Determination of the Stress Cracking Resistance of HDPE Geomembranes by using Accelerated Test Methods

Conference Paper · February 2015

5 authors, including:

Helmut Zanzinger
SKZ - German Plastics Center
69 PUBLICATIONS **112** CITATIONS
SEE PROFILE

Kurt Engelsing
SKZ - German Plastics Center
19 PUBLICATIONS **84** CITATIONS
SEE PROFILE

Mirko Wenzel
SKZ - German Plastics Center
19 PUBLICATIONS **153** CITATIONS
SEE PROFILE

Geosynthetics 2015
February 15-18, Portland, Oregon

Determination of the Stress Cracking Resistance of HDPE Geomembranes by using Accelerated Test Methods

Helmut Zanzinger, SKZ German Plastics Center, Wuerzburg, Germany, h.zanzinger@skz.de
Britta Gerets, SKZ German Plastics Center, Wuerzburg, Germany, b.gerets@skz.de
Mirko Wenzel, SKZ German Plastics Center, Wuerzburg, Germany, m.wenzel@skz.de
Sebastian Hausmann, SKZ German Plastics Center, Wuerzburg, Germany, s.hausmann@skz.de
Kurt Engelsing, SKZ German Plastics Center, Wuerzburg, Germany, k.engelsing@skz.de

ABSTRACT

Accelerated notched constant tensile load (ANCTL) tests were developed using alternative detergents and higher test temperatures, e.g. 70°C, to reduce the times to failure. Results showed the time to failure was reduced by a factor of 5 in comparison with standard notched constant tensile load (NCTL) tests. Another new method was used to determine the resistance against slow crack growth (SCG) by a tensile test performed at 80°C. The slope of such a tensile curve above its natural draw ratio (i.e. strain hardening modulus) correlates with the measured failure times determined by NCTL tests. A very good correlation between strain hardening modulus and failure times in single-point NCTL tests were found.

1. INTRODUCTION

The stress dependent End of Life (EoL) of HDPE geomembranes can be described either as ductile failure under high tensile stresses – this means HDPE geomembranes will fail under tensile stresses beyond the yield stress – or as brittle failure caused by tensile stresses less than its yield stress. The second one is called stress cracking (SC) failure. The EoL of HDPE geomembranes is furthermore limited by thermo-oxidative ageing.

This paper is devoted to stress cracking resistance (SCR). Stress cracking is caused by tensile stresses less than the materials short-time mechanical strength, in case of HDPE less than its yield stress. The failure occurs as a “sharp” break in a brittle failure mode – this means with no or nearly no elongation. SCR of a material depends on the applied stress. For high stresses beyond a certain “transition stress” the failure mode is ductile and those breaks occur rather fast. Underneath that “transition stress” the failure mode changes to a brittle failure and the failure time is more stress dependent than the ductile failure mode.

The traditional test in the field of HDPE geomembranes is the NCTL test (Hsuan and Koerner 1995). The NCTL test uses notched specimens, detergents and elevated temperatures to reduce failure times. With high stress crack resistant HDPE resins even the NCTL testing times nowadays become extremely long. Using the single point NCTL (SP-NCTL) tests (ASTM D5397, Appendix and EN 14576) HDPE geomembranes with modern resins may already last thousands of hours at a load ratio of 30% yield stress. The determination of the full stress-failure-curve may last extremely long. The test is described in ASTM D5397. Both methods use a detergent which consists of 10% by weight of surface-active agent such as nonylphenoxy polyethylene-oxyethanol, diluted with 90% by weight of deionised water. The test temperature of the test liquid shall be (50 ± 1)ºC for the whole time of the exposure.

The testing time for HDPE geomembranes depends on the notching, the temperature and the detergent used. For shorter test durations the described methods can be applied with the exception of test temperature and the detergent. The temperature can be increased and a more accelerating detergent can be used to shorten the times to failure. The test temperature of the test liquid shall be up to (80 ± 1)°C for the whole time of the exposure. The detergent shall consist of 10% by weight of surface-active agent such as lauramine oxide, diluted with 90% by weight of deionised water. This accelerated NCTL test is called ANCTL test (see section 3.1).

The methods are applicable to virgin and exposed HDPE geomembranes. The new ANCTL test might be useful for quality control tests. But even for SC tests for the determination of the full stress-failure-curve – at tensile loads less than 30% yield stress – the “real” field loads of a HDPE geomembrane can be simulated in a much shorter time by using the new detergent at higher test temperatures.

A further but extremely fast new method for determining the resistance of HDPE geomembranes against SCG is a tensile test performed at 80°C. The slope of such a tensile curve above its natural draw ratio (i.e. strain hardening modulus) correlates well with the measured failure times determined by conventional SCR tests. This strain hardening

(SH) test can additionally be used to determine the SCR of HDPE geomembranes. Very good correlations between strain hardening modulus <G_p> and failure times in SP-NCTL tests have been found (see section 3.2.3).

2. ROUTINE LABORATORY TEST METHOD: NCTL TEST

In the early 90's the NCTL test has been developed. A constant tension load is applied and stress relaxation cannot occur. The location of the highest stress is clearly defined as a sharp notch induces a high stress concentration at its tip. This accelerates the crack growth and shortens the testing time. Notching generates a plane-strain-condition in the specimen. For geomembranes the cross machine direction (CMD) is the more sensitive direction to SC.

If the SP-NCTL test is carried out at different laboratories it is known from the authors' experience that the absolute values of failure times may vary from one laboratory to another resulting often in inconsistent failure times. The reasons are the sensitivity of the test on e.g. the determination of the reference values in the tensile test, the notch quality, the age of the detergent, the flow conditions in the test medium and the temperature distribution in the bath.

Furthermore, if NCTL failure times of more than 1000 hours are reached, slowly thermo-oxidative ageing begins. So the failure is not only limited by SCG anymore, but by thermo-oxidative ageing of the material also. Because of the mentioned long testing times and great dependence of failure times on various test conditions there is a great need for faster, reliable and robust test methods to predict SCG behaviour.

3. ALTERNATIVE LABORATORY TEST METHODS

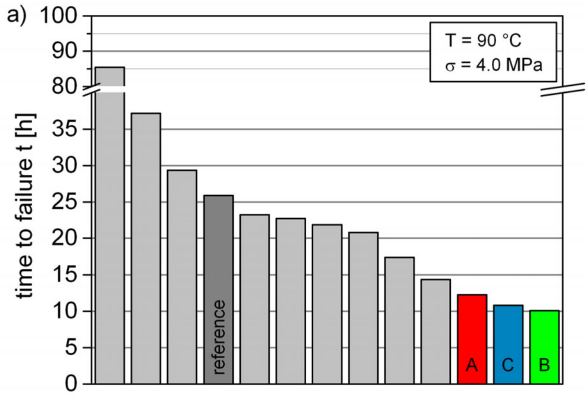
3.1 ANCTL Test

3.1.1 Screening Tests on Alternative Detergents

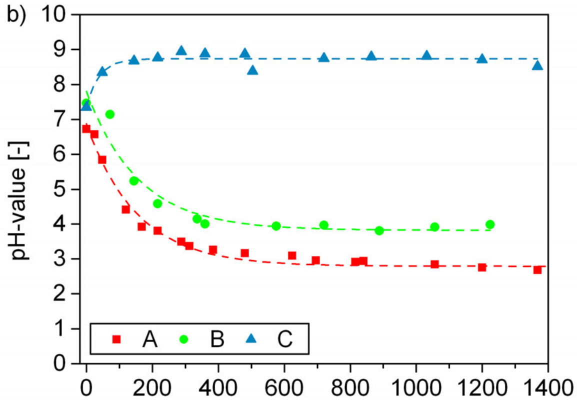
In an intercomparison study of different European laboratories it was found that detergents such as nonylphenolpolyglycolether lead to large scatter in time to failure measured by different laboratories. As no differences due to notching were found and the same detergent batch was used for all tests, the scatter seems to be caused by different flow conditions in the testing devices. Furthermore, the ageing behaviour of the commonly used detergent amplifies the problem. Therefore, detergents with less sensitivity to flow conditions and ageing are required. Additionally, with regard to materials with enhanced stress cracking resistance, "better" detergents should also lead to shorter testing times.

For this purpose screening tests on alternative detergents were performed. The detergents were used as aqueous solution with a concentration of 2% by weight. Specimens were prepared of a reference polyethylene grade in accordance with ISO 16770. The measurements were performed as accelerated full-notch creep test (AFNCT) with a testing temperature of 90°C and constant stress of 4.0 MPa. The resulting times to failure for the detergents under investigation and nonylphenolpolyglycolether as reference are presented in Fig. 1a.

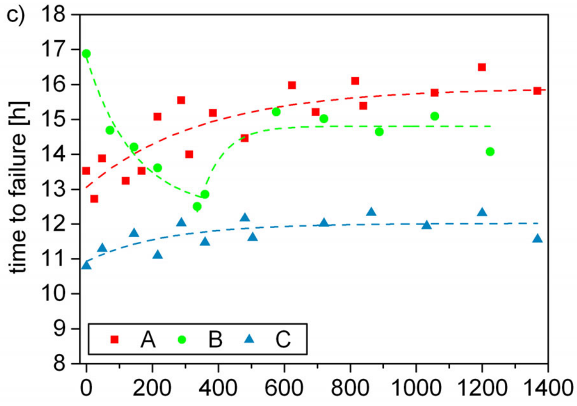
As secondary alkyl sulfonate (detergent A), sodium dodecylbenzenesulfonate (detergent B) and lauramine oxide (detergent C) showed considerable shorter testing times, they were chosen for further investigations: Surface tension was constant at about 30 mN/m for all detergents and, therefore, in the same range as polyethylene. In oven ageing testes at 80°C all detergents, except lauramine oxide, discolored brown. The effect was strongest for the reference and was confirmed in polydispersity measurements, clearly showing detergent degradation. While it is well known that the ageing of the detergent leads to an enlargement of testing time in the case of the reference, this has to be examined for alternative detergents. Therefore, AFNCT were performed using the same testing conditions as before, only varying the age of the aqueous detergent solution at the beginning of the test. Detergent A showed behaviour similar to that of the reference with a decrease in the pH-value correlating with an increase in resulting times to failure (Fig. 1b). In contrast the pH-value decrease leads to a decrease in testing time for detergent B. For detergent C only a slight increase in pH-value and time to failure is observed at the beginning before both remain stable. According to the presented results alternative detergents for the use in stress cracking resistance tests were found (Gerets et al. 2014). With these detergents and an increased testing temperature of 90°C testing of materials with enhanced stress cracking resistance is possible using AFNCT.



a) Time to failure of different detergents in accelerated FNCT



b) Influence of detergent age on pH-value in accelerated FNCT



c) Influence of detergent age on time to failure in accelerated FNCT

Figure 1. Behaviour of different detergents (detergent A (square), detergent B (circle), detergent C (triangle))

3.1.2 Comparison of NCTL Test Results and ANCTL Test Results

First ANCTL-tests are running with the alternative detergent C (lauramine oxide). It is planned to test a HDPE geomembrane under different test temperatures (50°C, 60°C, 70°C and 80°C). Actually the first results (Tab. 1) show that the use of detergent C at a test temperature of 70°C results in times to failure of 366 hours whereas the tests with the commonly used nonylphenoxy polyethylene-oxyethanol at a test temperature of 50°C failed after 2090 hours. In this example the testing time is 5.7 times faster than in the standard tests.

Table 1. Results on a HDPE geomembrane tested under different test conditions.

	Test method	Unit	Sample A	Sample B
Nominal thickness	EN ISO 9863-1	mm	1.0	1.0
Applied stress	EN 14576	-	30% yield stress	30% yield stress
Detergent, unaged	EN 14576	-	nonylphenoxy polyethylene-oxyethanol	lauramine oxide
Test temperature	EN 14576	°C	50	70
Time to failure (SCR)	EN 14576	h	2090	366

3.2 Strain Hardening Test

3.2.1 Slow Crack Growth in Polyethylenes

Resistance to SCG is considered when the applied stress on a product is much lower than the yield stress especially in the presence of bulk inhomogeneities (scratches, pigments, catalyst residues). The overall failure mode is brittle and it proceeds via a so-called craze crack mechanism that commences with a deformation zone or plastic zone formed at the tip of an advancing crack. Such a deformation zone consists of microscopic cavities (voids) that will grow and join up to form a cross-tied network of essentially fibrillar entities usually referred to as a craze. This craze crack mechanism, from the development of the plastic zone up to the fracture of the fibrils, within a craze is considered to proceed through three main stages, i.e. initiation, propagation and the craze crack transition (Fig. 2: left). The initiation step includes the formation of the deformation zone at local points of bulk inhomogeneities, which is strongly associated with the yield stress and stiffness of the material. The deformation zone will propagate and the material between the voids stretches into a network of strain hardened cross tied fibrils. The failure of the fibrils is governed by the effective entanglements in the strain hardened fibrils (disentanglement of (tie) molecules). This basically means that the resistance to craze initiation and failure in such a slow crack growth mechanism is primarily determined by the intrinsic strain hardening response of fibrils. This is why a simple tensile test measuring the strain hardening behaviour above the natural draw ratio (Fig.2: right) will be predictive of the slow crack growth resistance of the material.

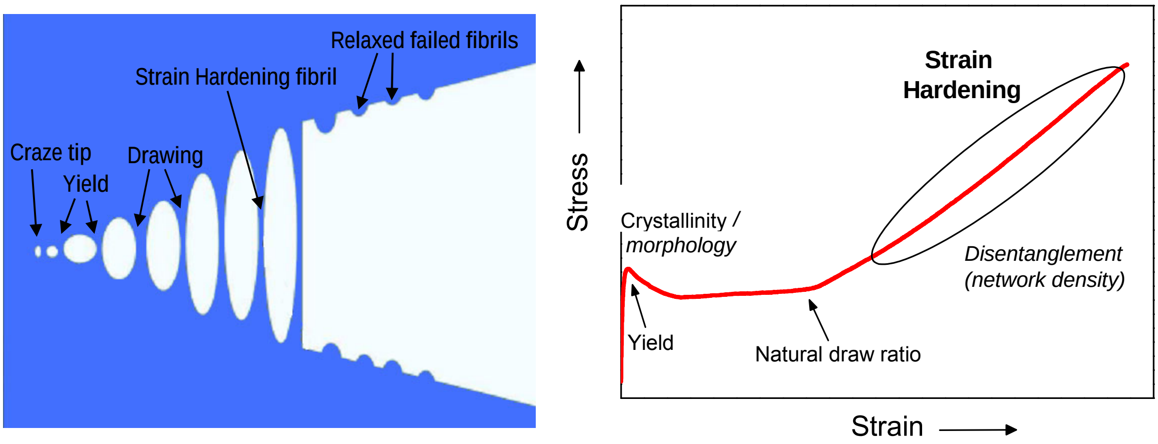


Figure 2. Schematic presentation of fibril strain hardening
in a craze and its relation to uniaxial tensile drawing (Havermans - van Beek et al. 2010)

3.2.2 Test Method

The strain hardening test (ISO/DIS 18488) is a tensile test at elevated temperature of 80°C and constant test speed performed in a temperature chamber. During testing the distance between the gauge marks and the resulting force are recorded. The strain hardening modulus characterising the SCR is calculated according to the following equations. The complete test lasts less than half a day.

The draw ratio λ is calculated on the basis of gauge length l_0:

$$\lambda(t) = \frac{l(t)}{l_0} \qquad [1]$$

where l_0 [mm] is the initial distance between the gauge marks and l(t) [mm] is the distance between the gauge marks during deformation.

The true stress σ_T is calculated from the force assuming conservation of specimen volume between the gauge marks:

$$\sigma_T = \frac{F(t)}{A_0} \cdot \lambda(t) \qquad [2]$$

where F(t) [N] is the measured force, A_0 [mm²] is the initial cross-sectional area of the specimen and λ is the draw ratio from Eq. 1.

The strain hardening modulus $<G_p>$ is defined as the slope of the $\sigma_T(\lambda)$ curve:

$$< G_p > = \frac{1}{N} \sum_{i=1}^{N} \frac{\sigma_{i+1} - \sigma_i}{\lambda_{i+1} - \lambda_i} \qquad [3]$$

For the calculation of $<G_p>$ the range between $\lambda = 8$ and $\lambda = 12$ is evaluated, using the Neo-Hookean constitutive model to fit and extrapolate the data.

We adapted the standardized method to the needs of geomembrane testing. The thickness of a geomembrane specimen is not limited. Any thickness of a HDPE geomembrane can be tested. Specimens of type 5B according ISO 527-2 were punched with a die directly from the geomembrane sample in CMD. Length of the specimen is 35 mm, length and width of the narrow parallel-sided portion are 12 mm and 2 mm respectively.

The test specimens were strained at constant crosshead speed of 10 mm/min until rupture of the test specimen. During the test, the load sustained by the specimen and the elongation were measured. The elongation was determined with an optical extensometer. Two reflecting and self-adhesive gauge marks were attached to the test specimens via a marking apparatus. The initial length (gauge length) between these marks (of about 10 mm) was determined after reaching the pre-load before each test. Prior to testing the specimens were kept for about 30 min in the temperature chamber at the test temperature to allow thermal equilibrium.

3.2.3 Results and Discussion

In Fig. 3, stress-strain-curves at 80°C for three HDPE geomembranes are given which show different strain hardening behaviour. The left diagram shows a standard plot of nominal stress versus nominal strain. It can be seen, that the three HDPE geomembranes do not only differ in yield strength but also in the strain hardening behaviour and that the strain hardening slope does not correlate with the yield strength. The right diagram shows a plot of true stress as calculated from Eq. 2 versus draw ratio. The strain hardening modulus $<G_p>$ was calculated according to Eq. 3 in the section of the draw ratio $8 \le \lambda \le 12$.

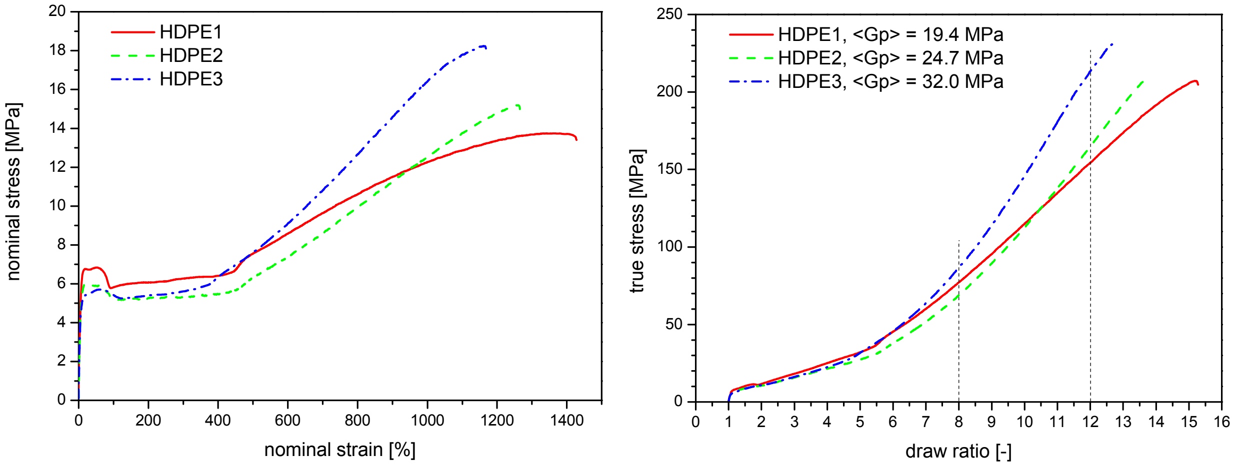


Figure 3. Nominal stress vs. nominal strain (left) and true stress vs. draw ratio (right) with indicated range for calculation of the strain hardening modulus $<G_p>$

Fig. 4 shows the strain hardening modulus – measured at our laboratory – plotted against the failure times in SP-NCTL tests – measured in three different other laboratories. A good correlation between SP-NCTL failure times and $<G_p>$ is found. The higher the value of $<G_p>$, the higher are the failure times in SP-NCTL tests. For the investigated HDPE geomembranes strain hardening moduli $<G_p>$ between 19.4 MPa and 32.0 MPa were found.

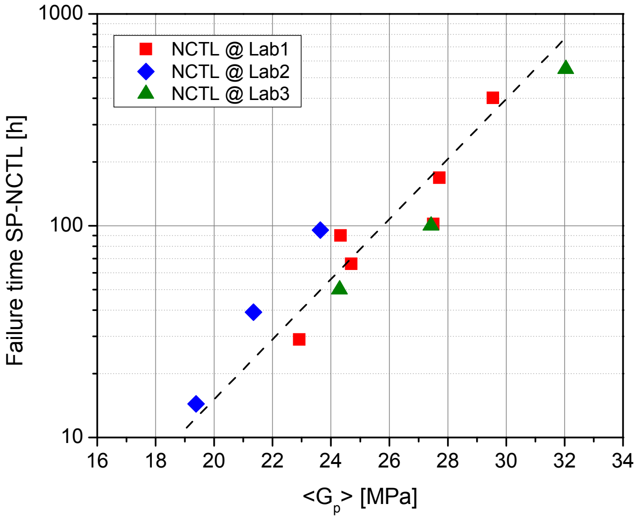


Figure 4. Strain hardening modulus <G_p> versus SP-NCTL failure times for different HDPE geomembranes (Engelsing and Zanzinger 2012)

4. SUMMARY

Established test methods for the characterisation of SCR of HDPE geomembranes are limited in terms of acceleration. So modern high SCR materials cannot be evaluated quantitatively as very long testing durations at high temperatures lead to thermo oxidative ageing before slow crack growth starts. Long testing times are also not acceptable with regard to the needs of quality control, research and development and economic aspects. With the ANCTL test a further development was made because higher test temperatures are applicable. Therefore this test leads to shorter test durations. Alternative test methods like the strain hardening test enable a quantitative differentiation and characterisation of highly SCR polyethylene qualities. They abstain especially from the use of detergents and avoid therefore the involved challenges. As the strain hardening test is a short term test results can be determined within a few hours. Both alternative methods can be used additionally to conventional NCTL tests for the evaluation of the SCR of HDPE geomembranes.

REFERENCES

ASTM D5397-07. Standard Test Method for Evaluation of Stress Crack Resistance of Polyolefin Geomembranes Using Notched Constant Tensile Load Test, *American Society for Testing and Materials*, West Conshohocken, Pennsylvania, USA.

EN 14576:2005. Geosynthetics - Test method for determining the resistance of polymeric geosynthetic barriers to environmental stress cracking, *CEN European Committee for Standardization, Brussels, Belgium.*

Engelsing, K. and Zanzinger, H. (2012). A new accelerated test method for stress crack resistance of HDPE geomembranes. Segundo Congreso Panamericano de Géosintéticos, *GEOAMERICAS 2012*, 01-04 de Mayo, 2012, Lima, Peru, 8p.

Gerets, B., Wenzel, M., Engelsing, K. and Bastian, M. (2014). Langsames Risswachstum von Polyethylen: Beschleunigte und alternative Prüfmethoden, *Grellmann, W. (edi.), Tagungsband, PolyMerTec 2014*, 25.-27.06.2014, Merseburg, ISBN 978-3-942703-31-4, 833-840 (in German)

Havermans - van Beek, D.J.M., Delieck, R., McCarthy, M., Kloth, R. and Kurelec, L. (2010). An elegant and fast method to predict the slow crack growth behaviour of high density polyethylene pipe materials, *Plastic Pipes XV Conference*, Vancouver, Canada.

Hsuan, Y.G. and Koerner, R.M. (1995). The single point - notched constant tension load test: A quality control test for assessing stress crack resistance, *Geosynthetics International*, Vol. 2, No. 5: 831-843.

ISO 9863-1. Geosynthetics - Determination of thickness at specified pressures - Part 1: Single layers, *ISO International Organization for Standardization, Geneva, Switzerland.*

ISO 16770 Plastics – Determination of environmental stress cracking (ESC) of polyethylene – Full-notch creep test (FNCT), *ISO International Organization for Standardization, Geneva, Switzerland.*

ISO/DIS 18488 Polyethylene (PE) materials for piping systems -- Determination of Strain Hardening Modulus in relation to slow crack growth -- Test method, *ISO International Organization for Standardization, Geneva, Switzerland.*