Ageing resistance of HDPE-geomembranes – Evaluation of long-term behavior under consideration of project experiences

Tarnowski, C. & Baldauf, S. GSE Lining Technology GmbH, Hamburg, Germany

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ABSTRACT: A question frequently asked about HDPE- geomembranes is "how long will they last?" After more than 30 years of experience with exposed (weathered) HDPE-geomembranes, the authors address the question from the designer's view point. The engineer needs to know, if available test methods tell us how long the geomembrane will last, and which properties must be specified and what are the minimum acceptable values?" The main focus of this paper is the practical approach for evaluation of the durability of exposed (weathered) HDPE-geomembranes. HDPE-geomembranes used in four projects after exposure in different climatic zones were tested for the amount of stabilization and for changes of their properties. After up to 30 years exposure to weathering these HDPE-geomembranes reveal a reduction of the antioxidant content, whereas the mechanical properties did not significantly change. The test results presented in this paper indicate clearly that the lining systems still work in their desired function and remaining antioxidants remain capability to provide further protection against degradation even after this time. On the basis of this research the appropriate properties required to evaluate the durability of HDPE-geomembrane lining systems are identified.

1 INTRODUCTION

The decisive criterion for the acceptance of HDPEgeomembranes is their durability. Since HDPE can be degraded to only a limited extent by most inorganic and many organic chemicals it is mainly oxidation (photo- and/or thermo-oxidative ageing depending on the application) that is the cause of degradation of the polymer. Under covered conditions (medium temperature/ absence of UVradiation) oxidation takes place only very slowly.

To absorb UV-radiation and for protection against oxidation carbon black and a specific antioxidantpackage are added to the polyethylene. Apart from oxidation (a chemical reaction) the other significant ageing mechanism is environmental stress cracking. Environmental stress crack resistance is also important when selecting appropriate resins.

Various scientific approaches for estimating geomembrane service-life in buried applications have been published (see paragraph 2 below). However, little information has been published to date in regard to field-experience with exposed geomembranes that have already undergone ageing processes.

In this paper test results on exposed HDPEgeomembranes are presented.

2 SERVICE-LIFE PREDICTION FOR BURIED GEOMEMBRANES

A well-known model is the elevated temperature Arrhenius incubation with modeling and extrapolation to lower site-specific temperatures carried out by the Geosynthetic Research Institute (GRI) in the United States. This has resulted in an estimated service lifetime of 265 years at normal conditions for a particular HDPE-geomembrane (Koerner 2005). Here three stages of ageing are described (stage A - antioxidant depletion time -200 years, stage B – induction time – 30 years, stage C, the actual change of the molecular structure accompanied by a deterioration of the decisive mechanical properties – 35 years).

A different approach was taken by members of the Federal Institute for Material Research and Testing (BAM) in Germany (Müller and Jakob 2003). HDPE–geomembranes were aged in ovens (air at 80°C, 13 years test time) and in de-ionized hot water (80°C, 6 years test time). It was found that ageing in water takes place more rapidly. Using this model the service lifetime was estimated conservatively to be at least 300 years.

3 TEST RESULTS ON FIELD EXPOSED HDPE- GEOMEMBRANES

Do field weathered HDPE-geomembranes follow the lifetime modeling above? Data available from the authors company on up to 30 years exposed HDPE-geomembranes are presented in the following. Table 1 gives a short overview about the projects. HDPE-geomembranes manufactured from different formulations/raw materials were used in the four projects. Therefore a direct comparison of the properties of the geomembranes between the projects would not be meaningful.

Table 1. Projects with HDPE-geomembranes

Project Y	ear of construction	Last test	Thickness
Galing 1 (Germany) 1974	2005	2.5 mm
Galing II (Germany	7) 1984	2005	2.5 mm
Sarchesmeh (Iran)	1975	2000	2.5 mm
Levante (Spain)	1994	2005	2.0 mm

In 1974, at the location Galing/Germany the first jarosite sludge deposit was built (Galing I). Jarosite sludge is the waste arising from the production process of zinc and lead. The sludge has a pH-value of ca. 2-4. The lined area was about 11 ha.



Figure 1. Jarosite sludge landfill Galing/Germany

In 1984, the second pond (12 ha) was built at the same location (Galing II). On the slopes, the HDPEgeomembranes are permanently exposed to weathering (annual solar energy ~ 90 kLy). For Galing II a comprehensive test program for the assessment of long-term behavior was realized by the Süddeutsches Kunststoffzentrum (SKZ), Würzburg/Germany already in 1984. The evaluation of properties was conducted 6 times in 26 years (SKZ, private communication).

Even after 26 years of exposure it was found that the tensile properties have not changed significantly within the limit of accuracy of the test methods. Fig. 2 shows the results on tensile stress at yield and at break (maximum tensile stress) in 6 test periods; Fig. 3 shows the elongation at yield and elongation at break (tested acc. to DIN 53455 superseded by ISO 527-3). The environmental stress crack resistance (ESCR) was originally measured according to ASTM D 1693, now superseded by the NCTL-test (ASTM D 5397). The initial value of 2000 hours acc. to the older test is still preserved. Evaluating the stress crack resistance acc. to ASTM D 5397 (SP-NCTL) the more stringent and state-of-the-art method, it was found that 60% of the initial value (52 hours) is still preserved.



Figure 2. Galing II, change in tensile stress at yield and at break (acc. to ISO 527-3)



Figure 3. Galing II, change in elongation at yield and at break (acc. to ISO 527-3)

A significant reduction of the oxidative induction time (OIT) was noted, a value which indicates the amount/depletion of antioxidants. In order to determine the remaining antioxidant content more precisely, in 2000/2005 the OIT measurements were taken at lower temperatures. The OIT-value decreased continuously over the years, which indicates that there is a slow consumption of antioxidants. The remaining antioxidants provide further ageing resistance. Fig. 4 shows the OIT results (full cross section of sample tested).



Figure 4. Galing II, OIT acc. to DIN EN 728 – Depletion of antioxidants

For Galing I after 31 years of exposure it was found, within the limit of accuracy of the test methods, that the tensile stress, the elongation at yield, and the tensile stress at break have not changed significantly, whereas only 30 % of the original elongation at break is retained (only transverse direction was tested). Time to failure in SP-NCTL-testing was only five hours. A significant reduction of the OIT-value (5 min) was found, but still small amounts of antioxidants are left.

To better understand the antioxidant depletion process, the OIT-testing was conducted separately on the top surface and the middle layer of the geomembrane. Table 2 gives the results.

Table 2. Antioxidant content in different layers measured in 1995/2005 - Galing I/ in 2005 - Galing II

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	Galing I (31 y)				Galing II (21 y)				
OIT [min]	1995		2005		2005				
Layer,	Тор	Middle	Тор	Middle	Тор	Middle			
~0.9 mm	layer	layer	layer	layer	layer	layer			
200 °C	7.1	8.8	0	4	5	65			
190 °C	13.8	20.9	0	8.5	10.5	148			

In the top layer of the 31 years weathered geomembrane no antioxidants could be detected. In the middle layer however, antioxidants are still present. Also in the top layer of the 21 years old geomembrane of Galing II a significant reduction of OIT could be detected whereas in the middle layer no reduction is found. We conclude that due to the carbon stabilization UV-radiation black the penetrates only the upper layer. The top layer is faster penetrated by oxygen since the availability in the geomembrane is essentially diffusion controlled. Therefore it can be concluded that the thickness has a major influence on antioxidant depletion and therefore on preservation of the barrier function. Outward migration of antioxidants has been demonstrated be to slower on а thicker geomembrane (Rowe et al, 2002).

The water reservoir in Sarchesmeh/Iran was built in 1975 at 3,000 m altitude. The lined area is about 40,000 m². The HDPE-geomembranes are permanently exposed to extreme weather conditions, as there are intensive UV-radiation (annual solar energy 290 kLy), high wind speed and annual differences in temperature from -26° C up to $+38^{\circ}$ C.



Figure 5. Water reservoir in Sarchesmeh/Iran

In September 2000 a sample was taken from the embankment above water level. Within the limit of accuracy of the test methods the tensile properties are unchanged compared to the initial state. There are no visual indications of the beginning of cracking or brittleness. It is planned to obtain further samples to determine the extent to which the antioxidants in the polymer are still effective. This test was not conducted in 2000.

The water reservoir in Levante/Spain (annual solar energy 160 kLy was built at the beginning of 1994. In September 2005 a sample was taken from the north/east slope, one half of the sample permanently exposed to UV-radiation, the other half at intermittent water coverage. This geomembrane is produced from a raw material (a linear polyethylene with α -olefin- C8 copolymer) that is still used in geomembrane manufacture today.

Both sections of the sample were tested. After 11 years the mechanical properties in neither area have changed significantly. Table 3 shows the SP-NCTL and the OIT-values and compares these with the initial values obtained on a retained sample that had been stored in the factory not exposed to light (values in brackets).

Table 3. SP-NCTL (ASTM D 5397) and OIT (DIN EN 728, 190°C) after outdoor weathering/initial properties (in brackets)

Levante	Above water level		Intermittent water level		
SP-NCTL [h]	581 ->1000* (>	1000)*	785->1000*	(>1000)*	
OIT [min]	56 - 67	(145)	107 - 110	(145)	

* tests were terminated after 1000 h

By comparing these SP-NCTL-values with the initial value of the geomembrane used in Galing II (52 hours, see above) it can be concluded that the geomembrane of the raw material still in use today offers even higher stress crack resistance. So one can conclude, that high quality HDPE-geomembranes made from linear polyethylene with C8 α -olefin comonomers provide even longer service-life.

The difference in OIT between the continuously UV-weathered section and that with intermittent water coverage is clearly demonstrated. In the wetted zone the geomembrane was protected against UV-radiation by the water and detritus. Consequently the depletion of antioxidants is lower compared to the permanently exposed section.

In terms of the GRI-model referenced in paragraph 2 above the test results on the projects show that the ageing processes are still in stage A, (anti-oxidant depletion without change in mechanical properties). The only exception is for the Galing I sample where a decrease of elongation at break was measured. All test programs are ongoing. In Europe the Construction Products Directive (CPD) imposes on consultants and engineers to consider the "working life time" and therewith the durability of the construction products with regard to the intended use of the product and the foreseeable service conditions. This led to the development of harmonized European Norms (EN) each including an extensive annex on durability. According to these new European norms for the use of geomembranes in waste disposal sites, reservoirs, dams, canals and tunnels (EN 13361, 13362, 13492, 13493, 13491) manufacturers are required to publish values for certain key-properties and a durability statement. Engineers should require the manufacturer's compliance statement to be presented.

EN-norms require the verification of change in mechanical properties after accelerated thermal ageing (EN 14575) and UV-exposure (EN 12224) to evaluate durability. The results of tests presented in this paper show that evaluation of durability using tensile properties after accelerated ageing is not as sensitive as evaluation using oxidative induction time. The figures presented in Section 3 demonstrate that at first a change of OIT was detected without changes in tensile properties. Even after 26 years of service life a decrease of 25% in tensile stress at break and elongation at break (durability criteria established by the EN standards) was not detected. The OIT measured after accelerated ageing also needs to be specified to get meaningful answers regarding the durability of HDPE-geomembranes.

Please note, the statement of just the initial OIT (immediately after manufacture) just confirms that there is a sufficient amount of antioxidants, but the efficiency is not verified. On the basis of the results presented herein it becomes clear that minimum properties for oxidative induction time (initial value and - even more important - retained OIT after photo- and thermo-oxidative ageing) together with minimum properties for stress crack resistance (SP-NCTL) are the properties to be well defined in every geomembrane specification.

Most important however is thickness. With regard to the intended use of the product and therewith the assumed design working life engineers should consider at first the thickness. It is clearly shown by the evaluation of OIT in different layers (see Section 3, Table 2) that a sufficient thickness is providing a longer service life.

A standard that states durability properties and proposes values as well as test frequencies for HDPE-geomembranes is the GRI-GM 13 of the Geosynthetic Research Institute. State-of-the-art HDPE-geomembranes can reach even higher values than required in GM 13.

5 CONCLUSIONS

This paper presents results of tests performed on field samples of HDPE -geomembranes that were exposed to weathering in different climatic zones for periods up to 30 years. The ability of the geomembrane continue functioning to as impermeable barrier could be verified. The correlation between the scientific approach (the stages of antioxidant depletion to material deterioration, as described in the literature (Koerner 2005) or (Müller and Jakob 2003) could also be found in the tested exposed materials.

Surprisingly, a decrease of mechanical properties could only be detected in the test specimen that was exposed 31 years, whereas antioxidant-depletion could be documented for three out of the four test specimen (the Iran project samples could not be verified).

After 31 years of exposure still traces of antioxidants could be found in the OIT-tests conducted. Using OIT-testing in particular layers of the geomembranes revealed that, as might be predicted the upper layer of the 2.5mm thick geomembrane (see Section 3, Table 2) had undergone accelerated ageing, compared to the middle laver that still shows an antioxidant content. therefore suggest The authors to consider geomembrane thickness a major criterion for durability, and to interpret thickness in context with other criteria such as chemical, UV, mechanical and other exposures.

Results presented herein indicate that OIT testing is a superior test parameter to determine durability and resistance towards ageing, at least in the early stages of degradation, and that this parameter should be used in addition to the tensile property testing to be conducted on weathered and/or buried geomembranes.

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