant grade. The tests demonstrate the applicability of the SENB test configuration and data processing method to determine the relative ESC resistance of materials which are similar to one another; additionally, the prospect of assigning an ESC parameter appears to be ever more realistic.

A considerable amount of scatter is still present across repeats of the same test. Further work includes attempting to understand and reduce the source of this scatter, in order to improve the repeatability and reproducibility of the fracture mechanics method for ESC testing. The method can also be further extended, as proposed by Andena et al. (2009), to provide life predictions for components, by integrating the rate of crack growth. Fractographic analysis of each stage in the G-crack speed curve is also envisaged, to help further explain the mechanisms contributing to ESC in each stage and thus assist in defining the rate-determining step(s) in the ESC process.

Acknowledgements

The authors gratefully acknowledge financial support from the Sultan Haji Hassanal Bolkiah Foundation, and also wish to thank Leonardo Castellani (Versalis S.p.A.) for the supply of test materials.

References

Andena, L., Rink, M., Frassine, R. and Corrieri, R., 2009. A fracture mechanics approach for the prediction of the failure time of polybutene pipes. Engineering Fracture Mechanics, 76(18), pp.2666-2677.

Andena, L., Castellani, L., Castiglioni, A., Mendogni, A., Rink, M. and Sacchetti, F., 2013. Determination of environmental stress cracking resistance of polymers: Effects of loading history and testing configuration. Engineering Fracture Mechanics, 101, pp.33-46.

Andena, L., Mendogni, A., Rink, M., Sacchetti, F., Castellani, L., Castiglioni, A. and Adib, A., 2012. Environmental crack initiation and propagation in polyethylene under different loading conditions. In: Deformation, Fracture and Yield of Polymers, 2012, the Netherlands.

ASTM Standard D1693, 2015. Standard test method for environmental stress-cracking of ethylene plastics. ASTM International, West Conshohocken, PA.

ASTM Standard E647, 2015. Standard test method for measurement of fatigue crack growth rates. ASTM International, West Conshohocken, PA.

Bakker, A., 1990. Compatible compliance and stress intensity expressions for the standard three-point bend specimen. Fatigue & Fracture of Engineering Materials & Structures, 13(2), pp.145-154.

De Vries, S.B., Horn, C.T., Ament, P.C.H. and Bakker, A., 2004. The adaptive da/dN method: a solution to the problem of processing crack growth data. Fatigue & Fracture of Engineering Materials & Structures, 27(7), pp.585-594.

Hansen, C.M. and Just, L., 2001. Prediction of environmental stress cracking in plastics with Hansen solubility parameters. Industrial & Engineering Chemistry Research, 40(1), pp.21-25.

ISO Standard 13586, 2000. Plastics – Determination of fracture toughness (GIC and KIC) – Linear elastic fracture mechanics (LEFM) approach. ISO, Geneva.

ISO Standard 16770, 2004. Plastics – Determination of resistance to environmental stress cracking (ESC) of polyethylene – Full-notch creep test (FNCT). ISO, Geneva.

ISO Standard 22088-3, 2006. Plastics – Determination of resistance to environmental stress cracking (ESC) – Part 3: Bent strip method. ISO, Geneva.

Williams, J.G., 1978. Applications of linear fracture mechanics. In Failure in Polymers (pp. 67-120). Springer Berlin, Heidelberg.

Williams, J.G., 1984. Fracture mechanics of polymers. Ellis Horwood Limited, Chichester.