

**The likely medium to long-term generation
of defects in geomembrane liners**

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Published by:

Environment Agency
Rio House
Waterside Drive, Aztec West
Almondsbury, Bristol BS32 4UD
Tel: 01454 624400 Fax: 01454 624409

ISBN: 1 84432 180 0

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R&D Technical Report P1-500/1/TR

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Statement of use:

This report presents a review of the processes and rates of geomembrane degradation reported from laboratory and field studies. It reviews landfill monitoring data and research from related fields to predict future rates of defect generation in geomembrane liners, for use in risk and performance assessment of new landfill sites. This report should be used in conjunction with the Agency's guidance on hydrogeological risk assessment for landfills and LandSim v2.5+.

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CONTENTS

EXECUTIVE SUMMARY	6
Acknowledgements	7
1 INTRODUCTION	8
1.1 OBJECTIVES	8
1.2 GEOMEMBRANES BARRIERS	8
1.3 DEFECTS IN GEOMEMBRANE LINERS	9
1.4 SCOPING STUDY	10
1.5 OUTLINE OF THE REPORT	10
1.6 HOW TO USE THE REPORT	11
2 LITERATURE AND DATA REVIEW PROCESS	12
2.1 LITERATURE SOURCES	12
2.2 SOURCES OF DATA ON ELECTRICAL LEAK LOCATION SURVEYS	13
2.3 SOURCES OF DATA ON LANDFILL TEMPERATURES	13
3 GEOMEMBRANE LINERS	14
3.1 BARRIERS	14
3.2 THE STRUCTURE AND PROPERTIES OF POLYETHYLENE FOR GEOMEMBRANE LINERS	15
3.2.1 <i>The structure of polyethylene</i>	15
3.2.2 <i>Structural properties of polyethylene geomembranes</i>	18
4 POLYETHYLENE DEGRADATION MECHANISMS	20
4.1 TYPES OF DEGRADATION	20
4.2 OXIDATIVE DEGRADATION MECHANISMS	22
4.3 ANTIOXIDANTS AND CARBON BLACK STABILISERS	26
4.4 ANTIOXIDANT DEPLETION	28
5 LABORATORY STUDIES INTO OXIDATIVE DEGRADATION	31
5.1 INTRODUCTION	31
5.2 TEST METHODS TO ASSESS DEGRADATION	31
5.2.1 <i>Oxidative Induction Time</i>	32
5.2.2 <i>Molecular weight</i>	34
5.2.3 <i>Tensile properties</i>	34
5.2.4 <i>Stress crack resistance</i>	34
5.2.5 <i>Crystallinity</i>	34
5.2.6 <i>Polymer structure</i>	35
5.2.7 <i>Chemiluminescence</i>	35
5.3 ARRHENIUS MODELLING	35
5.4 ASSESSING TIME FOR LONG-TERM GEOMEMBRANE DEGRADATION	39
5.4.1 <i>Hsuan and Koerner (1995 and 1998)</i>	40
5.4.2 <i>Sangam (2001)</i>	43
5.4.3 <i>Müller and Jakob (2003)</i>	48
5.5 REVIEW OF LABORATORY RESEARCH INTO GEOMEMBRANE DURABILITY	56
5.5.1 <i>Geomembrane materials</i>	56
5.5.2 <i>Test duration</i>	57

5.5.3	<i>Exposure medium and ageing temperature</i>	57
5.5.4	<i>Thickness effects</i>	58
5.5.5	<i>Absolute OIT values</i>	59
5.5.6	<i>Location of the OIT specimen</i>	60
5.5.7	<i>Activation energy</i>	60
5.5.8	<i>Three stage degradation process</i>	61
5.5.9	<i>Selecting the liner service temperature</i>	62
5.5.10	<i>Conclusions on HDPE geomembrane durability laboratory testing</i>	62
6	DURABILITY OF POLYETHYLENE LINERS IN FIELD APPLICATIONS	65
6.1	REPORTED FIELD PERFORMANCE	65
6.2	REVIEW OF REPORTED FIELD PERFORMANCE	67
6.3	EXPOSURE CONDITIONS	68
6.3.1	<i>Temperatures at the liner</i>	68
6.3.2	<i>Ultraviolet radiation</i>	76
6.3.3	<i>High energy radiation</i>	77
6.3.4	<i>Availability of oxygen</i>	77
6.3.5	<i>Exposure to leachate</i>	79
6.3.6	<i>External mechanical stresses</i>	82
7	STRESS CRACKING	83
7.1	INTRODUCTION	83
7.2	TESTING.....	85
7.4	FACTORS THAT INFLUENCE STRESS CRACKING	88
7.5	STRESS RELAXATION.....	89
7.6	IMPLICATIONS OF STRESS CRACKING TO SERVICE LIFE	90
7.7	AVOIDING STRESS CRACKING	91
8	PHYSICAL DAMAGE MECHANISMS	92
8.1	DAMAGE BEFORE WASTE PLACEMENT	92
8.2	MECHANISMS OF PHYSICAL DAMAGE AFTER LINER INSTALLATION.....	97
8.2.1	<i>Data from fixed ELL surveys</i>	97
8.2.2	<i>Sandy Lane landfill, Bromsgrove</i>	100
8.2.3	<i>Summary of findings from ELL surveys</i>	104
8.3	REVIEW OF LEAK DETECTION SYSTEMS IN DOUBLE LINERS	105
8.4	LARGE SCALE AND CATASTROPHIC FAILURE EVENTS.....	107
8.4.1	<i>Background</i>	107
8.4.2	<i>Slope instability</i>	107
8.4.3	<i>Down-drag effects of waste settlement</i>	109
8.4.4	<i>Excess basal movements</i>	110
8.4.5	<i>Seismic events</i>	112
8.4.6	<i>Landfill fires</i>	113
8.5	SUMMARY OF PHYSICAL DAMAGE MECHANISMS	114
9	ESTIMATION OF DEFECT GENERATION IN HDPE GEOMEMBRANE LINERS	116
9.1	INTRODUCTION	116
9.2	GEOMEMBRANE LINER SERVICE LIFE.....	116
9.3	STAGES OF HOLE GENERATION	117
9.4	ESTIMATION OF HOLE GENERATION.....	120
	<i>Stage 1</i>	120

Stage 2.....	121
Stage 3.....	121
Stage 4.....	121
Stage 5.....	123
Stage 6.....	123
9.5 WORKED EXAMPLES	124
9.5.1 Example 1	124
9.5.2 Example 2	125
9.6 FACTORS AFFECTING STAGE DURATION AND HOLE FREQUENCY	126
9.6.1 Design	126
9.6.2 Installation.....	128
9.6.3 Waste type, infilling practices and leachate management.....	129
9.6.4 Caps	129
9.6.5 Summary of actions to minimise geomembrane hole generation	130
9.7 FUTURE RESEARCH NEEDS.....	130
9.8 RISK ASSESSMENT MODELLING.....	131
REFERENCES.....	133
GLOSSARY AND ABBREVIATIONS.....	145
ANNEX 1: SCOPING STUDY: LONG-TERM DURABILITY OF NON-POLYETHYLENE SYNTHETIC LANDFILL LINERS.....	150
A1.1 INTRODUCTION.....	150
A1.2 GEOSYNTHETIC CLAY LINERS	150
A1.2.1 Background.....	150
A1.2.2 Literature review.....	151
A1.2.3 Mechanisms of failure.....	152
A1.2.4 Recommendations	153
A1.2.5 Material standards.....	154
A1.3 DENSE ASPHALTIC CONCRETE	155
A1.3.1 Background.....	155
A1.3.2 Literature review.....	156
A1.3.3 Hydraulic conductivity.....	157
A1.3.4 Design and installation guidance.....	157
A1.3.5 Degradation mechanisms.....	158
A1.3.6 Recommendations	159

EXECUTIVE SUMMARY

Geomembrane liners are widely used to contain wastes, contaminated soils and water in landfills, contaminated land remediation applications and water storage reservoirs respectively. This document presents a review of information on the medium to long-term generation of defects in geomembrane liners, with particular reference to their use at landfill sites.

The review and interpretation presented may be helpful when considering the generation of defects in synthetic geomembranes (or more formally polymeric geosynthetic barriers) during the potentially long polluting lifetime of modern landfills. The assessment (and management) of risks to human health and the environment over the entire duration of waste deposit and decomposition/storage in landfills is required by the Landfill Regulations 2002. The information present in this report may be used in conjunction with Agency guidance on hydrogeological risk assessment for landfills (Environment Agency 2003a) for the purposes of demonstrating compliance with the groundwater protection aspects of both the Landfill and Groundwater Regulations.

A six-stage model of hole generation with time throughout the service life of a geomembrane landfill liner is introduced. Hole generation can occur at the time of the initial installation of the liner and the early period during waste disposal when the geomembrane is particularly vulnerable to physical damage, and through the medium to very long-term period of material degradation when the degrading liner becomes increasingly more susceptible to stress cracking.

The model is described in Chapter 9 but it is necessary to read the preceding chapters to understand the basis of the model and how it may be applied for specific sites and circumstances.

Acknowledgements

The Environment Agency and EDGE Consultants UK Ltd would like to thank the following companies and organisations for contributing to this project (in alphabetical order):

BHF Environmental Ltd.
Cleanaway Ltd.
Cory Environmental
Federal Institute for Materials Research and Testing (BAM), Berlin, Germany
Geologger Systems GmbH
Geosynthetic Research Institute (GRI), Folsom, Pa., USA
Hesselberg Hydro 1991 Ltd.
Knox Associates (UK) Ltd.
SENSOR International
SITA UK
Walo UK Ltd.

1 INTRODUCTION

1.1 Objectives

The Environment Agency commissioned EDGE Consultants UK Limited in association with I-Corp International of Florida, Nottingham Trent University and Rapra Technology Limited of Shrewsbury, UK, to investigate and report on the generation of defects in geomembrane landfill barriers in the medium and long-term. The medium to long-term period extends from the completion of the liner construction and commencement of waste disposal, throughout the site operation and long post-closure periods of the landfill to the end of the service life of the geomembrane liner as an effective barrier to leachate and gas leakage.

The review considers both the degradation of the geomembrane material caused by inevitable chemical and physical processes in the geomembrane in the landfill environment, and damage caused by external mechanisms. This physical damage may be caused by factors such as puncturing, waste compaction plant operation, excess stresses leading to stress cracking, and large scale damage, for example by slope failures. Available literature has been reviewed together with an assessment of data from mobile and fixed electrical leak location (ELL) surveys for landfills in the UK and continental Europe and leakage information from double lined landfills in the USA.

Leakage by diffusion is not addressed in this report, but its significance in causing pollutant flux from landfills is reviewed elsewhere (Environment Agency, 2004).

Studies of landfill leachate chemistry and degradation processes suggest that landfills managed using typical current approaches will take hundreds, if not thousands, of years to stabilise (e.g., Hall *et al.*, 2003). It is only after this period that they will no longer pose a pollution hazard to their surrounding environment. The durability of engineering containment and control measures is therefore critical in assessing the long-term pollution potential.

The overall objective of the project is to provide guidance on the long-term effectiveness of geomembrane barriers in preventing groundwater contamination by leachate leakage, off-site gas migration and water ingress from cap infiltration or groundwater. The predicted performance with time of the geomembrane barrier may then be used in landfill risk assessments (Environment Agency, 2003a) and simulated in performance/risk assessment models, such as LandSim Release 2.5 or later versions. The conceptual model of hole generation presented in this report assumes a range in standards from excellent to moderate in the liner design, installation and other relevant factors. Where lower standards or defective design apply, then much greater defect frequencies may well apply.

A short scoping review has also been carried out on non-geomembrane synthetic barriers used in landfills to identify the need, if it exists, for research on the durability and generation of defects in these liners.

1.2 Geomembranes barriers

Geomembrane barriers used as basal and side slope liners, and in landfill caps are considered in this report. They are sometimes called flexible membrane liners (FMLs) or flexible membrane caps (FMCs) but the terms used throughout this report are geomembrane barrier, geomembrane

liner or, where the distinction is necessary, geomembrane cap. The geomembrane may be used as a single liner, or as a component of a composite or double liner in conjunction with other barrier materials such as compacted clay, bentonite enriched soil (BES) and geosynthetic clay liners (GCLs).

High density polyethylene (HDPE) is the material almost exclusively used for the geomembrane lining of the base and side slopes of landfill in the United Kingdom as well as in other countries across the developed world (Giroud and Touze-Foltz, 2002). The review of geomembrane degradation therefore concentrates on geomembranes made from HDPE. The durability of linear low density polyethylene (LLDPE), which is commonly used as a barrier in landfill caps and also occasionally on side slopes, is also addressed but to a lesser extent than HDPE because of the lack of published research on this material. Data from ELL surveys of damage caused to liners by external factors may be related to geomembrane materials other than HDPE and LLDPE.

While the focus of this project is on geomembrane barriers in landfills, much of the report will be of interest to regulators and practitioners using geomembrane liners for lagoons, ponds and reservoirs, and as barriers in contaminated land remediation and mine waste management projects.

1.3 Defects in geomembrane liners

Defects in the geomembrane liner arising after commissioning the landfill can take the form of fully penetrating holes such as tears, rips, pinholes, holes and cuts, together with a variety of imperfections that may subsequently develop into or cause fully penetrating holes. Holes may develop throughout the service life of the liner at existing partially penetrating defects and areas of weakness or stress, or may be the result of new damage. Numerous factors can lead to the generation of holes in this medium to long-term period, including:

- waste disposal activities (e.g. movement of plant, excessive insertion of survey and electrode stakes);
- articles of waste penetrating the liner (e.g. reinforcing bars);
- pressures arising from the waste load causing puncturing by cover gravel or protruding stones underneath the geomembrane;
- long-term degradation of protection geotextiles with a reduction in the protection afforded against drainage gravel puncture;
- stresses in the liner e.g. at wrinkles or poor quality seams leading to stress cracks or opening of partially penetrating defects;
- excessive stresses in the liner resulting from down-drag caused by waste settlement, sub-grade settlement or stress in the vicinity of the base of leachate wells;
- catastrophic events including slope instability and landfill fires;
- leachate / liner incompatibility in hazardous waste landfills; and
- long-term degradation of the liner material with a loss of physical properties.

Long-term degradation of polyethylene geomembranes is principally caused by oxidation processes after the removal of antioxidants added in the manufacture to delay the onset of oxidation. It causes embrittlement and a loss of physical properties, leading to the formation and enlargement of holes at locations of stress.

Many of the causes of holes, damage and stresses in a geomembrane liner result from poor design, mediocre standard liner construction and careless waste disposal practices. These defects and the premature development of fully penetrating holes in the liner are avoidable. The principal design, construction and operations factors leading to the generation of defects are identified in this report. Recognition of these factors will enable the adoption of high standard practices with the objective of maintaining the long-term integrity of the geomembrane barrier.

1.4 Scoping study

In addition to the principal objective of the project, a scoping study has been undertaken to identify the most important issues regarding the failure mechanisms and the generation of defects in other types of synthetic landfill liners. The study also assesses the extent and availability of relevant literature and other data on these issues. Geosynthetic clay liners (GCL) and dense asphaltic concrete (DAC) liners are the two non-geomembrane types of liners considered. The objective of the scoping exercise is to identify if there is a need for a project to investigate these aspects of GCL and DAC landfill liners and, if so, to recommend suitable scopes of work. The study is reported in Annex 1.

1.5 Outline of the report

The information in this report is presented in nine chapters including this introduction and one annex. A brief summary is provided below.

Chapter 2 describes the sources of literature obtained for review and the sources of data on ELL surveys and landfill temperatures.

Chapter 3 briefly covers the types of geomembrane barriers, and discusses the characteristics of polyethylene resins and the formulation of polyethylene geomembranes.

In **Chapter 4** the mechanisms of polyethylene degradation are introduced, followed by a description of oxidative degradation, the use of antioxidants to delay the onset of oxidation and the mechanisms of depletion of the antioxidants from the polyethylene geomembrane.

Chapter 5 describes the background of laboratory studies into oxidative degradation. While these studies are accelerated by the use of elevated temperatures, they are still long-term research programmes extending for many years. The three main published research studies are then discussed in some detail.

The durability of the liners in field applications is covered in **Chapter 6** with an extensive discussion on important exposure factors that influence the degradation process, particularly temperature, UV exposure, the availability of oxygen and the exposure to leachate.

The important damage mechanism of stress cracking is addressed in **Chapter 7**.

In **Chapter 8**, physical damage mechanisms are addressed by reference to extensive published information and data obtained for this project on the findings of ELL surveys, as well as technical literature with analyses of leakage in double lined landfills in the USA. Large scale and catastrophic failures affecting the lining systems are also addressed.

The findings of the project are then brought together in **Chapter 9**. A conceptual model is proposed which provides the basis for estimating the generation of defects in geomembrane liners in the medium to long-term for use in landfill risk assessments.

Annex 1 describes the scoping study introduced in Section 1.4 above.

1.6 How to use the report

The generation of defects in geomembrane liners arising from physical damage, material degradation and stress cracking can vary widely in the timing and extent of defects, as these depend upon numerous material, design, installation, operational and environmental factors. The conceptual model introduced in Chapter 9 sets out six stages of defect generation based on their causes and development with time as discussed in the preceding chapters. **Chapter 9 should not be read nor the conceptual model used without a clear understanding of the previous chapters. Without this understanding, the influence of the many factors on which the conceptual model is based will not be adequately appreciated, jeopardising the proper application of the conceptual model.**

The conceptual model is based on field and laboratory research and observations as far as possible. Where these are not yet available, extrapolations have had to be made beyond current knowledge in order to estimate defect generation throughout the full service life of HDPE geomembrane liners for use in landfill risk assessment models. These tentative predictions have been developed using reasonably conservative “educated” judgement but are subject to revision as the findings of more long-term research becomes available.

2 LITERATURE AND DATA REVIEW PROCESS

2.1 Literature sources

This report presents the findings of an extensive literature review carried out to establish the current state of knowledge with respect to:

- the mechanisms of degradation of polyethylene including antioxidant depletion;
- degradation of HDPE geomembranes in landfill exposure conditions from laboratory studies and field performance;
- stress cracking of HDPE;
- leachate effects on polyethylene geomembranes;
- failure of geomembrane liners by major events such as slope instability, waste settlement and basal settlement;
- the type, cause, location and frequency of holes in geomembrane liners as identified by mobile and fixed ELL surveys;
- leakage through double liner systems as evidence of holes in the upper liner;
- temperatures in landfills, especially at the liner, as temperature is a controlling factor in the rate of geomembrane degradation.

Over 250 technical publications were reviewed in this project primarily being obtained through the libraries of Nottingham Trent University and Rapra Technology. The literature came from a number of sources including journals, conference and symposia proceedings, books and technical reports on polymer chemistry, polymer degradation, geosynthetics, geotechnical engineering and landfill engineering.

The sources of publications gathered for this review included the following main journals:

Journal	Publisher
Polymer Degradation and Stability	Elsevier
Geosynthetics International	International Geosynthetics Society
Geotextiles and Geomembranes	Elsevier
Geotechnical Fabrics Report (GFR)	IFAI
Canadian Geotechnical Journal	National Research Council of Canada
Geotechnical and Geoenvironmental Engineering	American Society of Civil Engineers

Conference proceedings examined included:

- Geosynthetics 1987, 89, 91, 93, 95, 97, 99, 2001;
- International Conference on Geosynthetics 1982, 86, 90, 94, 98, 2002;
- European Geosynthetics Conference 1996, 2000;
- International Waste Management and Landfill Symposia (Sardinia) 1989, 91, 93, 95, 97, 99, 2001;
- GRI Conference Series 9, 10, 11, 12, 13, 14 and 15;
- Annual Technical Conference (ANTEC) of the Society of Plastic Engineers, 1992 to 2002.

Other sources included:

- Federal Institute for Materials Research and Testing (BAM), Berlin, Germany
- Geosynthetics Institute (GRI), Pennsylvania;

- Study of reference lists in publications;
- Textbooks;
- ASTM standards;
- BS/EN/ISO standards;
- Internet publications and websites.

2.2 Sources of data on electrical leak location surveys

In addition to the published information, data on the results of mobile and fixed electrical leak location (ELL) surveys were provided by two contractors undertaking these surveys and from one operator who had a fixed ELL system installed at one of its sites. The information came from landfills located in the United Kingdom and numerous countries in continental Europe including Germany, Czech Republic, Slovakia, Poland, Hungary, Croatia, Belgium and Spain. The fixed ELL systems were installed at 73 sites involving over 110 cells and 18 leachate lagoons and a total area of geomembrane liner in excess of 2,000,000m².

The data comprised the reports of surveys undertaken at some sites, and summarised results of surveys undertaken at numerous other sites. The monitoring reports for fixed ELL surveys at individual sites were particularly useful in providing detailed information on the type, frequency, causes and sizes of holes occurring in the geomembrane liner after the commencement of waste placement.

Published literature summarising mobile and fixed ELL surveys at many sites was reviewed together with the data acquired in the project to enable an assessment to be made of the occurrence of post-installation holes in geomembrane liners.

2.3 Sources of data on landfill temperatures

The temperature that a geomembrane liner is exposed to during its service life is critical to the rate of degradation of the polyethylene. To augment the available published literature that largely referred to temperatures in landfills outside the United Kingdom, landfill temperature data from the UK were obtained from several sources:

- An instrumented cell of a landfill in south east England, with thermocouples installed at the liner (Knox, 2003);
- Following a request to landfill operators, data on leachate temperatures were obtained from two operators on six representative landfills in England.

In addition, information on the ambient ground temperatures in the United Kingdom was obtained from the British Geological Survey (Rollin, 2003).

3 GEOMEMBRANE LINERS

3.1 Barriers

Geomembrane liners are used as barriers in landfills to contain waste materials and prevent or minimise the escape of leachate and gas generated in the decomposition of the wastes. They can also function to obstruct the inflow of groundwater into the waste where there is a groundwater level elevated in relation to the leachate level. Geomembranes are used to line the base and side slopes of a landfill, and are used to cap the landfill where the purpose is to control outward gas emissions and inward water infiltration. Usually the geomembrane is employed in conjunction with one or more other types of liners (e.g. compacted clay liner (CCL), bentonite enhanced soil (BES) or geosynthetic clay liner (GCL)) to form a single composite liner or a form of double liner. Examples of these liners are shown in Figure 3.1.

The term 'geomembrane' is used in this report to refer to any low permeability synthetic material designed for, and used in, landfill liner applications. It normally refers to HDPE geomembranes, and can be taken as having the same meaning as a 'polymeric geosynthetic barrier' as defined by the British Standards Institute.

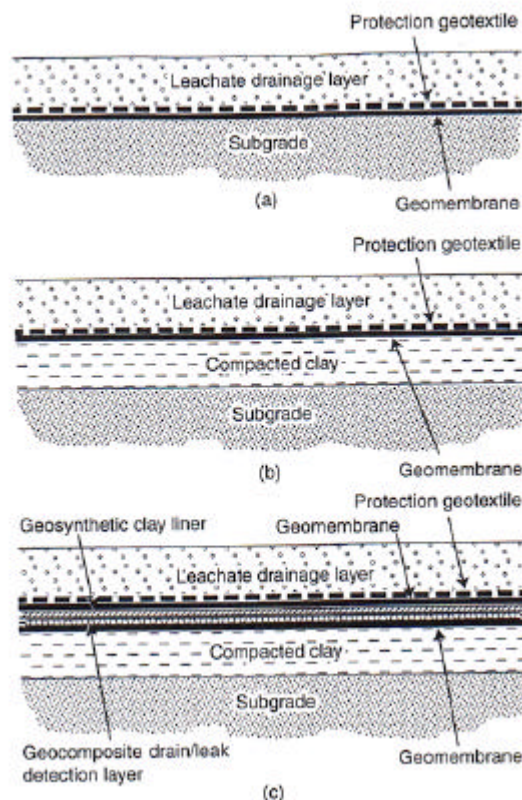


Figure 3.1 Typical liner arrangements for: (a) single liner; (b) composite liner; (c) double composite liner (from McQuade & Needham, 1999)

Geomembranes lining the base and side slopes of landfills are predominantly made from high density polyethylene (HDPE). HDPE liners are also sometimes used for capping applications. More flexible polyethylenes, sometimes generically termed very flexible polyethylene (VFPE), are more frequently used as landfill capping liners owing to their ability to undergo large strains

without failure. Occasionally, they are also used as side slope liners. Flexible polyethylene geomembranes include linear low density polyethylene (LLDPE), low density polyethylene (LDPE) and very low density polyethylene (VLDPE) although VLDPE is now not commonly available for landfill applications. LDPE is generally not used for landfill applications because of its lower durability and chemical resistance. Polyvinyl chloride (PVC) and polypropylene (PP) geomembranes are rarely used in the United Kingdom for landfill lining. The geomembrane sheet may be smooth or textured on both sides, or mono-textured with texturing only on one side, the texturing significantly increasing the interface shear strength with adjacent materials.

3.2 The structure and properties of polyethylene for geomembrane liners

3.2.1 The structure of polyethylene

A polymer (from the Greek *poly*, meaning many and *meros*, meaning part) is a long molecule consisting of many small units (monomers) joined end to end. Polyethylene (often also called polythene) is the simplest hydrocarbon polymer. It is a type of olefin (or alkene) hydrocarbon that has repeating units of two carbon atoms with four hydrogen atoms (called ethylene monomers $\{\text{CH}_2\text{-CH}_2\}_n$ or $[-\text{C}_2\text{H}_4-]_n$), which join together to form the molecule. The number of monomers in a molecule is usually in the order of 10^4 but may be as high as 10^6 or as low as 10^3 .

Depending on the polymerisation process used, side branched or linear polyethylene may be produced. The number of side branches may be varied during the production process by changing the polymerisation conditions. As the number of side branches per 100 carbon atoms is increased from zero there are pronounced changes in physical properties.

A conventional schematic drawing for polyethylene depicts the structure as a flat two-dimensional structure (Figure 3.2).

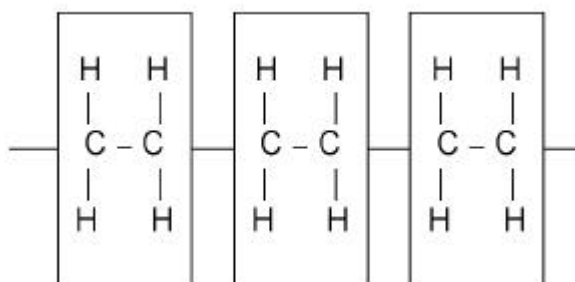


Figure 3.2 Repeating ethylene monomers in polyethylene

However, polyethylene actually has a long 'zigzag' carbon chain structure with hydrogen side groups (Figure 3.3).

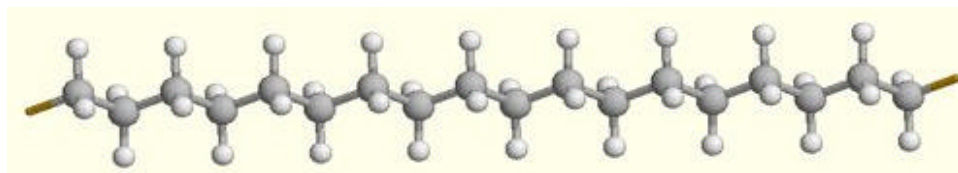


Figure 3.3 Polyethylene carbon chain with hydrogen side groups (from Harvey, 2003)

The regular hydrogen side group is interspersed with both long and short side branch chains (Figure 3.4). The generation of side branches can be controlled by the temperature and pressure

during the polymerisation process, while the degree of short side branching controls the final density of the polyethylene. A polyethylene with no, or very few, side branches is called a homopolymer. When the polyethylene has side branches, it is termed a copolymer, the components of the copolymer each being comonomers.

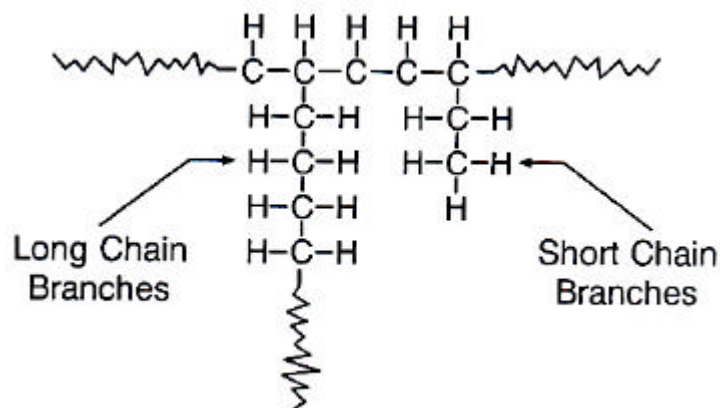


Figure 3.4 Polyethylene with a long chain branch and a butene comonomer short chain branch

Polyethylene is a semi-crystalline thermoplastic polymer consisting of two phases - crystalline and amorphous regions. It has a structure sufficiently symmetrical that, on cooling from the molten state, the polymer chains are able to orientate themselves into regions of alignment. These regions are in the form of lamellae that extend in flat sheets several microns in the x and y directions but are very thin (in the order of ten nanometres) in the third, z, direction. These crystalline lamellae are formed of tightly packed chains, with the polymer molecules repeatedly folding back and forth through the crystal region. The patterns of the crystalline lamellae are very complex, for example, forming a spoked-wheel type structure which, when extended into three dimensions forms a ball-like structure known as a spherulite.

The crystalline zones are separated by non-crystalline amorphous polyethylene, with the crystalline lamellae linked by polyethylene chains called tie molecules. The number, entanglement and distribution of these tie molecules are very important to the long-term performance of polyethylene geomembranes as they strongly influence the stress crack behaviour of the material. Tie molecules have a proportionately greater importance in a highly crystalline polyethylene such as HDPE as there are many more tie molecules linking the crystalline regions, compared to a less crystalline, more amorphous polyethylene such as LLDPE.

Lower density polyethylenes are highly side branched with both long and short-chained side branching. This degree of side branching hinders organisation and structure of the polymer on cooling producing a low density, poorly crystalline material. Linear low density polyethylenes (LLDPE) are virtually free of long side branches but contain short side branches due to co-polymerisation with higher alkenes. These branches interfere with the ability of the polymer to crystallise and low densities are produced.

In addition to side branching, the crystallinity of a polyethylene will also be controlled by its molecular weight. Higher molecular weight polymers have longer chain lengths and are less mobile when cooling. This lower mobility results in less organisation and structure and hence a

lower crystallinity. The mechanical and rheological properties of a polymer are largely dominated by the molecular weight and molecular weight distribution. The longer chain length of higher molecular weight polymers makes flow in the molten state more difficult and hence, high molecular weights are associated with higher viscosity polymer melts.

The melt index (MI) of a polymer (also called the melt flow index) is commonly used as an indirect measure of molecular weight. The MI is a measure of the flow of polymer at a certain temperature (190°C) through a defined die under the action of a certain weight (usually 2.16 kg) in a ten minute period (ASTM D 1238). The lower the MI value the higher the molecular weight. Commercial polymer grades are available in a range of MI values to match differing requirements of different processing techniques. HDPE geomembrane grades would typically have a MI in the region of 0.1 – 0.6 g/10 min (Hsuan, 2000). Brydson (1989) presents data (Figure 3.5) which show the relationship between MI and elongation at failure for a medium density polyethylene.

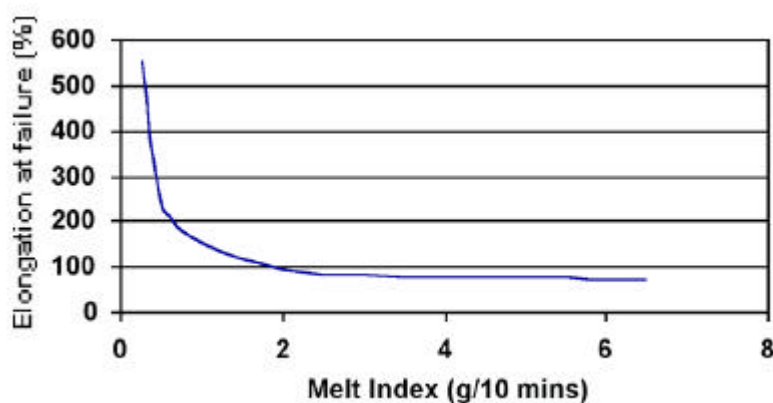


Figure 3.5 Effect of Melt Index on elongation at failure of MDPE (from Brydson, 1989)

Brydson (1989) also presents data (Figure 3.6) generalising the room temperature ductile-brittle transition region for polyethylene as a function of density and MI. For an MDPE resin with a density of 0.940 gcm^{-3} (note that the correct units in the SI system are kg l^{-1} but conventionally the density of geomembranes is given in gcm^{-3} which will be used in this report), the material would be expected to show brittle performance at MI values above approximately 0.8 g/10 min, illustrating the balance to be achieved between density, molecular weight and ductility.

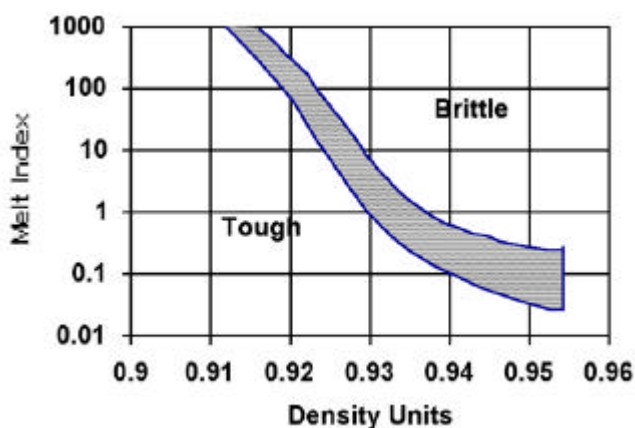


Figure 3.6 Polyethylene ductile - brittle behaviour in terms of Melt Index and density (from Brydson, 1989)

3.2.2 Structural properties of polyethylene geomembranes

Polyethylene resins are produced with a very wide range of densities, molecular weights, melt index values and other variables for different manufacturing processes and end products. For resins suitable for geomembranes, the values of density and melt index still vary widely which, with other variables, result in polyethylene geomembranes with very different physical properties and resistances to stress cracking and degradation.

Polyethylene geomembranes typically consist of 96% to 97.5% polyethylene resin, 2% to 3% carbon black and 0.5% to 1.0% antioxidant stabilisers (Hsuan and Koerner, 1995). The resin used is not a pure HDPE but a linear copolymer produced by adding α -olefin (butene, hexene, methyl pentane or octene) as comonomer to the dominant ethylene monomer. The amount of α -olefin has a direct effect on the density of the resin, the greater the amount of α -olefin added in the polymerisation resulting in a lower density polyethylene (Hsuan and Koerner, 1995). The comonomer forms short side branches along the backbone of the polyethylene chain.

Conventionally polyethylene is characterised into three density groups (ASTM D883 and ASTM D1248) as shown in Table 3.1:

Table 3.1 Classification of polyethylene

Resin Density (gcm ⁻³)	Description from ASTM D883	Description from ASTM D1248
0.910 – 0.925	Low density	Low density (Type I)
0.919 – 0.925	Linear low density	
0.926 – 0.940	Medium density	Medium density (Type II)
0.926 – 0.940	Linear medium density	
0.941 – 0.960	High density	High density (Type III)
0.961 and higher		High density (Type IV)

Note: Low and medium density polyethylenes are usually produced commercially by a process employing free radical polymerisation. Linear low and linear medium density polyethylenes are usually produced catalytically, i.e. by processes not employing free radical polymerisation.

HDPE geomembranes are commonly misunderstood as being manufactured from HDPE resin. This is not strictly correct. The base polymer is generally a linear medium density polyethylene (but normally abbreviated to MDPE) with a resin density in the range 0.932 to 0.940 gcm⁻³. As a result of the addition of carbon black, the density of the material is raised to the level of an HDPE (>0.941 gcm⁻³). Carbon black increases the density of the resin by 0.0044 gcm⁻³ for each 1% of carbon added (ASTM D3350). The geosynthetics community traditionally uses the density of the resin plus the additives to define the geomembranes as HDPE and the conventional term 'HDPE' is used in this report for such geomembranes. Density values of the resin in the region of 0.92 gcm⁻³ are typical for many commercial grades of LLDPE.

The crystalline zones of a polyethylene have a density of 1.004 gcm⁻³ while the density of the amorphous polymer is 0.853 gcm⁻³ (Apse, 1989). As polyethylene is a mixture of crystalline and amorphous polymer, there is a direct relationship between the density of polyethylene and its crystallinity. The crystallinity of the MDPE resin used in modern so-called HDPE geomembranes

is generally between 44% and 58% (Hsuan, 2000) and has reduced over the last twenty years to provide geomembranes with improved stress crack resistance. Sangam (2001) reports a crystallinity of 65.5% to 67.5% in a 14-year old HDPE lagoon liner, although the original values may have been modified (increased) by degradation processes. Apse (1989) noted that MDPE geomembrane resins in 1989 were typically 60% crystalline.

Hsuan *et al.* (1995) evaluated the properties of nine polyethylene geomembranes made from a range of different resins including LLDPE, MDPE and HDPE, and blends of these resins. The tensile modulus, yield stress and stress crack sensitivity (rather than resistance) all showed a decreasing trend as the low density component increased in the blends. This was mainly caused by the changes in the density and crystallinity of the materials. These show a decreasing trend as the low density component increases with respect to stress crack resistance as measured by the single point notched constant tensile load test (Figure 3.7).

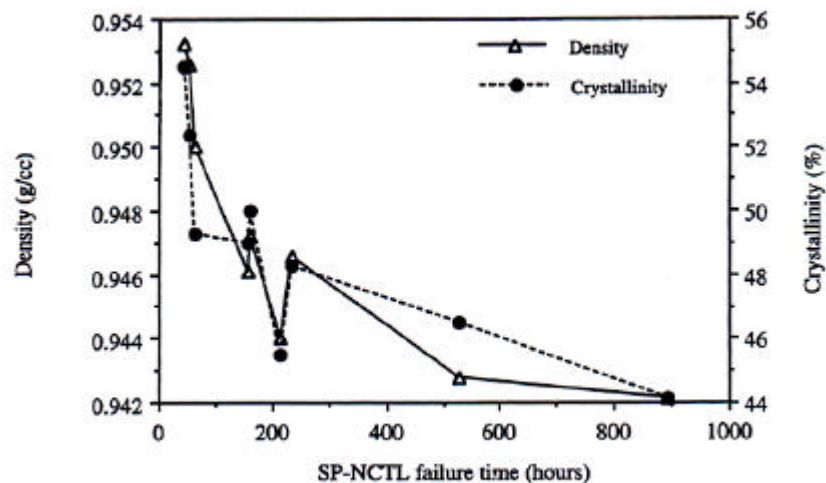


Figure 3.7 Density and crystallinity versus stress crack failure times (from Hsuan *et al.*, 1995)

4 POLYETHYLENE DEGRADATION MECHANISMS

4.1 Types of degradation

Degradation of natural polymers has been known since the earliest times with common examples being the deterioration of cellulose in wood and rubber in car tyres. Synthetic polymers similarly degrade with time. The types of degradation vary depending upon the structure of the polymer, the manufacturing history and the exposure conditions during its service life. Allen and Edge (1992) summarised the types of degradation processes, as set out below, identifying seven processes as being the most important in the majority of polymer uses.

- Thermal: this occurs during processing or use at elevated temperatures and may involve either oxidation or degradation.
- Mechanical: this occurs on the application of force or physical breakage. Chain scission may also occur here.
- Ultrasonic: the application of sound at certain frequencies may induce the polymer chains to vibrate and split.
- Hydrolytic: this occurs in polymers containing functional groups that are sensitive to the effects of water, especially those having a high moisture regain.
- Chemical: in this case, corrosive chemicals or gases, e.g. ozone, may attack the basic structural functionalities in the polymers causing chain scission and oxidation.
- Biological: this is specific to only a few polymers that contain functional groups that are attacked by micro-organisms.
- Radiation: on exposure to sunlight or high energy radiation, either the polymer or impurities within the polymer will absorb the radiation and induce reactions resulting in a loss of properties. In the case of high energy radiation the polymer chains will split directly.

Hsuan and Koerner (1995) describe the ageing process of HDPE geomembrane as a simultaneous combination of physical and chemical ageing. Physical ageing is associated with the slow process that takes place as the polymer attempts to establish equilibrium from its as-manufactured (non-equilibrium) state. For semi-crystalline polymers like HDPE, no primary bonds are broken and there is an increase in crystallinity. In contrast, chemical ageing indicates some type of degradation process (e.g. thermal oxidation or ultraviolet degradation - processes 1 and 7 above) involving the breakage of covalent bonds in polymer chains, intermolecular cross-linking and/or chemical reactions in the side groups or side chains. The process leads to a reduction in mechanical properties and eventually to failure. It follows that chemical ageing is the more important degradation mechanism to the long-term service life of a polyethylene geomembrane.

Koerner *et al.* (1990) describe the different types of degradation that an HDPE geomembrane may suffer as a consequence of the environmental conditions it is exposed to during its life in landfill and other containment applications. The degradation mechanisms identified include swelling, ultraviolet degradation, degradation by extraction, biological attack and oxidative degradation. In radioactive waste containment applications, degradation by high-energy radiation may also occur. The mechanisms described may be related back to the types of degradation processes identified by Allen and Edge (1992).

Swelling occurs when a geomembrane exposed to liquids (including leachate) increases in volume due to sorption of chemicals leading to the lubrication and disentangling of long tie-molecules in the amorphous phase between adjacent crystalline regions. This mechanism is largely reversible as desorption of the chemical can occur. The effect of swelling is to increase the permeation properties of the geomembrane and consequently, the material is more susceptible to degradation as chemicals can more readily reach polymer chains. Swelling is, in effect, a facilitator to other degradation processes rather than being a separate type of degradation as defined by Allen and Edge (1992). It is generally not a concern for HDPE geomembranes in most landfill applications provided that the leachate in contact with the geomembrane has low concentrations of the chemicals causing swelling (Rowe and Sangam, 2002). In lagoons storing certain chemicals or at hazardous waste sites where the leachate may contain relatively high concentrations of organic compounds, this would be a factor requiring consideration.

Ultraviolet (UV) degradation, also called photo-degradation, is induced by irradiation with UV or visible light and is a Type 7 degradation process (Allen and Edge, 1992). The wavelength of the sun's radiation extends from the infrared (>700nm) through the visible spectrum (about 400 - 700 nm) and into the UV range (<400nm) (Suits and Hsuan, 2003). When radiation strikes an exposed polymer surface, photons with energy similar to, or higher than, the chemical bond strength of the polymer cause a series of reactions that can lead to polymer chain scission and eventual degradation of polymer properties. In polyethylene and polypropylene, the degradation is controlled by photo-oxidation where free radicals created by the photon energy react with atmospheric oxygen.

Degradation by extraction occurs when one or more components, such as additives in the resin, are removed by exposure to chemicals or fluids. This is a degradation process of the geomembrane rather than one of the seven types of degradation of the polyethylene polymer itself. The effects of extraction become important when stabilisers and antioxidant additives are leached out, leaving the polyethylene susceptible to subsequent oxidative degradation.

Biological degradation (Type 6 degradation) arises when the polymer is attacked by micro-organisms where chemical reactions with the polymer are induced by enzymes produced by the micro-organisms. Koerner *et al.* (1990) indicated that this form of degradation is highly unlikely to occur in polyethylene geomembranes because of the high molecular weights (30,000 - 100,000) of polyethylene geomembrane resins.

As radionuclides in radioactive wastes decay, both energy from particles (alpha and beta) and gamma radiation energy are emitted (Badu-Tweneboah *et al.*, 1999). Of these, gamma radiation is the primary energy emitted. It affects polyethylene in a manner similar to UV radiation causing cross-linking in the amorphous regions (which increases crystallinity and density) and provides energy that can be used for oxidation (Type 7 degradation process).

The form of degradation that has the greatest detrimental effect on buried polyethylene geomembranes is thermal oxidative degradation (Type 1 in the list above). Polymer chains readily undergo chemical reactions with oxygen leading to important changes in molecular weight and molecular weight distribution. Oxidation is temperature dependant with the rate of oxidation increasing rapidly as temperature rises. As oxidation continues, the physical and mechanical properties of the polymer start to change eventually leading to failure of the geomembrane as an effective hydraulic barrier.

Rowe and Sangam (2002) noted that geomembranes might simultaneously experience several types of degradation processes during their service life. This would apply especially to liners for liquid impoundments where they are exposed to heat, UV radiation and thermally induced stresses. These different degradation mechanisms may have synergistic effects that could accelerate the overall rate of HDPE geomembrane degradation.

Oxidative degradation and means of retarding the process are discussed below.

4.2 Oxidative degradation mechanisms

It is generally accepted that the fundamental process underlying the oxidation of a polymeric material like HDPE is a free radical chain reaction. A free radical is a molecule with unpaired electrons which is therefore highly reactive. The mechanism is called autoxidation because such reactions often proceed automatically whenever polymers are exposed to oxygen. The autoxidation reaction was first reported by Bolland and Gee (1946). The results were rationalised as a free radical initiated chain reaction that can be regarded as proceeding by three types of reactions:

- chain initiation
- chain propagation
- chain termination.

When a hydrocarbon molecule breaks down (for example as a result of mechanical shearing during processing, exposure to heat or UV radiation, or attack by metal ions) two highly reactive radicals are released. This may be illustrated as shown in Figure 4.1 (Hsuan and Koerner, 1995).

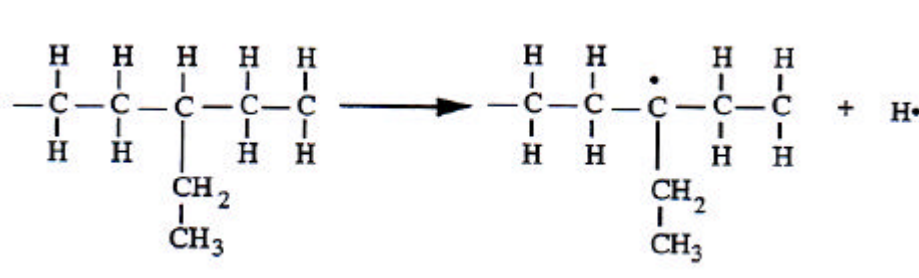


Figure 4.1 Formation of radicals (from Hsuan and Koerner, 1995)

The autoxidation process is described below where RH represents the polyethylene chains and the symbol R• represents a highly reactive free radical.

Chain initiation:



Chain propagation:

The free radical rapidly combines with oxygen to form a peroxide radical:



This peroxide radical will then react with a hydrocarbon to form an additional free radical and a hydroperoxide:



The radical will then react with a further oxygen molecule to produce an additional peroxide radical as per equation {4.2} whilst the hydroperoxide (ROOH) can either decompose by one of mechanisms {4.4i} or {4.4ii} below, or react as per equation {4.4iii}:

Decomposition:



Reaction:



Reactions {4.2} to {4.4} set up a series of accelerated chain reactions, the rate of which is governed by reaction {4.3}. This series of chain reactions will be terminated by one of the following mechanisms:

Chain termination:



The oxidation cycle is shown graphically in Figure 4.2 (Rowe and Sangam, 2002, modified from Grassie and Scott, 1985).

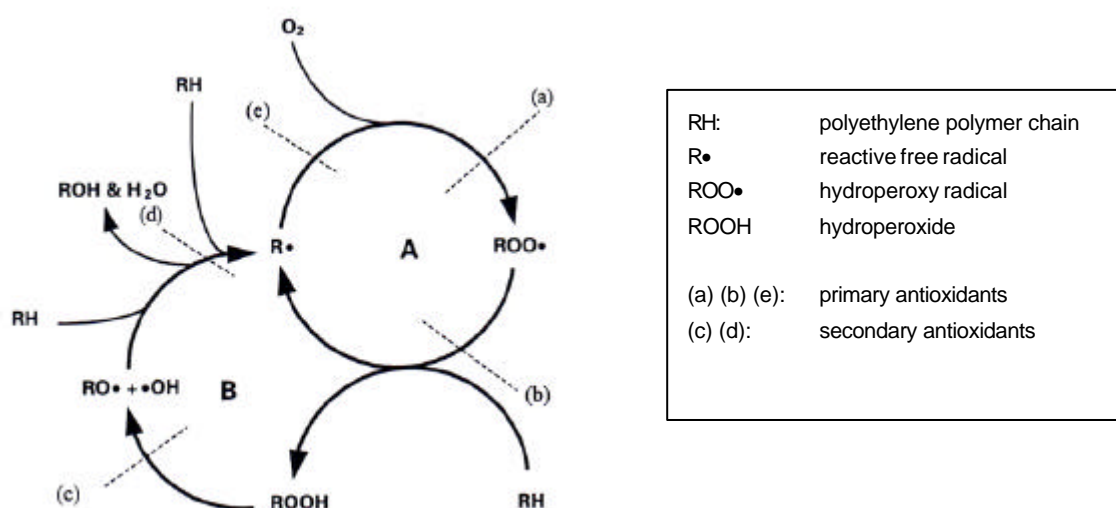
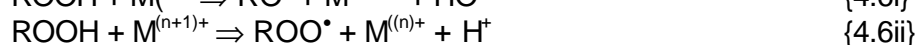


Figure 4.2 Oxidation cycles in polyethylene (modified from Grassie and Scott, 1985)

Hsuan and Koerner (1995) report that the oxidation reaction of polyethylene can be increased in the presence of transition metals, e.g. manganese, copper, aluminium and iron. The source of these elements usually comes from residual catalyst used to polymerise the resin (but these metals may also be present in leachate). Although the concentration of these elements is very low, they still can be a concern regarding the likely long-term durability of the polymer. The

transition metals break down hydroperoxides, creating an additional number of free radicals, as demonstrated in Equations {4.6i} and {4.6ii} where $M^{(n)+}$ and $M^{(n+1)+}$ are metal ions:



The oxidation reactions start slowly in an initial induction period. As described by Hsuan and Koerner (1995), in the induction period, little hydroperoxide is present and when formed, it does not decompose. Hence, the chain reaction cannot be initiated at first. As oxidation reactions gradually occur, additional hydroperoxide (ROOH) molecules are formed. The first formed oxidation products accelerate the further degradation. Once the concentration of hydroperoxide reaches a critical level, decomposition and reaction autoxidation occurs, signifying the end of the induction period and start of the acceleration period as shown on Figure 4.3.

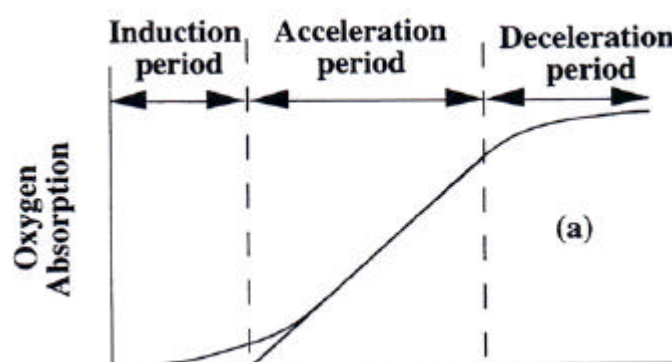


Figure 4.3 Stages of oxidation (from Hsuan and Koerner, 1995)

The chemical structure of a polymer strongly affects its ability to resist oxidative degradation. The rate of diffusion of oxygen into the polymer and the solubility of gases are much greater in amorphous regions than in the crystalline phase, such that the crystalline regions are inaccessible to molecular oxygen. It follows that the rate of oxidation is greater in amorphous than in crystalline polymers, and the oxidation rate of polyethylene is inversely proportional to the degree of crystallinity (Kelen, 1983). For short chain branches formed by comonomers, the particular carbon atom where the branch is attached to the polyethylene chain is surrounded by three other carbon atoms (Figure 4.1) and is termed the tertiary atom. The hydrogen atom attached to the tertiary carbon atom possesses a lower dissociation energy than other hydrogen atoms, thus free radicals are most likely to occur at these locations. Thus, polyethylene with greater branch density (e.g. LDPE and LLDPE) will generate more free radicals than those with fewer branches (HDPE) under the same conditions. Hence, LDPE and LLDPE will oxidise more rapidly than HDPE.

Of prime importance are those reactions that lead to a change in the molecular weight and molecular weight distribution of the polymer. For example, termination as shown by equation {4.5i} will result in chain scission. This will produce a reduction in the polymer chain length and a reduction in the molecular weight of the polymer and a consequential increase in the melt index. This increase in melt index will push the polymer closer towards the ductile-brittle transition region as shown in Figure 3.6. It follows that the oxidation reactions affect the polymer properties that determine the service life of the polymer and are the main reasons for material failure in an oxidative environment.

In semi-crystalline polymers such as polyethylene, oxidative degradation is a heterogeneous process in that the solubility and diffusion of oxygen from the surrounding environment occurs predominantly in the amorphous zones. As the mechanical properties of polyethylenes are determined to a large extent by the entanglement of the tie molecules, oxidative degradation in these regions leads rapidly to a loss of strength (Schwarzenbach *et al.*, 2001). The effect of oxidation on the tensile properties of the geomembrane is a decrease in break stress and break strain, and to a lesser extent the yield stress increases and the yield strain decreases. In time, the geomembrane material becomes brittle, with break strain occurring before the yield point is reached.

Hsuan and Koerner (1995) arbitrarily selected the limit of the 'service life' of the geomembrane as being reached when a specific design property has reduced in value by 50%. This is referred to as the 'half-life'. (In the context of the definition of "service life" used in this report, Hsuan and Koerner were referring to the material durability). However, although the, say, tensile strength or strain at break may have reduced by 50% and the geomembrane becomes brittle, the material remains intact and continues to act as a hydraulic barrier so long as it is not in a state of tensile stress and likely to suffer stress cracking. It follows that the "half-life" concept does not fit well with estimating the length of the service life of a geomembrane liner, as the end of the service life is reached when the geomembrane no longer acts as an effective hydraulic barrier, rather than when a tensile parameter reduces by 50%. The end of the service life, which is discussed further in Chapter 9, does not relate to a single property of the geomembrane. However, the "half-life" concept is useful when studying the oxidation stage of polyethylene durability, when testing has been conducted long enough to reach this stage

In the very limited oxygen environment that exists at the base of landfills, it is likely that slow oxidation of the geomembrane will still occur, if only from the underside of the geomembrane. Throughout oxidation, the HDPE geomembrane will continue to remain in place. It will be brittle and increasingly susceptible to stress cracking but there is no evidence that unstressed HDPE geomembrane fragments and disintegrates within a time frame of less than thousands of years (Müller, 2003). As noted in the laboratory research by Müller and Jakob (2003), once oxidation starts HDPE geomembranes can become brittle relatively quickly such that by bending 2.5mm thick specimens by hand, they easily break without yielding. However, without such stresses, the material remains essentially intact. Visually, these specimens appear like new and there is no indication of complete deterioration as is sometimes observed with polypropylene materials (Schwarzenbach *et al.*, 2001). In polypropylene and other polymers with branched alkanes as repeating units, the formation of hydroperoxide sequences on the polymer backbone readily occur, facilitating reactions with oxygen. This is why oxidative degradation happens more readily in polypropylene than in polyethylene.

Albertsson and Banhidi (1980) observed a small rate of oxidation of powdered HDPE during long-term exposure to water and to soil, resulting in a mass loss of 0.07% per year. In considering these results, Badu-Tweneboah *et al.* (1999) noted the extremely large surface area of the powdered sample compared to that of a 1.5mm thick HDPE geomembrane and estimated that the equivalent rate of mass loss was 0.00001% per year for the 1.5mm geomembrane. With this rate of mass loss, they estimated that it would take 10,000,000 years for the HDPE geomembrane to decay completely.

These observations lead to the conclusion that HDPE geomembrane can be considered, for the purpose of assessing the impact of leachate or gas leakages, as a 'permanent' material within a landfill lining system and will not disintegrate or disappear within a timeframe of interest, so long as they are not subjected to tensile or shear stresses. They are, however, susceptible to physical damage, tensile (ductile) failure under conditions of high stress, embrittlement with oxidation, and stress cracking at locations of only low levels of stress.

4.3 Antioxidants and carbon black stabilisers

Oxidative degradation in polyethylene geomembranes can be inhibited by suitable stabilisers called antioxidants which are introduced in small quantities to the polyethylene for the purposes of oxidation prevention and to ensure a long-term material durability. These complex compounds may be in the resin supplied to the geomembrane manufacturer or may be added (typically with the carbon black additive) during geomembrane manufacture. Antioxidants are usually supplied in proprietary packages of two or more antioxidants under trade names. The precise contents are commercially confidential information.

Antioxidant additives function by interrupting the autoxidation sequence described in Section 4.2 and shown on Figure 4.2. There are two principal generic antioxidant types:

- Chain breaking antioxidants (primary antioxidants) which interrupt the propagation cycle i.e. by breaking links (a), (b) and (d) in Figure 4.2 by reacting with both R^\bullet and ROO^\bullet radicals to produce their own termination reactions.
- Preventative antioxidants (secondary antioxidants) which decompose hydroperoxides, intercepting link (c) in the "B" cycle in Figure 4.2 forming inert reaction products.

Figure 4.2 illustrates the actions of primary and secondary antioxidants. A third group of additives referred to as oxidation retarders also exist. These react with R^\bullet and ROO^\bullet to produce new but slower propagation reactions thus slowing the overall chain reaction. Metal deactivators may also be used to trap and render trace metal ions inactive.

Antioxidants are used in quantities in the region of 0.02 to 1%. If used in excess, certain antioxidants may actually induce oxidation rather than extending the induction period. Usually, a combination of both primary and secondary antioxidants is used. Synergistic effects are noted where two antioxidants produce an effect greater than the sum of the individual components. Such synergistic effects can be produced either through the two stabilising systems operating by different mechanisms (i.e. peroxide decomposition and chain breaking) or through one antioxidant regenerating the second thus reducing depletion rates. Conversely, a mixture of two or more additives can result in a weaker stabilising effect, called antagonism, than would be expected from the sum of the individual components.

In addition to the categories of primary and secondary, antioxidants can be further classified into four chemical types within which many different products are included. Table 4.1 lists the chemical type of some of the commercially available antioxidants that can be used in polyethylene geomembranes. To ensure long-term durability, a manufacturer will use two or more types of antioxidants, at least one from each category (Hsuan and Koerner, 1998).

Table 4.1 Types of antioxidants (modified from Hsuan and Koerner, 1998, and Müller, 2001)

Chemical Type	Example of commercially available antioxidants
Primary antioxidants	
Hindered phenols	Irganox 1076; Irganox 1010; Santowhite crystals
Hindered amines (HALS*)	Various types of Tinuvin; Chimassorb 944; Various types of Hostavin and Uvinul
Secondary antioxidants	
Phosphites	Irgafos 168
Sulphur compounds (thiosynergists)	Dilauryl thiodipropionate (DLTDP); Distearyl thiodipropionate (DSTDP)

* Hindered Amine Light Stabiliser

The effective temperature range for each of these antioxidants differs and needs to be taken into account when selecting the antioxidant system to protect the geomembrane both at the high temperature during manufacture (and welding on site) and the significantly lower temperatures during its service lifetime. For the four chemical types listed in Table 4.1, the effective temperature ranges are shown in Figure 4.4.

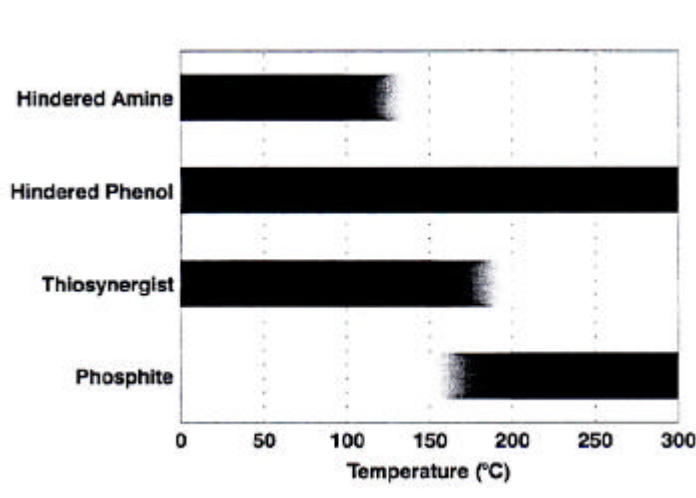


Figure 4.4 Effective temperature ranges of four antioxidant types (from Fay and King, 1994)

The figure shows that phosphites have an effective temperature range above 150°C and are used as processing stabilisers during geomembrane manufacture. Either hindered phenols, thiosynergists or hindered amines are added to the formulation to provide the low temperature service protection. The synergy of phenol and phosphite antioxidant systems has been assessed on HDPE by Allen *et al.* (2001). The inclusion of phosphites can prevent consumption of phenols during processing and hence extend the useful life of the phenol system. For a formulation consisting of hindered phenols, a wide range of temperatures is covered, from ambient to process temperatures. However, hindered phenols are only primary stabilisers and a

secondary antioxidant effective in service temperatures is also required which could be either a thiosynergist or a hindered amine (Hsuan and Koerner, 1998).

The most common type of UV protection for polyethylenes is provided by carbon black. Carbon black is a particulate form of industrial carbon consisting of very fine particles (the prime particles) fused together to form primary aggregates (Suits and Hsuan, 2003). The UV absorbing efficiency of the carbon black is governed by the average prime particle size. Primary aggregates composed of finer prime particles present greater surface area for optical absorption than those made from larger prime particles. Thus, UV absorption increases as prime particle size decreases. Carbon black has an antagonistic effect on many phenol and amine antioxidants but is synergistic with some phenolic sulphides, significantly enhancing the UV resistance of the resulting stabiliser package. For non-black geomembranes, UV screeners are added to the resin to protect the polymer from UV degradation.

While this subject is complex, it should be recognised that the polymer industry expends considerable effort and resources into antioxidant research and selection. There are many custom-designed stabiliser packages for each polymer product, including geomembranes, in order to accommodate a wide range of processing and service requirements.

4.4 Antioxidant depletion

Hsuan and Koerner (1995) describe the oxidative degradation of stabilised polyethylene as a three-stage process, the third stage being subdivided into the acceleration and deceleration periods, as shown on Figures 4.3 and 4.5.

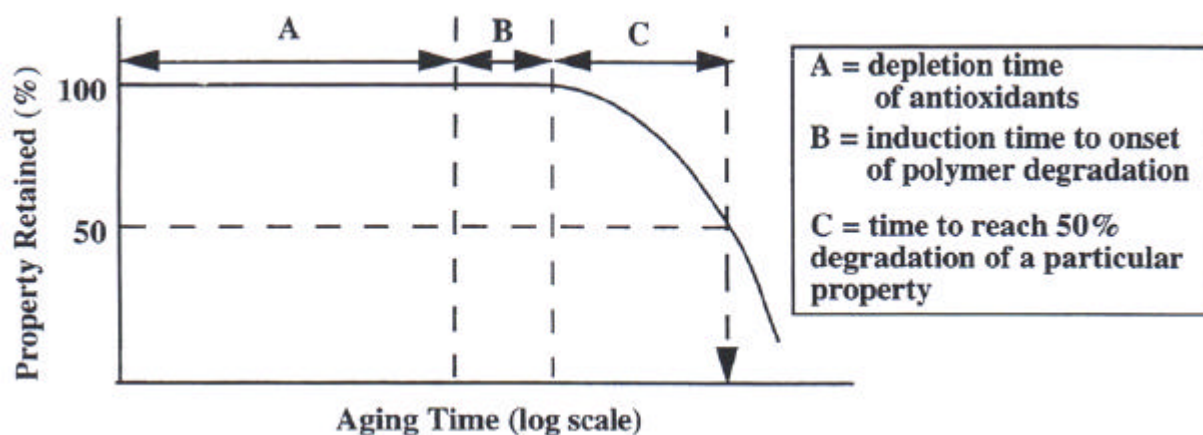


Figure 4.5 Three stage oxidative degradation of stabilised polyethylene (from Hsuan and Koerner, 1995)

The first stage is the depletion of the antioxidants and is due either to their consumption as a result of their chemical reactions with oxygen, free radicals and alkyl peroxides and/or to their physical loss by diffusion, extraction or volatilisation (Luston, 1986). Following depletion of the antioxidants, the oxidation reactions generally start very slowly in an initial induction period, as already described, followed by the third stage being the actual oxidation.

Depending on the operating mechanisms of an antioxidant system, the antioxidants may act sacrificially and be consumed during the service life of the geomembrane. Additive depletion

also occurs due to migration of the additive from the bulk to the surface where it can evaporate from the polymer into the surrounding air, or be leached out by a contacting liquid (such as landfill leachate or water). The rate of removal will be controlled by the migration by diffusion of the additive through the amorphous phase of the polymer. The molar volumes of typical additives are significantly greater than the volume of a crystal unit cell of a polymer, implying that additives cannot get into the polymer crystal. It has been shown that additives dissolve only in the amorphous phase of a polymer and are rejected from the crystalline phase during cooling from the melt during processing (Billingham, 2001). This is an advantage because oxygen does not enter the dense crystalline phase of typical polymers and therefore stabilising additives are concentrated in the regions where degradation occurs. It follows that the mobility of an additive (defined by the diffusion coefficient) within a semi-crystalline polymer is controlled by the free volume of the amorphous phase as the free volume defines the space available for an additive to move through the polymer. Foldes (1998) compared the diffusion rate of several antioxidants in several polyolefins and showed that a higher free volume allows more gaps in the polymer structure for the additive to occupy as it moves through the structure.

Polymer degradation can be very localised in nature with high rates of localised oxidation (Celina *et al.*, 1995). Hence, solubility and the homogeneous dispersion of an antioxidant within the amorphous phase of a polymer are vital to achieving stability of that polymer formulation.

When antioxidant loss takes place by evaporation of the additive from the surface of the polyethylene, this causes surface depletion and sets up a concentration gradient. Further loss can only occur by diffusion of the additive to replenish the surface from the bulk. The volatility of antioxidants is a thermally activated process and temperature changes affect not just the evaporation of antioxidants from the polyethylene surface but also their rate of diffusion from the interior to the surface. Exposed geomembranes subjected to high surface temperatures will be susceptible to antioxidant volatilisation. Antioxidant loss tends to be controlled by the evaporation rate if the sample is a thin film or fibre, where there is a large surface area / volume ratio but diffusion rates dominate as the sample becomes thicker (Billingham, 2001). Diffusion will tend to control antioxidant loss from 1.0mm to 2.5mm thick HDPE geomembranes.

As noted by Billingham (2001), if the polymer is in contact with a liquid that is a solvent for the additive, then the additive can be lost from the surface by extraction at a rate much higher than that into air, other factors such as temperature, being equal. In this case, the rate of extraction is controlled by diffusion of the additive through the polymer. The higher rate of depletion on exposure to liquids has been confirmed experimentally. Smith *et al.* (1992) performed a study by exposing a pipe to water internally and air externally. The antioxidant depletion across the thickness of the pipe was monitored using the oxidative induction time (OIT) test. They found that the depletion of antioxidants was three times faster in water than in air.

Sangam (2001) undertook a laboratory study into the degradation of a 2.0mm HDPE geomembrane (discussed in detail in Section 5.4). The geomembrane samples were incubated in air, water and a synthetic leachate at temperatures of 40°C, 55°C, 70°C and 85°C. The results from the OIT tests indicated that the depletion of antioxidants was 1.6 to 2.4 times faster in immersed samples in water than for samples exposed to air. For leachate-exposed samples, the depletion was about four times faster than in air and 1.6 to 3.2 times faster than in water. Sangam (2001) considered that the high antioxidant depletion rates measured in leachate may be attributable to the surfactants in the synthetic leachate acting on the geomembrane by increasing its "wettability" due to the reduction of the material surface tension. Consequently, the

antioxidants at the surface would be more quickly dissolved and extracted. A further reason may be the presence of transition metals in the synthetic leachate because transition metals are known to increase the consumption of antioxidants.

Müller and Jakob (2003) conducted an extensive series of tests to assess the depletion of antioxidants following oven ageing in air at 80°C and ageing in distilled water at the same temperature (discussed in detail in Section 5.4). They assessed rates of stabiliser loss from a number of commercially available geomembranes and found depletion rates to be significantly faster when geomembranes were aged in water. This effect is shown in Figure 4.6, which compares the reduction in oxidative induction time (OIT) during oven ageing at 80°C with that of ageing in water at 80°C. The oven ageing data are presented as an average plot for all samples assessed and the water ageing is for their sample 139.

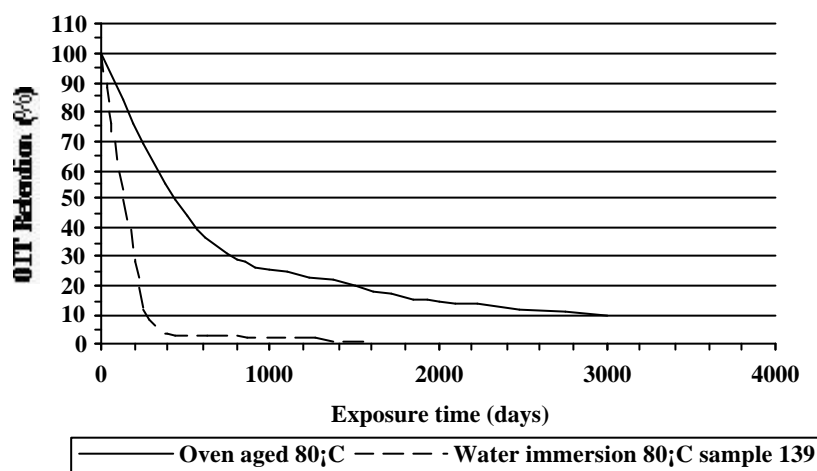


Figure 4.6 Comparison of OIT loss for air and water exposure, at 80°C (from Müller and Jakob, 2003)

Müller and Jakob (2003) also concluded that at 80°C, diffusion-controlled migration of antioxidant from the sample followed by evaporation or leaching was the dominant mode of antioxidant depletion and that consumption of the antioxidant by reactions with oxygen played only a minor role. This implies that if diffusion is too slow to replenish the antioxidant at the same rate as the removal is occurring, then oxidation may occur in a thin surface layer during antioxidant depletion. If thick enough, this layer may then be the initiation site for surface crazes which could then develop into stress cracks (Peggs, 2003).

5 LABORATORY STUDIES INTO OXIDATIVE DEGRADATION

5.1 Introduction

Since antioxidant loss and subsequent oxidation are very slow processes, case histories of field performance are not available to investigate the degradation mechanisms. Instead, laboratory testing accelerated by the use of elevated incubation temperatures is used to investigate the loss of antioxidants and oxidative degradation. Even so, these laboratory research projects still extend over many months and years. Several investigators have conducted laboratory tests into durability and degradation issues related to HDPE geomembrane liners. Of these, three important long-running research projects have been carried out into the long-term oxidative degradation of HDPE geomembrane liners. These are reported by Hsuan and Koerner (1995) and Hsuan and Koerner (1998) at the Geosynthetic Research Institute, Pennsylvania; Sangam (2001) and Rowe and Sangam (2002) then at the University of Western Ontario, Canada; and by Müller (2001) and Müller and Jakob (2003) at the Federal Institute for Materials Research and Testing (BAM) in Berlin, Germany. The findings of these extensive projects form the basis of the current understanding of antioxidant depletion and, to some extent, subsequent oxidative degradation of HDPE geomembranes under laboratory conditions.

5.2 Test methods to assess degradation

The laboratory testing involves ageing the geomembrane samples at elevated test temperatures to accelerate the degradation reaction. The geomembrane samples in the heated incubation devices are retrieved after pre-determined periods. The progression of the degradation process is monitored by the results of a set of physical, chemical and mechanical tests, as described below.

There are several properties of polyethylene geomembranes which either control the durability of the material or which may be examined to assess the progress of degradation. These are given in Table 5.1 together with the relevant test method. All of the methods are to ASTM specifications.

Table 5.1 Properties and test methods of HDPE geomembranes for assessing degradation

Property	Test procedure	Test method (all ASTM)
Density		D1505
Molecular weight	Melt Index	D1238
Crystallinity	Differential scanning calorimeter (DSC)	E794
Polymer structure	Fourier transform infrared spectroscopy (FTIR)	
Tensile behaviour		D638 or D6693
Stress crack resistance	Notched Constant Tensile Load (NCTL)	D5397
Oxidation	Standard Oxidative Induction Time (OIT) using DSC	D3895
	High pressure OIT	D5885
Chemical resistance	Sample incubation and specified comparative tests	D5322 and D5747
Ultraviolet degradation	Xenon arc	D4355

At present, the European Committee of Standardisation (CEN) has not released for publication by the national bodies, including BSI, the final versions of relevant test procedures, although several draft European standards have been published.

At the time of writing, relevant draft EN standards are as follows:

Standard	Title
pr EN ISO 10318	Geotextiles, geotextile-related products, geomembranes and Geosynthetic clay liners – Terms and their definitions.
pr EN ISO 13438	Geosynthetics – screening test method for determining the resistance to oxidation at elevated oxygen pressure.
pr EN 13493	Geosynthetic barriers – required characteristics for use in solid waste storages and waste disposals sites.
pr EN 14414	Geosynthetics – screening test method for determining chemical resistance for landfill applications
pr EN 14575	Geosynthetics – determination of the resistance to oxidation
pr EN 14576	Geosynthetics – determination of resistance of polymeric geosynthetic barriers to environmental stress cracking.

Most of the above tests are similar to their ASTM equivalents. Following their issue as EN standards, it is expected that in Europe, testing to the EN standards will become the norm.

5.2.1 Oxidative Induction Time

The oxidative induction time (OIT) test is used to measure the amount of antioxidant remaining in the incubated geomembrane specimens. The OIT value is the time required for the geomembrane sample to be oxidised under a specific temperature and pressure, therefore indicating the amount of antioxidant remaining in the test specimen. This test is non-specific with respect to the different types of antioxidant in the stabiliser package. Therefore, the concentrations of the individual antioxidants cannot be obtained. There are two OIT tests – the standard test (OIT) according to ASTM D3895 and the high pressure OIT test (HP-OIT) to ASTM D5885.

The standard test method covers a procedure for the determination of oxidative induction time (OIT) of polymeric materials by differential scanning calorimetry (DSC). The small geomembrane specimen (about 5mg) is heated at a constant rate in an inert gaseous environment (nitrogen). When the specified temperature has been reached, usually 200°C, the atmosphere is changed to oxygen and maintained at the same flow rate. The specimen is held at a constant temperature until the oxidative reaction is displayed on the thermal curve (Figure 5.1). The time interval from the first initiation of oxygen flow to the oxidative reaction is known as the induction period. The end of the induction period is marked by an abrupt increase in the heat or temperature of the specimen.

Although aluminium sample pans are generally used for geomembrane samples, copper sample pans can be used. Müller and Jakob (2003) state that typically the OIT values at a given testing temperature are about five times higher with aluminium pans than with copper pans. They also

used different OIT test temperatures (140 - 210°C), the lower values being used to reduce the high temperature effects.

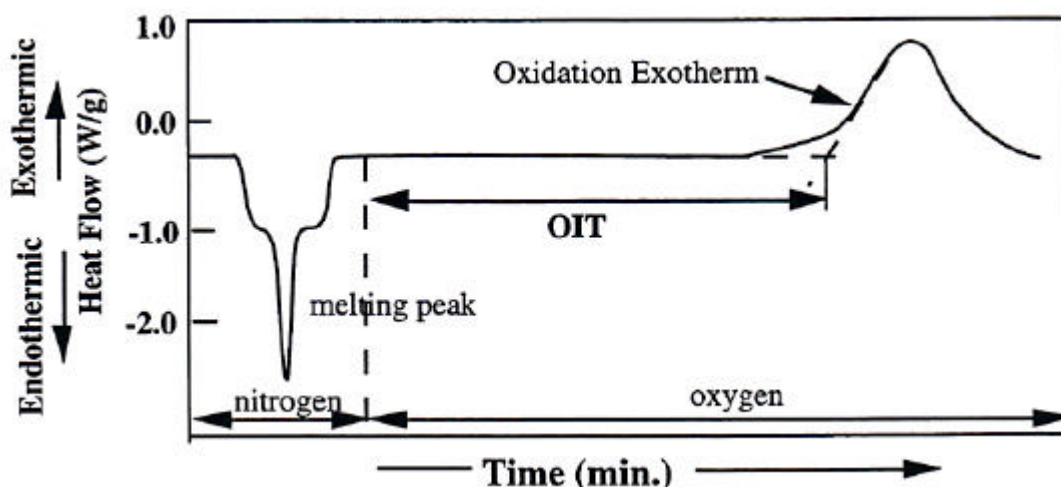


Figure 5.1 Thermal curve of a standard OIT test (from Hsuan and Koerner, 1995)

The HP-OIT test is also performed using DSC but with a different cell which can sustain a high pressure (3500 kPa test pressure). The test is conducted at the lower temperature of 150°C. This temperature is specified as HDPE reaches complete melting by 140°C, so at 150°C the thermal oxidation proceeds in a fully molten state. The lower temperature compared to the standard test would considerably lengthen the test duration but this is partly offset by the increased pressure. Nevertheless, testing times are several times longer than the standard OIT test. The main reason for developing the HP-OIT test is that the standard OIT test is unable to evaluate the stabilisation effect of hindered amine antioxidants, as these will rapidly volatilise at the test temperature of 200°C (Hsuan and Koerner, 1998). However, this is based on the premise that the HP-OIT test is able to detect the full presence of hindered amines at a test temperature of 150°C, which is above their effective temperature range (Figure 4.4). Hsuan and Koerner (1998) also note that the HP-OIT test is a less sensitive test, particularly for short OIT values, compared to the standard OIT test.

Depending on the antioxidant package used in a particular geomembrane, there may be a linear relationship between the standard OIT and HP-OIT test values. Hsuan and Koerner (1998) found a linear relationship for the geomembrane formulation they tested and commented that this implied that there were no hindered amines in the antioxidant package used for the geomembrane. Where a geomembrane contains hindered amines, they considered that a linear relationship between the two types of OIT tests is unlikely to occur. Their work suggests that either test procedure could be used to evaluate the antioxidant depletion where hindered amines are not present and, as the standard test is easier to perform, then it would be used in preference. Müller and Jakob (2003) argue that oven ageing or OIT testing at high oxygen pressures changes the method of antioxidant depletion from a migration and removal process (by leaching or evaporation) to one where the antioxidants are consumed by inhibiting the oxidation reaction chain. As antioxidant consumption is not relevant for typical landfill conditions, the results of HP-OIT tests have to be interpreted with caution.

5.2.2 Molecular weight

When oxidative degradation takes place in the geomembrane, the molecular weight of the polymer will be altered due to either cross-linking or a chain scission reaction. In general, cross-linking increases the molecular weight whereas chain scission causes a decrease. The melt flow index (MFI) or melt index (MI) test (ASTM D1238) is a qualitative method used to assess the molecular weight of the polymer, and therefore is an indicator of oxidation. The MI is defined as the amount of polymer extruded through a standard orifice at a given time and temperature. In principle, MI is inversely related to the molecular weight. Although several test conditions are specified in ASTM D1238, condition E is usually used for HDPE in which the amount of molten polymer at 190°C extruded through the standard orifice under a load of 2.16kg in 10 minutes is recorded. The result is expressed in units of g/10 minutes.

The test can also be performed using two different weights, for example, 2.16kg and 5kg at the same temperature. The resulting MI values give a ratio known as the Melt Flow Ratio (MFR) or Flow Rate Ratio (FRR):

$$\text{FRR} = \text{MI}_{5.0} / \text{MI}_{2.16}$$

High values of FRR, other things being equal, indicate broader molecular weight distributions. It should be noted that in Germany, the MI is determined according to DIN ISO 1133:1991 and the FRR is calculated using the MI for weights of 21.6kg and 5kg (Müller and Jakob, 2003).

5.2.3 Tensile properties

The tensile properties of the geomembrane can reveal the onset of embrittlement and is commonly used to assess oxidative degradation. The values of stress and strain at both yield and break are recorded. Of particular interest is the reduction in strain at break as this is sensitive to degradation. Several methods of tensile test are used, such as ASTM D638 and the recently introduced ASTM D6693, and BS EN ISO 527.

5.2.4 Stress crack resistance

Changes in the molecular weight of an HDPE geomembrane and embrittlement are likely to lead to a reduction in stress crack resistance. Stress cracking is discussed in Chapter 7. The test used to measure the effects of oxidative degradation on stress crack resistance is the single point notched constant tensile load (SP-NCTL) test according to ASTM D5397 Appendix. The test consists of introducing a controlled notch into one side of a dumbbell shaped geomembrane specimen, suspending it in a surfactant bath and subjecting it to a load of 30% of the yield stress. The standard requirement is a failure time greater than 300 hours (GRI GM13), recently increased from 200 hours.

5.2.5 Crystallinity

Changes in crystallinity have been noted in most semi-crystalline polymers as a result of ageing. In general, chain scission during oxidation produces smaller chains that can crystallise more easily, resulting in an increase in crystallinity as noted by Bandyopathyay *et al.* (1985). Although more sophisticated methods such as gel permeation and x-ray diffraction exist, the percent

crystallinity is usually evaluated using a differential scanning calorimeter (DSC) which measures the heat absorbed or released by a small sample while it undergoes thermal conditioning. In this technique described in ASTM E794, a specimen of known mass is heated at 200°C at a rate of 20°C/min and under nitrogen atmosphere (flow rate of 50ml/min). The amount of crystallinity is calculated from the heat of the fusion represented by the area under the endothermic peak. The percent is then calculated relatively to 100% crystalline HDPE.

5.2.6 Polymer structure

Most of the degradation of polymeric materials used in a geomembrane results from irreversible reactions of polymer molecules. Thus, evaluating changes in molecular composition provides a logical method for measuring the course of polymer degradation. As hydrocarbon polymers degrade, many different changes in polymer composition occur. Among available methods used to investigate these changes, the molecular composition is usually studied by Fourier transform infrared (FTIR) spectroscopy.

FTIR has long been recognised as a powerful tool for polymer characterisation and has been used successfully to study the degradation of polyethylene. It gives quantitative and qualitative information about physico-chemical composition of polymers, copolymers, polymer blends, composites and additives used in these materials. It is also used to study the functional groups (carbonyl, vinyl, vinylidene, alkene and hydroxyl). The method consists of taking thin slices of samples and submitting them to infrared light. The infrared spectra arise from the molecular transitions between quantum states of differing internal energies (Cernia *et al.*, 1963). The frequency of the emitted or absorbed radiation is related to energy differences and is associated with molecular vibrations and rotations characteristic of the chemical groups present (Hamid and Pritchard, 1988). In the case of oxidation, the carbonyl groups associated with the degradation products are detected by absorption bands between 1850 and 1750cm⁻¹. In general, the observations are reported as dimensional functional groups. For example, for polyethylene molecular degradation the functional group usually used is the carbonyl index as the ratio of the formed carbonyl peak (between 1850 and 1750cm⁻¹) and the peak of a stable compound.

5.2.7 Chemiluminescence

In the past few years, chemiluminescence (CL) has been gaining acceptance as a sensitive method of studying oxidative polymer degradation (Allen and Edge, 1992). CL is an analytical tool, which works by indicating the emission of photons resulting from the excitation of molecules by chemical reactions in solid and liquid systems, and has been known for forty years.

Researchers use proprietary apparatus designs to follow the degradation of unstabilised and stabilised polymers. Investigations include the kinetics of oxidation and the reaction (Setnescu *et al.*, 1998). CL has been shown to be sensitive enough to measure the thermo-oxidative differences of even individual polypropylene powder particles with sample weights in the microgram range.

5.3 Arrhenius modelling

Laboratory investigations into polymer degradation require accelerated tests in order to obtain results in a reasonable timeframe. Most chemical reactions for degradation are strongly

influenced by the temperature and the concentration of reagents. This dependence can be used advantageously to extrapolate short-term high temperature or high concentration data to predict longer-term service conditions. The predictive technique most widely used for polymer degradation is based on a time - temperature superposition principle called Arrhenius modelling. Koerner *et al.* (1992) and Shelton and Bright (1993) describe this technique for the prediction of geosynthetic degradation.

The rate constant of a chemical reaction (K_r) can be defined by the Arrhenius equation, which describes the temperature dependence of the reaction:

$$K_r = A e^{\left(\frac{-E_a}{RT}\right)} \quad \{5.1\}$$

or, by taking the natural logarithm of both sides, the following linear equation is obtained:

$$\ln(K_r) = \ln(A) - \left(\frac{E_a}{R}\right)\left(\frac{1}{T}\right) \quad \{5.2\}$$

where:

- A = constant
- E_a = activation energy (kJ/mol)
- R = gas constant (8.314 J/mol K)
- T = absolute temperature (Kelvin)

Comparing the latter form of the Arrhenius equation to the equation for a straight line, plotting $\ln(K_r)$ vs. $1/T$ will give a plot where the slope is $-E_a/R$ and the intercept is $\ln(A)$, as shown on Figure 5.2.

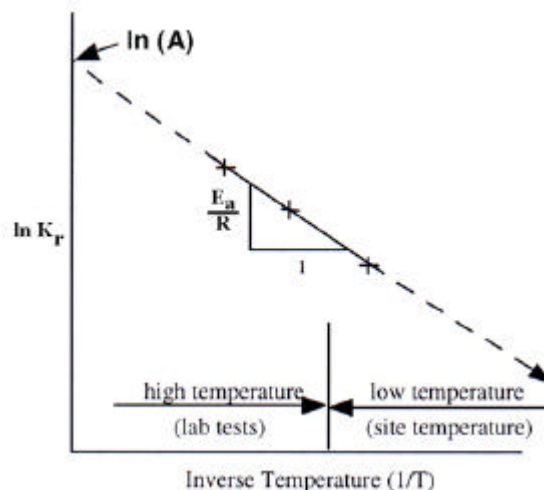


Figure 5.2 Typical form of Arrhenius plot (modified from Hsuan and Koerner, 1995)

Assumptions associated with the Arrhenius equation are:

- K_r is only a function of temperature
- A does not affect the temperature sensitivity of the reaction
- E_a remains constant over the time and temperature range of interest for evaluation, extrapolation and prediction

If the mechanism of the chemical reaction remains constant over a temperature range T_1 to T_2 , the change in rate constant over the temperature range can be assessed by defining an Arrhenius equation for each temperature and calculating the difference:

$$\ln(K_{r2}) - \ln(K_{r1}) = \left[\ln(A) - \frac{E_a}{RT_2} \right] - \left[\ln(A) - \frac{E_a}{RT_1} \right] \quad \{5.3\}$$

$$\ln\left(\frac{K_{r2}}{K_{r1}}\right) = \left(\frac{E_a}{R}\right) \left(\frac{1}{T_1} - \frac{1}{T_2}\right) \quad \{5.4\}$$

where, K_{r1} and K_{r2} are the rate constants at Kelvin temperatures T_1 and T_2 .

Data are commonly plotted as an Arrhenius plot whereby the logarithm of time is plotted against the reciprocal of temperature (Kelvin).

It is important to note from these equations that temperature has an exponential effect on the time required to produce a specific level of property loss. The activation energy will also have an exponential effect and careful selection of this parameter is essential.

The complexity of temperature dependence and the fact that measured values of the activation energy are subject to error means that extrapolation from high temperature data to predict service temperature behaviour needs caution. When measured over a wide enough temperature range, Arrhenius plots are often curved thus making linear extrapolations to lower service temperatures unreliable (Billingham, 2001). It is important to obtain as much high-temperature data as possible to ascertain if E_a is constant and to minimise the extrapolation to the lower, site-specific temperature (Koerner *et al.*, 1992).

Using the Arrhenius equation approach, the time for essentially full depletion of the antioxidants can be computed as follows.

The lifetime of an antioxidant package can be estimated using the depletion rate of the OIT values (Hsuan and Koerner, 1995). Figure 5.3 shows the natural logarithm of standard OIT data against incubation time from a set of samples incubated in test cells (as described in section 5.4 below) at different temperatures over a 24-month period. A set of linear response curves has been drawn and the slopes of the lines represent the OIT depletion rate at each particular temperature.

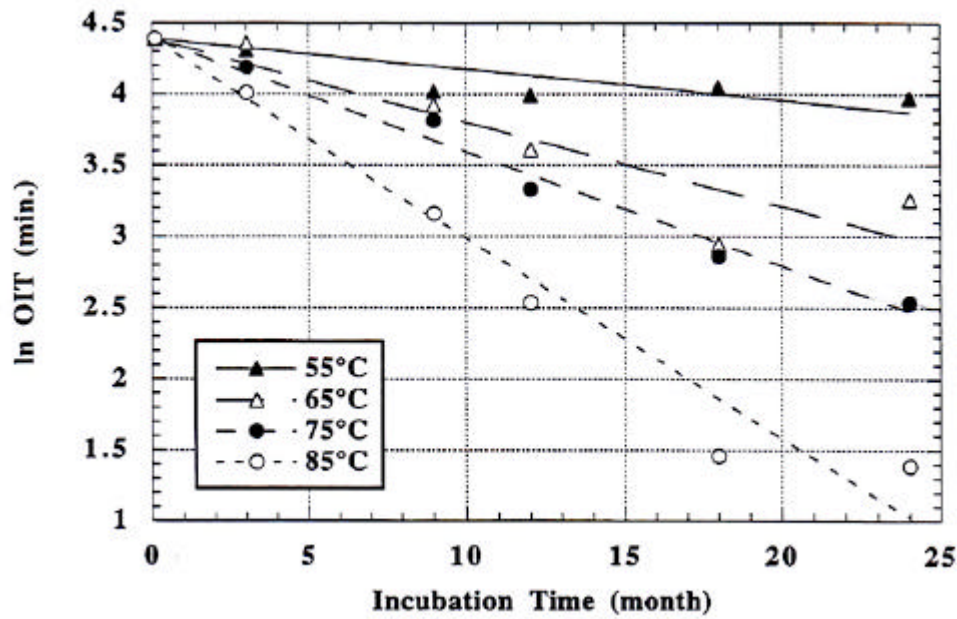


Figure 5.3 Natural logarithm (OIT) versus incubation time for OIT tests (from Hsuan and Koerner, 1995)

The generalised equation for each line is expressed by:

$$\ln(OIT) = \ln(P) - S.t \quad \{5.5\}$$

where:

- OIT = OIT time
- S = OIT depletion rate (slope of the line)
- t = depletion time
- P = constant (the original value of OIT of the geomembrane)

The OIT depletion rate then needs to be extrapolated to the lower, site-specific temperature using the Arrhenius equation approach:

$$S = A.e^{-E_a/RT} \quad \{5.6\}$$

$$\ln(S) = \ln(A) + \left(-E_a/R\right)\left(1/T\right) \quad \{5.7\}$$

A linear relationship is established between $\ln(S)$ and inverse temperature as shown on Figure 5.4. The activation energy is derived from the slope of the line. The OIT depletion rate (S) can thus be determined for any selected temperature. The value of S can then be used in equation {5.5} together with the original OIT value of the geomembrane and the OIT of pure unstabilised geomembrane resin. The time for complete depletion of the antioxidants (t) can then be calculated, giving an estimate for Stage A of the oxidative degradation process.

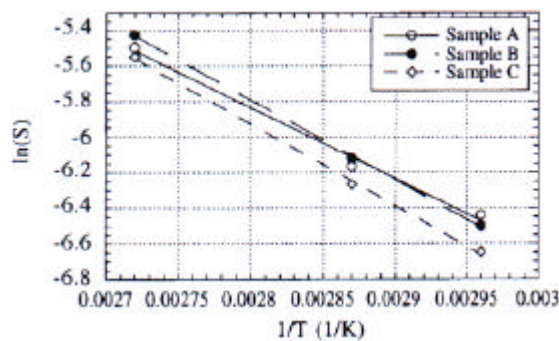


Figure 5.4 Typical Arrhenius plot for OIT test data (from Hsuan and Guan, 1998)

5.4 Assessing time for long-term geomembrane degradation

Several investigators have conducted laboratory tests to examine durability and degradation of geomembrane liners for landfills. Rowe and Sangam (2002) reviewed the published results of Duquennoi *et al.* (1995), Surmann *et al.* (1995), Cazzufi *et al.* (1995) and Maisonneuve *et al.* (1997, 1998) who had all carried out a variety of extended durability tests on HDPE geomembranes. The reviewers noted that none of these investigations identified or directly considered the three stages of degradation defined by Hsuan and Koerner (1995).

Three laboratory research projects have examined the depletion of antioxidants and estimated the long-term durability of HDPE geomembranes based on their findings (Hsuan and Koerner, 1995 and 1998; and for the lifetime prediction, Koerner and Hsuan, 2003; Sangam, 2001; and Müller and Jakob, 2003). Each of these projects is reviewed below.

In laboratory investigations into the degradation of HDPE geomembranes, the selection of the test temperatures is a key consideration. On one hand, the temperature should be as high as practical to accelerate the degradation process while on the other hand, the selected temperatures should not be so high such that the polyethylene changes its state from that at the service temperature. HDPE is a semi-crystalline thermoplastic, which means that it will undergo dramatic morphological changes at temperatures near and above the crystalline melting point. From a DSC curve, Thomas and Siebken (1997) identify that the very early stages of melting can be detected above 70°C although the melting temperature is 126°C. They suggest that it may be appropriate to test HDPE at temperatures as high as 80°C but that some changes will have occurred in the microstructure of the polymer. A concern is that a change in activation energy occurs when extrapolating high temperature tests down to the service temperature. If mechanical stress is also applied, this is another factor with an uncertain influence that has to be taken into account. For HDPE, the melt temperatures should not be passed in the accelerated test, so that the samples remain solid. Also requiring consideration in the selection of the test temperature are the types of antioxidants in the geomembrane, so that the temperature is not so high as to destroy or volatilise any part of the antioxidant package. For these reasons, incubation temperatures should be kept below 90°C (Sangam, 2001).

5.4.1 Hsuan and Koerner (1995 and 1998)

Summary

The work reported in these publications is Part I of an ongoing three-part research effort. Part I examines antioxidant depletion. Part II is investigating the induction time and onset of degradation while Part III is planned to cover half-life estimates and act as the summary report for the whole campaign. Publication of the results of Parts II and III is awaited.

The depletion of antioxidants in a single type of 1.5mm thick HDPE geomembrane was investigated by means of samples incubated at a range of elevated temperatures:

- in hot water baths, and
- in special cells at elevated temperatures with sand above and below the geomembrane sample under an applied 260kPa compressive stress, the upper sand having a 300mm water head and the lower dry sand having a connection to air.

Samples of the geomembrane were retrieved after pre-determined lengths of time and the samples tested to assess the progression of the ageing process and antioxidant depletion. The test results reported extend over a two-year period although testing is understood to have continued well beyond the two years but those results are not yet available.

Details

This project consisted of two series of tests in which samples of a single type of commercially available 1.5mm thick HDPE geomembrane were incubated at four elevated temperatures of 55°, 65°, 75° and 85°C. In the first series, samples were fully immersed in water baths. In the second, the geomembrane samples were placed between two 100mm thick layers of sand, saturated on top and dry at the bottom to approximate the conditions at the base of a landfill. The dry sand was vented to atmosphere. A water head of 0.3m was maintained above the geomembrane and a vertical compressive stress to simulate waste loading was applied. The incubation device (20 were used in the study) is shown on Figure 5.5.

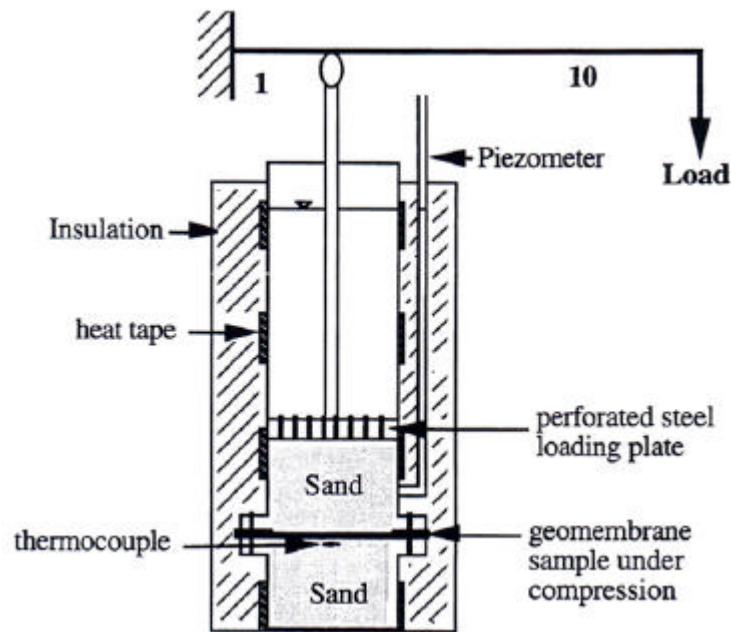


Figure 5.5 Schematic diagram of incubation device

The incubation arrangement in Figure 5.5 more closely represents a primary geomembrane liner underlain by sand in a double liner system. The use of water instead of a leachate differs from landfill exposure conditions but gives a consistent exposure medium as the effects of a leachate would be expected to differ depending on the composition of the particular leachate. The set up does not replicate a single composite liner, as commonly used in the UK, of an HDPE geomembrane over a near saturated compacted clay liner (CCL) in which the availability of oxygen or of pore water should be much lower than in the test arrangement.

From the second series, the response of four material properties at the highest incubation temperature of 85°C (which would show the greatest property changes compared to the lower temperatures) is shown on Figure 5.6.

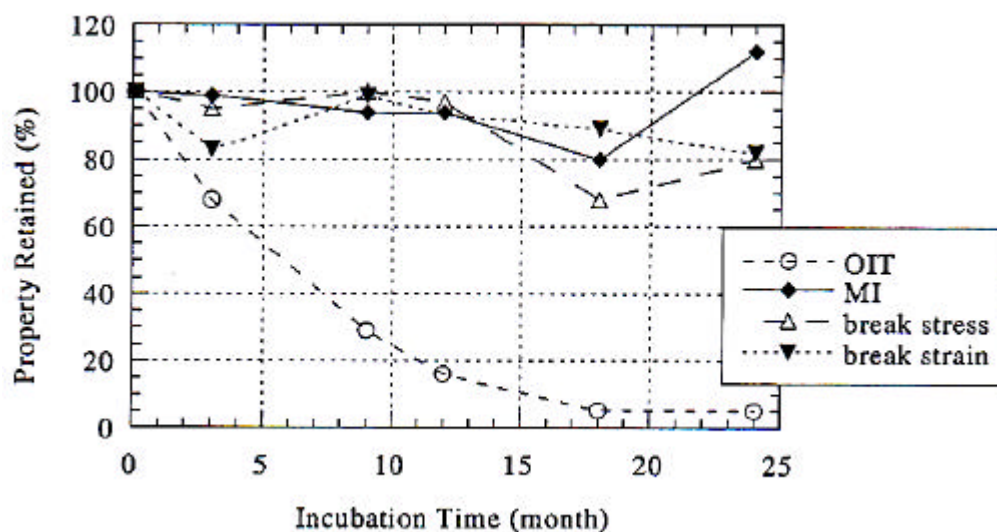


Figure 5.6 Changes in properties with incubation time at 85°C (from Hsuan, 2001)

The melt index and tensile properties show some variability over the 24 month incubation period but indicate that break stress and break strain have reduced by 20% and the MI, after reducing by 20% then increased by 12% above the original value at the end of the period. The OIT curve exhibits substantial change with time, decreasing to 5% of the original value. As the original OIT value reported was 80.5 minutes, the final OIT value was 4 minutes. This is still eight times larger than the OIT value (0.5 minutes) given by Hsuan and Koerner (1995) as representing the OIT value when all antioxidants are consumed.

The most sensitive mechanical properties to oxidative degradation are the stress and strain at break but the variability in the test results makes it difficult to assess if there is any indication that oxidation has commenced. As there is still a small amount of antioxidant remaining, then this is considered unlikely, as significant engineering property changes usually do not occur until after Stages A and B (antioxidant depletion and induction) are complete.

From the 85°C line on Figure 5.3 and using the Arrhenius equation approach given in Section 5.3 above, Hsuan and Koerner (1998) calculated the following relationship from equation {5.7}:

$$\ln(S) = 17.045 - 6798/T \quad \{5.8\}$$

Using a typical temperature at the base of two MSW landfills in the USA of 20°C (293K), the OIT depletion rate $S = 0.00212 \text{ month}^{-1}$ at 20°C. Inserting a final OIT value of 0.5 minutes when all antioxidants have been consumed and the original OIT value of 80.5 minutes into equation {5.5} gives:

$$\ln(0.5) = \ln(80.5) - 0.00212t \quad \{5.9\}$$

Hence, time for depletion of all antioxidants = $t = 2397$ months (200 years)

If a 5°C higher landfill temperature of 25°C was used, then "t" reduces substantially to 120 years, demonstrating the strong influence of temperature.

The reported results did not continue until all the antioxidants had been depleted and the induction period commenced. It can be calculated that the tests in the incubation cells at 85°C would have had to run for about 36 months before the antioxidants were fully depleted (OIT value of 0.5 minutes), assuming that the depletion rate does not change.

This is Part I of an ongoing three-part project by GRI, the other parts seeking to examine the effects of the induction and oxidation stages of geomembrane degradation. Results from Parts II and III are awaited. Thus, the results of the Part I project reported to date do not conclusively demonstrate the effects of complete antioxidant depletion on the geomembrane or if the rates of depletion (and activation energy) are applicable for the remaining period of antioxidants depletion. Koerner and Hsuan (2003) summarised their estimates of the three stages of oxidative degradation of HDPE geomembranes at a range of field temperatures as shown on Table 5.2. It is noted that Stage C representing the time for oxidation to reduce a material property (e.g. tensile strength) to 50% of the original value accounts for more than half of the predicted lifetime in virtually all cases.

Table 5.2 Lifetime prediction of HDPE at various field temperatures (from Koerner and Hsuan, 2003)

Field temperature (°C)	Stage "A" (years)		Stage "B" (years)	Stage "C" (years)		Total aver. years
	Std OIT	HP-OIT		Ref 1	Ref 2	
20	200	215	30	208	740	712
25	135	144	25	100	441	435
30	95	98	20	49	259	270
35	65	67	15	25	154	170
40	45	47	10	13	93	109

Notes: Stage "A" measured values from G. Hsuan research
Stage "B" estimated values from field samples
Stage "C" literature values from Martin & Gardner (1983) (Ref 1) and Viebke, *et al.* (1994) (Ref 2)

5.4.2 Sangam (2001)

Summary

Laboratory experiments of accelerated degradation of a geomembrane were undertaken within a project that was undertaken as partial fulfilment of the requirements for a PhD degree. Samples were incubated at several elevated temperatures in three exposure conditions - air, water and a synthetic leachate - over a three-year period. Retrieved samples were regularly tested to assess the progress of the degradation, in particular the antioxidant depletion. The test programme did not extend long enough to investigate the induction or oxidation phases. However, using the data from the research project, predictions were developed on the durability of the HDPE geomembrane in landfill conditions.

Details

A series of experiments conducted over three years was undertaken on samples of a single 2.0mm thick smooth HDPE geomembrane manufactured from an HDPE copolymer resin. The material had a crystallinity of 44%, an initial standard OIT of 133 minutes and a SP-NCTL stress crack resistance of 210 hours. Samples were exposed to three conditions:

- Air in forced air ovens (simulating a capping geomembrane)
- Tap water in heated water baths (simulating lagoon or reservoir application)
- Synthetic landfill leachate (changed at least every 2 weeks with fresh leachate).

The incubation temperatures were 40°C, 55°C, 70°C and 85°C. At various times, incubated samples were retrieved and evaluated by several tests to assess changes in key properties indicative of degradation:

- Standard OIT
- Crystallinity
- Stress crack resistance
- Tensile properties
- Melt Index

The variation of OIT value with incubation time at 85°C under different exposure conditions is shown on Figure 5.7

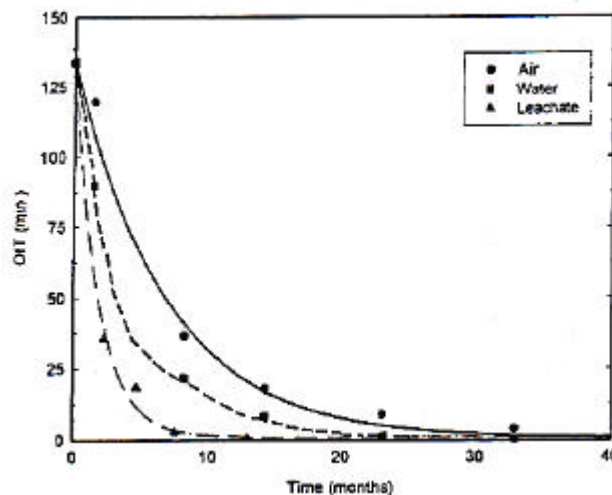


Figure 5.7 Variation of OIT with incubation time at 85°C under different exposure conditions

Sangam (2001) does not state if the OIT test specimen was taken from the whole thickness of the geomembrane or a particular part of geomembrane thickness. The antioxidant depletion rates at the 85°C incubation temperature for the three exposure conditions were:

Air	$0.1094 \text{ month}^{-1}$
Water	$0.1746 \text{ month}^{-1}$
Leachate	$0.4074 \text{ month}^{-1}$

Comparing these, it can be seen that the depletion rate for water was 1.6 times that for air while the rate for the synthetic leachate was the highest being 4 times that for air and 2.3 times that of water. Sangam (2001) considered that the antioxidants were depleted by extraction (rather than consumption) at least for the water and synthetic leachate exposure conditions. He also proposed that the high rate of depletion with leachate was due to the effects of the surfactant in the synthetic leachate causing an increase in the “wettability” of the geomembrane. This enabled quick dissolution and extraction of antioxidants between the core of the sample and the surface, leading to an increase in the diffusion flux of the antioxidants. In addition, the presence of transition metals in the synthetic leachate may also have accelerated the loss of antioxidants by reacting with the antioxidants.

The Arrhenius plots of antioxidant depletion rate, $\ln(S)$ against the inverse of absolute temperature (K) were then calculated from the data (as described in Section 5.3 above), as shown on Figure 5.8. The activation energies obtained were 53.9, 52.4 and 43.3 kJ/mol for air, water and synthetic leachate exposure respectively.

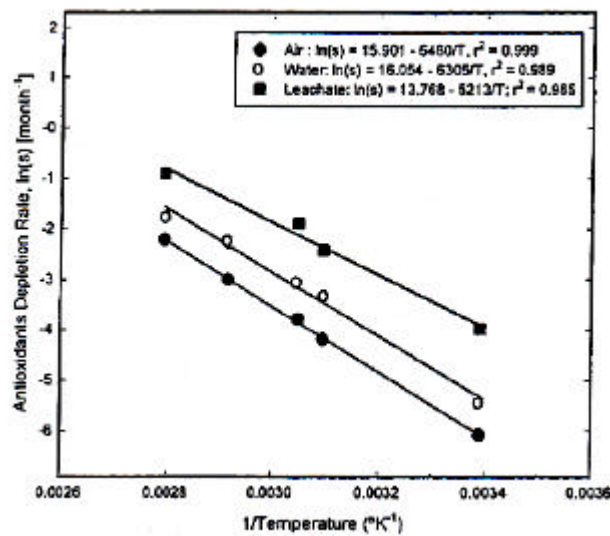


Figure 5.8 Arrhenius Plot of antioxidant depletion rate for different exposure conditions

The variations in molecular and tensile properties were determined for each test condition. Looking at the results for the severest condition, synthetic leachate at 85°C, shown on Figures 5.9 and 5.10, it can be seen that the OIT value had reduced to a very small percentage, the crystallinity had increased by over 40% and the melt index had dropped by 10%. The increase in crystallinity may be attributed to the breaking of polymer chains in amorphous regions that then crystallise. However, there was no change to the stress crack resistance or to the stress or strain at break or to the strain yield. The yield stress had increased by about 18%.

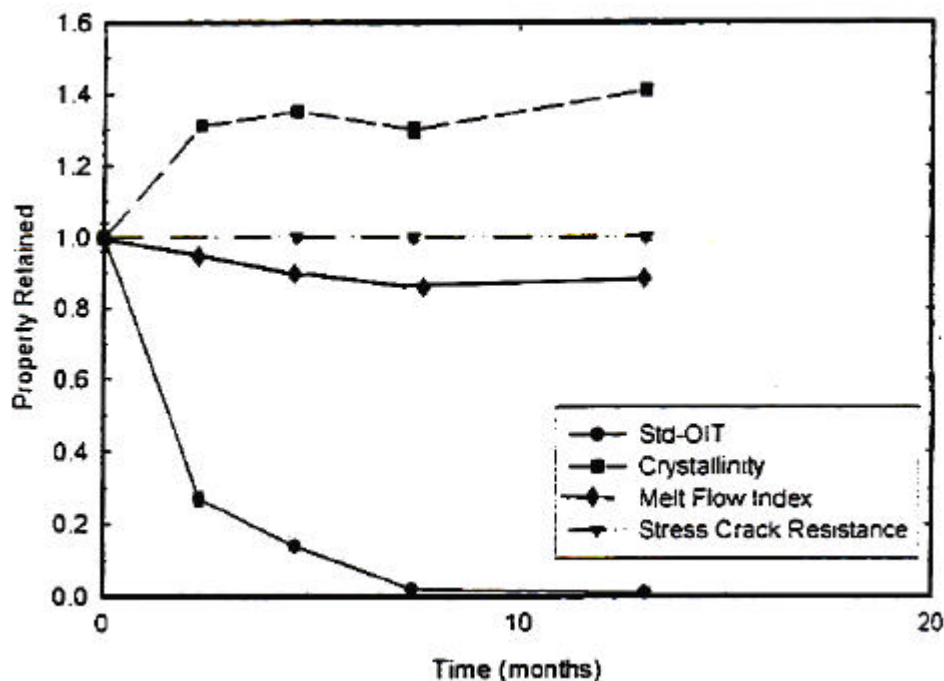


Figure 5.9 Variation in molecular properties with time during incubation in leachate

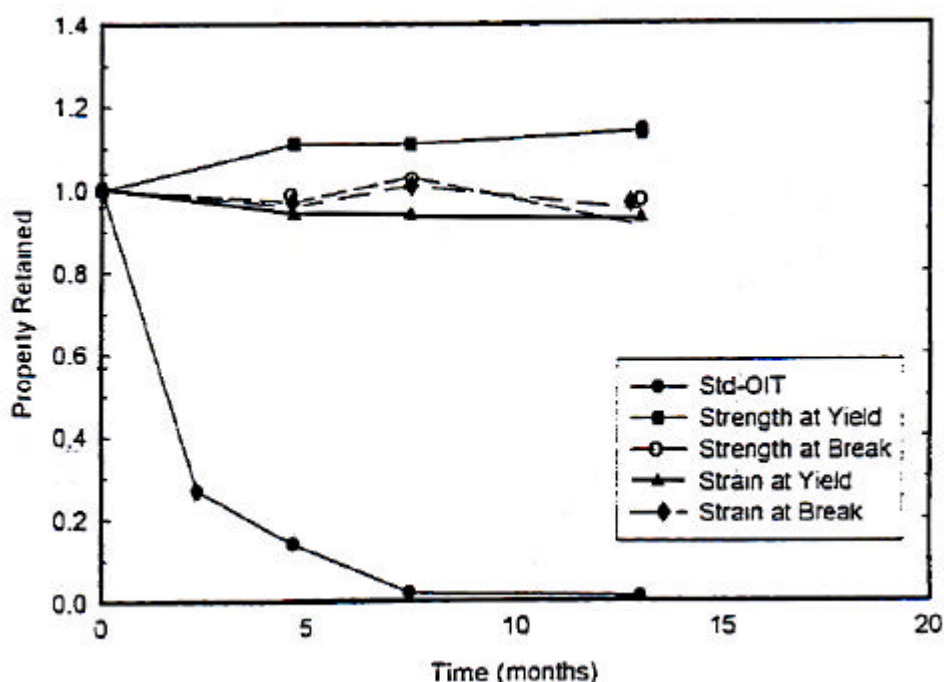


Figure 5.10 Variation in tensile properties with time during incubation in leachate

Although the OIT value indicated that antioxidant depletion was complete, or very nearly complete, there was no evidence of any embrittlement as would have been shown by a reducing stress crack resistance or a reduction in strain at break. Sangam (2001) concludes that for the samples at 85°C, antioxidant depletion had been completed and oxidation was in the induction stage as proposed by Hsuan and Koerner (1995). For the tests run at lower incubation temperatures, he considered that these samples were still in the antioxidant depletion stage. As the test programme did not extend into the third, oxidative degradation stage, the length of the induction period could not be determined and the long-term durability of the HDPE geomembrane could not be directly estimated from the results.

Sangam went on to estimate the long-term durability of HDPE geomembranes in landfill applications, based on the three-stage degradation process proposed by Hsuan and Koerner (1995) – antioxidant depletion, induction and oxidative degradation. The laboratory accelerated testing was unfortunately of insufficient duration to allow an assessment of the length of the induction period or the oxidative degradation stage. He estimated the induction period by reference to the work by Viebke *et al.* (1994) from which an activation energy of about 75kJ/mol was derived for an unstabilised pipe made from HDPE resin exposed internally to hot water and externally to air at temperatures of 70°C to 105°C. The oxidative degradation stage to “failure” (which was not defined) was simply taken as at least 25 years based on the observation that unstabilised 25-year old HDPE plastic had been found in landfills (Rowe, 1998).

The steps in the estimation of the long-term durability of HDPE geomembrane were given as:

- establish the service conditions (temperature, exposure media);
- estimate the time for antioxidant depletion (t_1 - Stage A in Hsuan and Koerner);
 - calculate the antioxidants depletion rate at the service conditions using Arrhenius modelling (Section 5.3);

- calculate the time for the complete depletion of antioxidants using equation {5.5};
- estimate the induction time (t_2 - Stage B in Hsuan and Koerner) by extrapolation to service temperature using Arrhenius modelling;
- estimate the oxidation degradation time (t_3 - Stage C in Hsuan and Koerner) as 25 years;
- calculate the long-term durability to “failure” as $D = t_1 + t_2 + t_3$

The different exposure conditions of the primary and secondary HDPE geomembrane liners in a double liner were considered separately. The primary liner was assumed always to have leachate on the top and nearly saturated compacted clay on the underside, similar to a single composite liner. As suggested by Rowe (1998), the long-term durability was taken to be the average between that calculated for leachate exposure and for unsaturated clay on the underside. The leachate condition had been tested in the laboratory but for the unsaturated compacted clay, he estimated that this condition could be estimated by taking the average of the results for air and water exposures. On this basis, the long-term durability of the primary HDPE geomembrane was calculated for several exposure temperatures as summarised in Table 5.3.

Table 5.3 Estimated durability of primary HDPE geomembrane liners (modified from Sangam, 2001)

Temperature (° C)	Antioxidant depletion time (t ₁)			Induction time (t ₂) (years)	Time to failure (t ₃) (years)	Durability (D)		
	(years)					(years)		
	(1)	(2)	(3)			(1)	(2)	(3)
13	214	113	164	30	25	270	170	220
15	184	98	141	26	25	235	150	190
20	130	74	102	15	25	170	115	140
25	91	49	70	12	25	130	90	110
33	52	28	40	5	25	80	60	70

- (1) Leachate / air conditions
 (2) Leachate / water conditions
 (3) Leachate / unsaturated soil conditions

For a landfill with a service temperature of 20°C, the durability would be 140 years before “failure”. It was considered that the durations are conservative since they assume that the leachate strength remains constant while, in reality, the leachate strength will decrease with time so that the antioxidant depletion rate will also decrease. Additionally, the methanogenic conditions, if they prevailed in the landfill, would mean very little oxygen is available for oxidation to proceed and therefore the induction stage may be much greater than the estimates given in Table 5.3. It is noted that by replenishing the leachate in the tests every two weeks, the leachate would contain dissolved oxygen and would not replicate methanogenic leachate, which has no oxygen. The assumption that the condition of a nearly saturated compacted clay liner can be approximated by averaging forced air and water bath exposure appears conservative.

The secondary liner exposure conditions were chosen as unsaturated compacted clay below and unsaturated granular soil secondary leachate collection system above until the primary geomembrane “fails” when the secondary liner would come in continuous contact with leachate. Using the same approach as for the primary liner, Sangam presented a series of tables summarising the expected durability of the secondary liner for different temperatures (lower than

the primary liner temperatures), different averaged exposure conditions (air/air, water/water and unsaturated soil/unsaturated soil) and different periods to “failure” of the primary geomembrane. Table 5.4 shows the results for the condition of unsaturated soil above and below the liner before the failure of the primary geomembrane. After failure, there are four potential exposure conditions as shown in Table 5.4.

Table 5.4 Estimated durability with unsaturated soil / unsaturated soil pre-failure conditions

PGM failure (years)	Temperature (°C)	Antioxidant depletion time (t ₁) (years)				Induction time (t ₂) (years)	Time to failure (t ₃) (years)	Durability (D) (years)			
		(1)	(2)	(3)	(4)			(1)	(2)	(3)	(4)
220	7	371	318	442	383	41	25	440	380	510	450
	10	319	286	361	328	35	25	380	350	420	390
	15	251	242	265	255	26	25	300	290	320	310
270	7	400	355	455	410	41	25	470	420	520	480
	10	346	321	379	353	35	25	410	380	440	410
	15	279	276	283	280	26	25	330	330	330	330

1. Leachate / unsaturated soil

2. Leachate / water

3. Water / unsaturated soil

4. Water / water

PGM Primary geomembrane liner

While the durability is extended compared to the primary liner, the assumed temperatures are considerably lower and Rowe (1998) indicated that the temperature would be similar to ambient groundwater temperatures.

Sangam concludes by stating that if the landfill temperature is maintained no higher than 15°C, the primary geomembrane will last at least 200 years but at 33°C the durability is estimated at about 70 years. For a secondary liner at the ambient groundwater temperature in Ontario of 7°C to 10°C, it was estimated that the secondary HDPE geomembrane liner would last at least 400 years.

5.4.3 Müller and Jakob (2003)

Summary

The long-term durability of a variety of HDPE geomembranes, all 2.5mm thick, was tested by oven-ageing in air for more than 13 years and by water immersion tests for 6 years, at a test temperature of 80°C. The mechanical properties and OIT values of the samples were monitored throughout the project. They found that the ageing behaviour in hot air ovens was different from that in hot water. During oven ageing, a slow, exponential decrease in OIT was observed but even after 13.6 years, there was no indication of oxidative degradation of the mechanical properties. With hot water immersion, a rapid reduction in OIT occurred within the first year, after which the depletion rate levelled off. Oxidative degradation was found to start when very low OIT values had been reached after about 5 years at which time the mechanical break strength rapidly fell to values below the yield point. Referring to the Hsuan and Koerner (1995) three-stage

degradation concept, both the antioxidant depletion and the induction stages would have been completed and the third stage was underway. Müller and Jakob estimated that under normal ambient conditions in landfills, many centuries would have to pass before the functional mechanical properties of modern HDPE geomembranes would be reduced below acceptable limits by oxidative degradation.

Details

The German building authorities have defined two categories of durability of landfill lining materials:

- long-term where no relevant change in the functional engineering properties will occur over a period of 50 – 100 years
- permanent, where the period is “several hundred years.”

Müller and Jakob of the Landfill Engineering Laboratory of the German Federal Institute for Materials Research and Testing (BAM) undertook the research to assess the durability of HDPE geomembranes in relation to this regulatory classification.

Nine commercially available HDPE geomembranes using seven different resins, all 2.5mm thick as required in Germany, were tested. Samples 1 and 48 were of the same resin, same geomembrane manufacturer but several years difference in date of manufacturing; similarly with samples 12 and 136. Table 5.5 gives details of the melt index, melt flow ratio, density and various OIT values determined at different temperatures and using copper and aluminium pans (see OIT testing in Section 5.2 on use of different pans).

Table 5.5 Summary of sample properties

Sample	MI ^a 190/5 g/10min	FRR ^b	Density g/cm ³	Crystallinity %	OIT (minutes)			
					Cu Pan		Al Pan	
					160°C	180°C	200°C	210°C
1	0.85 +/- 0.15	19	0.946	-	61	7	-	-
48	0.85 +/- 0.15	19	0.946	-	72	13	11	-
12	1.6 +/- 0.2	9	0.942	-	-	48	79	-
136	1.6 +/- 0.2	9	0.942	51	-	123	138	-
82	23 +/- 3 ^c	18	0.947	52	-	113	81	-
123	(0.5)	(26)	(0.950)	52	-	56	102	-
139	2.5 +/- 0.4	9	0.943	48	-	203	81	-
146	0.8 +/- 0.15	23	0.950	54	-	46	70	-
257	(1.7)	(9)	(0.940)	-	-	-	-	41

^a Note MI at 190/5 not 190/2.16 as generally used from ASTM D1238

^b Note FRR is ratio between 190/21.6 and 190/5 values, not 190/5 and 190/2.16 as generally used from ASTM D1238

^c MI from 190/21.6 test conditions

Values in brackets are measured values rather than those specified by the manufacturer.

Most of the resins in the geomembranes used were ethylene - α - olefin copolymers. The FRR reflects the width of the molecular weight distribution and the type of copolymer. Considerable differences in the initial OIT values are seen which results from the different antioxidant stabiliser packages and the intrinsic polymer properties.

The geomembrane test sheets for oven ageing were hung in ovens at 80°C with relatively static air (gravity air connection) except for sample 257 which was aged in an oven with forced air circulation and 47 air changes per hour. The water-immersed samples were placed in closed flasks with de-ionised water, changed every 3 months, and placed in ovens at 80°C. The tensile properties of samples taken at various times from the test sheets were determined according to EN ISO 527-3, using Type 1B specimens. The relative value of the elongation at break at any time t compared to the initial value was determined:

$$\delta e_B(t) = \frac{e_B(t)}{e_B(o)} \quad \{5.10\}$$

The melt index and density was also determined for each recovered sample. The standard OIT values were measured from a small section taken from the centre of a 3mm diameter specimen punched out of the geomembrane. Both copper and aluminium pans may be used in ASTM D3895 for geomembranes but Müller and Jakob stated that OIT values using aluminium pans are roughly 5 times longer than for copper pans. By using copper pans, they could use lower OIT test temperatures that were close to the melting temperature of the HDPE geomembrane while still having reasonably short OIT testing times. The relative OIT value at any time t compared to the original value at time zero was determined using:

$$\delta OIT(t) = \frac{OIT(t)}{OIT(o)} \quad \{5.11\}$$

The results of oven ageing for over 5000 days are shown on Figure 5.11.

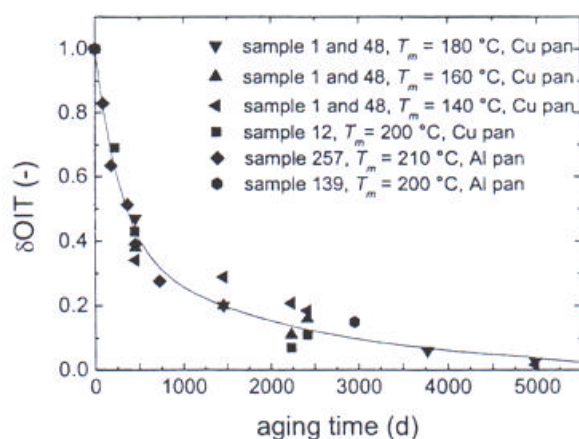


Figure 5.11 Relative change in OIT value during ageing in air at 80°C of various samples

Although very low, OIT values were still being recorded after 5000 days, and no significant change in the tensile properties was seen. A reduction in the melt index was observed for samples 1 and 12, which had been aged for the whole 13.6 years. A small increase in crystallinity was also noted. Figure 5.11 illustrates that the relative OIT values show a

reasonably common decline as a function of ageing time, independent of the resin or OIT testing temperature. The forced air circulation condition on sample 257 tested for 730 days seems also to follow the same curve.

The behaviour of the geomembrane samples immersed in water (Figure 5.12) shows a sharp decline in relative OIT values in the first 200 days of immersion and then levels off to a low rate of reduction.

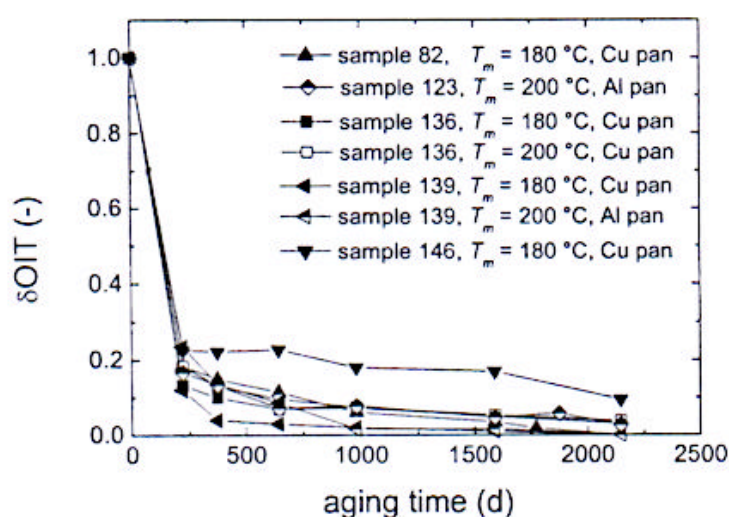


Figure 5.12 Relative change in OIT value during immersion in water at 80°C of various samples

Results showing the actual OIT values and relative tensile break strain with time from three of the samples (82, 136 and 139) are shown on Figures 5.13 to 5.15. Figure 5.16 shows the rapid reduction in relative OIT and tensile break strains with immersion time for sample 48.

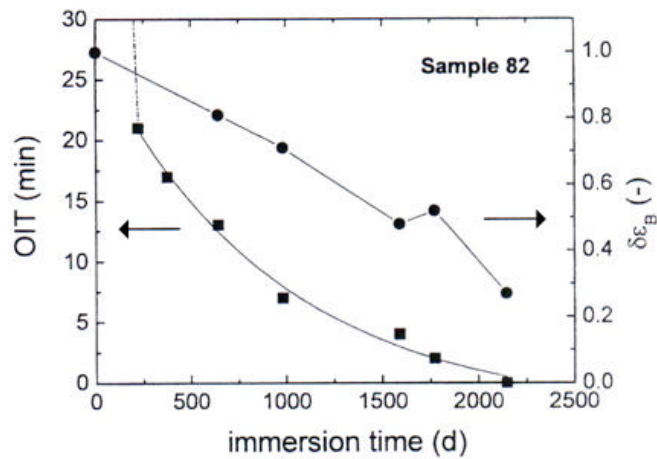


Figure 5.13

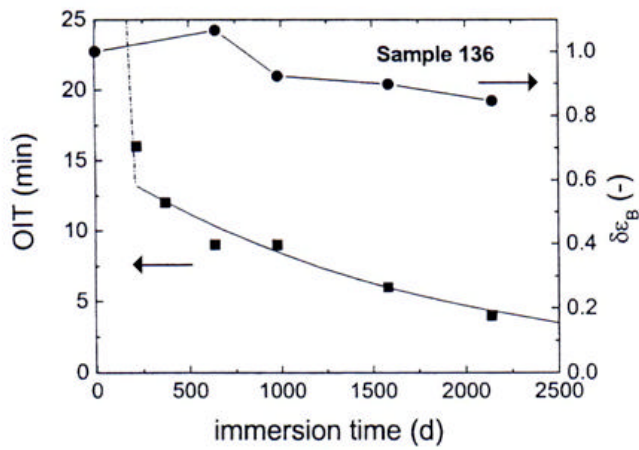


Figure 5.14

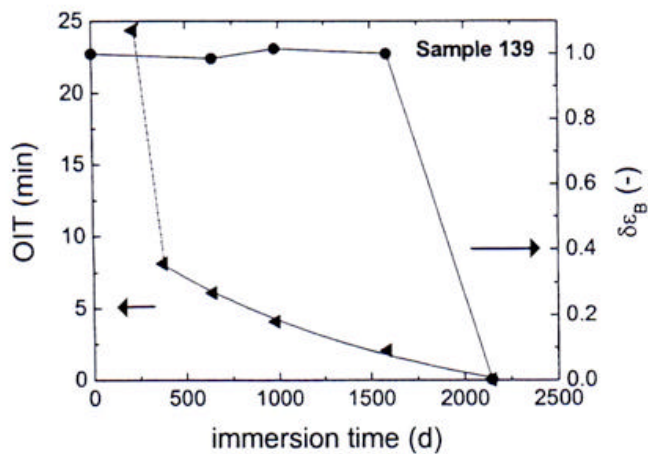


Figure 5.15

Figures 5.13 - 5.15 Measured OIT values ($T_m = 180^\circ\text{C}$, Cu pans) and relative elongation at break during immersion in water at 80°C for samples 82, 136 and 136.

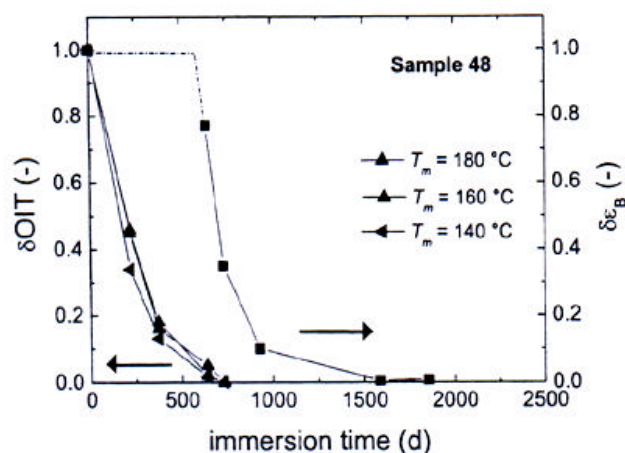


Figure 5.16 Change in OIT value and relative elongation at break during immersion in water at 80°C for sample 48. The OIT test temperatures (T_m) are indicated.

For sample 139, the decline in the OIT value occurs much more slowly than for sample 48 after the initial rapid reduction. After 2147 days (about 6 years), the OIT value has dropped below the detection limit. The elongation at break remains reasonably constant until the OIT value is below the limit of detection but then rapidly reduces within several months to values below the elongation at yield due to oxidative degradation.

The progression of the stress / strain curves with time for sample 139 is shown on Figure 5.17.

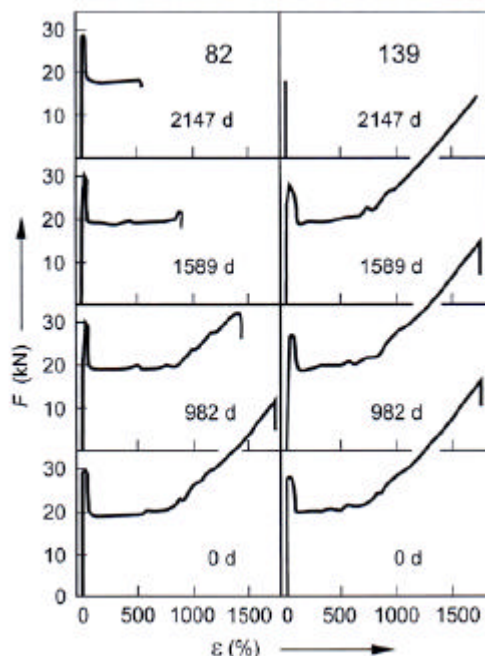


Figure 5.17 Examples of tensile force (F) versus strain (ϵ) diagrams from tests on samples 82 and 139 after immersion in hot water

The behaviour of the mechanical properties of sample 82 was quite different from the other samples (Figures 5.13 and 5.17). The decline in the OIT of this sample was accompanied by a continuous decline of its tensile properties and MI over several years, but the elongation at break

was still well above the yield point. The reasons for this particular geomembrane displaying atypical behaviour are not known but must relate to morphological changes during ageing and/or the particular antioxidant package used.

For sample 136, the OIT value has declined but after 2147 days, it was still 3 minutes (test temperature (T_m) 180°C, copper pan). No change was observed in the tensile properties (Figure 5.14), MI or density.

Sample 48 was made from a poorly stabilised resin that is no longer used for geomembrane manufacture. After about 2 years immersion of sample 48, no OIT was measurable at all three OIT test temperatures. At 580 days, the relative OIT value was very low and rapid reductions in the elongation and stress at break were recorded, although there was no change in the yield stress or strain. This was indicative of the onset of oxidative degradation (Figure 5.16 for sample 48 shows a dashed line at the step change in relative strain at break without justification from test results, but this step was known from earlier extensive testing with pipes made from this resin). The oxidation process continued and after 6 years immersion, the elongation at break is well below the yield point, i.e. the sample has become very brittle. It can be seen that the break strain had reduced to about 50% of its original value approximately 100 days after the estimated start of oxidation.

The stress crack resistances of the different geomembranes as measured by the SP-NCTL test (ASTM D5397 Appendix) were interesting in comparison to the latest GRI GM13 specification value of 300 hours. Sample 48 was very sensitive to stress cracking, failing in about 10 hours. Sample 136 was also low at 80 hours, samples 82 and 146 had good resistance at about 400 hours, and samples 139 and 123 had excellent resistance with failure not occurring until much longer than 400 hours had elapsed (Müller, 2003).

Discussion

Müller and Jakob considered that the service lifetime (i.e. durability) of an HDPE geomembrane could be written as:

$$D = t_1 + t_2 \quad \{5.12\}$$

where D = service lifetime (i.e. material durability)

t_1 = antioxidant depletion time (Stage A of Hsuan and Koerner)

t_2 = the induction time of the autooxidation (Stage B of Hsuan and Koerner)

(N.B. In their paper, they use t_1 as the induction time and t_2 as the antioxidant depletion time but, to be consistent with Sangam (2001) and with normal numerical progression, t_1 and t_2 are used in this report as defined above).

They did not include any time for oxidation (Stage C of Hsuan and Koerner) within the estimate of durability.

Müller and Jakob concluded that the depletion of antioxidants was controlled by a combined process of migration of the stabiliser by diffusion from the bulk of the HDPE geomembrane to the surface driven by a concentration gradient, followed by evaporation or leaching and possibly accompanied by chemical degradation of the stabiliser. They did not consider that consumption of the antioxidant stabiliser in inhibiting the oxidative chain reaction was significant. They also argued that in the OIT test, which uses a pure oxygen atmosphere, the antioxidant depletion is

due to consumption (not migration) because of the high rate of free radical initiation and a large oxygen supply. This deduction was confirmed by the higher activation energy of the antioxidant consumption in the OIT test which was calculated as 140kJ/mol while the activation energy of the antioxidant migration process during oven ageing, immersion or service conditions is much lower.

The sharp decline in OIT values in the water immersion tests was considered to reflect the rapid loss in hot water of the phosphite component of the stabiliser package. The phenolic stabiliser then determines the long-term antioxidant depletion time. The phosphite stabiliser, which forms the largest part of the antioxidant package, substantially determines the initial OIT value of a HDPE geomembrane. Therefore, a high initial OIT does not necessarily correlate with a good long-term oxidation stability. Instead, it is the relative rate of reduction in OIT values under ageing conditions that is important.

As Müller and Jakob conducted their accelerated tests at a single temperature, 80°C, rather than over a range of temperatures as had Hsuan and Koerner (1995, 1998) and Sangam (2001), they could not create Arrhenius plots of the logarithm of the depletion rate against the inverse of temperature. Consequently, they could not derive the activation energy from the slope/s of the plots, so they used a different deduction process to estimate the durability of the HDPE geomembrane material.

They proposed that the change in OIT during ageing, ΔOIT , could be fitted by an exponential decline with amplitude A and offset B, and a depletion rate S:

$$\Delta \text{OIT} = Ae^{-S \cdot t} + B \quad \{5.13\}$$

From the oven ageing results (Figure 5.11), they noted that the data are fitted by a superposition of two exponential declines, each described by the form of equation {5.13}. They calculated a short-term depletion rate of $S = 0.11 \text{ month}^{-1}$ and a long-term rate of 0.014 month^{-1} for the oven ageing. In the case of water immersion, the superposition of two different depletion rates is obvious. Here, the short-term depletion stage is so short that the long-term behaviour is essentially described solely by the long-term component with a depletion rate of $0.015 - 0.03 \text{ month}^{-1}$. Müller (2003) indicated that the short-term depletion rate was estimated as approximately 0.2 month^{-1} . Müller and Jakob compared these depletion rates to that obtained by Hsuan and Koerner (1995) at 85°C of 0.14 month^{-1} for ageing conditions of air (dry soil) below and water saturated soil above the geomembrane measured over a 2-year period. In addition, Müller and Jakob's findings may be compared to the depletion rates obtained by Sangam (2001) at 85°C in air ($0.1094 \text{ month}^{-1}$), water ($0.1746 \text{ month}^{-1}$) and synthetic leachate ($0.4074 \text{ month}^{-1}$) over a 2-year period. They may also be compared to the water immersion tests undertaken by Hsuan and Koerner (1995) in which the depletion rate of $0.1765 \text{ month}^{-1}$ at 85°C was obtained, being very similar to Sangam's value for the same conditions.

From the form of the Arrhenius equation in equation {5.4}, the temperature dependence of the antioxidant depletion time $t_1(T)$ can be written as:

$$t_1(T) = t_1(T_1) \cdot e^{\left(\frac{E_a}{R}\right) \left(\frac{1}{T} - \frac{1}{T_1}\right)} \quad \{5.14\}$$

From their testing, oxidative degradation did not start until after 5 years of immersion in water at 80°C, which they took as a lower bound of the depletion time at that temperature. Taking the time for complete antioxidant depletion, $t_1(T_1)$, = 5 years at $T_1 = 80^\circ\text{C}$ and a low activation energy of 60kJ/mol (giving $E_a/R = 7200\text{k}$) under ambient landfill conditions of $T = 20^\circ\text{C}$, the antioxidant depletion time $t_1(T)$ is computed as being 325 years which they have taken as a lower limit of the geomembrane durability. This value ignores any contribution from the induction time (t_2). For a larger activation energy of 100kJ/mol (giving $E_a/R = 12000\text{k}$), which they argue is more realistic, a durability of more than 5000 years is obtained at an average landfill temperature of 20°C. As the estimations of antioxidant depletion time are so long and more than meet the regulatory requirement for geomembrane durability (D), they have not reported an estimated length of time for the induction period (t_2) or for the subsequent oxidation stage.

Müller and Jakob conclude that, with respect to the German building authority requirement for “permanent” durability being no relevant change of functional engineering properties over “several hundred years”, HDPE geomembranes meeting the BAM certification requirements will have a durability that satisfies the building authority criterion. The durability of such HDPE geomembranes could therefore be classed as “permanent”. Further work on predicting the service life of geomembranes as a hydraulic barrier beyond the regulatory period was therefore not considered necessary and the research programme is being terminated.

5.5 Review of laboratory research into geomembrane durability

Several factors were identified as having an influence on the results of the laboratory ageing tests and, therefore, also on the estimation of the long-term durability of the HDPE geomembranes. These are:

- geomembrane materials;
- test duration;
- exposure medium – air, water, leachate or soil (saturated and dry) and if these are static or flowing, and the availability of oxygen in these media;
- ageing temperature/s;
- thickness of the geomembrane;
- use of absolute OIT values;
- location of the OIT specimen;
- activation energy;
- method of assessment of the three stages of the degradation process;
- extrapolation of the laboratory results to a long-term durability also requires the selection of an average service temperature that reflects the effect of the range of temperatures at the liner that are likely to occur throughout the life of the landfill.

These factors are considered below. The results of the research programmes are then compared.

5.5.1 Geomembrane materials

The American and Canadian researchers both tested only one HDPE geomembrane material, 1.5mm and 2.0mm thick respectively. The work by Müller and Jakob was on 9 samples (7 resins) all 2.5mm thick. Geomembranes formulated from different resins and using different antioxidant packages are unlikely to have the same antioxidant depletion performance. This can

be seen in the nine geomembranes tested by Müller and Jakob, which exhibited different OIT depletion rates and tensile characteristics in response to the water incubation. However, as seen in Figure 5.12, the change in OIT with time of most of the geomembranes occurred within a relatively tight envelope. However, the curves of some of the samples are not shown (nos. 1, 12 and 257), and sample 48 (see Figure 5.16) is atypical by commencing oxidation at an early stage. It is understood that GRI has also found variable OIT depletion response patterns for different geomembrane formulations, including two-stage OIT depletion as at BAM, but also has example/s of rapid, complete OIT depletion (Koerner, 2003).

It is apparent that prediction of the long-term OIT depletion performance of a geomembrane to a typical response pattern currently has to be made with caution unless supported by long-term testing. The criteria of OIT time and OIT retention after oven ageing, as required by both the GRI-GM 13 (GRI, 2003) and BAM specifications (BAM, 1999), do give some confidence. However, there is the need for published information comparing the results obtained from testing to these specifications against the long-term OIT depletion performance.

5.5.2 Test duration

The Müller and Jakob (2003) test programme extended much longer than that of Hsuan and Koerner (1995) and Sangam (2001). They identified a two stage antioxidant depletion process giving a much slower long-term OIT depletion rate and therefore a longer estimated durability than found by the other researchers. GRI may also have noted this type of response but results have not been published. Examination of Figure 5.7 from Sangam (2001) showing $\ln(\text{OIT})$ with respect to time for the three exposure conditions at 85°C indicates that he may have started to measure a slow long-term depletion rate for the leachate and water exposure conditions in addition of a faster, initial rate. However, as the tests were terminated before further data points could become available, this aspect was not investigated.

5.5.3 Exposure medium and ageing temperature

Hsuan and Koerner (1995), Sangam (2001) and Müller and Jakob (2003) all investigated ageing in air but the first two teams used forced-air ovens at four temperatures while the German researchers used essentially static air ovens, except for their sample 257, all at 80°C.

All the research teams used ageing in water, the German team in de-ionised water at 80°C, Hsuan and Koerner (1995) in tap water at 55°, 65°, 75°, and 85°C and Sangam (2001) in tap water at 40°, 55°, 70°, and 85°C, the temperatures he had employed for the oven ageing. Hsuan and Koerner (1998) also used a 'compressive stress – water saturated sand/dry sand' incubation procedure to simulate landfill conditions while Sangam (2001) used a synthetic leachate, both teams using the same temperatures they employed for the water immersion.

As Müller and Jakob (2003) ran their tests at only one ageing temperature, they could not make an Arrhenius plot to obtain the activation energy and had to use estimated values obtained from tests by other researchers.

The OIT depletion rates determined by the three teams for the different exposure conditions and ageing temperatures are shown on Figure 5.18. Several effects stand out:

- Reducing the exposure temperature resulted in a very marked decline in the depletion rate.
- The large reduction in the two-stage depletion rates found by Müller and Jakob (2003) in their very long-term tests.
- The depletion rates for water incubation found by Hsuan and Koerner (1995) were very similar to those for water immersion of Sangam.
- The early depletion rate in air obtained by Müller and Jakob (2003) agrees reasonably well with those obtained by Hsuan and Koerner (1998) and Sangam (2001) but the initial depletion rate in water found by Müller and Jakob was significantly faster than that of the other researchers.
- Immersion in synthetic leachate resulted in a much higher depletion rate than any of the other exposure media.
- The much longer duration testing by Müller and Jakob showed a very slow antioxidant depletion process with a rate of 0.015 – 0.03 month⁻¹ in water. Therefore, the long-term durability of the HDPE geomembrane may be significantly longer than that estimated by Hsuan and Koerner (1998) and Sangam (2001), depending on the geomembrane formulation.

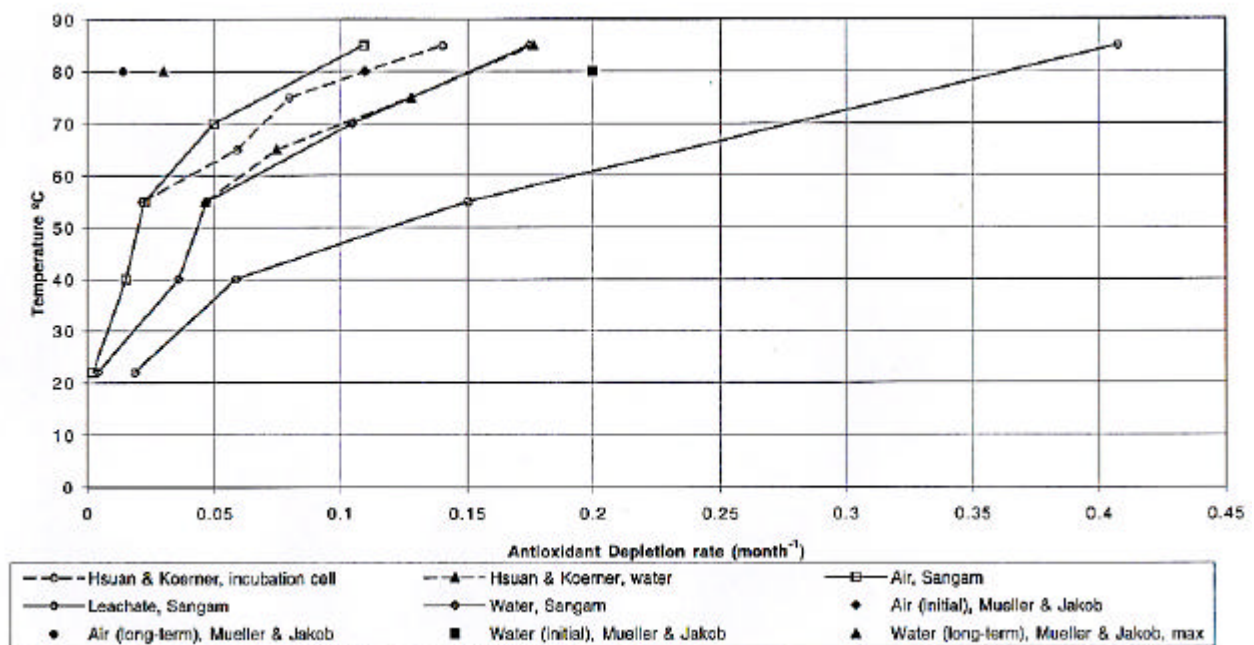


Figure 5.18 Comparison of antioxidant depletion rates by different researchers

5.5.4 Thickness effects

Since outward antioxidant migration and inward oxidation of the polymer chains are controlled by the rate of diffusion through the polymer, the thickness of a polymer film should influence the rate of degradation. As noted by Müller *et al.* (2003), the overall rate of antioxidant loss from a geosynthetic is proportional to its surface area and the total amount of stabiliser in the geosynthetic is proportional to its volume. Thus, the antioxidant depletion time should be proportional to the thickness of the material.

Ram *et al.* (1990) considered the effects of the thickness of stabilised and unstabilised LDPE subjected to accelerated weathering. In the stabilised films they state that the increased thickness has the effect of providing a reservoir of antioxidants for replacing depleted zones on the external surface. From this, lower degradation rates would be expected for thicker films. This effect has also been observed by Kelen (1983), who found that the rate of oxidation decreases with increasing polymer film thickness, and that thick films displayed longer induction time than thin films. Schwarzenbach *et al.* (2001) report from research by Gijsman (1994) into the embrittlement of polypropylene that samples 3mm thick reached a lifetime of 76000 hours at a test temperature of 80°C, reducing to 48000 hours for 1 mm thick samples and 20000 hours for 0.5 mm thick samples. Sangam (2001) also reports that Lopes *et al.* (1998) observed a similar thickness dependence of geomembrane degradation from laboratory investigations where a greater reduction was seen in the tensile strength of a 1.0mm geomembrane compared to a 2.0mm geomembrane.

Keegan and Ramsey (1998) undertook an investigation into accelerated UV weathering of polyethylene film (0.06mm and 0.13mm) and liner samples. They found that the 0.06mm film consistently deteriorated faster than the 0.13mm film samples. They concluded that the ability to withstand UV radiation damage is directly related to the thickness of the sample. Although they did not test liner samples (0.5mm to 2.5mm thick), they further concluded that UV radiation would take much longer to damage normal geomembrane liner (say 2mm thick) than the thin film tested.

Hence, it would be expected that the thicker 2.5mm sheet tested at BAM would have slower relative antioxidant depletion and induction-stage oxidation rates compared to the 1.5mm sheet samples used by Hsuan and Koerner (1995) or the 2mm sheet used by Sangam (2001). This may be a reason for some of the differences seen in the results of these research projects but the effects of the different geomembrane resins have also to be taken into account. Thus, at present, there is insufficient research information demonstrating that a quantitative increase in durability can reliably be achieved by increasing the geomembrane thickness. It follows that the use of a thicker geomembrane (e.g. 2.5mm instead of 2.0mm) at present cannot provide a quantifiable increase in liner durability. However, if a geomembrane less than 2 mm thick is to be used (e.g. in a cap), then the reduced thickness should be taken into account by a proportional reduction in time from the durability estimate for a 2 mm thick geomembrane.

5.5.5 Absolute OIT values

High initial OIT values need not correlate with long-term oxidation stability. Certain antioxidants, such as phosphites, will markedly increase the initial OIT value but are ineffective below about 150°C (see Figure 4.4) and do not contribute to long-term oxidation stability at normal operating temperatures. Thus, polyethylene geomembrane specifications should not stipulate only the initial OIT value.

The GRI and BAM specifications combine OIT test values with oven ageing procedures to provide a measure of antioxidant stability. The GRI GM13 standard requires an initial standard OIT value of 100 minutes at a test temperature of 200°C (GRI, 2003). The reduction in OIT value must be less than 45% following 90 days oven ageing at 85°C. BAM certification requirements (BAM, 1999) specify initial OIT values at 200°C of >20 minutes and >10 minutes after 6 months of air oven ageing at 80°C; continuing the ageing for one year, the relative change in OIT value between 6 months and one year must be less than 30%.

The GRI specification also has a requirement for limiting OIT reduction to 50% of the HP-OIT test when the HDPE geomembrane sample has been subjected to a specified UV exposure. This will be important for geomembranes exposed to the atmosphere for extended periods of time.

5.5.6 Location of the OIT specimen

Müller and Jakob (2003) tested specimens taken from the centre of the geomembrane thickness while Hsuan and Koerner (1998) used the whole thickness giving an average value. Sangam (2001) does not report the location of the sample. Karlsson *et al.* (1992) undertook experimental investigations of antioxidant variations across the 2.2mm wall thickness of HDPE pipes. They showed there to be considerable variations in the OIT values across the thickness of the pipe wall, even in pipes that had not been exposed to accelerated ageing, as shown on Figure 5.19.

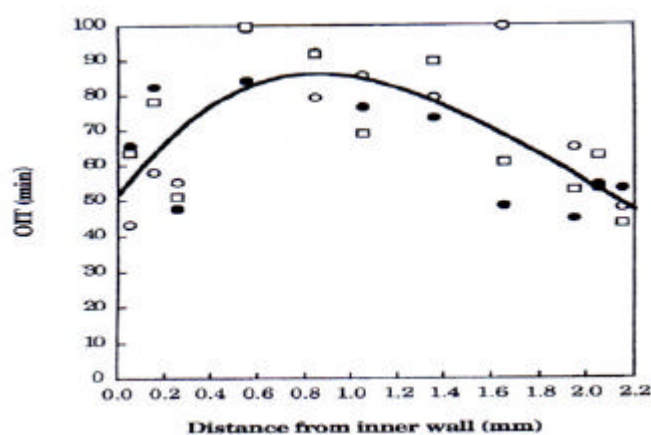


Figure 5.19 Variation of OIT across the wall of a 2.2mm HDPE pipe (Karlsson *et al.*, 1992)

When the pipes were subjected to long term internal water / external air exposure at elevated temperatures, the OIT values in the inner part of the pipe wall were significantly higher (two to three times greater) than on the faces. As exposure continued, the OIT values on the internal and (to a lesser extent) external faces reduced to zero while there remained a significant OIT value in the inner part of the pipe wall.

It follows that differences in OIT values would be expected between Müller and Jakob (2003) and Hsuan and Koerner (1995), other factors being equal, as the average OIT value across the geomembrane thickness would be less than the value obtained from a sample taken from the central part of the geomembrane. Samples taken from the central part should result in a longer estimate of Stage A (antioxidant depletion) compared to an estimate based on an average OIT value, but would result in a shorter estimated induction time. The time to oxidation would not be affected by the OIT sample location as this is separately determined based on the reduction in the tensile test break strain

5.5.7 Activation energy

The activation energy (E_a) reflects the necessary minimum energy of the antioxidant depletion process and will depend on the characteristics of the polyethylene resin, the antioxidant package and the exposure conditions in which the antioxidant loss is occurring. As the rate of antioxidant

depletion is exponentially dependent upon the activation energy, it is important to assess the appropriate value to use in the estimation of antioxidant depletion times. Sangam (2001) found activation energies of 53.9, 52.4 and 43.3 kJ/mol in air, water and synthetic leachate conditions respectively for the antioxidant loss experienced by the single geomembrane material tested. Hsuan and Koerner (1995) deduced activation energy values of 43.3 and 56.5 kJ/mol for water and the landfill simulation incubation cell respectively, again for a single geomembrane.

Müller and Jakob (2003) could not derive activation energy from their laboratory studies at the single temperature but refer to published values of activation energy. They report that the activation energy of the diffusion process of organic molecules with high molecular weight in HDPE bulk material is typically in the order of 100 kJ/mol. For example, the activation energy of the diffusion of Irganox 1010, a phenolic stabiliser typically used for geosynthetics, was determined to be 115 kJ/mol in LDPE, 100 kJ/mol in PP homopolymer and 113 kJ/mol in a PP-PE copolymer. Smith *et al.* (1992) found an activation energy of the overall migration process of a combined phosphite and phenolic antioxidant package of 80 kJ/mol for ageing in air and 100 kJ/mol for immersion in water.

Viebke *et al.* (1994) report an activation energy value for MDPE pipe of 105 kJ/mol in pressure tests and noticed that this value was very similar to that for the diffusion of the antioxidants found in earlier tests. In tests on unstabilised MDPE pipe (i.e. with no added antioxidants), they found that in the induction period before detectable oxidation occurred, the activation energy was 75 kJ/mol. These results indicate a decrease in the activation energy occurring from the antioxidant depletion stage to the induction period. They report that the activation energy value for the induction period was corroborated by other researchers (71-113 kJ/mol for HDPE and 67 kJ/mol for LDPE).

Billingham (2001) presents the activation energies for antioxidant diffusion for a range of common antioxidant additives, with values for polyethylenes of 40-115 kJ/mol but predominantly in the range 75-100 kJ/mol.

Müller and Jakob (2003) expected that activation energies relevant to the second, slow antioxidant depletion stage found in their long-term tests would be much higher than found by Hsuan and Koerner (1998). As it appears that antioxidant depletion is controlled by the rate of diffusion, then use of the activation energy values derived from studies of antioxidant diffusion appears justified. It is tentatively inferred that the lower activation energies found by Hsuan and Koerner (1995,1998) and Sangam (2001) reflect faster diffusion of more easily depleted antioxidants, rather than slower diffusion of the residual antioxidants, which provide the very long-term antioxidant protection.

This tentative antioxidant depletion / activation energy model for each individual geomembrane formulation will clearly depend on the actual composition of the antioxidant package used and the polyethylene resin characteristics. For this model, values of activation energy of 60 – 75 kJ/mol appear a reasonable, conservative estimate.

5.5.8 Three stage degradation process

Müller and Jakob (2003) combined the antioxidant depletion and induction Stages (Stages A and B) and once noticeable oxidative degradation (by a significant reduction in elongation at break) occurred (Stage C), then the limit of durability was considered to have been reached. However,

in their calculations of durability, they neglected the second stage as they found that the first stage was sufficiently long to meet the regulatory requirements in Germany. They also found that when oxidation started in Stage C, the decrease in mechanical properties such as break strain proceeded relatively rapidly.

Both Hsuan and Koerner (1995, 1998) and Sangam (2001) attempted to estimate each of the three stages separately. However, no clear point between Stages A and B could be discerned particularly as definite completion of Stage A was not demonstrated except possibly for leachate at 85°C in the Sangam work.

As shown on Table 5.2, Koerner and Hsuan (2003) estimated Stage B to be 10 to 30 years based on estimated values from field samples. From pipe and other research (Martin and Gardner, 1983 and Viebke *et al.*, 1994) they estimated Stage C, the time for a mechanical property to reduce by 50%, to be 200 to 740 years for a liner temperature of 20°C. This is much longer than indicated by the laboratory studies of Müller and Jakob (2003).

Sangam (2002) used work by Viebke *et al.* (1994) to estimate Stage B as 5 to 30 years depending on temperature, and took Stage C as 25 years as suggested by Rowe (1998), based on the observation that unstabilised 25-year old intact plastic has been found in landfills, so it is not unreasonable to allow at least 25 years for the oxidation stage.

5.5.9 Selecting the liner service temperature

The strong temperature dependency of the HDPE geomembrane degradation has been shown. A review of temperature data recorded at landfill is discussed in Section 6.3.1.

5.5.10 Conclusions on HDPE geomembrane durability laboratory testing

All three research projects provide very valuable results which, when considered together, give a technical basis for the derivation of a reasonable estimation of HDPE geomembrane durability in landfill conditions. The estimation includes:

- slow long-term OIT depletion rates from Müller and Jakob (2003);
- the increased rate of depletion for leachate exposure found by Sangam (2001);
- the effects of a confined sample under comprehensive stress sandwiched between saturated sand and dry sand, as investigated by Hsuan and Koerner (1998);
- measuring durability of the geomembrane in terms of the tensile test (but not service life as a hydraulic barrier).

The slow, long-term OIT depletion rate obtained by Müller and Jakob (2003) was 0.03 minute⁻¹ at 80°C for water immersion. Sangam (2001) found that OIT depletion was 2.3 times faster in leachate compared to water immersion at 85°C, making the long-term depletion rate = $(0.03 \times 2.3) = 0.069$ minute⁻¹. Sangam (2001) describes an approximation process to relate laboratory ageing conditions to the actual exposure conditions of a composite landfill liner (see Table 5.6).

Table 5.6 Estimated antioxidant depletion time (years) for a primary HDPE liner (modified from Sangam, 2001)

Temp.	Air	Water	Leachate	Unsaturated soil [§]	Leachate/air	Leachate/water	Leachate/unsaturated soil	Leachate/landfill ratio
°C	(1)	(2)	(3)	(4)=[(1)+(2)]/2	(5)=[(1)+(2)]/2	(6)=[(1)+(2)]/2	(7)=[(1)+(2)]/2	(3)/(7)
13	388	186	40	287	214	113	164	0.24
15	332	160	36	246	184	98	141	0.26
20	233	121	26	177	130	74	102	0.26
25	160	76	22	118	91	49	70	0.31
33	91	44	12	68	52	28	40	0.30

§ This is an average of the values of air and water alone and does not really account for the presence of the soil particles (which could have a beneficial effect since contact with soil particles reduces contact with air and / or water). This also assumes that the air contains 21% of oxygen whereas in reality the oxygen concentration may be lower than 21%.

The last column in Table 5.6 has been added, from which it can be seen that the estimated antioxidant depletion time under landfill conditions (leachate above and unsaturated soil (compacted clay) below) is 0.24 to 0.31 times that of leachate immersion in the laboratory within the service temperature range 13° to 33°C. Using this range of factors, the OIT depletion rate becomes (0.069×0.24) to (0.069×0.31) for the landfill liner exposure model, i.e. 0.016 to 0.021 month⁻¹.

These depletion times may be compared to the faster 0.03 month⁻¹ obtained by Müller and Jakob (2003) for water immersion. They estimated that oxidative degradation commenced after 5 years in water immersion. Factoring the 5 years by the ratio $\frac{0.03}{0.016} \sim 2$ and $\frac{0.03}{0.021} \sim 1.4$, it can be estimated that for the landfill liner exposure model, oxidative degradation would start between $(1.4 \times 5) = 7$ and 10 years.

Müller and Jakob (2003) could not derive activation energies. In view of the much slower long-term OIT depletion rate, they expected that an activation energy much higher than that estimated by Hsuan and Koerner (1998) of 56 kJ/mol would apply. However, selecting a conservative activation energy value of 60 kJ/mol, and a time for complete antioxidant depletion $t(T_1)$ of between 7 and 10 years at 80°C, the durability of HDPE geomembrane can be determined for different service temperatures using equation {5.14}. Increasing the activation energy to 70 kJ/mol has a strong impact on estimated durability. As discussed in Section 5.5.7, Müller and Jakob (2003) argued that a higher activation energy of 100 kJ/mol was appropriate. Typical results are given in Table 5.7 below:

Table 5.7 Estimates of HDPE geomembrane material durability in landfill conditions derived from laboratory research projects (Stage A - antioxidant depletion only)

Average landfill temperature (°C)	Activation energy (kJ/mol)	Estimated durability (years)
20	60	450 - 650
20	70	900 - 1300
35	60	140 - 200
35	70	220 - 320
35	80	370 - 530

The estimates in Table 5.7 are based on a 2 mm or 2.5 mm liner. As noted in Section 5.5.4, although longer durability may be expected for a thicker geomembrane, current research does not justify refining the estimates for 2.0 and 2.5 mm thick geomembranes. However, where liners less than 2.0 mm thick are proposed, then the durability estimates in Table 5.7 should be reduced in proportion to the reduction in thickness below 2.0 mm.

The estimates above rely on the results from different research projects and will benefit from confirmation by further laboratory investigation.

There are a number of conservative assumptions in the landfill liner exposure model which result in the estimate of durability given above being lower than may occur in practice:

- the durability estimates are based only on the depletion of antioxidants (Stage A) and neglect the induction period (Stage B of the oxidative degradation process);
- the leachate strength remains constant in the laboratory tests whereas it will decrease with time and the rate of antioxidant depletion will probably also decrease with time;
- oxygen will not be available in methanogenic landfill conditions (where these persist after the long antioxidant depletion stage) for oxidation to proceed on the upper surface of the liner, so the induction period would be expected to lengthen;
- the presence of soil particles in contact with the underside of the geomembrane reduces the contact of the geomembrane with air or water. In addition, the low availability of oxygen within a partially saturated or saturated compacted clay liner would restrict both antioxidant depletion and oxidation.

The durability estimates refer to the onset of increasing brittleness (and reducing stress crack resistance) in the HDPE geomembrane and do not directly relate to the continuing ability of the geomembrane to act as an effective hydraulic barrier, if not previously damaged by other factors.

6 DURABILITY OF POLYETHYLENE LINERS IN FIELD APPLICATIONS

This chapter reviews case study literature describing the performance of geomembrane liners over extended periods of time under various exposure conditions with information on measured changes to the material properties. The exposure conditions that strongly influence the geomembrane durability are then reviewed. These comprise:

- liner temperature;
- ultraviolet radiation;
- high energy radiation;
- availability of oxygen;
- exposure to leachate;
- external mechanical stresses.

Temperature has an exponential effect on geomembrane degradation and data from publications as well as from several UK sites are considered in some detail to provide a resource of information on this subject.

6.1 Reported field performance

Schmidt *et al.* (1984) reported that polyethylene liner used in salt works in South Africa in 1960 in exposed and submerged conditions had not substantially deteriorated in fifteen years. Eight case studies of polyethylene liner were presented and the differences between the original specifications and the exposed specimens were given. The results show that in all cases, the geomembranes were still providing satisfactory service (up to 16 years). Reductions in elongation and breaking strength of up to 48% were observed, indicating stiffening of the polymer, but reportedly without having a negative effect on the functionality of the geomembrane. Buried or unexposed geomembranes showed smaller reductions in these properties when compared to exposed membranes. The geomembranes were subject to mechanical damage, proving to be the major cause of failure.

Hsuan *et al.* (1991) studied the effects of exposure on a 7-year old HDPE geomembrane for leachate storage in a surface impoundment. Samples were obtained from four different locations in the lagoon, ranging from areas continuously exposed to the atmosphere to those at the bottom of the lagoon, continuously covered by leachate. The results of the testing showed that the engineering properties (yield strength, peel and shear strength of the seams) were not affected by the different levels of exposure. The physical properties (stress crack resistance, OIT) did show some degree of alteration. The OIT of the exposed samples was much lower than the OIT of the samples from the covered and partially covered locations. While the stress crack resistance (SCR) of samples from the sheet from the different locations was similar, the SCR of seamed samples, formed by hot air welding, from the exposed location was somewhat lower than that of unexposed samples, suggesting that surface embrittlement caused by photo-oxidation, heat from the sun accelerating diffusion and possibly complete removal of antioxidants from the surface may have accelerated crack initiation.

A study by Brady *et al.* (1994) as reported by Sangam (2001) examined the behaviour of HDPE geomembranes in different environments over a period of 30 years. The results of tests on un-aged and 30-year old samples showed that there were no substantial changes in density, water adsorption value, and water extractable matter content. There was no significant change in impact resistance over a 15½-year period but a reduction of 50% was observed over 30 years. The data from the tensile tests showed that over the 30-year period, the tensile strength

remained essentially constant but there was a reduction in the strain at the peak (yield) strength indicating that the HDPE became stiffer with time.

The performance of an HDPE geomembrane after 7 years in a landfill application was reported by Rollin *et al.* (1994). The 2.0mm geomembranes were part of a composite liner (clay/geomembrane/clay) system used to contain a contaminated soil in a landfill constructed in 1983. A 1.5mm HDPE geomembrane was used in the landfill cap. The soil was highly acidic (pH ~ 1.2) and contained mainly heavy metals (cyanides, zinc, copper and arsenic), phenolic compounds and heavy hydrocarbons (oil). The contaminated soil was removed to another site in 1990 and the geomembrane liner exposed, sampled and tested. Tensile tests revealed that the strength at yield increased slightly (0 to 8%). However, both the tensile strength and elongation at break decreased significantly with average decreases of 16% for samples from the sloping sides, 25% for samples from the cap and 60% for samples taken from the basal liner. At the seams, the elongation at break values were lower, with reductions from the original sheet values of 70% to 96%, indicating ageing along the edge of the seams.

Eith and Koerner (1997) described a case in which an HDPE geomembrane was used as a part of a double liner system for a municipal waste landfill constructed in 1988 and which was exhumed in 1996. During the 8 years of service, the geomembrane had been exposed to various concentrations of leachate constituents. The physical, mechanical and endurance test results indicated no apparent degradation of the HDPE geomembrane properties since they were still within the range of data generated for the original material at the time of the installation.

Sangam (2001) reviewed the evaluation of a 1.0mm HDPE smooth geomembrane after 11 years use in a wastewater treatment facility, as reported by Adams and Wagner (2000). The wastewater was from a fruit concentrate processing plant with a pH in the range 5 to 8. Two samples collected from above and below the wastewater level were tested and the measured properties compared to the original properties. The results indicated that while the strengths at yield and break had not changed significantly, the elongation at break of the samples from above and below the wastewater was reduced by about 21% and 14% respectively. The OIT had decreased by about 14% in the sample from above the wastewater and to a greater amount, 24%, in the sample from below. The study showed the sensitivity of the elongation at break to the assessment of the ageing process.

In the UK, an HDPE geomembrane exposed for periods of 2 and 6 years at intercell bunds was tested prior to welding to the new HDPE geomembrane for the adjacent cell. Conformance tests including SP-NCTL stress crack and tensile tests showed that the material remained within the original specification criteria, although the original values for the geomembrane when it was new were not available.

A detailed study was carried out by Rowe *et al.* (2003) on a 1.5mm HDPE geomembrane overlying a 3m thick compacted clay liner used for a leachate lagoon. Detailed mapping of the liner revealed many wrinkles and defects (cracks, holes and patches). Cracks observed on the slopes where the HDPE geomembrane liner was not covered by leachate and therefore exposed to sunlight and climate extremes were typically oriented down the slope and located near either seams or patches. The cracks were similar to field observations reported by Peggs and Carlson (1989) and Hsuan (2000) who attributed the cracks to high thermal contraction stresses along the top of the slope adjacent to the anchor trench where the geomembrane was completely restrained from contraction. Some of the cracks were 300mm long suggesting high susceptibility to cracking and that the geomembrane was relatively brittle. An undefined number of the holes were considered to be the result of lagoon maintenance activities. In summary, 82 cracks, holes and patches (repaired former holes or cracks) having different forms, patterns and sizes were observed in the geomembrane over an area of 1552m². This yields an average of 528 defects

per hectare over the 14-year period of operation. Of these, 70% (3851 defects/hectare) were above leachate level and 30% (178 defects/hectare) were below, a more than twenty-fold higher frequency for the exposed liner.

They observed very low OIT values for the exposed geomembrane of 1.8 minutes compared to OIT values in new, modern geomembranes typically of 100 minutes. Rowe *et al.* (2003) observed that the OIT value is close to the OIT value of 0.5 minutes reported by Hsuan & Koerner (1995) for an unstabilised (without any antioxidant) HDPE geomembrane and the possibility that oxidative degradation may already have started could not be excluded. The OIT values for the samples from below leachate level, although somewhat higher at 5 - 6 minutes, were still very low. The initial OIT value at the time of installation was unknown but using a typical value from material available in the 1980s, they estimated that the antioxidant depletion proceeded at approximately 0.24 year^{-1} for the exposed samples and 0.15 year^{-1} for samples covered by leachate. The density of the HDPE resin (excluding additives) was measured at about $0.954 - 0.955 \text{ gcm}^{-3}$, being higher than typical values for copolymer HDPE resin and much higher than MDPE resins as used in modern HDPE geomembranes. The crystallinity was commensurately high at 65 - 67%. This suggested a stiffer geomembrane at the time of installation than modern geomembranes. The initial stress crack resistance was unknown but the values in the recovered samples were remarkably low with all specimens failing within 4 hours in the single notched constant load test (SP-NCTL) compared to 200 or 300 hours specified for a new modern HDPE geomembrane. The samples from above the leachate level had earlier failure times than for the covered samples. The Melt Index (MI) values also indicated that the exposed samples had experienced the greatest degradation.

Using the contaminant profile through the underlying 3m thick clay liner, Rowe *et al.* (2003) modelled the leakage through the geomembrane liner and concluded that the geomembrane had failed to function effectively at some time between construction and 4 years after construction. They also concluded that to protect the geomembrane liner, maintenance of the lagoon should have been part of the planning and design of the facility. While the original properties of the geomembrane and the installation details were not available to the authors, the investigation provides a useful study on the degradation of "old" HDPE geomembrane liners which is helpful in assessing future degradation of modern HDPE geomembranes.

6.2 Review of reported field performance

Most of the field performance case histories relate to the use of HDPE geomembrane in storage impoundments for salt evaporation, leachate and wastewater storage rather than landfills. The particular effects of UV radiation (photo-oxidation), heat and repeated expansion and contraction stresses on sheet exposed to the elements are much more severe exposure conditions compared to a landfill liner at a relatively constant and lower temperature buried below waste. Suits and Hsuan (2003) note that antioxidant depletion occurs significantly faster by photo-oxidation than by thermal oxidation. The outdoor exposure conditions may well be synergistic, accelerating the oxidation process particularly in the exposed surface layer of HDPE liner. It was noted for geomembranes in exposed conditions, physical damage from installation, maintenance and operational activities was generally the major cause of defects.

Of the two cases reviewed where liners were exhumed from landfills, only one was from a municipal waste site while the other was from a site containing heavily contaminated soil. In the former, the liner was reported to have performed well with no apparent degradation while in the latter, significant degradation was reported as shown by the reduction in elongation at break in tensile tests.

It is difficult to assess the implications of the results of these case studies for several reasons:

- The HDPE geomembrane used is not directly comparable with modern geomembranes, particularly with respect to stress crack resistance, crystallinity, density and OIT (initial values and response to oven ageing).
- The range of tests carried out at the time of installation was generally limited and did not include OIT and SP-NCTL (which had not been introduced). As a result, only restricted comparisons can be made of the original and aged samples.
- The range of exposure conditions is very varied and predominantly relates to exposed liners for surface impoundments.

Polyethylene geomembranes for lining lagoons, ponds and reservoirs are much more susceptible to degradation than landfill liners, as well as being particularly vulnerable to physical damage. Koerner (1998) discusses the design of HDPE geomembrane liners for liquid impoundments and describes the benefits of overlying protection to the liner but this can add design, operational and cost implications. The case study by Rowe *et al.* (2003) underlines the benefit of a composite liner for liquid impoundments rather than relying solely on a single geomembrane.

6.3 Exposure conditions

The oxidation reaction in polyethylene is substantially influenced by the conditions it is exposed to throughout its service life. Haxo and Haxo (1989) itemise in detail and discuss qualitatively the environmental conditions and stresses that geomembrane liners may encounter during construction and service in MSW and hazardous waste landfills, and in surface impoundments. Any condition that removes antioxidants and other additives, provides oxygen or accelerates the formation of free radicals, particularly the decomposition of hydroperoxides, increases the rate of degradation. Antioxidant depletion is accelerated by exposure to simulated landfill leachate and to a lesser extent water compared to air, while oxidation requires a supply of oxygen. As noted earlier, Allen & Edge (1992) identified elevated temperatures, sunlight and high-energy radiation as primary activators of polyethylene degradation, and the effect of these factors can be enhanced by mechanical stress. For an exposed HDPE geomembrane, sunlight coupled with heat and tensile contraction stresses create an environment where there is great potential for free radical formation and relatively rapid degradation of the polyethylene. In a landfill situation where the geomembrane is buried below waste, the temperature of the liner throughout the life of the geomembrane will be a controlling influence on the rate of degradation, as seen from the Arrhenius plots and the laboratory-predicted liner lifetimes in the previous chapter.

6.3.1 Temperatures at the liner

The best quality and most reliable data of liner temperatures in operating conditions come from temperature measurements taken by instruments at the liner, followed by temperature of leachate measured in the leachate collection system. Temperatures of recovered leachate samples recorded at ground level are less reliable, while measurements taken in the body of the landfill will not necessarily reflect the temperature at the liner. The temperature in a landfill is influenced by a large number of factors some of which are waste types, biodegradable content, rate of filling, gas extraction activities, moisture content, depth of waste, leachate mounding, leachate recirculation, waste pre-treatment, waste density, location in the landfill and time since waste deposition. In view of the large number of variables, it is not surprising that a wide range of temperatures is reported in the literature.

Exposed geomembrane liners

Where a geomembrane liner is exposed to the sun, surface temperatures have been recorded at 80°C in the USA while Aversch and Schicketanz (2000) reported temperatures on the

geomembrane surface of 60°C. Philip *et al.* (2002) report temperatures on exposed geomembrane from a test cell at a site in SW England recorded during July and August reaching over 44°C with a diurnal range typically of 16° - 42°C. These figures may be compared to the typical air temperature range of 12° - 26°C recorded during the same period.

Koerner and Koerner (1995) recorded surface temperatures using thermocouples on exposed HDPE geomembrane test panels at a site in Pennsylvania, over different seasons of the year. Both black and white geomembranes were tested, as well as smooth and textured sheet. The peak temperature responses are summarised in Table 6.1. It is interesting to note the reduction in surface temperature achieved by the white geomembrane.

Table 6.1 Results from exposed geomembrane study (modified from Koerner and Koerner, 1995)

Season	Maximum ambient temperature (°C)	Black geomembrane max. temperature (°C)	White geomembrane max. temperature (°C)
Winter	5	13	2
Spring	22	46	38
Summer	30	70	57
Autumn	18	35	28

Solid waste landfills - published data

Koerner and Koerner (1995) also installed thermocouples in a MSW landfill cell with eight gauges directly on the geomembrane, one in the gravel leachate collection system above the geomembrane and one in the solid waste at a higher elevation. Once covered by the protection geotextile, the geomembrane temperature was 36°C compared with an ambient temperature of 42°C; after placing the gravel leachate collection layer, the liner temperature was 26°C compared to the maximum ambient temperature of 40°C. After waste placement, the geomembrane temperature stabilised at about 21°C.

Monitoring at the Pennsylvania landfill continued, as well as on geomembranes at sites in Florida and California over periods of 6, 3 and 5 years respectively. Over the period monitored, the temperature on the liner remained relatively constant within the following ranges: Pennsylvania 18° - 23°C, Florida 20° - 30°C and California 10° - 30°C. Subsequent information on the Pennsylvania landfill is available from the Geosynthetic Research Institute (GRI) website extending the period of monitoring to 8.7 years and the minimum, maximum and average temperatures recorded at the liner are 17°, 38° and 27°C respectively. The cell is classed as a conventional 'dry' cell with no additional liquids added. Temperatures have also been recorded in another cell at the same site but which is a "wet" cell operated as a bioreactor landfill. As reported in the June 2003 issue of the Newsletter from GRI available on their website, the geomembrane liner was at an average temperature of 25°C (5°C higher than the dry landfill) from the start. It has gradually risen over the 2.5-year monitoring period to an average temperature of 40°C (approximately 10°C higher than the dry landfill). This indicates a relationship between the temperature at the liner and the moisture content of the waste.

Rowe (1998) reviewed landfill temperatures reported in the literature, noting that a wide range of temperatures was reported. High temperatures (50°C to 70°C) have been reported in a number of continental European landfills (Ramke, 1989; Lechner and Lahner, 1991). Brune *et al.* (1991) reported temperatures ranging from 24°C to 38°C in a leachate drain beneath 4 - 6 year old waste at one German landfill, which had been filled rapidly at a rate of 10 - 20m per year.

Although it was in a methane forming phase, the leachate was still acidic and very strong. In contrast, at the Venneberg landfill in Germany, the temperature in the drain only ranged from 14° to 20°C. This landfill had been filled much more slowly and the leachate was only lightly loaded with both organic and inorganic contaminants.

Figure 6.1 (Rowe, 1998 from data published by Collins, 1993) shows the variation of temperature with depth at two locations in the old City of Hanover landfill (filled 1936 - 1980). The temperature increases with depth to a peak at 30m below surface and then decreases towards the base of the landfill. The base temperature in 1990 (i.e. 10 years post-closure) ranged from 30°C to 60°C in this landfill where the leachate level was reported to be 4 - 6m above the base. The landfill increased the temperature of the groundwater at 15 m below the base of the waste by 4° - 7°C above the normal 12° - 15°C.

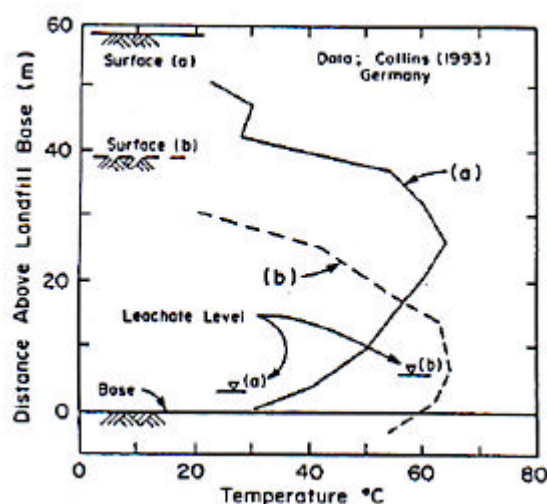


Figure 6.1 Temperature variations (Rowe, 1998)

A similar landfill temperature profile is also reported by Rowe (1998) for a 30m deep Japanese landfill. Landfilling took place between 1976 and 1979 and, in 1987 and 1988, the basal temperature was 50°C. The very high leachate mound was typically 20 - 25m above the base.

Rowe (1998) also discussed the temperatures recorded by Koerner and Koerner (1995) at the three sites in Pennsylvania, Florida and California. He considered that the reason for the relatively low temperatures compared to those recorded at other sites was due to lower leachate mounding and moisture content of the waste. The recently reported higher temperatures in the "wet" cell support this conclusion.

Barone *et al.* (1997) monitored temperatures at the base of a MSW landfill in Toronto, Ontario using thermistors in vibrating wire piezometers from 1983 to 1996. In areas 12-13 years old (Stage 1) the average temperature recorded was 33°C with a maximum of 40°C, with leachate levels ranging from 4 - 6m. As discussed by Rowe (1998), examining the data for one Stage 1 monitoring point (Figure 6.2) it can be seen that for the first 8 years, the head remained low and the temperature increased from a few degrees up to 12°C. However, after 1992 the leachate head increased to 5.4m and the temperature rose to over 40°C.

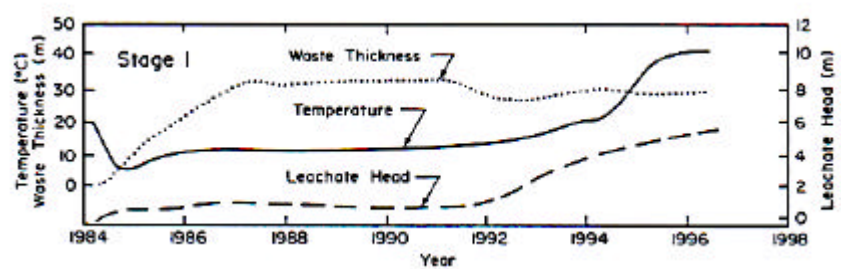


Figure 6.2 Variation in temperature, leachate head and waste thickness above base of landfill (Rowe, 1998 modified from Barone *et al.*, 1997)

In areas less than 7 years old, temperatures were recorded as 10° - 15°C, with leachate levels of less than 0.3m. Barone *et al.* (1997) also reviewed temperatures from other MSW landfill sites indicating temperatures ranged from 20° to 55°C, the higher temperatures reported for sites having leachate levels greater than 10m above the liner. Figure 6.3 from Rowe (1998) shows the data presented by Barone *et al.* (1997) plus some additional data. It shows a strong correlation between the temperature at the base of a landfill and the leachate head above the base.

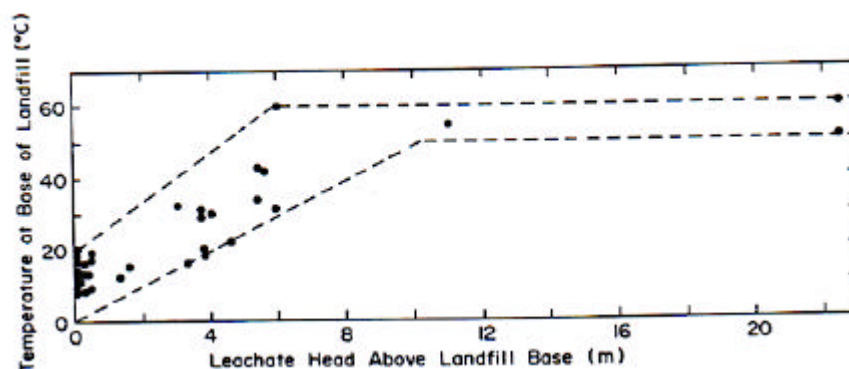


Figure 6.3 Variation in temperature at landfill base with leachate head for a number of landfills (Rowe, 1998 modified from Barone *et al.*, 1997)

Di Molfetta and Sethi (2001) reported temperatures in leachate collection wells at between 11°-27°C from a MSW landfill in the north of Italy. The groundwater temperature was recorded as 18.5°C being 4°C higher than the background temperature of the groundwater, a difference they considered to be due to heat transport from the landfill.

In the detailed review of the composition of leachates from domestic wastes in UK landfill sites (Robinson, 1995), temperature measurements at a few of the landfills were recorded. These were at five sites, four of the landfills being classed as “large landfills with a high waste input rate, deep, wet and bioreactive.” The results are summarised below:

Aveley Landfill, Essex

Landfilling took place from 1971 to 1987. Leachate monitoring was regularly undertaken from 1981, giving 13 years data up to the preparation of the report. The waste depth is 30m. The leachate level rose 1-2m per annum in the first few years of disposal with records of temperatures of 45°-50°C. The leachate depth by 1994 had reached 20m. The leachate rise was due to both surface infiltration and groundwater inflow (no engineered liner had been provided). It is recorded that leachate temperatures of about 40°-45°C have been sustained from 1979 to 1994. Work by Rees and Grainger (1982) demonstrated that based on measured rates

of methane production, heat released during the process of methanogenesis was high enough to account for the observed temperature rises.

L-Field Landfill, Bedfordshire

This is a co-disposal site that accepted waste from 1976. Water entry has been by rainfall infiltration rather than by groundwater inflow and a complex, heterogeneous water distribution was found within the waste compared to the rising leachate level as seen at Aveley. The saturated depth in the waste is substantial. The leachate temperatures have generally been 25°-28°C. The lower temperature at L-Field compared to Aveley was attributed to the surface infiltration of water rather than by basal inflow.

Warnham Landfill, West Sussex

Infilling of domestic, commercial and industrial wastes took place between 1982-1992; the average depth of wastes is 25m. The leachate level rose to a depth of 10-15m, caused both by groundwater inflow and surface infiltration. Temperatures of leachate samples from wells were 41°-44°C. This site was considered to be similar to Aveley Landfill.

Withnell Landfill, Lancashire

This site is located in a former quarry near Blackburn in an area of very high incident rainfall (>1200mm). Local wind patterns and the high wall of the quarry increase quantities of direct incident rainfall and there are reportedly significant groundwater flows into the quarry. Infilling began in 1981 comprising 90% domestic waste and was due to continue to the late 1990s. The leachate depth was limited by extraction to 14m arising from high surface infiltration and some groundwater inflow. Leachate temperatures of 31-34°C recorded in 1990 are indicative of bioreactive conditions. They are 10-15°C lower than other sites where waste saturation has occurred largely by ingress of groundwater from beneath (Aveley and Warnham). The lower temperatures were attributed to most of the leachate arising from high surface infiltration of the very high quantities of incident rainfall at this site.

Holiday Moss Landfill, Merseyside

This was classed as a large landfill with high waste input, deep, wet and "cold." The site was operated as a containment site with a liner and leachate collection system installed since about 1988 taking 75% industrial and commercial waste, 25% domestic waste. The maximum waste depth was 15m. Recorded leachate temperatures were 20°-23°C, measured in 1991.

Solid waste landfills - additional temperature data

To augment the published data, measurements recorded at sites in England have been obtained and reviewed:

- Data from ongoing monitoring of six landfills (Sites 1 to 6) has been made available by two commercial operators. All the landfills accepted MSW. The temperatures were recorded at the time of sampling of leachate samples recovered from leachate wells.
- Results from October 2000 to May 2003 from 19 thermocouples installed at the liner of Beddington landfill near Croydon (Knox, 2003). Readings were recorded every 15 minutes.

Sites 1 to 6

Summary information on four of the sites is as follows:

Site 1

Data from closed landfill cells.

Leachate recirculated.

Took Household (1000t/day max), Industrial (100t/day max) and Difficults (100t/day max and 6000t/year max).

Open since approximately 1991.

Temperature data from extracted leachate samples.

Site 2

Data from closed landfill cells.

Leachate not recirculated.

Took Industrial, Commercial and Putresible wastes (quantities not available).

Temperature data from extracted leachate samples.

Site 3

Data from closed landfill cells.

Leachate recirculated.

Co-disposal site (takes flux waste from incinerators)

Open since approximately 1990.

Temperature data from extracted leachate samples.

Site 4

Data from closed landfill cells.

Leachate recirculated and extracted.

Took Household, Industrial, Inert and Commercial including Difficult (Group E Clinical).

Planning permission from 1985, although landfill waste may include older material.

Waste to approximately 50m depth.

Temperature data from extracted leachate samples.

Sites 5 and 6 both accepted MSW as well as commercial and industrial waste. The leachate samples from all six sites were removed from the wells before temperature testing. This technique may introduce temperature changes in the samples. A summary of the recorded temperatures is given in Table 6.2.

Table 6.2 Temperature monitoring information for Sites 1 to 6.

Site	Number of monitoring points	Monitoring	Monitoring frequency	Temperature (°C)				
		From	To		Min.	Max.	Median	Temp. range
1	19	03.03.98	28.11.02	Monthly	8	28	13	20
2	11	31.01.01	26.02.03	Monthly	14	30	20	16
3	3	10.01.01	18.02.03	Monthly	20	46	31	26
4	>75	10.02.99	29.08.02	Monthly	6	65	18	59
5	11	08.01.02	16.12.02	Monthly	8	32	11	24
6	17	16.01.02	02.12.02	Monthly	10	40	22	30

The temperature values obtained vary from 6°C to 65°C. The temperature range is between 16°C and 30°C for Sites 1, 2, 3, 5 and 6. Site 4, however, has a temperature range of 59°C and this site also possesses the lowest and highest temperature readings and, by a significant margin, the greatest number of monitoring points.

Erratic temperatures were recorded at Sites 1 – 4 with temperatures at times changing by up to 15°C in a single month. The data are unlikely to accurately reflect temperature variations at the base of the landfills which would be expected to change only slowly from month to month and must be considered to indicate that temperature modifications took place during the sampling and measurement procedure. Records at Sites 5 and 6 are much more consistent with time but showed variations of over 17°C and 20°C across the site respectively.

While average temperatures of 20°C and 15°C could be assigned to sites 1 and 2 respectively, locations within sites 3, 4, 5 and 6 show temperatures of about 30° to >40°C.

Beddington Landfill

An experimental study into the hydraulic response of landfills was conducted at Beddington bioreactor landfill starting October 2000 (Knox, 2003). Twenty vibrating wire piezometers with thermocouples were installed in the base of Cell 1B of the landfill, an area of approximately 1 ha. Readings of leachate head and temperature were recorded on a data logger every 15 minutes from 5th October 2000 on the 19 functioning instruments. Placement of MSW began in October 2000 and by the end of January 2001 there was about 9m of waste in the cell. The final thickness of waste exceeded 20m, attained in autumn 2001 and a clay cap was installed during November and December 2001. Injection trenches to facilitate leachate recirculation were installed in the top of the waste just prior to placement of the clay cap. Monitoring of temperatures continues and the results from October 2000 to May 2003 are shown on Figure 6.4.

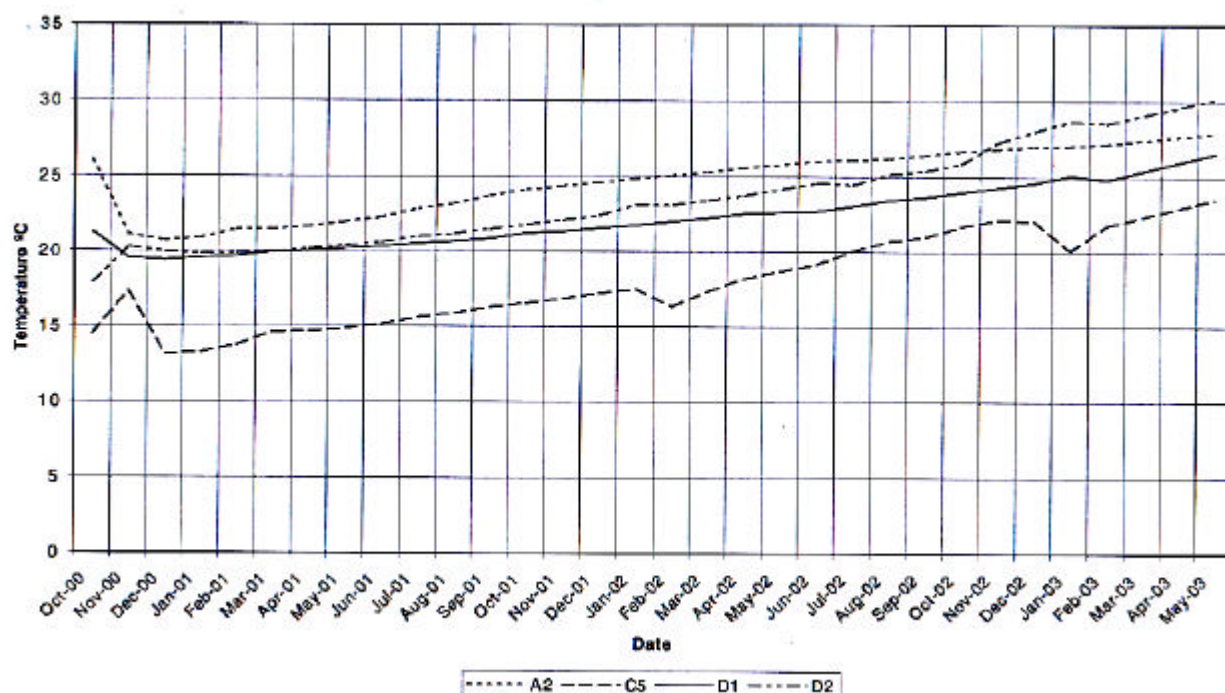


Figure 6.4 Graph of temperature against time for Beddington landfill

Results from four of the thermocouples representing the lowest, highest and typical intermediate results are shown in Figure 6.2. There is a temperature range of about 7°C between the highest and lowest measurements. After the initial period, temperatures increased gradually at rates of 0.25°C to 0.4°C per month as decomposition proceeded. Recorded leachate levels have been affected by intermittent extraction and also a drift in the piezometer instruments, so that a leachate depth/ basal temperature relationship cannot be determined. The moisture content of the wastes will be elevated as a result of the recirculation.

Ambient ground temperatures in the UK

The temperature of a landfill would be expected to reduce eventually to the ambient temperature of the ground in the vicinity of the site. From temperature data reviewed above, the period before this happens will be very long and will depend on numerous landfill-specific factors. It should also be noted that the landfill influences the temperature of the surrounding ground and groundwater.

The mean equilibrium sub-surface temperature for the UK is 12°C (+/- 2°C) at 100m below ground level with a range of 7 - 15°C, decreasing to 11°C at 50m depth (Rollin, 2003). At 15m depth, the ground temperature is close to mean annual air temperature which is controlled by elevation, aspect and location. Variations from the mean ground temperature are related mainly to thermal conductivity, heat flow and groundwater movement.

Concluding remarks on landfill temperatures

The considerable variation in temperatures recorded at the base of landfills both within individual sites and between sites demonstrates the difficulties in predicting an average service temperature to use in estimating the long-term degradation of HDPE geomembranes. A significant factor affecting basal temperatures in biodegradable wastes is the moisture content of the waste and, in particular, the saturated thickness of the waste. No records have been seen which show the rise and eventual fall towards ambient values of basal temperatures, so the duration of elevated temperatures is also difficult to predict. Sites that are active bioreactors, practise leachate recirculation and / or have elevated leachate levels should be expected to have temperatures at the higher end of the range. A reasonable long-term average for such landfills is estimated as between 30°-35°C. This recognises that mean temperatures across the landfill may be about 10°C higher than this for about a decade but will subsequently reduce, bringing down the long-term average. For "dry" landfills or those with a low biodegradable content, a long-term average temperature of 15° - 20°C may be appropriate.

Reductions in biodegradable content instigated by the Landfill Directive should result in reduced temperatures at the liners. Further research using temperatures obtained from well documented landfills with instruments at the basal liner will be necessary before reliable predictions of liner temperatures can be made from landfills with these wastes. An average temperature over the expected life of the liner should be selected based on the particular characteristics for the site. The longer the estimated durability of the geomembrane, it is probable that the liner temperature will decrease closer to the background ambient temperature of the surrounding ground. This will reduce the average temperature over the geomembrane service life. Hence, an iterative cycle of estimates may be necessary to select the average temperature.

There is a trend in continental Europe towards aerobic remediation and acceleration of stabilisation which could result in significantly higher waste and leachate temperatures in landfill for limited periods of time.

6.3.2 Ultraviolet radiation

Short-wavelength energy in the UV range from sunlight can penetrate the polymer structure of an HDPE geomembrane causing the formation of free radicals and chain scission as discussed in Chapter 4. This will be relevant where a geomembrane is left exposed for a considerable number of years. Koerner (1998) states that the temporary covering of geomembranes prior to placement to protect from UV degradation is not generally necessary. The consequences of long-term exposure include surface cracks, discoloration, brittleness and deterioration of mechanical properties. Permanently exposed geomembranes such as exposed side slopes of landfills and lagoons or reservoirs may be covered with soil or a sacrificial material, for example, a replaceable geotextile. A 0.15m soil cover has been reported as being sufficient to protect a geomembrane from UV light (Koerner *et al.*, 1990). Where polyethylene geomembranes are exposed for a number of years (e.g. upper parts of lagoons and reservoirs, floating covers in reservoirs or sometimes at landfill intercell bunds), then UV degradation will be a major design life issue. However, exposure of HDPE geomembranes to UV radiation should always be minimised as hardening and embrittlement of the surface may occur (Tavares *et al.*, 2003) potentially leading to microscopic crazing which could, in the long term, become initiators for future stress cracking.

The susceptibility of HDPE geomembranes to UV degradation is countered by the addition of carbon black or chemical-based light stabilisers to the resin that prevent the light from penetrating the polymer structure. The carbon black pigment in HDPE geomembranes is added at between 2 – 3% w/w to act as a screening or blocking agent to minimise photo-oxidation. The effectiveness of the carbon black depends on several factors including concentration, particle size and dispersion (Allen & Edge, 1992). Both the concentration and particle size influence the dispersion of the carbon black in the polymer matrix and consequently its screening efficiency. The 3% content of the pigment provides the maximum opacity level above which no significant improvement in UV resistance occurs. In fact, higher concentrations can start to have an adverse effect on the physical properties of the polymer. The efficiency of carbon black is shown by the fact that polyethylene samples containing only 1% of the pigment have been found to be stable even after outdoor exposure in Florida for over 30 years (Allen and Edge, 1992).

6.3.3 High energy radiation

Radioactivity above 10^6 to 10^7 rads is quite likely to cause polymer degradation by chain scission (Koerner, 1998) so that containment of high-level radioactive waste would not be likely to use HDPE geomembranes as barriers. Low-level radioactive waste has much lower activity and HDPE may be suitable for use in containment systems for these materials. Kane and Widmayer (1989) describe a number of HDPE lined radioactive waste containment uses and reported that HDPE geomembrane lining had been used since 1979 for high-integrity containers for low-level radioactive waste disposal. They also noted that results from research show that HDPE under stress becomes brittle following exposure to radiation but they gave no details of the tests, and conclude that considerable further research is required to assess the resistance of geosynthetics to degradation caused by radiation effects over a 300 to 500 year life in a radioactive waste disposal environment. Badu-Tweneboah *et al.* (1999) evaluated the predicted long-term (500 years) performance of two polyethylene components of a low-level radioactive waste disposal landfill and concluded that they would perform their containment functions during the 500 years design period.

As Koerner (1998) notes, there are very few references in the open literature on radioactive degradation of geomembranes and that further published research should be undertaken.

6.3.4 Availability of oxygen

The concentration of available oxygen is an essential component of any oxidation reaction. Hsuan and Koerner (2002) reported a recent durability study by Elias *et al.* (1999) in which the oxidative degradation of polyolefin geotextiles and geogrids was evaluated at oxygen concentrations of 8% and 21%. Figure 6.5 shows the strength retained at 70°C and 80°C at the two oxygen concentrations after being incubated in forced air ovens. Clearly, the availability of oxygen had a major effect, one that would also be expected to apply in a modified manner to polyethylene geomembranes.

In a recent study on oxidative resistance of polyolefin geotextiles by Müller *et al.* (2003), the induction and oxidation stages of the geotextile fibres were examined in some detail. The much greater surface to volume ratio of the fibres compared to geomembrane sheet gave much shorter test durations, enabling the research to be conducted on these aspects. It was noted that these processes were strongly dependent on the rate of oxygen supply. The lower the available oxygen, the longer the induction stage and the slower the rate of oxidation.

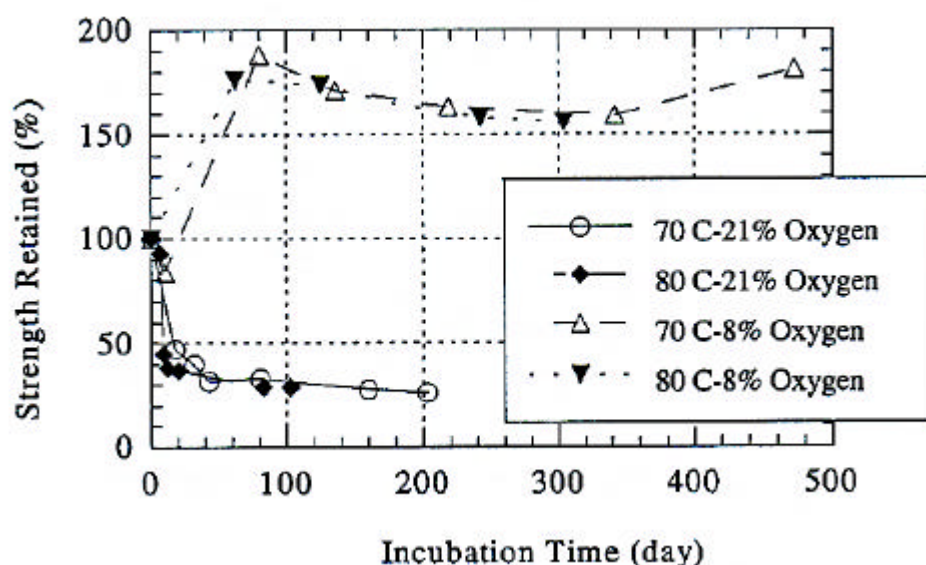


Figure 6.5 Tensile strength retained at 21% and 8% oxygen environments (from Elias *et al.*, 1999)

Exposed geomembranes will have a readily available supply of oxidation on the upper surface and, to a lesser extent, on the underside.

The removal of antioxidants can progress in an anaerobic environment, for example, by extraction into leachate. Once removed, oxidation commences but only if oxygen is available. The availability of oxygen depends upon the location of the geomembrane in the landfill (or other facility). Table 6.3 summarises the potential availability of oxygen in landfill and lagoon / reservoir applications (Hsuan & Koerner, 1995).

Table 6.3 Oxygen availability to geomembranes (from Hsuan and Koerner, 1995)

Application	Location	Geomembrane surface	Oxygen availability
Surface impoundment (liner)	top of slope	top	high
	top of slope	bottom	moderate
	base of slope	top	low
	base of slope	bottom	moderate
Landfill liners	beneath waste	top	nil (after 5 years)
		bottom	low
Landfill and waste pile covers	above waste	top	high to moderate
		bottom	nil (after 5 years)

However, once anaerobic conditions have been established in a methanogenic landfill, the availability of oxygen will effectively be zero in either free form or as oxygen in nitrates or sulphates (Robinson, 2003). Whether these conditions persist into Stages B and C after the long antioxidant depletion process requires consideration. Oxygen will be present to some degree on the underside depending on the saturation of the contact material and groundwater conditions. When the geomembrane is part of a composite liner, the underlying contact material may be compacted clay, BES, or a GCL, probably in a hydrated condition, where a substantially reduced oxygen supply compared to open exposure may be considered to be available.

Viebkke *et al.* (1994) reported on the degradation of MDPE pipes without antioxidant additives. Pipes of 2mm wall thickness filled with pressurised stagnant de-ionised water and circulating air as an external medium at different temperatures were examined. From OIT measurements, it was reported that oxidation occurred only near the outer wall surface.

Thus, for a basal geomembrane liner, the lack of oxygen available to the upper surface exposed to methanogenic leachate will prevent the induction and actual degradation stages of oxidation proceeding on that surface. It is possible that oxygen will gain access to parts of the liner along the leachate collection and removal system. Once the degradation of the landfill has progressed such that oxygen is again present at the liner, then oxidation can proceed on the upper surface after complete antioxidant depletion. On the underside of the geomembrane, the presence of oxygen should be limited where there is good geomembrane to mineral liner contact. This will retard both the antioxidant depletion and subsequent oxidation induction stages. On side slopes and caps, oxygen availability to the inner as well as the outer surfaces of the geomembrane is likely to be higher.

Measurement of oxygen concentrations on the inner and outer surfaces of geomembrane barriers throughout their service life are scarce and efforts to obtain these data would be beneficial, as would further study on the rate retarding effects of depleted oxygen levels on the oxidative degradation processes.

6.3.5 Exposure to leachate

Geomembrane liners in landfill, lagoon and contaminated land applications will be exposed to the liquids of the lagoon, leachates derived from the wastes or contamination, and contaminated groundwater. The selection of suitable geomembrane material that will provide the required chemical resistance needs to be made based on established experience and published chemical resistance data or, where this is not available, by a programme of testing. The field performance data on HDPE geomembranes are discussed in Section 6.1 above. Laboratory studies on leachate effects on HDPE geomembranes have been undertaken by, among others, Artieres (1991); Overmann *et al.* (1993); Bernhard *et al.* (1994); Duquennoi *et al.* (1995); Surmann *et al.* (1995); Castaldo *et al.* (1996); Carroro *et al.* (1997); Maissonneuve *et al.* (1997); Lopes *et al.* (1998); Lodi and Bueno (2002) and Maia and Vilar (2002). In all of these studies, no significant deterioration in physical or mechanical properties was recorded. In the USA, leachates from MSW landfills are considered relatively benign with respect to HDPE geomembranes such that chemical resistance tests are now rarely required. Many EPA 9090 tests have been conducted with MSW leachates and none have been shown to damage the geomembrane (Konrath and Ballod, 2001).

Candidate liner testing remains a useful selection tool for geomembranes of new materials or where unusual leachates are involved. Such testing, for example to ASTM D5322, with actual or synthesised leachate, lagoon liquid or contaminated groundwater may be necessary (Koerner, 1998). This incubation process would then be followed by a series of physical and mechanical property tests (e.g. to ASTM D5747) over varying times to determine if the original geomembrane properties had changed during the incubation period. The EPA 9090 method of candidate liner testing may alternatively be used. There are no established criteria on the allowable variation from the original test properties but Koerner (1998) has published suggested values for various geomembranes including HDPE. He reports that specific values are not available for VFPE (e.g. LLDPE) and flexible polypropylene, and suggests that the criteria for thermoset and thermoplastic polymers be used, as opposed to the more onerous HDPE criteria that reflect the higher chemical resistance of HDPE.

HDPE geomembranes are recognised as having excellent chemical resistance against leachates derived from municipal, industrial and commercial wastes from landfills in the UK and elsewhere, as well as co-disposal waste sites in the UK. A significant proportion of the UK solid hazardous waste streams is currently co-disposed without prior treatment. Leachate quality from these sites has been well characterised by studies over many years (e.g. Robinson, 1995). It has been shown to be similar in most respects to the leachate quality from MSW landfills, that is, it is dominated by the biological processes responsible for the degradation of the organic compounds of MSW.

An HDPE geomembrane in contact with leachate may potentially be affected by swelling and plasticising, accelerated loss of additives, as well as minor changes to tensile strength and elongation at yield and failure. Some organic contaminants at low concentrations within leachate and more concentrated organic liquids can migrate through the amorphous regions of HDPE geomembranes by diffusion (Rowe and Sangam, 2002). While these substances are absorbed by the HDPE geomembrane causing it to swell and soften, they are not considered to cause a continuing and permanent degradation. If the liquid environment is removed, the organic vapours should volatilise out of the geomembrane, which then should recover to its original condition. An effect of the swelling is that antioxidant depletion may be facilitated and accelerated.

Environment Agency research: Post-Landfill Directive leachate chemistry

The European Council Directive on the landfill of waste, 1999/31/EC known as the Landfill Directive, will ultimately prohibit the co-disposal of hazardous wastes with biodegradable municipal wastes. The amount of biodegradable municipal waste deposited at landfill will also have to reduce progressively over a 15-year period to only 35% of the total amount produced in 1995. Furthermore, the Landfill Directive also requires member states to pre-treat wastes prior to landfill disposal to reduce the quantity or hazard. Hazardous wastes will have to be deposited in hazardous waste landfills (or in a non-hazardous waste landfill following treatment, so long as new acceptance criteria are satisfied and biodegradable wastes are not deposited in the same cell).

The Agency has commissioned a research project to examine the potential impacts of the Landfill Directive on the quality of leachates in future UK landfills (Environment Agency R & D Project P1-494). Preliminary results of the project have been published by Bone *et al.* (2002a, 2002b). The project has examined leachate quality data from research work and landfills in EU member states where the treatment of biodegradable MSW has been required for a number of years, as well as from hazardous waste landfills where waste acceptance criteria are similar to those expected in the UK.

For future non-hazardous waste landfills in the UK which will accept minimally treated biodegradable wastes and wastes subjected to mechanical and biological pre-treatment (MBP) and/or incineration, effects on leachate quality may be summarised in the following way (Bone *et al.*, 2002a).

- ash residues from incineration of biodegradable MSW can produce highly variable leachates which have an organic strength similar to or lower than a dilute methanogenic leachate from a conventional landfill;
- dissolved solids are significantly higher than in typical UK leachates;
- leachates produced by residues from mechanical pre-treatment can have a high polluting potential but biological treatment of these residues can avoid the peak acetogenic phase

of decomposition and produce leachates that are similar to or of lower polluting potential than methanogenic leachates from conventional landfills;

- leachates will require long-term management over similar timescales to leachate from untreated MSW.

The summary of findings from the leachate data from hazardous waste sites were (Bone *et al.*, 2002b):

- the leachates at most of the sites exhibited little or no evidence of being affected by biological activity;
- the leachates were highly variable due to site-specific and waste-specific factors (compared to UK MSW / co-disposal leachates);
- certain generic characteristics were identified;
- salinity was several times greater than MSW / co-disposal leachates;
- ammoniacal nitrogen was at levels similar to a dilute / medium strength MSW leachate;
- total Organic Carbon was typically less than 100 mg/l;
- there were elevated levels of site-specific heavy metals;
- concentrations of oil, PAH, VOCs and PCBs were no higher than at current UK landfills;
- the leachates reported in the study were considered unlikely to have a deleterious effect on geomembrane liners.

Consideration of the published findings of the research project identifies three factors that may influence HDPE geomembrane degradation.

Firstly, the reduction in the biodegradable activity at some sites, especially at the hazardous waste sites, would be expected to result in lower temperatures of the waste and, therefore, also at the geomembrane liner compared to liners in landfills with untreated MSW. Certain chemical processes including carbonation may be significantly exothermic but are likely to be more localised and of shorter duration than the biodegradation of MSW. Unfortunately, no temperature data are available from the leachate research project to confirm this expectation. Further temperature data records are necessary to assist in predicting geomembrane liner temperatures in future landfills.

Secondly, the elevated metals content of the leachate from MBP wastes, MSW incineration residues and pre-treated hazardous waste sites may be instrumental in accelerating geomembrane degradation. The susceptibility of polyolefins to metal ions is well reported. This has been noted both when a polyolefin is in contact with metal foil or metal contaminated soil, and when metal ions in leachates are present. The degradation of the HDPE geomembrane in the field case study reported by Rollin *et al.* (1994) in Section 6.1 may have been linked to the high concentrations of metals including transition metals. A thermo-oxidation study on polypropylene immersed in metal-rich aqueous solutions undertaken by Van Langenhove (1990) showed that degradation was accelerated by a factor of 10 or more depending on the metal. Ezrin *et al.* (2000) describe failure of an HDPE cable sheath. The cable had been wrapped in copper foil and cracking of the samples occurred where the HDPE was in close contact with the copper (at regions of foil overlap) and the foil had become wet, converting some copper to copper ions. OIT tests showed zero antioxidant to be present within the cracked regions indicating consumption by locally accelerated degradation rates.

The presence of transition metals such as Cu, Mn and Fe in leachate may significantly enhance the oxidation rate of a geomembrane by breaking down the hydroperoxides present in the geomembrane and creating additional free radicals. The transition metals in the presence of moisture or liquid can diffuse into the geomembrane (Rowe and Sangam, 2002). Antioxidant stabilisers could include metal deactivators to help protect the polyethylene from oxidation driven

by reactions with transition metals. No published information has been seen on whether the antioxidant packages currently used in HDPE geomembranes include metal deactivators. The long-term effect of transition metals in leachates on the service life of HDPE geomembranes with different antioxidant packages is unknown and more research is required in this area.

The third factor apparent from the study is that the timescale before waste reaches final storage quality is likely to be similar to that of current UK landfills. It appears at present that no reduction in the required design service life of HDPE geomembrane liners can therefore be expected from waste complying with the Landfill Directive.

6.3.6 External mechanical stresses

Allen and Edge (1992) identified mechanical stress as a chemical degradation process of polymers in which chain scission may occur. However, the level of stress required to initiate chemically degrading mechanisms is quite large. Sangam (2001) reported the presence of a safe stress level for polypropylene rods in the temperature range 80° – 130°C below which no degradation occurred following the application of the stress. Above this stress level, an appreciable acceleration of polymer embrittlement was observed. Little has been reported regarding this effect of stress on the degradation of HDPE geomembranes. Laboratory investigations conducted by Surmann *et al.* (1995) on HDPE geomembranes stressed at 5.6% to 8% revealed no accelerating effects when immersed in leachate. Similar observations were reported by Maisonneuve *et al.* (1997) who strained the geomembrane to 5%.

Stress cracking, as discussed in Chapter 7, is a form of mechanical degradation arising from relatively low levels of external stresses.

7 STRESS CRACKING

7.1 Introduction

Stress cracking is a brittle cracking phenomenon defined in ASTM D883 as “an external or internal crack in a plastic caused by a tensile stress less than its short-term mechanical strength”. These brittle cracks were first observed in HDPE geomembranes by Peggs and Little (1985) since when the phenomenon has been extensively studied (e.g. Peggs and Carlson, 1989, 1990; Halse *et al.*, 1989, 1990; Peggs and Kanninen, 1995; Thomas, 1998; and Hsuan, 2000).

The microstructure of an HDPE geomembrane is a series of crystalline lamellae of folded molecules with side branches, molecule ends, and cilia (loose loops), dangling outside the lamellae and often entangled in the adjacent lamellae. As presented by Lustiger and Rosenberg (1989), when a high stress is applied to this structure (Figure 7.1) these linking chains called tie molecules remain frictionally entangled in the adjacent lamellae and break the lamellae into fragments that produce the common necking (yielding) and elongation characteristics of a conventional uniaxial tensile test. However, when a low stress is applied (Figure 7.2), the linking chains have time slowly to disentangle themselves so that separation of lamellae occurs, generating a smooth break in comparison to the previous yield / elongation ductile break.

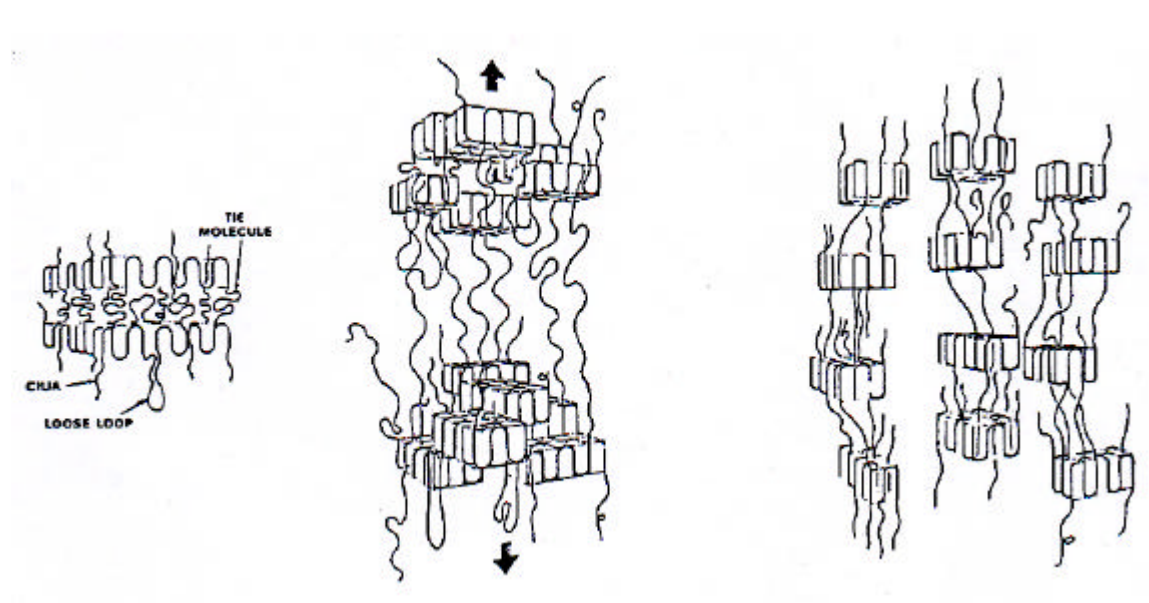


Figure 7.1 Lamellar structure of HDPE and how it yields (Lustiger and Rosenberg, 1989).

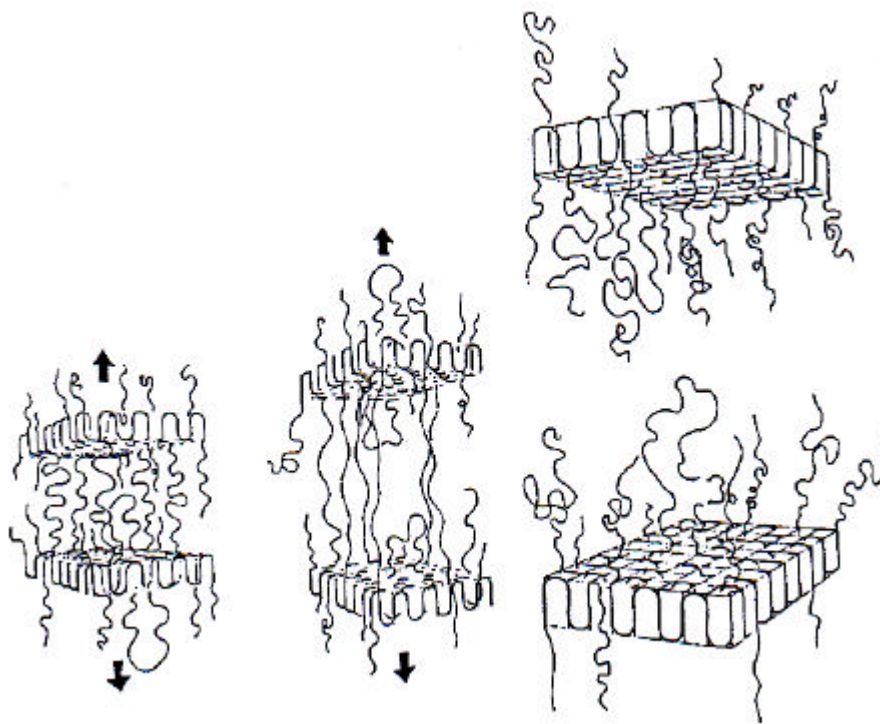


Figure 7.2 Stress cracking at low applied stress (Lustiger and Rosenberg, 1989)

To be effective, tie molecules need to be long enough to bridge the inter-lamellae distance when randomly coiled. This distance is normally taken as twice the lamellae thickness plus the inter-lamellae (amorphous) thickness (Wright, 1996). The density of tie molecules and resistance to stress cracking increases with increasing molecular weight, and is influenced by both the degree of crystallinity and crystal size.

The post-yield tensile performance of polyethylene has a strong influence on stress crack resistance. Types that exhibit strain hardening after yielding (such as LLDPE) offer the better resistance. Such materials have higher molecular weights and stronger craze fibrils (Wright, 1996). Thus, a decreasing Melt Index (MI) is indicative of improving stress crack resistance. Another important influence is the presence of side chain-branches. These suppress crystallisation, decrease the lamellae thickness and increase resistance to dis-entanglement of the tie molecules. Copolymerisation with alkenes is, for these reasons, particularly effective as a means of enhancing stress crack resistance and has been used to improve this property in modern HDPE geomembranes.

In the field, stress cracks typically occur along the edge of extrusion seams in the lower sheet, along the peaks of wrinkles, or as “star” cracking at protruding stones. The cracks are usually quite short, perhaps up to 50mm long. Long cracks are often made up of many individual short cracks that link together to form the long one. Eventually, the extent of the cracking could lead to excessive leakage through the HDPE liner and the end of the service life of the geomembrane as an effective hydraulic barrier. The single unbranched cracks grow slowly by “slow crack growth” (SCG). If the cracks become of a critical size and reach a critical growth rate they can propagate very rapidly and branch many times to produce the appearance of shattering, (Peggs and Carlson, 1990). This type of propagation is termed “rapid crack propagation” (RCP). RCP is usually associated with exposed HDPE geomembranes in sub-zero temperatures but it is

believed that it will not occur without a precursor SCG crack. No cases of RCP have been reported in landfills where the geomembrane liner is confined below solid waste. To achieve a long service life for an HDPE geomembrane liner, a primary objective is to minimise the possibility of SCG occurring.

Elevated temperatures accelerate the process of stress cracking. Stress cracking can also be accelerated by the simultaneous action of stress and contact with specific fluids and is called environmental stress cracking. Typical stress cracking agents are detergents, alcohols (e.g. methanol, ethanol, and propanol), acids and chlorinated solvents. Stress cracking agents are understood to cause reductions in yield strength locally within micro-yielded or stress dilated zones, leading to premature embrittlement. The threshold concentrations of chemicals that cause stress cracking in HDPE geomembranes are not well established. While some of the components of leachate at high concentrations may individually accelerate stress cracking in HDPE, it is unlikely that they have a significant effect at the concentrations present in MSW leachates. Higher strength leachates in some hazardous waste sites or particular liquids in lagoons may have an effect on the stress cracking resistance of HDPE geomembranes.

7.2 Testing

The procedure for testing an HDPE geomembrane material to determine its fundamental resistance to stress cracking is ASTM D5397 *Standard test method for evaluation of stress Crack resistance of polyolefin geomembranes using notched constant tensile load test*. This test uses a small 'dog bone' specimen similar to a uniaxial tensile test specimen. The specimen contains a notch in one face (not the edge) 20% through the thickness of the geomembrane. It is subjected to a constant tensile load at 50°C in a surfactant, both of which accelerate the failure but without changing the fracture mechanism. The notch generates plane strain conditions at its root, similar to the conditions that occur in the field where the material cannot contract in a direction normal to the direction of the principle stress to produce the yielding and elongation that occur in a tensile test specimen (plane stress). Tests are performed at several loads to generate a stress rupture curve similar to one of those shown in Figure 7.3. These curves were generated by testing five different commercially available geomembranes in 1992 (Hsuan *et al.*, 1993).

Developing a stress rupture curve requires the testing of numerous samples and is not suitable for material specification or quality assurance purposes. The Single-Point Notched Constant Tensile Load Test (SP-NCTL) as described in the appendix of ASTM D5397 is used instead. In this test, only one tensile load of 30% of yield stress is carried out, with the test results expressed in terms of failure time in hours. The GRI test method GM13 specifies a minimum requirement of 300 hours (recently revised from 200 hours).

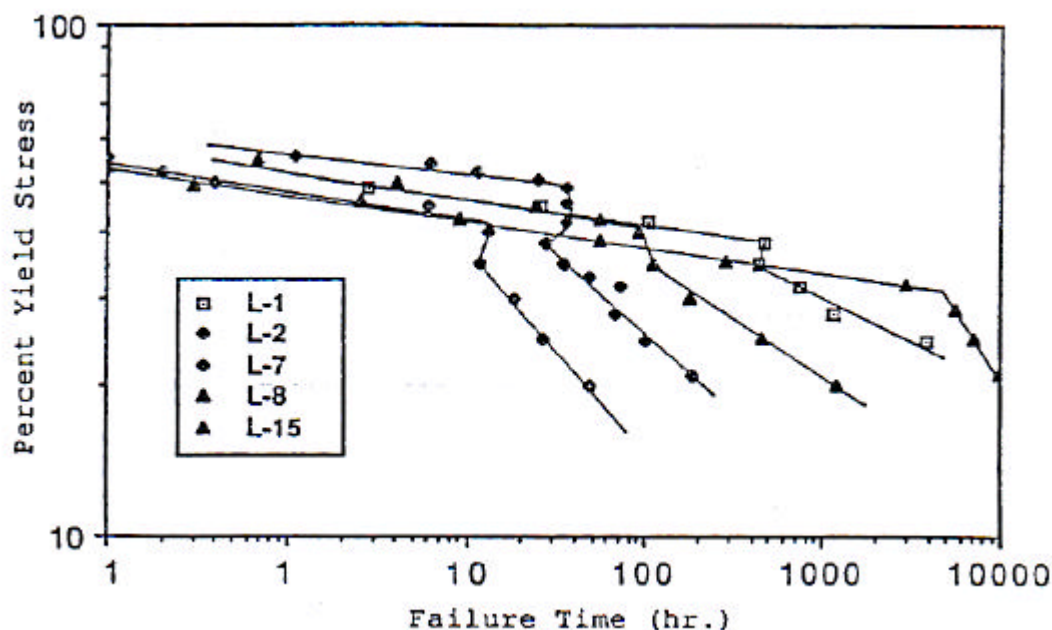


Figure 7.3 HDPE geomembrane stress rupture curves (Hsuan *et al.*, 1993)

The NCTL (and SP-NCTL) tests are only suitable for smooth sheet. On textured sheet, the test must be run on specimens from smooth edges of textured rolls or from smooth sheets made from the same formulation as the textured sheet. However, these tests do not take account of the effects of the texturing. An alternative test for textured materials is the test procedure developed by BAM in Germany (BAM, 1999). The BAM criterion for its test is for failure to occur in more than 700 hours.

Above the “knee” in the curve in Figure 7.3 the failure is ductile, below the knee it is quasi-brittle, with essentially no macro-ductility. The point at which the change in slope occurs (the knee) is termed the ductile / brittle transition point. Below the knee the material fails by stress cracking, hence the ductile segment cannot be extrapolated to determine a maximum service stress for a required service lifetime. There is a factor of about 1000 difference between ductile / brittle transition times of these five geomembranes. In all other respects these five geomembranes would have essentially the same mechanical properties (yield, break, puncture, tear). Only a measure of stress crack resistance differentiates the long-term mechanical durability of the different HDPE geomembranes. Hsuan *et al.* (1993) also showed that the ASTM D1693 bent strip environmental stress cracking resistance test does not show these differences. This test should no longer be performed on HDPE geomembranes.

Figure 7.3 shows a two-stage stress rupture curve. Durability research by Hoechst in Germany (Hessel, 1990) identified a third stage (Figure 7.4) in which the curves for one material at different test temperatures showed a second knee followed by an even steeper, almost vertical, slope. The second knee was defined as the stage at which all antioxidant stabiliser has been consumed and any applied stress will cause cracking. As the stress crack sensitivity has markedly increased, it appears likely that the induction stage has also passed and oxidation has commenced.

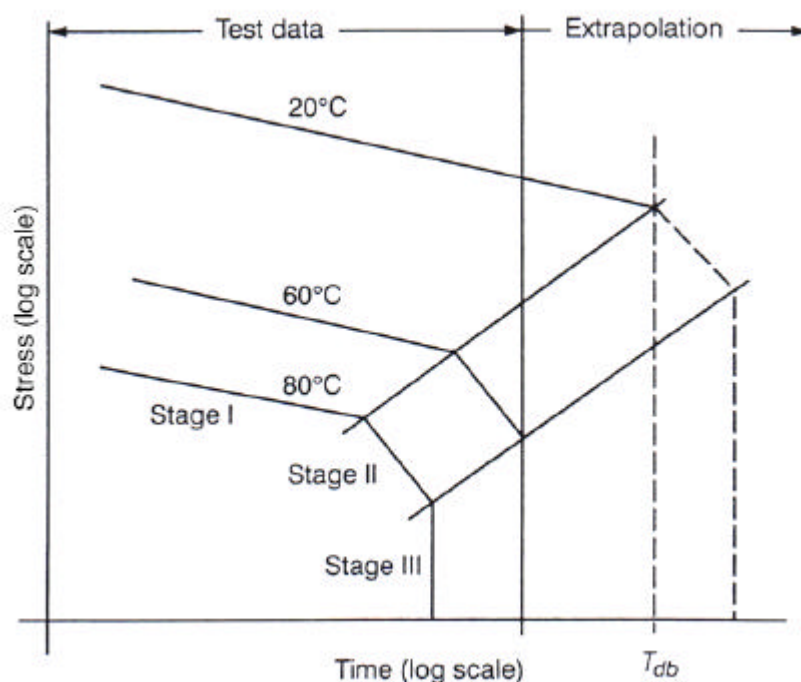


Figure 7.4 Three-stage stress rupture curve (from Hessel, 1990)

At higher stresses close to the yield point, the material fails in a ductile mode before oxidation occurs. At intermediate stresses, premature (compared to ductile region extrapolation) brittle break will occur before oxidation occurs, this being the stress cracking region. However, brittle fracture is even more premature at lower stresses when the antioxidants are fully depleted and oxidation occurs before the extrapolated stress rupture curve is reached. Therefore, in a stressed area there is a competition between the oxidative degradation process and the initiation of stress cracking as to which causes cracking first.

While stress cracking resulting from oxidation is termed “premature”, it would still take a long time to occur, but it is premature compared to extrapolations of failure times at the higher stresses. The three-stage model illustrates that when a geomembrane is under tensile stress or has shear stresses imposed on the surface (e.g. in textured geomembranes) at the same time as oxidation is occurring, the kinetics of degradation are more complex than the simple models used in laboratory studies where the samples are not subjected to tensile or shear stresses. Although stress cracking is normally associated with HDPE geomembranes, it will also occur readily in LLDPE under stress once oxidation commences (which will occur earlier than with HDPE, other factors being equal).

Localised concentration of stress due to local geometrical features (cuts, welds, wrinkles, etc.) will not only increase stresses but also promotes micro-yielding which is known to be the activation process for both craze initiation and crack growth. Stress cracks are initiated at a craze, which opens and finally propagates through the geomembrane. Oxidation within a continuously propagating and opening crack tip will further accelerate the crack growth rate. If oxidation occurs on the surface of a geomembrane during antioxidant depletion (as discussed in Sections 4.4 and 5.5.6), it would cause embrittlement of the surface layer so that surface stress cracks may appear. From the research by Karlsson *et al.* (1992), it appears likely that antioxidant depletion will occur more rapidly on exposed surfaces than the central core of the geomembrane. Oxidation may then occur on the surface while the bulk of the geomembrane is still in the stage of antioxidant depletion, allowing crazing and shallow stress cracks to form. The

likelihood of crack propagation into the bulk of the geomembrane would be determined by the fracture toughness of the bulk material, i.e. its ability to absorb energy, blunt the crack tip and prevent crack propagation.

The significance of the rates of initiation of stress cracks on the surface of a geomembrane followed by crack propagation into the body of the geomembrane was shown by Cadwallader (2001). He found that co-extruded textured material made with a surface layer of low stress cracking resistance (SCR) (apparently recycled) polymer would cause the accelerated cracking of core material with otherwise high stress crack resistance. Thus a core material that had a SP-NCTL stress cracking resistance of over 1000 hours failed in 324 hours in a BAM stress crack test (i.e. it failed the BAM criterion) when co-extruded with a textured surface layer made with inferior quality resin. The cracks were easily initiated in the textured surface layer but did not slow down when they met the core layer. Thus it is easier for a crack to propagate into a core layer than it is for a crack to initiate and propagate within that material alone. It was concluded that surface texturing might reduce the SCR of the basic smooth geomembrane.

7.4 Factors that influence stress cracking

Stresses can be established in geomembrane liners from numerous factors including:

- wrinkles and folds in the liner
- stresses near the base of leachate wells or under leachate pipework
- protruding stones under the liner
- inadequate protection from drainage gravel
- differential settlements in the sub-grade
- down-drag from settling waste
- restraining movement of cover materials on slopes
- residual stresses in welds
- bridging of the liner at small radius corners
- thermal (contraction) stresses where the liner cools down after installation at high temperatures.

Defects in the liner will lead to areas of stress concentration such as at scratches, gouges and cuts as well in weld areas from the weld geometry, excessive preparation for extrusion welds and overheating. The stress crack resistance in welded areas can be reduced by overheating during welding which increases the crystallinity of the adjacent geomembrane material and consumes antioxidants, both of which are detrimental to stress crack resistance. The stress crack resistance of the welding rod used in fillet extrusion welds has often been found to be poor in relation to that of the sheets being welded together (Thomas, 1998). He also confirmed that extrusion welds are more likely to fail by stress cracking than fusion seams.

The use of double textured liners on side slopes where there is a higher shear resistance on the top surface than on the underside of the geomembrane leads to the geomembrane becoming a load bearing member of the system due to the induced shear stress. As indicated by Thomas and Woods-De Schepper (1993), the presence of the surface texture can cause a reduction in the SCR of the geomembrane itself, to different degrees depending on the type of texturing.

Thomas *et al.* (1995) studied the effects of different degrees of blemish-inducing wedge and wheel combinations from the equipment used in hot wedge welding. Although geomembranes with high SP-NCTL test values substantially increased stress crack failure times for both high and

low surface blemishing wedge / wheel combinations, more dramatic improvements were obtained in stress crack failure times simply by reducing the blemishing of the surface.

Higher temperatures at the liner will reduce the time to the brittle failure condition (Figure 7.4).

7.5 Stress relaxation

The kinetics of stress crack initiation and propagation increase at elevated temperatures as shown schematically in Figure 7.4. However, stress relaxation also increases as temperature increases resulting in a permanent race between stress cracking and stress relaxation as to which will prevail. If the induced stresses can be sufficiently reduced before cracking is initiated, cracking will not occur. Also to be factored into this process is oxidation of the geomembrane. Once oxidation starts, only small stresses will be sufficient to cause fracture.

While the benefits of stress relaxation are apparent it is not a topic that has been thoroughly studied for geomembranes. Soong *et al.* (1994) investigated stress relaxation in a 1.5mm thick HDPE geomembrane with initial stresses of 40, 50, and 60% of yield stress (at test temperature) and initial strains of 1, 3, and 5% at temperatures between -10° and 70°C . These were quasi-biaxial tensile tests using 100mm wide by 50mm long “wide width” tensile specimens. Initial loading was done quite quickly to minimise stress relaxation on loading. Whatever the starting conditions, there was a trend to a very narrow range of final, but still significant stress, after about 100 days. The relaxation modulus curves (stress / strain as a function of time) for a given starting condition could be superimposed into a master curve for a given relaxation temperature, as shown in Figure 7.5 for an initial 3% strain and a temperature of 10°C .

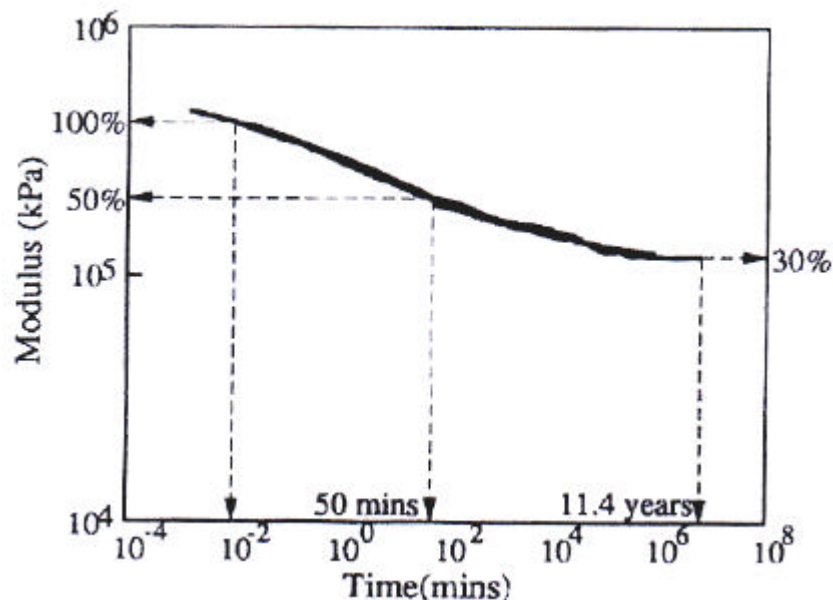


Figure 7.5 Master stress relaxation curve for 3% strain at 10°C (Soong *et al.*, 1994)

In this case, 50% of the applied stress was removed by relaxation after 50 minutes with final equilibrium being achieved at about 30% of applied stress after 11.4 years. At higher temperatures as would apply in landfills, the stress relaxes more quickly. At 30°C , the time to reach 50% stress relaxation was 30 minutes and the residual stress at equilibrium was 35% after only 0.8 years. Note that the strain in the test was applied far more quickly than will generally

occur in field conditions, so in the landfill significant stress relaxation will occur during deformation. Soong *et al.* (1994) stated:

“Trial tests were performed initially to determine the suitable loading rate. The results suggested a rate of 12.7mm / min as being appropriate..... At slower rates a very significant amount of stress relaxation occurred during the loading process.....”

Also, note that Soong *et al.* (1994) concluded:

“..... other HDPE geomembranes will undoubtedly respond differently than the HDPE studied”

Further testing of a range of geomembranes will be necessary to develop a broader understanding of the stress relaxation behaviour of HDPE geomembranes.

These stress relaxation rates compare well with those generated by Soong and Koerner (1997) for stress relaxation in wrinkles in HDPE geomembranes under a uniform vertical loading. After 1000 hours at temperatures of 23, 42, and 55°C they found stresses relaxed between 60% and 78% leaving residual stresses of between 1% and 22% of the yield stress. Recollect that stress crack testing is done at 30% of the yield stress, not much higher than some of these residual stresses.

However, these tests were done under semi-confined conditions (waves raised off a flat support surface) while the Soong *et al.* (1994) tests were done under unconfined conditions. Under semi-confined conditions the residual stresses were lower than for unconfined specimens, possibly as a result of the stress relaxation occurring during loading. Under fully confined conditions as would apply on a basal liner, the residual stresses would probably be even lower.

These reported studies indicate that where applied stresses on the liner are not maintained, then stress relaxation will reduce the stress substantially but there is likely to remain a significant residual stress which could lead eventually to the development of stress cracks. In cases of constant stress where relaxation cannot occur, then stress cracking should be expected to occur. Flaws in the geomembrane will lead to concentrations of stress which can be several times higher than the stress in the surrounding liner, providing the conditions for premature stress cracking.

7.6 Implications of stress cracking to service life

Even when the antioxidants in HDPE geomembranes have been fully depleted and oxidation commences, the geomembranes remain in place as effective hydraulic barriers unless physically damaged or they develop holes. Setting aside physical damage, holes through the liner should only develop or enlarge as a result of stress cracking unless stresses are so high as to cause ductile tensile failure. Oxidative degradation embrittles HDPE geomembranes making them much more susceptible to stress cracking so that at locations in states of even low tensile or shear stress, it is likely that stress cracks will occur.

The extent of stress cracking will largely depend on the SCR of the geomembrane, the states of local and global stress over the liner, the temperature at the liner and the onset of oxidative degradation. Where tensile and shear stresses in the liner can be avoided, the liner should

remain intact, for practical considerations, indefinitely. Where the liner is under stress, whether from residual stresses or in a state of constant stress, cracking will occur leading to leachate and/or gas leakage outwards, and inward water leakage. Once oxidation commences, additional stress cracks will occur at the locations where the liner stresses had been too low already to have triggered cracking. The service life of the geomembrane liner will end once excessive leakage for that site occurs. If physical damage can be limited to acceptable levels, then the service life of the geomembrane liner as an effective hydraulic barrier depends on the development of stress cracks leading to excessive leakage.

7.7 Avoiding stress cracking

The development of stress cracks can be avoided or, at least, inhibited by good design and installation practices, and avoiding stress-inducing activities during the disposal of the first layer of waste. Important factors in avoiding stress cracking are summarised in Box 7.1.

Box 7.1 Important factors in avoiding stress cracking

- Specify a geomembrane with an adequate stress cracking resistance. The latest GRI GM13 Specification (revision 6) requires in the ASTM D5397 Appendix test a single point (30% room temperature yield stress) test break time exceeding 300 hr. A ductile / brittle transition time consistent with the criticality of the containment function at the particular site could be specified, up to a maximum, presently, of about 1000 hr.
- The OIT characteristics of the geomembrane should meet the GRI GM13 Specification as a minimum, or the BAM Specification, Table 3, Part I.
- Note that stress cracking will readily occur following the onset of oxidation in LLDPE and other polyethylenes with high stress crack resistance.
- Design the liner for minimum stress. Downdrag and strain incompatibility on slopes, settlements of the subgrade, corners and changes in slope, leachate wells and if present, at liner penetrations (e.g. gas wells through caps) are all likely locations or causes of liner stresses.
- Avoid the use of double textured liners where long-term shear stresses may be imposed on the liner, especially on side slopes.
- Allowance by the designer to accommodate interim stresses between installation and operating conditions.
- Specify adequate damage protection for the geomembrane that will have a service life comparable to that of the geomembrane.
- Specify geomembrane placement procedures that reduce the likelihood of wrinkles (waves) (Averesch and Schicketanz, 2000) but do not impose long-term contraction stresses in the liner.
- Avoid the use of fillet extrusion welds and ensure that the wedge welding equipment causes minimal wedge/wheel combination blemishing on the geomembrane.
- Minimise the exposure of the geomembrane to UV radiation to avoid the potential for microscopic crazing of the geomembrane surface.
- High standard construction quality assurance (CQA) provided by experienced and knowledgeable engineers and liner construction by competent, qualified installers to ensure good seams, smooth subgrade without stones, intimate contact with the subgrade, lack of wrinkles and to avoid scratches, cuts and other damage.
- Careful placement of cover layers to avoid causing and trapping wrinkles and folds and minimising movement on cover layers to avoid creating stresses in the liner.

8 PHYSICAL DAMAGE MECHANISMS

8.1 Damage before waste placement

Geomembrane liners, being relatively delicate construction materials, are susceptible to damage at all stages from manufacture to the end of their service life. Table 8.1 (McQuade and Needham, 1999) gives some examples of types of holes or other defects that a geomembrane can suffer and the possible causes. Manufacturing quality control is a fundamental component of the manufacturing process with the objective of ensuring that the geomembrane meets the manufacturer's stated product specification. Delivery and handling of the geomembrane rolls prior to installation can lead to damage, typically scuffing, tears and cuts. Effective construction quality assurance (CQA) should identify such defects prior to placement of the liner and, by laboratory conformance tests, should demonstrate prior to installation that the geomembrane liner meets the required material parameters.

Table 8.1 Typical defects and possible causes (McQuade and Needham, 1999)

Stage	Type of defect	Possible cause / concern
Manufacture	Pinholes, excessive thickness changes, poor stress crack resistance	Unusual now for procedures with good quality control. Poor resin
Delivery	Scuffing, cuts, brittle cracks, tears, punctures	Unloading with unsuitable plant or lifting equipment. Impact. Poorly prepared storage areas
Placement	Scratches, cuts, holes, tears, crimps	Dragging sheet along ground, trimming of panels, rough subgrade, use of equipment on top of sheet without protection layer, wind damage, large wrinkles, folds, damage by lifting bars
Welding	Cuts, overheating, scoring, poor adhesion, crimping	Careless edge trimming, welding speed or temperature incorrect, excessive grinding, dirt or damp in weld area, excessive roller pressure
Cover placement	Tears, cuts and scratches, holes, stress in membrane	Action of earthmoving plant, insufficient cover during placement, careless probing of cover depth. Contraction of sheet due to ambient temperature reduction
Post-installation	Holes, tearing, slits, cracks	Puncture from drainage materials, puncture by items of deposited waste, opening of partial depth cuts, pulling apart of poor quality welds, down-drag stresses caused by settling waste, differential settlement in the base

This project concentrates on damage to geomembrane liners occurring after installation of the lining system (the sixth stage in Table 8.1) but often these may arise as a result of defects caused during liner installation. Many of the publications describing leak location surveys discuss holes identified on liner completion as well as after waste placement has commenced. Thus, it is instructive to consider damage (and the causes) identified during both of these periods.

Most damage to geomembranes, causing holes or non-penetrative defects, has been found to occur in the installation and seaming of the liner, and as a result of the placement of the overlying drainage or cover material. Analysis of the causes of geomembrane liner damage has now been extensively reported from the results of electrical leak location (ELL) surveys, (e.g. Laine (1991); Nosko *et al.* (1996); McQuade and Needham (1999); Rollin *et al.* (1999); and Nosko and Touze-Foltz (2000)). The ELL surveys have been undertaken using mobile and fixed ELL systems. The techniques of ELL testing of geomembranes have been reviewed in an Environment Agency research project (Bishop, 2002). Mobile ELL surveys are carried out on completion of liner construction including placement of the cover material, but are unable to detect holes after waste disposal commences. Surveys using fixed (or “permanent”) ELL systems can be performed at any time after the system installation, continuing well into the operational and post-closure phases of the landfill. The general configuration of the mobile system is shown in Figure 8.1 and a schematic drawing of the fixed system is shown in Figure 8.2.

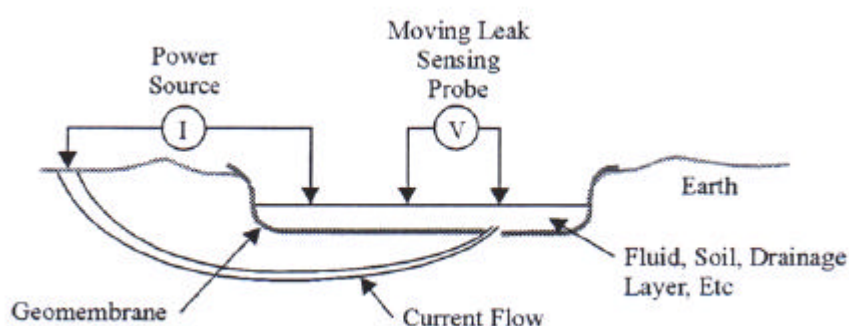


Figure 8.1 General configuration of the electrical leak location technique for landfills and fluid impoundments (Bishop, 2002)

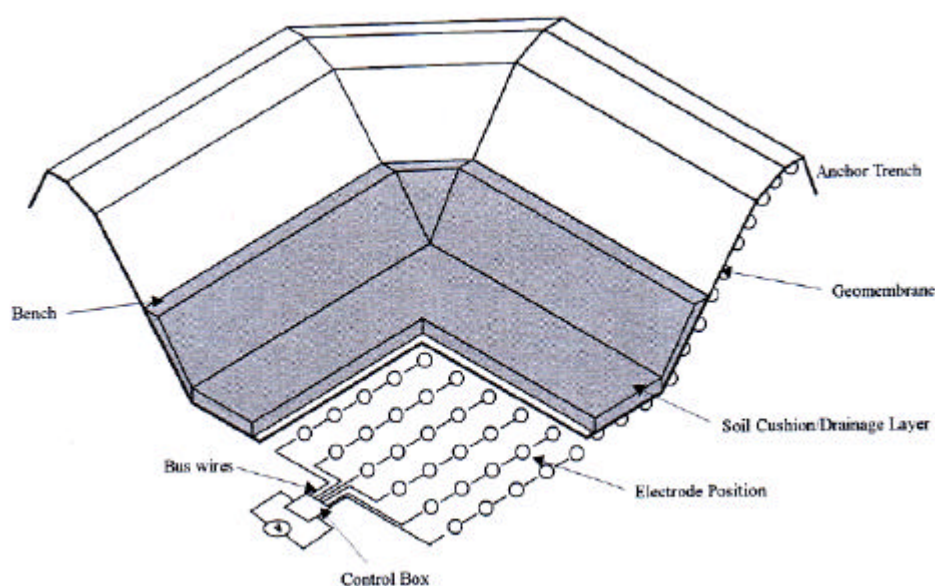


Figure 8.2 Schematic drawing of a permanent leak detection system beneath a landfill (Bishop, 2002)

The ELL surveys have primarily been carried out on sites in numerous European countries, including the UK, as well as the USA and Canada. Inevitably, there are variations in installation and CQA standards in different countries and there has generally been an improvement in these factors over the last 10 years, which has been reflected in the results of the surveys.

The survey by Nosko *et al.* (1996) on the detection of localised defects in geomembranes indicated that:

- 24% of the holes were caused during installation of the FML.
- 73% of the holes were due to mechanical damage occurring during placement of the cover soils.
- Only 2% occurred during the post-construction phase.
- 1% were test holes

Of the 24% caused during liner installation, the breakdown of the causes were found to be:

- 61% at welds mainly from extrusion welded T-joints and Y-joints, or extrusion welds at other locations.
- 18% from overheating or melting faults.
- 17% from stone penetration during the deployment of the sheet.
- 4% caused by cuts.

Of the 73% of the total number of holes occurring during cover placement, this was sub-divided into the following mechanisms:

- 68% punctured by stones
- 16% punctured by heavy machinery
- 16% punctured by stakes.

Nosko *et al.* (1996) report that stone puncture was caused by sharp or large stones, incorrect stone spreading techniques and the lack of a geotextile between the geomembrane and the cover material. Incorrect spreading can lead to the pulling apart of overlapped protection geotextiles, exposing the geomembrane to direct contact with the cover soil. Non-penetrating damage may be caused at construction stage by the cover or drainage material, leading to failures in the medium term. Lack of attention by the plant operator or incorrect level controls can lead to very large tears by construction plant. Stakes used to measure cover thickness, to earth electrical equipment or to secure items to the cover surface also lead to punctures. Apart from noting that the hole size varied from 10cm² to several metres, no analysis of the hole size related to the cause was published.

A more recent survey by Nosko and Touze-Foltz (2000) summarised the results of surveys at more than 300 sites in 16 countries covering more than 3,250,000m² of geomembrane liner. The results of the survey are shown in Tables 8.2 and 8.3. The study examined three criteria being the locations, sizes and causes of the damage. They summarised the cause of damage in relation to the hole size, as shown in Table 8.2.

Table 8.2 Cause of damage versus size of hole (adapted from Nosko & Touze-Foltz, 2000)

Size of damage (cm ²)	Stone	%	Heavy equipment	%	Welds	%	Cuts	%	Worker directly	%	Total	%
< 0.5	332	11.1	-	-	115	43.4	5	8.5	-	-	452	10.8
0.5–2.0	1720	57.6	41	6.3	105	39.6	36	61.0	195	84.4	2097	50.0
2.0-10	843	28.2	117	17.9	30	11.3	18	30.5	36	15.6	1044	24.9
> 10	90	3.0	496	75.8	15	5.7	-	-	-	-	601	14.3
Amount	2985		654		265		59		231		4194	
Total	71.17%		15.59%		6.32%		1.41%		5.51%			

Of the total 4194 holes detected, they identified the location of the damage and the cause of the holes in each of these locations (Table 8.3).

Table 8.3 Locations and causes of damage (adapted from Nosko and Touze-Foltz, 2000)

Location	Frequency of defect by location	Frequency of cause at each location					Total
		Cause of Failure					
		Stones	Heavy equipment	Worker	Cuts	Welds	
Flat floor	78%	81%	13.2%	4%	1%	0.8%	100%
Corner, edge, etc	9%	59.2%	18.9%	3.5%	0.9%	17.5%	100%
Under drainage pipe	4%	30.3%	14.3%	14.5%	13.7%	27.2%	100%
Pipe penetration	2%	0%	0%	8.5%	0.6%	90.9%	100%
Other	7%	20.6%	43.4%	19.3%	0%	16.7%	100%
Total	100%						

The mechanisms and relative importance of physical damage resulting in fully penetrating holes as identified by the ELL surveys can be seen from inspection of Tables 8.2 and 8.3. The predominant size of stone-related damage was typically 0.5 – 2.0cm². Damage resulting from heavy equipment resulted in holes larger than 10cm², while holes in faulty welds were typically found to be less than 0.5cm² in size. Holes caused by cuts or direct worker damage were typically found to be 0.5 – 2.0cm².

McQuade and Needham (1999) reviewed the results of 111 mobile ELL surveys from sites in the UK and, where available, identified the range of hole sizes obtained in each survey and the causes of the holes. The larger holes (> 100mm long) were generally the result of damage by plant during placement of the cover materials. Relatively few of the holes were found in seams.

Phaneuf and Peggs (2001) tabulated the hole size and type found in a 640,000m² uncovered liner (Table 8.4). As it was uncovered, the recorded holes do not include damage incurred during cover placement. It is seen that punctures were the most frequent type of hole but the largest holes were caused by lack of bond at seams and by scrapes.

Table 8.4 Holes found in 640,000m² uncovered liner (Phaneuf and Peggs, 2001)

Size (mm)	Punctures	Gouges	Cuts	Tears	Burns	Scrapes	Lack of bond	Seam
< 1	10	1	2			1	1	1
2-10	28			1	8	7	4	1
11-50	7	11	7	2		3	2	1
51-100		1	3	1		1		3
101-500	1		1			1		1
501-1m							1	2
> 1m						2	1	1
Unknown	4	3		1		2	1	2
Total	50	16	13	5	8	17	10	12
% total	38.2	12.2	9.9	3.8	6.1	13	7.6	9.2

Rollin *et al.* (1999) reported the results of ELL surveys at 11 sites in France and Canada constructed between 1994 and 1998 for landfills and liquid impoundments, covering a total of 241,000m². The summary of the number and type of leaks found is shown in Table 8.5. Sites 7 and 8 were lined with a prefabricated bituminous geomembrane, Site 9 with a polypropylene geomembrane and the remainder by HDPE geomembranes.

Table 8.5 Number and type of located leaks on uncovered geomembranes (Rollin *et al.*, 1999)

Site ID	Prospected area (m ²)	Number of detected leaks				Leak density (leaks per hectare)
		Holes	Knife cuts	Seam failures	TOTAL	
1	18 500	0	0	5	5	2.70
2	14 926	4	0	2	6	4.02
3	13 480	1	1	1	3	2.23
4	11 652	1	2	2	5	4.29
5	8 200	0	0	0	0	0.00
6	9 284	0	1	0	1	1.08
7	67 100	3	0	2	5	0.75
8	66 150	1	1	7	9	1.36
9	11 460	2	2	2	6	5.24
10	18 135	0	3	3	6	3.31
11	2 021	0	0	3	3	14.84
TOTAL	240 908	12	10	27	49	
		24.5%	20.4%	55.1%		Average 2.03

All of their sites were surveyed using ELL techniques on uncovered geomembranes, so did not include damage caused during cover placement, and the high relative percentage of seam failures (55%) has to be seen with this in mind. It was noted that all of the holes in the seams were in fillet extrusion welds on the HDPE geomembrane after they had already been tested for leaks by vacuum box, raising concerns on the effectiveness of the vacuum box testing.

The data from the published papers are weighted towards mobile ELL surveys which identified holes and their causes on completion of construction. These holes would then be uncovered, repaired and re-tested so that they do not remain throughout the service life of the geomembrane liner. However, the repairs may themselves be stress concentrating features susceptible to the development of future stress cracks especially where patches with fillet extrusion welds have been used.

The surveys reveal the causes of holes in geomembrane liners occurring before waste disposal commences and identify the mechanisms of physical damage. Where ELL surveys are not conducted, then it must be expected that holes of the types identified will be present in these liners. In addition, ELL surveys can only identify fully penetrating holes. Partially penetrating damage in a liner from the same causes may develop into holes after the survey at some time during the life of the geomembrane liner, by stress cracking or tensile failure. The strain at which HDPE geomembrane reaches yield failure is much reduced from the yield strain of intact material by partial depth cuts (Giroud *et al.*, 1993). For example, a half-depth cut will reduce the yield strain to only 16% of the intact yield strain.

The frequency of holes identified by ELL surveys at the end of construction that is of most relevance to UK practice is presented by McQuade and Needham (1999). They found that the range in frequency of holes was large, from zero to the equivalent of 120 holes/ha. Of the 111 surveys undertaken, 48% found no holes at all, while 16% found in excess of 10 holes/ha. The 111 surveys covered approximately 790,000m² of geomembrane liner and detected 331 holes. While this gives an overall frequency of 4.2 holes/ha, the median frequency was 0.7 holes/ha which they considered to be more representative of the standard that is readily achievable with competent installation teams and thorough CQA. Where they knew that thorough CQA had been conducted (14 of the 111 surveys), an improved hole frequency of zero to 5.7 holes/ha was apparent with a mean value of 0.8 holes/ha and a median frequency of zero.

The data from the ELL surveys illustrate where efforts should be concentrated to minimise geomembrane liner construction defects, both fully penetrating and latent damage. The activities warranting particular care and attention are as follows:

- placement of the protection and drainage materials on top of the geomembrane. [CQA should be continued throughout this activity];
- movement of plant on top of protection layers should be minimised;
- the quality of the subgrade surface (e.g. of the compacted clay liner);
- adequate protection should be provided to the geomembrane commensurate with the grading and angularity of drainage gravel and the future waste load;
- fillet extrusion welding should be minimised to the extent practical, and thorough non-destructive testing undertaken on 100% of all those welds;
- installation and CQA personnel need to be well trained to recognised standards;
- geomembranes should be installed in suitable weather conditions.

8.2 Mechanisms of physical damage after liner installation

8.2.1 Data from fixed ELL surveys

Nosko *et al.* (1996) is the only published paper that specifically identifies holes found by fixed ELL surveys after commencement of waste disposal. As noted above, they report that only 2% of the total number of holes identified occurred in the post-construction phase during waste filling. The causes were found to be:

- 67% were accidental damage by trucks/compactors, etc.
- 31% occurred from the installation of pipes, drainage systems, sumps, haul road access, etc.
- 2% from weather damage or other unplanned calamities e.g. fire.

No records of holes caused by geomembrane material deterioration were recorded.

Data has been received for this project from ELL survey company A on their fixed ELL surveys conducted on 17 commercial sites with a total liner area of about 800,000m². The countries where the sites were located are not available. The maximum period of monitoring for leaks after liner installation was 6 years. The summary data are given in Table 8.6.

Table 8.6 Summary details of fixed ELL surveys

project	start	years in operation	surface area (m ²)	leaks during installation	leaks during operation
A	1998	5	13000	0	1
B	1997	6	30000	1	1
C	1999	4	55000	0	0
D	1998	5	140000	0	1
E	1998	5	6550	0	0
F	1999	4	500	0	0
G	2000	3	42000	1	2
H	2000	3	88000	1	1
I	2001	2	94000	1	1
J	2001	2	1000	0	0
K	2002	1	30000	1	0
L	2002	1	55000	0	0
M	2002	1	94600	0	0
N	1998	5	8500	7	2
O	2000	3	36000	4	2
P	2000	3	20000	4	4
Q	2000	3	80000	2	1
TOTALS					
A...M			649650	5	7
N...Q			144500	17	9
All			794150	22	16
TOTALS				leaks/hectare	
A...M				0.1	0.1
N...Q				1.2	0.6
All				0.3	0.2

A to M : projects with a higher level of CQA procedures

N to Q : projects with a lower level of CQA procedures

Details of the stages or causes of the holes are not available but it was noted that for the holes occurring during the operational phase, the causes related to physical damage and not deterioration in the geomembrane liner material. It is seen that of the total holes detected (38), 42% (16) occurred during the site operation, a much higher proportion than the 2% found by Nosko *et al.* (1996). The clear benefit of higher quality CQA is evident from the much lower frequency of holes being caused when the CQA procedures were of a higher standard (0.1 holes/hectare). This same frequency was recorded both at the end of installation and during site operation, and can be compared to the frequencies of 1.2 and 0.6 holes/hectare respectively for poor CQA procedures.

Results were also provided by ELL survey company B from surveys using fixed ELL systems over the 7-year period 1996 – 2003. The data was obtained from 88 cells and 18 leachate lagoons at 55 landfill sites in Eastern Europe, Belgium and the UK. The total area monitored was approximately 1,022,000m². The total number of fully penetrating defects was 1460, with 1080 (74%) located during the initial leak survey at the end of liner construction and 380 (26%) holes being located in subsequent monitoring surveys. This frequency is intermediate between those reported by Nosko *et al.* (1996) and the records of ELL survey company A. The causes of the post-installation damage were reported as:

- stone puncture (total 298 (78%) of which only 21 were under waste);
- compactor (37 on a side slope or bund, 4 on floor area; 34 in total below waste);
- waste articles (iron bars) punched through the geomembrane (3);
- stakes or rods (2);
- pump (2) – not under waste;
- cracks on wrinkles (2);
- side slope stability – land slide (2);
- stability of construction layers or waste (2);
- stress crack – near the top of the side slope, not under waste, geomembrane in tension (1);
- fire (1);
- netting pylon (1);
- not uncovered (25) so cause not determined (6.6% of 380 holes).

The most common cause of damage was from stone puncture resulting from traffic movement over empty cells, illustrating the vulnerability to damage of geomembrane liners in completed but unused cells.

To augment this information, the detailed results of ELL surveys using fixed systems at six landfills in the UK have been examined. The results of the surveys are given in periodic monitoring reports held on the public record. One of the sites (Sandy Lane) has been monitored since June 1995 and is considered separately below. Of the remaining five sites, their results are included in the summary details from ELL survey company B given above. All of the fixed systems were used initially to check on the presence of holes occurring up to the completion of liner construction. Most of the sites have only undertaken two surveys – an initial survey following placement of the drainage or cover layer and a second, final survey following placement of one layer of waste. Subsequent surveys, although possible, have not been undertaken.

At one of the five sites, only the first survey had been conducted by the time of this project and is not considered further as it does not have post-installation survey information.

Of the four remaining sites, three have had only the two surveys carried out while one site has had regular monitoring surveys over three years. The results of the ELL surveys during the operation phase at these four sites are summarised in Table 8.7. Where known, the hole sizes are given.

Table 8.7 ELL surveys after commencement of waste filling

Site	Area (m ²)	Survey findings
A	29500	1 stone puncture (repaired). Size not recorded.
B (Cells 2A and 2C)	21950	No holes
B (Cells 2B and 2D)	27000	5 holes in one area caused by metal rods each approx. 30mm dia. (repaired)
C (Cell 1)	21600	80 x 80mm hole, unknown cause (repaired)
C (Phase 4, Cell A)	11000	No holes
C (Phase 4, Cell B)	11000	No holes
D (Cell 1)	11860	No holes detected on any of the 8 surveys over 3 years in two cells since operations commenced.

Significant damage was detected in three of the seven cells / areas surveyed but repairs were completed in each instance.

8.2.2 Sandy Lane landfill, Bromsgrove

A fixed ELL system has been installed at this site since 1995. The stated primary aim of the system is to track any leakage from the site but this aim has yet to be tested in practice as there has been no detectable leakage. A secondary and very practical benefit has been the ability to identify holes in the HDPE geomembrane liner.

The ELL system at this site is modular in design enabling additional electrodes to be attached to the system as new cells were built, thereby enabling monitoring of the entire site throughout the construction, landfilling and post-closure stages. The system comprises a grid of electrodes installed just beneath the composite liner (HDPE geomembrane over 300mm BES) providing full coverage across the base and the side slopes. The basic electrode grid uses a 20m spacing, whilst alternate east-west lines have electrodes positioned at 10m spacing. Geophysical measurements are undertaken remotely from the weighbridge. Cell 5 is the latest and final cell of this site. Surveys have been conducted now for almost eight years on a quarterly basis. A diagrammatic section of the location of the geophysical electrode grid is shown on Figure 8.3.

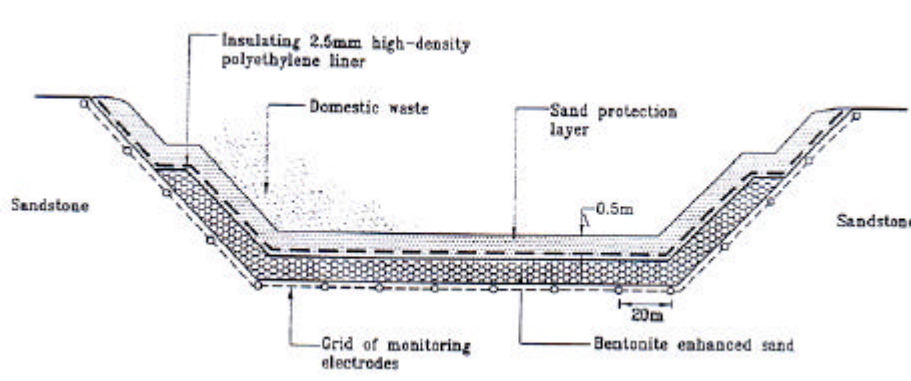


Figure 8.3 Diagrammatic section through the landfill showing the position of the geophysical electrode grid

Features of the fixed ELL system relating to the identification of holes from recorded electrical anomalies include the following:

- a large electrical anomaly in the survey results can mask other, weaker, anomalies that only become apparent following repair of the first anomaly and re-survey;
- an electrical anomaly may actually comprise more than one hole;
- there is no relationship between the magnitude of an electrical anomaly and the size of the hole because the anomaly signal is strongly dependent on the distance of the hole from a sensor in the electrode grid;
- a mobile ELL survey is used to pinpoint the position of a leak identified within a $10 \times 10\text{m}$ area by the fixed system;
- electrical isolation of the cell being monitored is very important to the sensitivity of the survey results. Variable degrees of isolation (e.g. caused by changes in moisture conditions or influence of adjacent cells) can lead to new anomalies being detected although the associated holes existed undetected during earlier surveys;
- knife cuts clearly detected by the fixed system were less easily located by the mobile ELL system.

A typical contour plot of normalised potential difference and associated orthographic surface plots of the survey data are shown on Figures 8.4 and 8.5, illustrating three electrical anomalies.

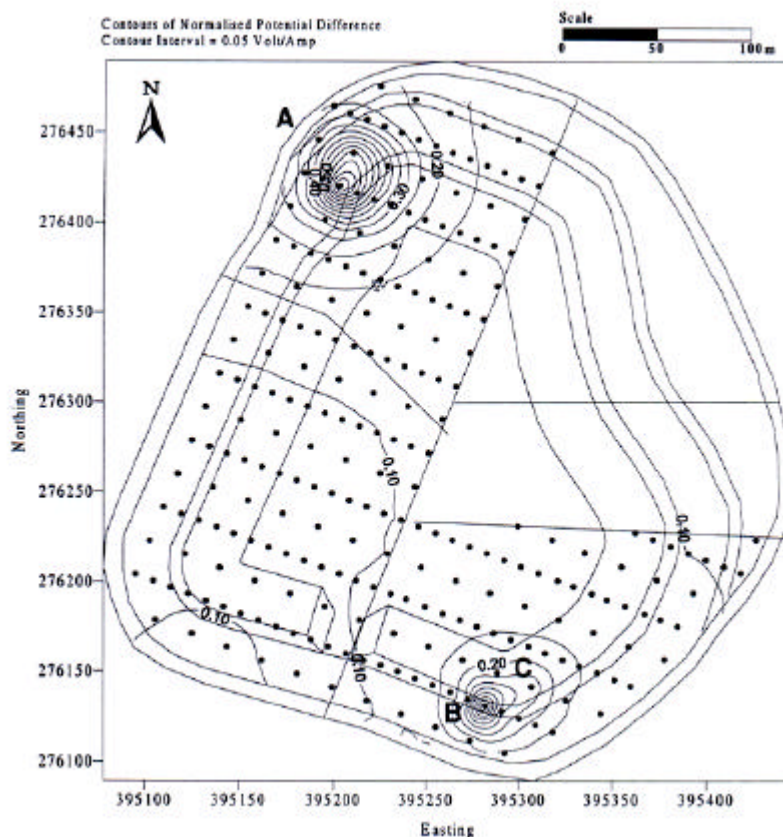


Figure 8.4 Contoured integrity test data

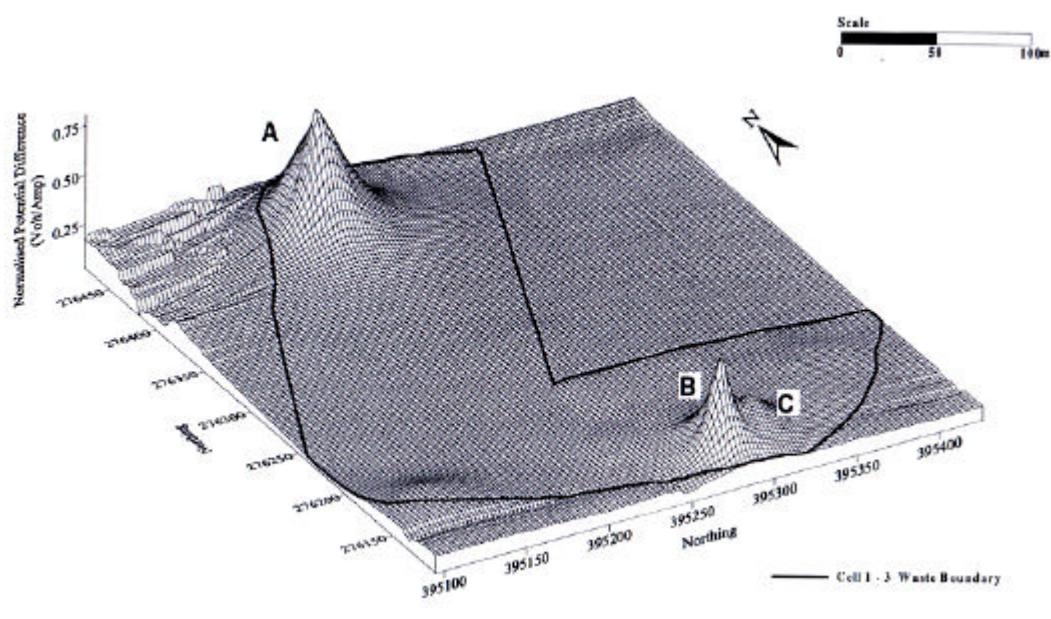


Figure 8.5 Orthographic surface plot of integrity test data

Most holes identified during the ELL surveys have been accessible, enabling repairs to be made. The holes occurring after the liner had been installed and surveyed, and repaired if necessary, are summarised in Table 8.8. While the great majority of these holes were repaired, had a fixed ELL system not been in place, then these holes would not have been detected. A large proportion of the holes were located on slopes or on top of bunds where the presence of a leachate head in the future is less likely. Fully penetrating holes through the geomembrane liner in these locations would not necessarily result in leachate leakage.

Routine monitoring has been undertaken quarterly with additional surveys at the end of cell construction. The majority of holes occurred during the construction of the liner and these were repaired prior to waste disposal. Holes discovered after completion of the liner are summarised in Table 8.8.

Table 8.8 Summary of detected electrical anomalies

1997	
Cell 1	No holes developed since cell commissioned
Cell 2	13 holes detected: 6 cuts, 10 – 75mm long; 1 hole below waste, inaccessible, details unknown; 5 defective welds; 1 tear, 100mm, on rain flap weld due to weight of wet cover sand.
1998/99	
	Summary: A total of 60 holes identified in 5.5ha, including liner construction holes. Six holes repaired. Cell 1 (3 holes), Cell 2 (20 holes) and Cell 3 (17 holes). Only 2 of the 6 unrepaired holes are not on a slope and may be subject to a leachate head. Two of the holes programmed for repair in 2000.
Cell 1	No new holes
Cell 2	One new hole on side slope (inaccessible) plus previous inaccessible hole still present
Cell 3	6 holes post completion. 3 holes were in the side slope berm and 3 holes above this level. Of the 6 holes: <ul style="list-style-type: none"> i). 2 caused by waste material penetrating liner through the 500mm sand layer causing 63cm² and 30cm² holes. ii). 1 caused by 40mm tear on rain flap iii). 3 holes inaccessible due to waste depth, unrepaired at this time.
Cell 4	Completed October 1999. 21 holes had been found at end of construction, all repaired. One anomaly in berm at junction of Cells 1, 3 and 4 being several actual holes, all repaired.
2000	
Cells 1 – 4	Five of the six anomalies previously identified and unrepaired were again recorded. One new anomaly caused by earthing spike near intercell bund – repaired. Three anomalies were repaired leaving 2 anomalies in Cell 2.
2001	
Cell 1	No new anomalies
Cell 2	Two unrepaired anomalies from 1997/98 remain. No new anomalies
Cell 3	No anomalies
Cell 4	Two new anomalies <ul style="list-style-type: none"> i). 5 holes caused first anomaly. 3 caused by landfilling, one knife cut and one was a cluster of 3 small holes at defective weld. Repaired. ii). 2 holes associated with second anomaly, occurred prior to placement of sand layer. Unclear why not previously detected. Repaired.
2002	
Cells 1 – 5	Full system now installed comprising 14 cables, 340 electrodes, and 3 above liner electrodes.
Cells 1 and 2	No new anomalies
Cell 3	Large rips 0.5 – 1.0m and 10 – 12m long on upper bench adjacent to haul road.

The records are not always clear whether the hole occurred in the fallow period between the acceptance of a liner free of electrical anomalies and the commencement of waste placement, or subsequent to waste placement starting. The main findings from the review of the extensive monitoring at the Sandy Lane site are as follows:

- Two holes remain unrepaired in the liner, one being on the basal area, one on a side slope, both estimated as being very small. No detectable leachate leakage has occurred.

- A total of 74 anomalies were detected, with approximately 90 holes being associated with these anomalies over a liner area of 5.5ha. This gives a frequency of 16 holes/ha. Of these, 27% of the anomalies appear to have been detected after completion of the liner (i.e. during the fallow period or after landfilling commenced). Within this figure, some are new anomalies found as the degree of electrical isolation improved but which relate to pre-existing holes.
- The sizes of holes found after liner completion ranged from pinholes in welds to major rips caused by plant movement.
- Over the 1997 – 2002 period, holes were rarely found to develop as a result of stresses imposed by the waste alone, and these occurred soon after waste disposal. Almost all new holes detected after the onset of waste disposal resulted from the early activities of landfilling (e.g. stakes, puncture by waste materials and plant movement).
- The provision of rain flaps extrusion welded to the extensive side slopes at this site led to several instances of liner holes.
- Frequent (quarterly surveys) allowed the early identification of defects and their repair. Infrequent surveys would have meant the waste was too deep to locate and repair holes.
- There has been no evidence of the gradual development of holes.

8.2.3 Summary of findings from ELL surveys

A number of conclusions can be drawn from the ELL survey data review as summarised below.

- The provision of either mobile or fixed ELL surveys at the end of liner construction enables the completion of effectively hole-free liners, if the ELL survey is correctly undertaken in favourable conditions. If the conditions are not favourable, particularly with respect to the electrical isolation of the area under test and the moisture content of the overlying soil or geotextile cover material, then the survey can miss holes. Knife cuts and holes on top of wrinkles are also more difficult to detect as the void will not conduct electricity well. Leak location surveys of double lined sites require particular care. The grid spacing of fixed systems is important, with the sensitivity reducing as the grid spacing increases (Taylor *et al.*, 1999).
- While holes present on completion of liner construction can be located and repaired, partially penetrating damage, poor quality welds, wrinkles, and stress concentrating features will not be detected by an ELL survey.
- The provision of effective CQA is seen to result in a much lower incidence of holes and it is reasonable to conclude that the number of non-penetrating and other defects should also be commensurately less.
- Areas of liner left exposed or only lightly protected for an extended period before being covered by waste, e.g. at bunds, cell margins and benches on slopes, are susceptible to damage from a range of activities.
- Side slope liners are more susceptible to damage than basal liners. They are often exposed for long periods of time, stressed by overlying cover materials and are vulnerable to plant movements. Also, cover placement during site operation may take place without CQA monitoring.
- Holes detected by ELL surveys may occur where there will not be an effective leachate head, e.g. on bunds or slopes, but gas leakage may be of concern. However, at sub-groundwater (hydraulic trap) landfills, high external groundwater and leachate levels could allow leakages through these holes.

- Damage to the liner is likely to occur during landfilling operations. Estimates of the comparative frequency of holes located after completion of the liner against the total number of holes identified during liner construction and after completion vary from 0% to 68%. A reasonable estimate would be approximately 30% - 35% occurring after liner completion.
- The provision of a fixed ELL system with an adequate grid spacing (10-20m) and effective analytical software allows identification of leaks during operation and enables their repair if the waste depth is not excessive. If inaccessible, the hole position is known reasonably accurately, leachate leakage may be tracked using the ELL system and other monitoring techniques (or remote repairs) can be targeted on the known defect.
- Without a fixed ELL system, then an allowance for operational defects in the geomembrane should be made.
- Current experience is accumulated from ELL surveys conducted for 7 - 8 years since liner completion. There is no evidence of the development of holes in a liner after completion of waste disposal activities in that area over this period. However, subsequent stress cracking failure may occur at locations of stress.

8.3 Review of leak detection systems in double liners

Double liner systems (see Figure 3.1(c)) are commonly used in the USA to meet regulatory requirements. The upper liner (a single geomembrane liner or a geomembrane/GCL composite liner) is the primary liner and the lower liner (usually a composite liner comprising a geomembrane and compacted clay or GCL liner) is the secondary liner. In between the primary and secondary liners is a drainage layer - the leakage detection system (LDS) - and the volume of leakage is monitored to ensure it does not exceed the permitted action leakage rate, typical values being 180 – 200 litres per hectare per day (lphd). The results of such leakage monitoring over time are useful in assessing whether holes in the primary liner are developing or increasing with time.

The interpretation of the leakage monitoring is not straightforward as the liquid measured may comprise liquid from sources other than leachate leakage. Besides leakage through the primary liner, liquid may enter the LDS as:

- infiltration during construction of the lining system;
- water from consolidation of the clay liner or other mineral layer; and
- groundwater infiltration from outside the landfill.

Leakage rates must therefore be estimated taking these additional potential sources of liquid into account. Leachate leakage depends upon the actual leachate head above any holes which is not necessarily the same head as measured at a monitoring well. It is also difficult to assess the numbers, sizes and locations of any holes.

Rowe (1998) summarised some of the studies on LDS measurements. He reports that Bonaparte *et al.* (1996) examined data for 26 landfill cells containing geomembrane / GCL composite primary liners. Three periods were identified during which there may be very different flows in the LDS – (a) the initial period of operation, (b) the active period of operation and (c) the post-closure period (after the final cover has been constructed). Table 8.9 summarises the mean and standard deviations of the flow (lphd) in the leachate collection layer above the primary liner (PLCS) and in the leakage detection system (LDS).

Table 8.9 Flow in PLCS and LDS in lphd from 26 landfill cells (after Bonaparte *et al.*, 1996, from Rowe, 1998)

Period	No. of cells	Average flows				Peak flows			
		PLCS		LDS		PLCS		LDS	
		Mean	s.d.	Mean	s.d.	Mean	s.d.	Mean	s.d.
Initial period	25/26	5350	3968	36.6	68.5	14964	11342	141.8	259.9
Active period	18/19	276	165	0.7	1.1	752	590	7.7	13.7
Post-closure	4	124	-	0.2	-	266	-	2.3	-

The mean and peak LDS flow rates were small during the active operation and reduced further during the post-closure period, from which it is reasonable to conclude that there was no evidence of increasing holes in the primary liner.

A recent study sponsored by the US EPA reviewed the performance of upper liners at sites containing double lining systems (Bonaparte *et al.*, 2000 from Koerner *et al.*, 2000). The reported data are from a survey of 287 individual cells in 91 double-lined landfills with leak detection systems which had up to 10 years of service performance (Figures 8.6(a) and 8.6(b)). The figures show average leakage rates from double liner systems using different types of primary liner, being geomembrane GM, GM/CCL and GM/GCL primary liners with both sand and geonet as leak detection materials.

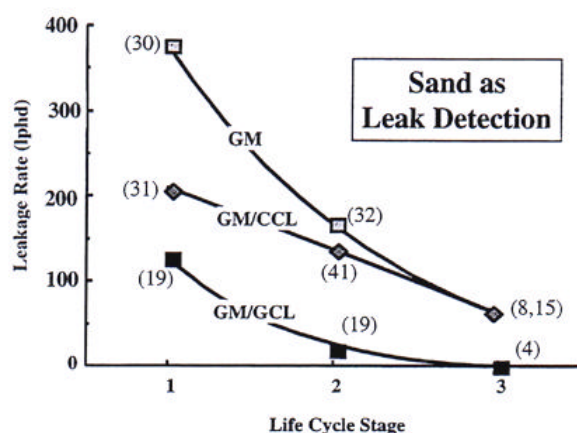


Figure 8.6(a)

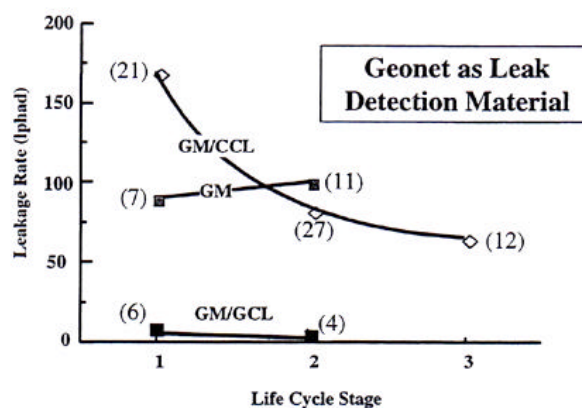


Figure 8.6(b)

Figure 8.6(a & b) Leakage rates in different lining systems at various stages of landfill lifetime (sand and geonet as LDS material) (from Koerner *et al.*, 2000)

Key:

- Life-cycle stage
- (1) Initial stage
 - (2) Active stage
 - (3) Post-closure stage

The numbers in brackets show the number of sites in the study that fell into that category

It is seen from Figures 8.6(a) and 8.6(b) that in most cases the average leakage flow reduced from the Initial Life Stage to the Active Life Stage. The single exception was for the single

geomembrane liner and geonet combination when there was a slight increase that probably reflected a rise in leachate level. For the four types where there were data, the flows in the Post-closure Stage were the least. It is likely that the leachate head was reduced following capping of the cells leading to reduced leakage but the data do indicate that new holes were not being generated in the primary liners after completion of landfilling.

Other experience in the USA confirms that there is no known instance of leakage increasing in a double lined landfill cell after it had been placed in service that was not due to external influences. There have been a number of cases where the leakage rate increased after some service period but these have generally been found to be due to the new presence of or an increase in leachate head above original defects in the liner. Leakage then commenced or increased in such cases. Increases in leakage flow rates have also been found to be caused by rainwater accumulation in an anchor trench “wicking” through the geotextile in a geonet / geotextile composite drain down the side slopes. Koerner (2003) is not aware of any HDPE geomembrane landfill liner that has developed a hole in service from anything other than an external influence.

8.4 Large scale and catastrophic failure events

8.4.1 Background

Large-scale and catastrophic failures of geomembrane liners and caps may be caused by a variety of mechanisms including:

- slope stability failures and mass movements of the waste body;
- down-drag effects of waste settlements on geosynthetics on side slopes or steepwalls, and on leachate or gas wells;
- excess basal movements including heave and settlement;
- excess differential settlement effects on capping systems;
- seismic events; and
- landfill fires.

By definition, these are major events and not directly related to durability although the geomembrane liner may be subjected to severe damage. The potential impacts on geomembrane liners of large-scale and catastrophic failures are reviewed below.

8.4.2 Slope instability

Numerous case histories have been presented in the literature of large-scale and catastrophic failures within landfills (Jones & Dixon, 2003, also references within their report). Failures can occur during construction of the liner, on initial filling with waste, as waste height reaches its maximum, or after landfill completion.

Jones & Dixon (2003) reviewed UK case histories of stability failures at landfills. On the basis of a questionnaire to Environment Agency officers (26 responses received, each describing a single failure), some broad trends emerged as follows:

- most failures were on sites underlain by a cohesive soil, none on granular soil or low permeability rock;
- many of the failures (11 of 26) were experienced at sites with side slopes greater than 30° inclination;
- 50% of sites with failures had composite (mineral-geomembrane) lining systems although there was a broad range of lining systems employed at the sites;
- the majority of failures (over 75%) involved the side slopes rather than base;
- 48% of failures were in the lining system and 32% in the subgrade, the remainder being in the capping, the waste or at an interface;
- 30% of the cases of instability were in the side slope liner, 26% were in the subgrade, 17% in the geomembrane and 13% were basal heave;
- the timing of failures was spread between the construction phase (8 instances), prior to waste placement (10), during waste placement (7) with only one after waste placement.

Instability of waste slopes that involve elements of the lining system is the most common mode of stability failure and will probably, but not inevitably, cause severe, widespread damage to any geomembrane liner in the lining system. Factors that control the stability of waste slopes involving geomembrane liners include the following (Jones and Dixon, 2003).

- Interface shear strengths between geosynthetics and geosynthetic / soil interfaces. The measurement of interface shear strength and the selection of the appropriate value to use in specific cases are complex.
- Leachate conditions, where increased levels result in higher destabilising forces and reduced effective stresses, which reduce shear strengths.
- Groundwater pressures acting on the lining system and subgrade.
- Surface water infiltration on to geomembrane caps.
- Gas pressures on capping systems and on the lining of existing waste slopes.
- Location and slope of the potential shear surface which are controlled by the weakest layers and interfaces.

With respect to geomembrane liner durability, the primary aspects of concern are to ensure that slope failure will not occur and to avoid as far as practicable the imposition of stresses in the geomembrane liner. Adjacent materials and different areas of an interface can have different mobilised strengths at the same time. Materials mobilise their shear strengths at different strains and this strain incompatibility can lead to stresses being imposed on geomembrane liners or caps.

The self-weight (and any imposed loading) of cover soils placed above a geosynthetic layer on a slope is transferred through the various geosynthetic components in shear. Shear stresses mobilised at the upper surface of a geosynthetic are transferred to its lower surface by shear until the maximum shear strength of the lower surface is reached, the remaining stress then being taken in tension in the geosynthetic. The amount of tension developed in the geosynthetic depends on the interface shear strength between the various materials. The integrity of the lining system is assessed by comparing the stress transferred into the geosynthetic to its tensile strength.

An example of large displacements developing at geosynthetic interfaces is given by Gourc *et al.* (1997). A field experiment was conducted to measure the displacements at geosynthetic

interfaces due to the placement of a gravel layer on a 1V:2H side slope. At the interface between a protection geotextile and a geomembrane liner it was found that:

- 70mm displacement occurred in response to 6m of gravel placement;
- this increased to 170mm in response to the removal of gravel from the toe region; and
- a further increase to 650mm of displacement in response to placing gravel to 8m up the slope.

While a slope analysed by normal limit equilibrium methods may show there to be an adequate factor of safety against slope failure, as a result of strain incompatibility there may still be significant relative movements between geosynthetic elements. This may lead to long term stresses being imposed on the geomembrane liner which will eventually lead to the generation of stress cracks in HDPE geomembrane liners.

The use of double-sided textured geomembranes to retain cover soils on side slopes or caps should be avoided as these will impose tensile or shear stresses on the geomembrane liner, sharply increasing the onset and extent of stress cracking if the stress is maintained. As indicated in Chapter 7, the presence of the surface texture may also cause a reduction in the stress crack resistance of the geomembrane. Reinforcing geosynthetics e.g. geogrid or high strength geotextiles should be employed to provide the necessary stability and to protect the underlying geomembrane liner. A mono-textured liner with a smooth upper surface assists in reducing tensile stresses in the liner.

8.4.3 Down-drag effects of waste settlement

Waste settlement from consolidation and degradation of the wastes leads to downdrag forces on side slopes and on leachate and gas wells.

Side slopes

Large settlements can compromise the integrity of the lining system by causing a loss of protection to the geomembrane through failure or distortion of overlying layers, and may lead to failure of the geomembrane by tension or, in the longer term, by stress cracking. The provision and location of a low shear interface is important so that settlement of the waste body is not transferred as tension to the geomembrane liner (Gallagher *et al.*, 2003). Movement at the low shear interface should not compromise the other functional elements of the lining system. This can be achieved by the provision of a dedicated low shear interface and locating this to the waste side of the geomembrane, overlying drainage layer and protection geotextile. In this way, the integrity of the drainage layer and protection geotextile remains intact, as well as the stresses in the geomembrane liner minimised.

Locating the low shear interface at the smooth upper face of a smooth or mono-textured geomembrane will cause overlying layers including any drainage and liner protection materials to settle and distort with the waste mass, compromising their integrity. The large differential movements on the geomembrane face may lead to scratches, gouges and other defects in the geomembrane, which are likely to be detrimental to the durability of the liner.

Stress on a side slope liner will be at a maximum in the upper parts of the slope, potentially leading to rupture. Tears or other damage on side slope liners only permit leakage if leachate (perched or otherwise) or external groundwater rises to these levels in the future, possibly after leachate management at the landfill ceases. Landfill gas escape may remain a concern.

Waste settlements on slopes can be reduced by the placement of a buttress zone of less compressible wastes, where available, adjacent to the slopes (Gallagher *et al.*, 2003). This will have the additional benefit of reducing the differential settlement in the cap around the perimeter of the landfill where the cap crosses from natural ground to settling waste.

Down-drag on wells

Settling waste is likely to cause down-drag stresses (or negative skin friction in geotechnical terminology) on vertical leachate and gas wells in the waste, although settlement-compensating wells are available which should reduce the stresses.

If the wells are supported on the base of the landfill, the down-drag forces can be transferred to the base, potentially placing additional stresses on the basal liner. As leachate extraction wells are at low points and the leachate head is greatest at these locations, the design of the area around the base of the well, and the lining system in particular, are of critical importance. The design should avoid the imposition of additional tension stresses in the geomembrane liner elements and the need for complicated welding involving fillet extrusion welds.

8.4.4 Excess basal movements

Excess basal movements are likely to induce tensile stresses in a basal geomembrane liner and arise from compressible subgrade, cavities in the subgrade and basal heave (Jones and Dixon, 2003).

If a highly compressible subgrade is present such as peaty soils, then the total and differential settlements caused by the waste load can be quite large. It is the size of the differential settlements and angular distortion (differential settlement ÷ distance) which are important to the tensile stresses in the geomembrane liner and measures to reduce differential settlements need to be taken to ensure that stresses are limited to acceptable values.

BAM in Germany places a limit of 3% long-term strain on HDPE geomembrane liners to avoid stress-cracking problems for a period of at least 100 years (Seeger and Müller, 2003). Peggs (2003) has recommended maximum strains for different materials as follows:

- | | |
|--|-----|
| • HDPE smooth SCR <1500 hr | 6% |
| • HDPE smooth SCR >1500 hr | 8% |
| • HDPE random texturing | 4% |
| • HDPE structured profile | 6% |
| • LLDPE density <0.935 g/cm ³ | 12% |
| • LLDPE density >0.935 g/cm ³ | 10% |
| • LLDPE random texture | 8% |
| • LLDPE structured profile | 10% |
| • PP unreinforced | 15% |

The measurement of strain is used as an indirect measure of the stress that exists in a geomembrane that might result in stress cracking. While this is clearly important for HDPE, it is not as significant for other materials that are not susceptible to stress cracking, unless oxidised. The objective is to limit stress to a sub-critical value where stress cracking will not be a practical problem. However, in a confined situation the stress will be applied very slowly to the geomembrane as the adjacent soils move and the geomembrane will be able to relax resulting quite rapidly in geomembrane stresses reducing to maybe 50% of the value implied by the deformation. The maximum strain values given by Peggs (2003) for HDPE are higher than the BAM limit on the basis that stress relaxation will reduce the stresses (although this is also taken into account by BAM (Seeger and Müller, 2003)). Recognition should be given to the benefit in using HDPE geomembranes with higher SCR.

Compressible sub-grades can also underlie geomembrane caps and at vertical and lateral landfill extensions, where a liner is placed on the upper surface or sloping face of existing waste deposits. With increasing difficulty in obtaining permission to develop new landfills, maximising the capacity of existing sites is an attractive land-use approach. This can lead to the development of “piggy-back” landfills which, with the advent of the soil and water protection requirements of the Landfill Directive, presents technical challenges to the landfill designer. The predicted settlements, particularly future differential settlements, need estimation and may require methods to reduce such settlements e.g. reinforced mattresses of geosynthetics and granular material, or dynamic compaction.

Geomembrane caps have to accommodate the maximum settlements of the waste body. In response to this, flexible geomembranes such as LLDPE are usually used for landfill caps, although in Germany, HDPE is still the required material but with a doubled strain criterion of 6% compared to the basal HDPE liner. Jones and Dixon (2003) review methods of waste settlement calculation. Caps have the ability to be inspected, repaired and even replaced in future years should leakage through defects become excessive but they are also much more susceptible to physical damage arising from human activities and burrowing animals than are basal liners.

Natural and artificial cavities (voids or loosely filled) can develop in the subgrade e.g. resulting from solution features in limestone or collapse of old mineshafts. The significance of a cavity is a function of its size in relation to its depth below the lining system, and the strength and stability of the material between the cavity and the lining (Jones and Dixon, 2003). The calculation of stresses imposed on geomembrane liners overlying cavities has been addressed by Giroud *et al.* (1990). However, in conditions where cavities may be present, the objective is to minimise any additional stresses on the liner and precautionary measures should be taken to eliminate the potential for cavities or to include reinforcing elements to provide the necessary bridging. Parnell (2003) describes ground treatment methods to address the problem of chalk solution features at a landfill at Beaconsfield.

The mechanisms of basal heave caused by excess porewater pressure in an underlying stratum exceeding the total overburden stresses are well known to geotechnical engineers. The formation of an excavated void leads to a reduced total stress at the formation level, potentially triggering piping (upward movement of groundwater) and softening of the subgrade.

Heave can also be caused by expansive soils either gaining access to water to develop a swelling pressure or due to precipitation of new minerals. Such effects are relatively uncommon in the UK. If it occurs, heave is likely to be a problem arising during liner construction. However,

heave will be counter-balanced by the weight of waste once landfilling commences. In principle, smectite/illite or smectite rich soils could be susceptible to volumetric changes due to the influence of water and, in particular, cation-rich groundwater. Under certain circumstances, ingress of air and water to soil or weak rocks may result in the oxidation of sulphide minerals present. Dependent upon the mineralogy, re-precipitation of secondary mineral phases, such as gypsum, causes volumetric expansion leading to heave. Heave can cause additional pressure on the geomembrane liner potentially compromising its integrity and in the worst case affecting liner stability.

8.4.5 Seismic events

In the United Kingdom, data on seismic events have been collected for a period of over 1,200 years. Earthquakes generate shear, pressure and surface waves. These waves are transmitted through the Earth's core with differing characteristics. Seismographs now record the magnitude of these waves, which can be used to calculate the intensity of earthquake energy released (Richter value) at any point on the surface of the Earth. An alternative is the EMS-92 Intensity Scale which records the intensity of an earthquake as felt at a particular point on the Earth's surface.

The largest three UK seismic events were recorded in Dover Straits (1580), Barrow-in-Furness (1865) and Colchester (1884). The greatest intensity was 8 EMS. According to the British Geological Survey, 6.0 is the maximum Richter magnitude seismic event in the UK and this is at a frequency of approximately 1 in 1,000 years (Musson, 1990).

The EMS description of surface effects of intensity 7 EMS events, in a single instance, includes landslips occurring in roads cut into steep slopes. At 8 EMS, small landslips may occur in road embankments on steep slopes. On this basis, basal and sloping earthworks in landfills are unlikely to be affected by instability by seismic events with a felt intensity less than 8 EMS. The stability improves on completion of landfilling from the support offered by the placed and compacted waste.

Orr and Finch (1990) examined the effects of a Richter 7.1 event in 1989 on Californian landfills. Generally only minor damage was caused, most common being minor cracking of landfill slope surfaces, seen to be little different from normal settlement cracks. Gas collection systems were affected, albeit temporarily with all leachate and gas systems reportedly repaired and operational within 24 hours. Seed and Bonaparte (1992) considered the principal failure modes at lined quarries and pits to be instability of the waste in the temporary, partly filled condition and failure of relatively steep capping by slippage. The mode of failure of a fault line movement at surface level across the landfilled area is not considered a normal seismic design condition, even in California.

In summary, the probability of significant damage to a geomembrane liner at a UK landfill from a seismic event is extremely low.

8.4.6 Landfill fires

Landfill fires are uncommon, but increasing, occurrences in the UK but can be significant, complex events of extended duration that require careful management to extinguish. Landfill fires appear to occur more frequently in the USA, which has been a source of useful guidance and case histories (Sperling and Henderson, 2001). Potential causes of landfill fires include direct sources of combustion, both deliberate, i.e. arson, and accidental such as embers within deposited waste, careless cigarette smoking and methane flash from equipment spark. Spontaneous combustion is another significant cause. Fires occur in the near surface (i.e. within about 1.5m) and underground. The latter are more difficult to control and have greater significance in terms of potential damage to the basal and side slope lining system.

Methane is a significant factor in the ignition of many landfill fires. Waste materials, particularly construction and demolition wastes, contain a significant proportion of combustible materials and may locally contain highly inflammable material, possibly deposited in breach of regulations, which can initiate a fire. A waste body that is poorly compacted or contains large bulky items, which are difficult to compact will have voids of air which can provide the oxygen required to sustain a fire. A common cause of underground fires is a combination of locally increased oxygen concentration causing increased biological activity, hence raised temperatures, coming in contact with a pocket of methane. Such underground fires can then smoulder undetected for weeks or months.

Dealing with these fires requires a careful, systematic approach to protect the fire crews from hazardous gas and to prevent exacerbating the problem (Federal Emergency Management Agency, 2002). The management of underground landfill fires is complicated because they are buried and thus not readily defined, difficult to reach and not always responsive to standard fire fighting techniques. Landfill fires can lead to the sudden collapse of large voids posing additional hazards to fire fighters. The fire has also to be dealt with in a manner that is consistent with minimising further environmental damage. Flooding the fire with water is not always successful and can merely lead to the creation of large volumes of leachate, which can overwhelm the leachate drainage system. In the intensity of the fire, the geomembrane liner can be compromised and the underlying clay liner can be severely desiccated.

French *et al.* (1998) describe a case history of a fire involving tyre chips used in the leachate and gas collection systems on the base of the landfill cell. A 300mm thick layer of tyre chips was underlain by 300mm sand over a 340g/m² protection geotextile over a 1.5mm HDPE geomembrane and GCL composite liner. The leachate collection pipework was surrounded by coarse aggregate in place of the 300mm of sand. The fire was believed to have been caused by a “hot load” of waste placed on top of the tyre chips and it spread across the exposed tyre chips. Water application failed to control the fire but it was rapidly extinguished about 7 hours after ignition by smothering the fire with over 3000m³ of soil. Six hours after the fire had been put out, temperature measurements were taken to assess the possible effects on the HDPE geomembrane liner. The readings at the top of and within the sand layer under the tyre chips are shown on Table 8.11. The maximum temperature recorded in the tyre chips was 53°C.

Table 8.11 Temperature readings after fire in tyre chips (from French *et al.*, 1998)

Depth	Test hole 1	Test hole 2	Test hole 3	Test hole 4
Top of sand	23°C - 28°C		40°C	35°C
5cm		18°C	32°C - 35°C	
10cm	18°C - 19°C	probing stopped		17°C
15cm	probing stopped			probing stopped

Note: Temperature probing stopped when a 17°C temperature was encountered.

Ten days after the fire, the trial holes were excavated in four places for visual examination of the lining and leachate collection systems. Damage to the HDPE geomembrane and polypropylene protection geotextile had been avoided by the protection given by the 300mm moist sand layer. The HDPE leachate drainage pipes and geotextile surround were damaged in the areas of the cell where the fire had burned longest. The rapid response by the fire fighters prevented the damage being much worse, as shown by the damage in areas that had burned longest.

Adams *et al.* (1997) reported a case study of a fire damaged lining system at a hazardous waste landfill containing industrial waste sludges and other chemical manufacturing waste. Based on thermocouple measurements installed in the vicinity of the fire, the temperature near the liner system may have approached 800°C. It took 11 months from its discovery to the fire being finally extinguished. Visible damage to the double liner (single 2mm HDPE liner over leakage detection geonet over 2mm HDPE geomembrane / 0.9m CCL composite liner) was observed over 300m².

Although the fire burned for almost a year, the affected area was not very large. Damage to the geosynthetic components in the fire affected area ranged from complete disintegration, to melting and fusing of the various components further from the centre of the fire, to rippling and stretching of the materials along the perimeter of the visibly damaged area. In several areas, evidence of melted geosynthetic materials was observed within desiccation cracks in the CCL.

The stability and integrity of a landfill lining system and the waste body should be reassessed after any significant fire. Remedial action must then be determined on a case-by-case basis. Landfill fires can to a very large extent be prevented by good operational practice - appropriate waste acceptance procedures, adequate compaction and break-down of waste, sufficient inert cover material, together with ongoing monitoring of landfill gases and waste body temperatures. These measures will reduce the risk and provide the data to initiate intervention should the waste body start to become a fire risk.

8.5 Summary of physical damage mechanisms

Physical damage mechanisms and the development of holes in geomembrane liners in the medium term are summarised below.

- i) Pre-existing non-penetrating defects, and holes if no ELL survey is undertaken, will be present in liners at the start of their operational life from unidentified damage caused during liner installation.
- ii) Liners are susceptible to damage during the interim period before waste placement. An average of about 30-35% of all holes caused in the construction and medium term

operational stages may be expected to occur during the latter period, although there can be considerable variation in this estimate. The installation and regular monitoring of a fixed ELL system can enable the identification of these holes if they occur and, in most cases, their repair.

- iii) The commonest mechanisms for damage have been found to be plant movements, puncture by stakes used by personnel or from articles in the waste (e.g. reinforcing bars), and small holes opening up in defective welds.
- iv) The locations experiencing the greatest frequency of defects are exposed or poorly protected liner at the margins of a cell, on bunds and benches, and on slopes.
- v) Effective liner protection at all stages will reduce the susceptibility of geomembranes to damage.
- vi) The evidence from monitoring of landfill liners and leakage rates for up to 10 years shows no evidence of the development of damage from degradation, ductile failures or stress cracks.
- vii) Once a liner is covered by several metres of waste, the agents for the future development of holes in the liner are limited. Thus, holes are unlikely to occur for at least the first decade of the service life of the geomembrane liner, and probably much longer. Agents that would lead to the generation of new defects in a geomembrane liner in the medium term are:

- the continued imposition of stresses on the liner caused by, for example, waste settlement down-drag on welds and side slopes and, for landfill caps, settlements of the underlying waste;
- delayed brittle fracture (stress cracking) of stressed parts of HDPE geomembrane liners at low residual stress levels;
- degradation and settling of waste allowing sharp / hard waste articles to move downwards and puncture the liner;
- external factors such as drilling in new gas / leachate wells or, in landfill caps, excavation in cover soils;
- catastrophic events as discussed in Section 8.4;
- inadequate initial design of protection materials;
- degradation of protection materials e.g. geotextiles, allowing new stresses to be imposed on the liner, or puncturing by drainage gravel.

9 ESTIMATION OF DEFECT GENERATION IN HDPE GEOMEMBRANE LINERS

9.1 Introduction

This chapter brings together the findings from the earlier chapters with the purpose of developing a model of the development of defects in geomembrane liners from installation to the end of the liner's service life, which can then be applied to landfill risk assessments. The model described in this chapter takes account of physical damage as discussed in Sections 8.1 to 8.3, inevitable material degradation and stress cracking. It does not include large scale and catastrophic failure events (Section 8.4) as these occur either as a result of poor design or operational practices that are avoidable failings, or highly unusual natural phenomena.

The model, therefore, assumes an adequate standard of design that avoids slope instability, down-drag stresses caused by settlement, and excessive differential settlements of the subgrade below geomembrane liners and caps. It is the responsibility of designers to address such issues effectively. Where this is not demonstrably achieved, then additional defects should be added to those derived from the model described in this chapter. Such additional defects, by their nature, are likely to be large and may dominate the total area of geomembrane defects.

9.2 Geomembrane liner service life

The service life of a geomembrane liner or cap can be defined as the length of time the geomembrane continues to act as an effective hydraulic barrier for the purposes of the site under consideration. Clearly, this will depend upon the circumstances at the site and, for groundwater quality, the acceptable amount of leakage of specific contaminants in the leachate. Factors influencing this are:

- the number and sizes of holes in a geomembrane liner overlain by leachate;
- is the geomembrane liner a single liner or part of a composite or a form of double liner?;
- the quality of contact between the geomembrane and the underlying liner within a composite liner;
- leachate head before and after effective leachate control ceases;
- the types of contaminants in the leachate, their concentrations and variations in these with time;
- site sensitivity with respect to groundwater quality (e.g. groundwater vulnerability);
- attenuating properties of the soil/rock between landfill and water table (i.e. the geological barrier); and
- for below groundwater level landfills (hydraulic traps), the respective levels of the external groundwater and the leachate.

With regard to the acceptable degree of landfill gas escape, the gas composition and respective partial pressures, ease of migration in the external ground conditions and the distance to receptors will also be important factors.

A geomembrane may have a certain number and sizes of holes, and at one site be deemed acceptable yet at another, more sensitive site, the leakage may be considered unacceptable.

The generation of holes in geomembrane liners, whether by physical damage mechanisms, stress cracking or material degradation is only one factor in the prediction of the service life of that liner.

9.3 Stages of hole generation

The development of perforations in HDPE geomembrane basal and side slope liners can be seen to occur at six stages, as proposed by the following conceptual model and as shown diagrammatically in Figure 9.1:

- | | |
|---------|---|
| Stage 1 | The first stage is the number and sizes of holes remaining in the liner after construction of the liner and placement of the cover material, wells and drainage system. Where an ELL survey has been properly carried out, then the detected holes would be repaired and zero or a very small number of holes may be considered to remain at the end of Stage 1. |
| Stage 2 | This second stage represents the holes caused before or during waste filling operations by physical damage mechanisms resulting in either new damage or the opening of defective welds or other latent defects. Where a fixed ELL system is in place and regularly monitored, and detected holes repaired, then a small number of holes may be considered to remain at the end of Stage 2. |
| Stage 3 | After completion of waste disposal above the liner and capping, there will be no increase in the applied load. Present evidence shows that holes are not seen to develop for at least the next 10 years. Where the landfill has been well designed and large-scale stresses on the liner are avoided, then there is generally no agent to cause holes, either by physical or degradation mechanisms. It is therefore reasonable to predict based on data presented in Chapter 8 that Stage 3 would comprise a period of at least 10 years during which no further holes develop. Any duration beyond 10 years to an assumed maximum value of 50 years would reflect better geomembrane material properties, efficacy of liner protection, design quality and standard of installation. |
| Stage 4 | Stage 4 represents the main period of antioxidant depletion, and continues until both antioxidant depletion and the initial induction stage of oxidation are complete (Stages A and B, Hsuan and Koerner, 1995). Oxidation causing embrittlement is deemed not to have commenced in Stage 4. Stress cracks will develop within the stressed areas of the geomembrane. The number of cracks will depend on the estimated extent of areas under stress. Geotextile protectors may be expected to become largely ineffective during this stage because of degradation of the fibres, potentially exposing the geomembrane to stresses from drainage gravel. In this case, stress cracking is assumed to become widespread, depending upon the size and angularity of the gravel. The level of stress, the stress crack resistance of the geomembrane and the prevailing temperature at the liner would control the time to the initiation and growth of stress cracks. This incidence of new or enlarging stress cracks may reasonably be expected to continue until the onset of oxidation. |
| Stage 5 | Stage 5 represents the oxidation stage of the geomembrane liner (Stage C, Hsuan and Koerner, 1995). The geomembrane will embrittle and further stress cracking damage is predicted to occur relatively rapidly at all locations in the geomembrane remaining under any significant level of tensile stress. Where geotextile protectors have been used and the geomembrane is exposed to multiple locations of stresses applied by drainage gravel, then it is reasonable to conclude that the |

extent of the cracking of the embrittled geomembrane would be so great that the geomembrane liner may be considered non-existent (if not already considered so in Stage 4).

Stage 6

The terminal stage is Stage 6 when it is assumed that further generation or extension of holes in the geomembrane continues at a steady rate related to the total number of holes present at the end of Stage 5. For poorly protected liners, complete failure of the geomembrane barrier is considered to occur in Stages 4 or 5, Stage 6 never being attained. In Stage 6, the geomembrane will continue as a "leaky", degrading barrier with the HDPE geomembrane away from cracks and other holes being brittle but remaining intact.

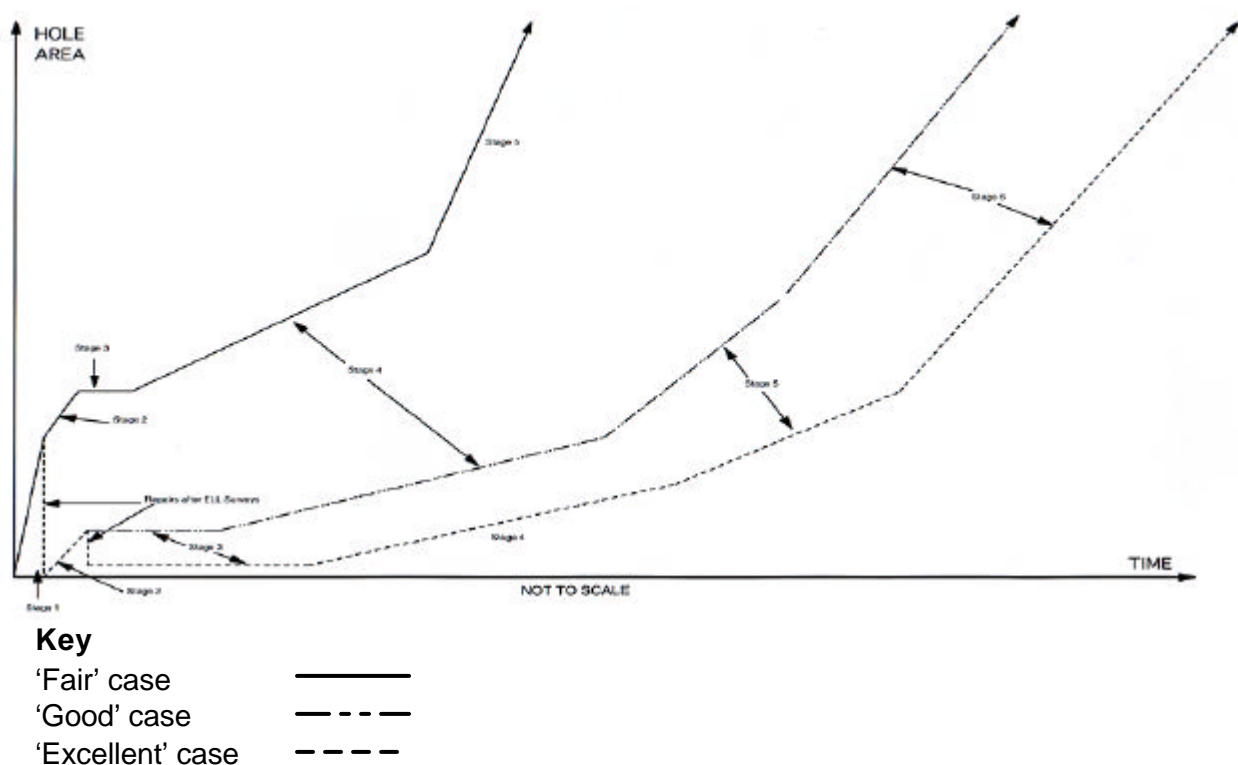


Figure 9.1 Diagrammatic representation of the conceptual model of hole generation with time

Figure 9.1 shows three conditions. The "excellent" case only applies where the highest standard of materials, design, installation (including CQA to the highest standard) and operation is assured. The features of the "excellent" category include:

- the geomembrane material meets the requirements of the GRI GM13 specification with the added criterion that the stress crack failure time should be well in excess of the 300 hours currently specified in GRI GM13;
- the OIT value and oven ageing procedure should meet the GRI GM13 and BAM certification requirements (see Section 5.5.5);
- the design has been subjected to independent design quality assurance;
- mineral geomembrane protection is provided;
- installation of the liner system by installers certificated under the British Geomembrane Association / The Welding Institute scheme together with experienced CQA supervision until the first layer of waste has been placed;

- a fixed ELL system is installed and monitored quarterly (or more frequently) until completion of waste disposal.

The “good” case will apply where the following conditions are met:

- the geomembrane material meets the requirements of the GRI GM 13 specification;
- the design meets in full current Environment Agency guidance with mineral protection provided where drainage gravel is employed;
- installation of the liner system by installers certified under the British Geomembrane Association / The Welding Institute scheme together with experienced CQA supervision until the first layer of waste has been placed;
- a mobile ELL survey is carried out after placement of the protection and drainage materials.

The “fair” case applies the geomembrane liner does not attain any one of the criteria for the “good” case above, and assumes that no ELL survey is undertaken. However, the quality of the design is still considered to be reasonable and avoids the large scale or catastrophic physical damage discussed in Section 8.4.

In the eventuality of poor design and/or construction, or if a catastrophic failure event did occur, then a much faster generation of holes and hole area than that shown in the “fair” case would apply. The need to achieve high standards of design, material specification and installation is emphasised, otherwise the generation of holes is likely rapidly to increase from those estimated for the “good” case.

The stage at which the geomembrane liner ceases to be an effective hydraulic barrier also depends on the other circumstances relevant to the site, as noted in Section 9.2 above. The geomembrane liner may remain as a partially effective hydraulic barrier. Alternatively, the predicted leakage may be so large that the geomembrane can be assumed not to exist and that the barrier function has to be fulfilled by other elements of the liner system. For a composite liner, this would mean that the leakage through the composite liner would be the same as for the mineral (clay) or GCL component alone.

Deterioration will not occur evenly across the liner. Different stress levels, localised damage, different exposure conditions (e.g. temperature variations) and stress concentrating features will lead to different rates of deterioration over the liner. The conceptual model is an averaged, idealised representation of the evolution of holes over the whole liner.

Where LLDPE geomembrane is used, the stages will be different. LLDPE has much greater resistance to stress cracking, but oxidation will commence earlier than for HDPE, other circumstances being equal. Stage 4 may be omitted. Stress cracking of oxidised LLDPE geomembrane would occur in the same pattern as Stage 5 for HDPE. Currently available research results do not permit predictions to be made on the duration of the three stages of oxidative degradation of LLDPE and the consideration of a case with LLDPE will require a review of the data at that time to develop a modified conceptual model for the case involved.

For HDPE geomembrane landfill caps, the stages of hole generation will be similar, in qualitative terms to HDPE liners. In exposed conditions, for lining water reservoirs and liquid storage/treatment lagoons, or as floating covers, the effects of photo-degradation and the much greater

risk of physical damage need to be taken into account. For chemical or waste storage/treatment lagoons, selection of the appropriate geomembrane material to meet chemical resistance criteria may lead to material other than HDPE or LLDPE being specified. The mechanisms of physical damage and material degradation discussed in this report will assist designers of lagoons and reservoirs to assess the stages and timeframe of deterioration of geomembrane liners for these purposes.

The range of uses for geomembrane barriers in other applications including contaminated land remediation or leakage containment is very broad (e.g. in vertical cut-off walls, mine waste containment, horizontal capping barriers or as protection against migration of chemical leakage/spillage). The chemicals involved may also be present in high concentrations and the interaction of these substances with the geomembrane material should be given specific consideration. For these applications, the factors leading to geomembrane liner deterioration discussed in this report will be helpful in estimating the geomembrane service life.

9.4 Estimation of hole generation

The development of holes in HDPE geomembrane liners in landfills, at the various stages of liner deterioration, is estimated below. The hole sizes have been subdivided into the three types as used in LandSim (Environment Agency, 2001) plus stress cracks which are defined as longitudinal slits (whereas LandSim assumes all defects including tears are circular in shape for the purpose of leak calculation).

Pin holes	0.1 - 5 mm ²
Holes	5 - 100 mm ²
Tears	100 - 10,000 mm ²
Cracks	10 - 1,000 mm ²

Individual stress cracks are slit-shaped and estimated to be 1 mm wide by a minimum of 10 mm long (i.e. 10 mm²) and increasing to a maximum length of 1000 mm (1000 mm²) in the confined conditions at the base of a landfill.

The estimated hole frequencies in Stages 1 and 2 are given as probability density functions (PDFs) of three values representing the minimum, most likely and maximum estimated values. In the later stages, information is not available to estimate PDFs and tentative ranges of values are proposed.

Stage 1

The number of holes at the end of this stage will depend on whether or not an ELL survey has been properly undertaken, and on the standard of installation and CQA. Where those standards are demonstrably high and the ELL survey completed (together with any repairs), the installation may be classed as "excellent" or "good", both with zero holes. The "fair" case has been selected on consideration of the data presented in Section 8.1. No stress cracks are included in Stage 1. The estimated hole incidence for these three grades is given in Table 9.1.

Table 9.1 Stage 1 - Estimated hole frequencies per hectare

Category	Pinholes	Holes	Tears
"Excellent" case	0,0,5	0,0,2	0,0,0
"Good" case	0,10,15	0,5,10	0,2,5
"Fair" case	0,20,30	0,10,20	0,5,10

Stage 2

At the end of Stage 2, the critical consideration is whether a fixed ELL system is installed and frequently monitored during waste placement. Three grades of site are identified for Stage 2 – the "excellent" case with a fixed, regularly monitored ELL system and the "good" and "fair" cases where no fixed ELL system (or other effective hole detection system) is provided. The hole frequencies for the three grades given in Table 9.2 are derived from a review of the data presented in Section 8.2. The cumulative hole frequencies of Stages 1 and 2 are given in Table 9.2.

Table 9.2 Stage 2 – Estimated hole frequencies per hectare

Category	Pinholes		Holes		Tears	
	Stage 2	Cumulative	Stage 2	Cumulative	Stage 2	Cumulative
"Excellent" case	0,0,3	0,0,8	0,0,1	0,0,3	0,0,0,2	0,0,0,2
"Good" case	1,5,8	1,15,23	0,2,5	0,7,15	0,1,2	0,3,7
"Fair" case	2,10,15	2,30,45	1,5,10	1,15,30	1,3,5	1,8,15

Stage 3

During this stage, no additional perforations develop in the geomembrane liner. The duration of this stage may be modified depending on the stress crack resistance of the geomembrane and the quality of design and installation. The period ranges from 10 years (based on currently available monitoring data) to a reasonable extrapolated maximum of 50 years for the "excellent" category. The prerequisite of reasonable design standards, avoiding large-scale stresses and catastrophic failures, as previously discussed, remains.

Stage 4

No field or laboratory data are available to assist in predicting the development of stress cracks in this and subsequent stages. The actual evolution of these holes will depend on the factors identified in Section 7.7 and how these may relate to a specific site. Individual cracks could combine to form much longer cracks where larger scale stresses are imposed. A tentative "excellent" case estimate of stress crack development may be made on the basis of:

- approximately 1900m of seams with minimum use of fillet extrusion welding per hectare;
- simple cell geometry;
- the provision of high standard geomembrane protection (discussed in Section 9.4 below);
- careful procedures to minimise wrinkles in the covered liner (discussed in Section 9.4 below); and
- design of the landfill to avoid large-scale stresses as discussed in Section 8.4.

The tentative estimate of the incidence of stress cracks per hectare throughout Stage 4 for "excellent", "good" and "fair" cases is given in Table 9.3. For the "excellent" case, this is based on 20 – 40 stress cracks developing, mainly on seams or wrinkles, with a 50/50 apportionment between the minimum and maximum sizes. It is more difficult to select a tentative "fair" case estimate as there are many factors that could lead to significant long-term stresses in the liner and the development of numerous stress cracks. If widespread stresses caused, for example, by waste settlement, stability restraint or geotextile protector failure were present in the liner, this could lead to progressive stress cracking, crack propagation and liner failure as an effective barrier. Thus, in these circumstances, Stage 4 could be the end of the liner service life. However, if widespread stresses can be demonstrably discounted, then a tentative estimated "fair" case incidence of stress cracks per hectare in Stage 4 is given in Table 9.3. This assumes that:

- the liner protection quality was not of the highest standard;
- wrinkles and folds have not been prevented during liner construction;
- the CQA quality was only of a moderate standard;
- a relatively high amount of fillet extrusion welding has been necessary;
- the cell geometry is complicated; or
- the leachate drainage system design/construction causes abnormal liner stresses.

The "good" case is an intermediate condition. Although many stress cracks may be associated with existing holes and enlarge those holes, for the purpose of the model and for leakage calculation, the stress crack frequencies given in Table 9.3 are for new holes, additional to those arising in Stages 1 –3. The duration of Stage 4 extends from the end of Stage 3 to the estimated time of the onset of oxidation of the geomembrane.

Table 9.3 Stage 4 – Tentative estimate of stress crack incidence throughout the stage (defects per hectare)

Category	Stress crack size	
	10 mm ²	1000 mm ²
"Excellent" case	10 - 20	10 - 20
"Good" case	20 - 50	20 - 30
"Fair" case	50 - 100	30 - 40

Note that the total number of stress cracks for each category is the addition of the 10 mm² and 1000 mm² cracks. The selection of these two sizes of stress cracks is obviously a simplification for the conceptual model and stress cracks will vary between these two sizes and beyond.

Stage 5

The time to the onset of oxidation is addressed in Section 5.5.10. This length of time depends critically on the activation energy of the antioxidant loss and the predicted average landfill temperature. From Section 5.5.10, the periods to the commencement of oxidation from the time of liner installation are conservatively estimated for different temperatures and an activation energy of 70 kJ/mol as:

15°C	1500 – 2100 years
20°C	900 – 1300 years
25°C	570 – 800 years
30°C	350 – 500 years
35°C	220 – 320 years
40°C	150 – 210 years

The range of values for each temperature comes from the antioxidant depletion model derivation discussed in Section 5.5.10 (Table 5.6) and represents the upper and lower time limits for the complete depletion of the antioxidants, neglecting any oxidation induction period.

Extensive additional cracking and increasing length of existing cracks are assumed to occur whenever the liner is under even low stress. Indicative predictions of additional stress cracks that would occur over a relatively short time period are given on Table 9.4. The stress crack frequencies given in Table 9.4 are for new cracks, additional to the holes arising in Stages 1 - 4. It is emphasised that there are no research results on which to base the assumed incidence and it is made as a reasonable judgement, subject to revision when further data become available. Similarly, there are no reliable research data on which to base the duration of Stage 5. From Rowe (1998), a period of at least 25 years was obtained from the observation that unstabilised 25 year old intact plastic (of undefined type) had been found in landfills. Based on this observation and the research results of Müller and Jakob (2003) who observed the oxidation of some of the samples tested, a figure of 50 years has been selected as a reasonable estimate.

Table 9.4 Stage 5 - Tentative estimates of stress crack incidence during oxidation (defects per hectare)

Category	Stress Crack Size			
	10 mm ²		1000 mm ²	
	Stage	Cumulative	Stage	Cumulative
"Excellent" Case	20 - 40	30 - 60	10 - 20	20 - 40
"Good" case	40 - 60	60 - 110	20 - 30	40 - 60
"Fair" Case	60 - 140	110 - 240	30 - 70	60 - 110

Stage 6

The conceptual model assumes that further holes will be generated in Stage 6 although the primary cause of additional holes is expected to be stress cracking. It is tentatively estimated

that the number of cracks, pinholes, holes and tears present at the end of Stage 5 will again occur every 100 years. It is believed that this assumed rate of growth of all types of holes is reasonably conservative. A limiting condition will be met for composite liners when the holes, cracks, etc. in the geomembrane are so numerous that the wetted areas beneath the holes overlap and the leakage through the composite liner is limited to that through the mineral liner.

It is emphasised that there is no available research regarding hole generation in Stage 6 on which to rely, so the assumptions given above are subject to change when suitable research findings become available.

9.5 Worked examples

To illustrate the evolution of the four types of holes through the geomembrane using the conceptual six-stage model described above, two landfill scenarios are considered. In these examples, the most likely value within the PDFs has been used as are the averages of the range of hole incidence in Stages 4 and 5.

9.5.1 Example 1

The first is a new, high profile commercial site developed to the highest standards for acceptance of hazardous wastes. A fixed ELL system is to be installed, the design is to be undertaken by engineers with known expertise in landfill design and the design is to be subjected to a third party quality assurance review. A single composite liner of a geomembrane over a CCL is proposed. The 2.5mm thick HDPE geomembrane liner will meet the GRI GM 13 specification and will have a SP-NCTL value for SCR of >1000 hours and OIT also meeting the BAM criteria. The geomembrane will be well protected with a geosynthetic and mineral protection layer. Installation will be by experienced, certified installers working to a specification including detailed placement requirements to minimise wrinkle formation. CQA will be by independent, trained and experienced CQA engineers working to a detailed CQA plan prepared for this high standard site. Cover material placement and deposition of the first layer of waste will be continuously monitored. The ELL system will be monitored quarterly until completion of waste placement and at least annually thereafter. The waste materials have an estimated average temperature at the liner of 25°C, and the time to commencement of oxidation is estimated as 650 years.

This site is classed in the "excellent" category. The estimated lifetime of the landfill is 1500 years after which time it is considered that the wastes will have stabilised to a benign condition. The duration of each stage is estimated as shown in Table 9.5.

Table 9.5 Stage duration in "excellent" case scenario

Stage	Duration (years)	Cumulative (years)	Notes
1	0	0	Landfill construction
2	2	2	Period of landfilling
3	48	50	No hole generation during this stage
4	600	650	Oxidation commences 650 years after construction
5	50	700	Period of further stress cracking during oxidation
6	800	1500	Continuing deterioration

The estimated development of holes per hectare is given in Table 9.6.

Table 9.6 Development of holes per hectare in "excellent" case scenario (by stage and cumulative)

Hole type	Stage											
	1		2		3		4		5		6	
	Stage	Cum.	Stage	Cum.	Stage	Cum.	Stage	Cum.	Stage	Cum.	Stage	Cum.
Pinholes	0	0	1	1	0	1	0	1	0	1	8	9
Holes	0	0	1	1	0	1	0	1	0	1	8	9
Tears	0	0	0	0	0	0	0	0	0	0	0	0
Cracks	0	0	0	0	0	0	30	30	45	75	600	675

The number of stress cracks, pinholes and holes developing in Stage 6 are (1 x 8) times the cumulative holes at the end of Stage 5, as Stage 6 is 800 years long.

9.5.2 Example 2

The second scenario is a landfill for the use of a single waste producer to accept non-hazardous waste. Waste quantities are relatively low and each completed cell of the landfill will take 4 years to fill, with the liner exposed to a greater risk of damage during this period. A single composite liner of 2.0mm HDPE geomembrane over a CCL is proposed. No fixed ELL system will be installed but a mobile ELL survey will be undertaken at the end of liner construction. The design will be carried out by consultants with some experience in landfill design but the site has some complex geotechnical conditions (weak sub-grade and high groundwater table). The geomembrane will meet the GRI GM13 specification except that the SP-NCTL value of the geomembrane is 150 hours. CQA is to be provided by the designer who has no site staff with significant CQA experience. No particular criteria have been specified for the installers. The biodegradable content of the wastes is very low and the estimated average liner temperature is 15°C.

This site is judged to be in an "intermediate" category, closer to "fair" than "good". In view of the low temperature, the onset of oxidation is not estimated to commence until approximately 1500 years has elapsed after installation. The estimated lifetime of the landfill is 750 years (from an assessment of waste degradation rates) after which the wastes are deemed to be in a "stabilised" condition. The estimated duration of each stage is shown in Table 9.7 and the estimated development of holes per hectare is given in Table 9.8.

Note that the landfill is deemed to have stabilised at 750 years, half way through Stage 4. Stages 5 and 6 do not apply as the HDPE geomembrane degradation will not have reached these stages by the end of the 750 years.

Table 9.7 Stage duration in this "intermediate" case scenario

Stage	Duration (years)	Cumulative (years)	Notes
1	0	0	Landfill construction
2	4	4	Period of landfilling
3	6	10	No hole generation during this stage
4	1490	1500	Oxidation does not commence until 1500 years after construction. Landfill stabilises during Stage 4.
5	n.a.	n.a.	Landfill stabilised at 750 years
6	n.a.	n.a.	Landfill stabilised at 750 years

n.a. = not applicable

Table 9.8 Development of holes per hectare in this "intermediate" case scenario (by stage and cumulative)

Hole type	Stage											
	1		2		3		4		5		6	
	Stage	Cum.	Stage	Cum.	Stage	Cum.	Stage	Cum.	Stage	Cum.	Stage	Cum.
Pinholes	5	5	10	15	0	15	0	15	-	-	-	-
Holes	2	2	5	7	0	7	0	7	-	-	-	-
Tears	0	0	3	3	0	3	0	3	-	-	-	-
Cracks	0	0	0	0	0	0	40	40	-	-	-	-

9.6 Factors affecting stage duration and hole frequency

Factors that can influence the stages of geomembrane barrier deterioration as a hydraulic barrier and the incidence (including sizes) of holes are addressed below. Action taken on these issues may justify the selection of less conservative parameters, resulting in a longer duration of some stages, lower computed leakages of leachate or gas emissions and an extended geomembrane liner service life.

9.6.1 Design

The principal concerns of the designer with respect to liner integrity and durability include:

- the specification of a geomembrane with SCR and OIT performance characteristics appropriate for the application;
- avoiding instability or large scale stresses or strain incompatibility;
- providing suitable protection for the geomembrane with a durability compatible with the desired service life of the geomembrane liner;
- as far as practicable, avoiding features that may lead to stresses in the liner.

Design quality assurance, where a design is reviewed by an experienced third party landfill designer competent in geosynthetics design, can provide important, additional confidence in the design.

Geomembrane specification

Currently, HDPE geomembranes for landfill liners in the UK are commonly offered by manufacturers to comply with, or close to, the GRI GM13 specification. This specifies a set of minimum properties to be met or exceeded. The recent 50% rise in the stress cracking SP-NCTL criterion to 300 hours may cause a difficulty to some geomembranes. The GRI GM13 or BAM OIT performance criteria (see Section 5.5.5) should be met including the OIT (standard) value and the oven ageing criteria. For exposed geomembranes, the UV resistance criteria should also be achieved.

It is logical that the liner specification should be appropriate for the application. It is not reasonable practice to specify the same material requirements for a small water reservoir and for a hazardous waste landfill. It may be appropriate to specify a thicker sheet for enhanced survivability, a substantially higher SP-NCTL criterion and an improved OIT performance for the more critical sites. The typical value of the SP-NCTL should be provided by the manufacturer rather than merely that the geomembrane exceeds the GRI GM-13 criterion, as the time to failure can vary by more than a factor of ten from one liner to another, although both may pass the 300 hour criterion.

Instability and large scale stresses

As well as ensuring stability against slope failure, the designer should be aware of the potential effects of strain incompatibility between different, adjacent natural or synthetic materials. Waste settlement is inevitable to some degree but the effects can be addressed by sensible application of interface shear strengths. Cap and basal settlements on yielding sub-grades can also be satisfactorily addressed by design, for example, using ground improvement techniques and geosynthetically reinforced layers to reduce differential settlements and angular distortions.

Geomembrane protection

Geomembranes are relatively delicate construction materials so that protection of the geomembrane from damage is essential at all stages of manufacture, installation and their service life. The degree of protection necessary for the installed geomembrane should be commensurate with the potential for physical damage and the imposed stresses, particularly from the drainage medium, the subgrade and other adjacent materials. Protection from waste articles can be afforded by drainage gravel and by the placement of selected waste in the first lifts of waste.

Design of geotextile protection material is currently based on the cylinder test (Environment Agency, 1998). The evaluation criteria previously used in the UK (0.25% indentation strain) has recently been confirmed to be specific to geomembrane and drainage stone characteristics as used in Germany. Thus, a review of the criteria used in the UK may be necessary (Seeger and Müller, 2003).

The design of the protection needs to ensure that the durability of the protection is comparable with the desired service life of the geomembrane liner. The long-term durability of geotextile protectors has not received the same level of investigation as HDPE geomembranes but it is clear that their durability is significantly less than that of HDPE geomembranes (Müller *et al.*, 2003). To ensure reliable long-term protection from overlying drainage gravel, mineral protectors such as sand layers will be necessary.

Stress inducing features

Leachate wells, sharp changes in liner direction, slope drainage flaps welded on to geomembrane lined slopes and inadequate bedding of drainage pipework are examples of features with the potential to induce stresses. Consideration should be given to means of minimising or mitigating stresses transferred to the liner.

9.6.2 Installation

The primary influencing factors on the quality of the finished installation are:

- a well crafted technical specification;
- installation by well trained, experienced personnel in reasonable weather conditions;
- independent CQA by well trained, experienced personnel; and
- ELL surveys.

Commercial constraints and inclement weather can place undue pressure on site staff and particular attention to maintaining a high standard of installation is essential in these circumstances.

The presence of holes or wrinkles in the geomembrane will substantially increase the leakage rate. Rowe (1998) describes a method of calculating leakage through holes in wrinkles. Evidence now available (Soong and Koerner, 1998) indicates that wrinkles will not flatten out completely and are likely to remain even under high landfill pressures and elevated operating temperatures. Wrinkles in a liner at construction stage can become folds under the weight of the overlying waste (Koerner *et al.*, 1999). Wrinkles not only give the potential for increased leakage but can also cause local tensile stresses that may lead to post-installation holes, for example, by stress cracking. Placement of a wrinkle-free geomembrane is difficult and time-consuming, and relies on reasonable weather conditions and careful scheduling of procedures and resources (Averesch and Schicketanz, 2000). However, the objective of a wrinkle-free liner in good contact with the subgrade is achievable by a high standard installation.

The quality of geomembrane panel seaming is another aspect of installation where improvements in techniques and equipment have reduced the incidence of defects over the years. The change from extrusion to fusion welds has been a primary reason for this improvement. Fillet extrusion welding demands a high level of skill and the reduced amount of this type of welding now undertaken means that the necessary skill and experience are even more difficult to obtain. The result may be that where fillet extrusion welds are required, achieving high quality welds is becoming more difficult. While holes in welds have reduced, less

obvious latent damage can be caused in forming hot wedge fusion welds by unsuitable equipment (Thomas *et al.*, 1995) and a high standard installation would avoid such damage.

Considerable research into developing a practical means of improving quality control of hot wedge welds has been undertaken at BAM (Lüders, 2002). Consideration of the findings of that research and the potential for introducing improvements in the quality control of fusion welds in the UK may lead to greater confidence in the long-term integrity of these seams.

The benefits of high standard CQA are well known. The difficulty is actually obtaining this standard as CQA has become a commodity service, often suffering from excessive commercial constraints and high staff turnovers, which can make the provision of adequate training problematic. To achieve the "excellent" case condition, high standard CQA would have to be demonstrated by the provision of an independent, "hands-on" experienced and well-trained CQA team. The evidence of the benefit of providing ELL surveys at the end of construction has been amply proven by numerous authors, with Section 8.1 summarising many of their findings.

9.6.3 Waste type, infilling practices and leachate management

It is difficult to ensure the adoption of best practice throughout waste placement to prevent physical damage to the geomembrane. The provision and frequent monitoring of a fixed ELL system is the most effective means currently available of detecting penetrative damage to the liner, enabling repairs to be made. The costs of this facility have to be weighed against the benefits gained from the use of a greater hole frequency in the risk assessment.

The types of waste, placement rate and leachate management will have a major influence on the service temperatures at the geomembrane liner. There may be means available at a proposed site to reduce liner temperature, which would then lead to a longer time before the onset of stress cracking (Stages 4 and 5) and oxidation (Stage 6).

Leachate management and the duration of control of leachate levels will have a direct impact on leakage quantities passing through the geomembrane liner. Rising leachate levels following the cessation of leachate pumping will create a greater head, while liner defects at higher levels will become submerged by leachate for the first time.

9.6.4 Caps

The characteristics and operational conditions of geomembrane caps differ from those of a basal liner in many respects:

- they are usually thinner (e.g. 1.0 or 1.5 mm) and often made of LLDPE rather than HDPE;
- they are often single barriers rather than an element of a composite barrier;
- temperatures are likely to be lower but the availability to oxygen will be higher;
- the susceptibility to damage from future human activities is much higher;
- the underlying waste will settle, potentially causing long term stresses in the geomembrane;
- there is the potential to replace (or cover over) the geomembrane with a new cap in the future;

- the head of water on top of the cap will depend upon the continued effectiveness of the overlying drainage, as well as on the amount of rainfall and the site afteruse; and
- the effectiveness of the geomembrane cap in controlling infiltration into the waste (coupled with the leachate management) will dictate the eventual leachate head and leakage at the base, unless groundwater inflow is also occurring.

Referring to the six stage model of the service life of liners, Stages 1 and 2 essentially combine into one installation and construction stage for a cap. The quiescent Stage 3 will not apply as the landfill settlement is likely to cause stresses to be imposed on the geomembrane cap. Stages 4, 5 and 6 will apply in modified form in view of the different exposure conditions and the potential for additional physical damage to the cap from external activities.

The antioxidant depletion time of an HDPE geomembrane cap may be estimated from the discussion in Section 5.5 using air exposure and reducing the time proportionally if the thickness is less than 2.0 mm. There is little published information on physical damage to caps caused during the construction of the cap and less on post-construction damage; some geomembrane caps are installed with panel overlaps rather than welded seams. Human interference is likely to be a major cause of new holes generated during the effective lifetime of the cap, resulting from activities associated with the long-term after-use of the landfilled area. Stress cracking will occur where the geomembrane is under long term stress. The majority of the landfill settlement is likely to take place in the early part of the oxidative degradation of the geomembrane and the resulting stresses could potentially initiate stress cracking at an early stage in the geomembrane cap. Residual settlement or other stresses will eventually lead to stress cracking.

Geomembrane caps of LLDPE are unlikely to be subject to stress cracking until the LLDPE reaches the oxidation stage, when it will then become brittle and susceptible to cracking. However, the LLDPE will degrade more quickly than HDPE, reaching the oxidation stage (Stage C, Hsuan and Koerner (1995)) much earlier than HDPE. Results of laboratory research on the oxidative degradation of LLDPE geomembranes are not yet available, preventing estimates of material durability to be made on a similar basis to those on HDPE geomembranes.

9.6.5 Summary of actions to minimise geomembrane hole generation

A summary of the key actions to take to minimise the generation of defects in geomembrane liners is given in Box 9.1 at the end of this chapter.

9.7 Future research needs

As given below this project has identified several issues where there would be clear benefits of further research effort to the understanding of polyethylene liner durability.

- Long-term monitoring of temperatures at the landfill liner (this may equally be done at sites without geomembrane liners) using thermocouples at the liner. Waste types, placement history and leachate management would be key variables.
- Confirmation or otherwise of the two-stage antioxidant depletion observed by Müller and Jakob (2003), and the relevant activation energy values.
- Assessment of the duration of the oxidation induction period in different exposure conditions.

- Antioxidant depletion and oxidation induction of LLDPE.
- Long-term durability of geotextile protectors.
- The application of improved liner installation and welding quality control techniques in the UK.
- Reliability of the OIT performance criteria in reflecting the long-term antioxidant depletion behaviour.
- Antioxidant depletion in incubation cells which seek to replicate geomembrane/compacted clay composite liner.
- The effects of metal content in leachates on antioxidant depletion and the ability of geomembranes with metal de-activator additives to resist this form of antioxidant depletion.
- Long term monitoring of oxygen concentrations directly above and below geomembrane barriers on the base, side slopes and caps.
- Linkage of stress crack resistance and OIT performance to a material durability factor (MDF) as proposed by Peggs *et al.* (2002).

9.8 Risk assessment modelling

The conceptual model of defect generation presented in this report can be applied to probabilistic as well as deterministic groundwater risk assessment models. LandSim is a commonly used computer model for assessing risk to groundwater quality from landfill sites (Environment Agency, 2001). LandSim 2.5 (Environment Agency, 2003b) includes the facility to simulate the longevity of the engineered containment system, including both the landfill cap and the artificial sealing liner.

The liner degradation assumptions of some models, including LandSim 2.5, are likely to require modifications to be made to the hole generation model developed from this research project in order to fit the restricted input requirements of the risk assessment model. However, it will often be the case that increased leakage caused by rising leachate levels once leachate management has ended will be the principal factor controlling the impact on groundwater quality.

Box 9.1 Key actions to minimise geomembrane hole generation

- Select a geomembrane that exceeds the GRI GM 13 specification.
- Specify a thicker geomembrane to attain greater survivability and resistance to oxidative degradation.
- Design the containment system to ensure that the geomembrane liner is not exposed to avoidable stresses and that large-scale failures do not occur.
- Have design quality assurance undertaken by a suitably experienced third party engineer.
- Ensure that the durability of geomembrane protection materials is at least comparable to the desired service life of the geomembrane.
- Prepare a well-crafted, site-specific technical specification and CQA Plan.
- Install the liner in fair weather conditions using installers certified to the British Geomembrane Association / The Welding Institute scheme and monitored by well experienced CQA inspectors with the objective of achieving a wrinkle-free and undamaged liner.
- Full-time CQA monitoring of placement of protection and drainage materials, and the first layer of waste.
- Install a fixed ELL system and monitor quarterly until the completion of waste disposal and annually thereafter.
- The working practices at the site to hold the maintenance of the integrity of the geomembrane liner as a priority.

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GLOSSARY AND ABBREVIATIONS

Definitions provided are for the common use of terms and acronyms in polymer degradation, geosynthetics and geotechnics in landfill engineering.

Activation energy	The minimum energy required for a chemical reaction to take place. It is the energy barrier that has to be overcome for the reaction to proceed.
Ageing	(1) the effect on materials of exposure to an environment for an interval of time. (2) the process of exposing materials to an environment for an interval of time.
Antioxidant	A substance or chemical used to halt an oxidation reaction.
Asphalt	Well graded aggregate with a bituminous binder and filler.
Asphaltic concrete	One of the hot-mix types of construction material, which consists of a dense-graded mix of stone and sand aggregate, filler and bitumen.
Autoxidation	An oxidation reaction which proceeds only when another oxidation reaction is occurring simultaneously in the same system.
BES	Bentonite enriched soil. Bentonite is added to soils (usually sand) in order to produce a low-permeability lining material.
Basal heave	Upward movement of the base of the landfill / excavation.
Bitumen	A semi-solid product (at room temperature) obtained from a refined asphaltic – base crude oil.
Branched polyethylene plastics	Those containing significant amounts of both short-chain and long-chain branching and having densities in the 0.910 to 0.940 gcm ⁻³ range.
CCL	Compacted clay liner.
CGB	Clay geosynthetic barrier.
Chain reaction	A reaction that is self-sustaining as a result of the products of one step initiating a subsequent step usually involves free radicals as intermediates.
Copolymer	A polymer formed from two or more different monomers.
CQA	Construction quality assurance. The process of checking the quality of materials, construction and compliance with the design.
Crazing	Apparent fine cracks at or under the surface of a plastic. The crazed

areas are composed of polymeric material of lower density than the surrounding matrix.

Crosslinking	The formation of a three dimensional polymer by means of inter-chain reactions resulting in changes in physical properties.
Crystalline region	Region of a polymer having the regular internal arrangement of atoms, ions or molecules characteristic of crystals.
DAC	Dense asphaltic concrete. A dense graded mix of stone and sand aggregate, filler and bitumen with an air voids ratio <3%.
DSC	Differential scanning calorimeter.
Diffusion	The migration of atoms, molecules, ions, or other particles as a result of a concentration gradient.
Degradation	A deleterious change in the chemical structure, physical properties, or appearance of a plastic.
E_a	Activation energy.
ELL	Electrical leak location – acronym used in this document to cover the various terms for this survey method.
FML	Flexible membrane liner.
FRR	Flow rate ratio. A ratio of two different melt index values.
Film	In plastics, an optional term for sheeting having a nominal thickness not greater than 0.25mm (0.01 in.).
Free radical	Molecule or ion with impaired electrons and hence generally exceedingly reactive.
Free volume	The space between polymer molecular/chains. Due to the regions of high structure, semi-crystalline materials have a relatively low free volume. The random spaghetti-like structure of amorphous materials produces a much higher free volume.
GCL	Geosynthetic clay liner.
Geocomposite	Manufactured, assembled material using at least one geosynthetic product among the components.
Geomembrane	An essentially impermeable geosynthetic composed of one or more synthetic sheets.

Geosynthetic	A planar product manufactured from polymeric material used with soil, rock, earth, or other geotechnical engineering related material as an integral part of a man-made project, structure, or system.
Geological barrier	The <i>in situ</i> geological formation below the constructed liner that provides sufficient attenuation to ensure that no unacceptable discharges are made to groundwater.
Geosynthetic clay liner	A low permeability sheet constructed from a thin layer of clay bonded together with one or two layers of geosynthetic material that is used as a liquid and vapour basin in geotechnical and civil engineering applications.
Geotextile	A permeable geosynthetic comprised solely of textiles.
Glass transition temperature	The temperature at which a phase change occurs within polymers changing their characteristics from relatively hard, and glass like to soft and rubbery. This change occurs due to increased chain mobility at higher temperatures.
GM	Geomembrane.
GRI	Geosynthetic Research Institute.
HDPE	High density polyethylene.
HP-OIT	High pressure oxidative induction time.
High density polyethylene	Those linear polyethylene plastics, having a standard density of 0.941 g/cm ³ or greater.
Homopolymer	A polymer resulting from polymerisation involving a single monomer.
Hydroperoxide	A class of compounds containing the hydroperoxyl group, -OOH.
Linear low density polyethylene plastics (LLDPE)	Those linear polyethylene plastics, having a standard density of 0.919 to 0.925 g/cm ³ .
Linear medium density polyethylene plastics (LMDPE)	Those linear polyethylene plastics, having a standard density of 0.926 to 0.940 g/cm ³ .
Linear polyethylene plastics	Those containing insignificant amount of long-chain branching but which may contain significant amounts of short-chain branching.
Low density polyethylene plastics (LDPE)	Those branched polyethylene plastics having a standard density of 0.910 to 0.925 g/cm ³ .

MBP	Mechanical and biological pre-treatment of wastes.
Medium density polyethylene plastics (MDPE)	Those branched polyethylene plastics having a standard density of 0.926 to 0.940 g/cm ³ .
Melting point	The temperature at which the solid and liquid phases of a substance are in equilibrium at a specified pressure (normally taken to be atmospheric unless stated otherwise).
MI	Melt index.
MFI	Melt flow index.
Monomer	A low-molecular-weight substance consisting of molecules capable of reacting with like or unlike molecules to form a polymer.
MSW	Municipal solid waste.
OIT	Oxidative induction time.
Olefin	Aliphatic hydrocarbon of the general formula C _n H _{2n}
Oxidation	Process whereby a loss of electrons normally involves the combination of oxygen with another element to form one or more new substance.
PDF	Probability density function
PP	Polypropylene.
PVC	Polyvinyl chloride.
Peroxide	Compounds of structure ROOR in which R may be any organic group.
Photo-oxidation	Oxidation induced by light or ultraviolet radiation.
Polyethylene	A polymer prepared by the polymerisation of ethylene as the sole monomer.
Polymer	A substance composed of molecules of high relative molecular mass (molecular weight), the structure of which essentially comprises the multiple repetitions of units derived, actually or conceptually, from molecules of low relative molecular mass. A single molecule of a polymer is called a macromolecule.
Polymerisation	A chemical reaction in which the molecules of monomers are linked together to form polymers.

Polyolefin	A polymer prepared by the polymerisation of an olefin(s) as the sole monomer(s).
Polypropylene	A polymer prepared by the polymerisation of propylene as the sole monomer.
R_x	Reactive free radical.
RH	Polymer chain.
ROO_x	Hydroperoxy radical.
ROOH	Hydroperoxide.
S	Antioxidant depletion rate.
SCR	Stress cracking resistance.
Side chain	A chain that is attached to the main chain of the polymer.
Std-OIT	Standard OIT
Stress crack	An external or internal crack in a plastic caused by a tensile stress less than its short-term mechanical strength.
Thermoplastic	A plastic that repeatedly can be softened by heating and hardened by cooling through a temperature range characteristic of the plastic, and that in the softened state can be shaped by flow into articles by moulding or extrusion.
Thermoset	A plastic that, after having been cured by heat or other means, is substantially infusible and insoluble.
Tie chain	Molecule chain contained in amorphous region that links crystal lamellae.
Transition metals	Elements characterised in the periodic table by a partially filled "d" sub-shell. The First Transition Series comprises Sc, Ti, V, Cr, Mn, Fe, Co, Ni, Cu. The Second and Third Transition Series include the lanthanides and actinides, respectively.
VFPE	Very flexible polyethylene

ANNEX 1: SCOPING STUDY: LONG-TERM DURABILITY OF NON-POLYETHYLENE SYNTHETIC LANDFILL LINERS

A1.1 Introduction

There are a number of non-polyethylene synthetic liners available for use as landfill liners including:

- geosynthetic clay liners (GCLs);
- dense asphaltic concrete (DAC);
- bituminous membranes;
- other polymeric geomembranes such as polypropylene (PP) and PVC liners.

Bentonite enriched soils (BES) and Trisoplast (a proprietary BES with polymer additive) are classed as mineral rather than synthetic liners. For the purposes of this report, DAC is classed as a synthetic rather than mineral liner. Of the list above, only GCLs and DAC are currently used in the United Kingdom to a significant extent and have been selected as the subject of a scoping study to determine the availability of data on defects and degradation of liners made from these materials. The objective is to provide recommendations on an appropriate investigation of failure mechanisms and defects of these alternative liners, if it is considered it would be beneficial to the understanding of the medium to long-term behaviour of landfill liners made from these materials.

The exclusion of the other lining materials from the scoping study in no way pre-judges their suitability or otherwise as landfill liners.

A1.2 Geosynthetic clay liners

A1.2.1 Background

Geosynthetic clay liners (GCLs), also known as Clay Geosynthetic Barriers (CGBs), may currently be categorised into four types:

- geotextile-encased, adhesive-bonded type (unreinforced);
- geotextile-encased, stitch-bonded type (reinforced);
- geotextile-encased, needle punched type (reinforced);
- geomembrane-supported, adhesive-bonded type (unreinforced).

The encasing geotextiles may be non-woven or woven, they are usually made up of polypropylene fibres and the bentonite is usually sodium bentonite in granular form.

With respect to the durability of GCLs, the durability of each constituent of the GCL must be considered as well as the GCL as a whole. The medium to long-term failure mechanisms of GCLs can be summarised as follows.

- Effects of permeating liquids on the long-term permeability of the bentonite.
- Thinning of the bentonite core by uneven loading causing bentonite migration.
- For caps, repeated wet-dry and, to a lesser extent in the UK, freeze-thaw cycles leading to increased permeability.

- Chemical degradation (e.g. oxidation) of the upper and lower polyolefin geotextiles leading to disintegration and loss of confinement of the bentonite.
- Mechanical failure of the stitch-bonding or needle-punching connecting the upper and lower geotextiles when under long-term shear stress, either before or as a result of chemical degradation.
- Physical damage (puncture) or pulling apart of adjacent GCL panels.
- Biological effects in GCLs include potential root action on GCLs in caps and the effects of microorganisms on the GCL itself. Work has been conducted into these effects and further research is not considered a priority.

The large surface area / volume ratio of geotextile fibres compared to geomembrane sheet, as well as the fact that many of the encasing geotextiles are of polypropylene rather than polyethylene mean that the geotextiles potentially will degrade significantly more rapidly than HDPE geomembrane liners.

A1.2.2 Literature review

There is a substantial body of literature on GCL use, applications, testing and design. Key sources for a literature review would include the following.

- Geotextiles and Geomembranes (journal).
- Geosynthetics International (journal).
- Geotechnical Fabrics Report (trade journal).
- Geosynthetics Institute (GRI), including proceedings of the annual GRI Conference series; the 14th Conference (2000) has a session on GCL durability and lifetime.
- Munich Technical University's specialist research group "Geosynthetics in geotechnical engineering", which is also the base for the German Chapter of IGS
- Geosynthetics '97 conference proceedings and later.
- Proceedings of Sardinia 93 and later.
- Proceedings of International Geosynthetics Society (IGS) 6th Conference and later.
- EA Guidance on the use of GCLs in Landfill Engineering – useful bibliography.
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- Melchior – numerous articles in German & English on long-term trials conducted at the Georgeswerder Landfill, Germany.
- GRI Generic Specification – GCL3 – in preparation.
- Ongoing test programmes by BAM (Germany) and GRI.

A1.2.3 Mechanisms of failure

Manmade and physical

Manmade failures can occur at several stages including manufacture, delivery, offloading, site handling, deployment of the GCL, deployment of overlying layers and in the early operational filling of the site. Manufacturer quality control (QC) is undertaken to identify and address failures in the production stages. CQA procedures are designed to identify potential and actual failures during the physical works on site, including poor site practices.

GCLs may be subject to settlement of the subgrade, particularly when used as landfill caps. LaGatta *et al.* (1997) conclude that GCLs perform better than compacted clay under differential settlement, citing for example GCL panels subject to 1% to 10% tensile strains with in-plane slippage of 25 to 100mm still maintaining their hydraulic conductivity. Gross movements of the waste body post-placement can induce tensile strains in GCLs. These mechanisms should certainly be assessed as part of the design process (particularly on side slopes where settlement of the waste body is unavoidable) and appropriate allowances made. The waste body itself needs to be carefully managed throughout the operational phase to avoid mass movements due to, for example, over-steep, exposed waste slopes or poor leachate management triggering stability failures.

Physical degradation of GCLs can occur due to degradation of the bentonite core itself. Two mechanisms have been investigated - freeze/thaw effects and desiccation (hydration/dehydration) cycling. Freeze/thaw effects appear less damaging than repeated hydration/dehydration cycles. These mechanisms can directly affect capping GCLs, particularly where appropriate design measures (such as positioning of the GCL below an overlying geomembrane and/or ensuring adequate thickness of overlying soil) have not been incorporated.

These failure mechanisms are considered to be well understood and further research in this area is not considered a priority.

The thickness of GCLs with respect to the potential for puncture is a concern and is a reason for the limited use of GCLs in the UK, despite the positive attributes of GCLs. However, there is little published data on damage to these materials in practice that would benefit from a separate study into this aspect. Focus by designers and installers should be on protecting the GCL so as to prevent penetrative damage being an issue.

Chemical degradation of the bentonite

The core bentonite is commonly natural sodium bentonite, but it can also be calcium bentonite. Calcium bentonite can be modified by addition of soda ash (Na_2CO_3) to increase its swelling potential; this is termed “activated” sodium bentonite. Activated sodium bentonite tends to have higher initial hydraulic conductivity and degrades more quickly than natural (unmodified) sodium bentonite. Activated sodium bentonite has a significantly lower swell index with time and undergoes ion exchange much earlier than unmodified / natural sodium bentonite.

The suitability of GCL as an “adsorption filter”, the ability to bond heavy metal ions based on exchange of sodium ions of the montmorillonite for higher value cations, means it is susceptible to changes in its hydraulic conductivity. Shear strength increases while swell potential decreases, leading to increased hydraulic conductivity and decreased effectiveness as a barrier.

The relatively limited thickness of GCLs in turn limits the amount of adsorption that can take place.

The sensitivity of GCLs to leachate exposure should be assessed as part of the design. High concentrations of neutral polar organics and some organic and strong mineral acids (albeit these are not usually encountered in MSW landfills in high concentrations) can adversely affect the clay structure. There are likely to be compositional changes to “typical” landfill leachates as the implications of the Landfill Directive work their way through the waste cycle. How these permeants will impact upon GCL susceptibility to ion exchange is currently unclear.

Considerable work has been undertaken both at laboratory level and with large-scale field trials, particularly in the USA and Germany. The mechanisms of leachate and bentonite core chemical interactions are generally well understood. However, it would be appropriate to bring together the various strands of work already undertaken in this area in one collated review.

Chemical degradation of the geotextile fibres

There has been limited work on the process of degradation of geotextiles used to form GCLs. The implications of ageing and loss of strength of the reinforcing fibres by oxidation of the polymers within GCLs are significant. The polymer predominantly used in the geotextile elements of GCLs is polypropylene but polyethylene geotextiles are used in at least one product. The bentonite core depends upon the restraint given by the encasing geotextiles and by the reinforcing fibres to provide the confinement of the clay necessary to maintain a low permeability. This confinement enhances the process of pore size reduction that occurs on swelling of the montmorillonite and prevents the core bentonite reaching the gel stage, which is characterised by high moisture content, relatively high permeability, low shear strength and a tendency to migrate away from pressure points. In side slopes incorporating a GCL, the overall stability of the slope ultimately depends upon the reinforcing fibres within the GCL. Long-term reductions or loss of strength, including creep effects, may push the factors of safety below acceptable values.

Compared to HDPE geomembranes, there is generally less attention given by specifiers to the antioxidant protection package used in the manufacture of GCL geotextile fibres. Antioxidants are used as an intrinsic part of the geotextile manufacturing process, but their addition to geotextile resins is less than to HDPE geomembrane resins (Müller *et al.*, 2003). The lower antioxidant protection is exacerbated by the greater surface area to volume ratio of geotextile fibres compared to geomembrane sheet making these fibres more susceptible to early depletion of the antioxidant from the bulk material, leading to oxidation of the fibres and tensile failure.

Some research including laboratory test programmes has been initiated at BAM in Germany and at GRI, USA into long-term degradation of GCL reinforcing fibres. There is a clear need to investigate and quantify likely service life implications of ageing of reinforcement fibres and confinement of the geotextile layers within GCLs for all landfill related applications.

A1.2.4 Recommendations

Considering the gaps in current knowledge, research into the long-term degradation of GCLs is desirable in three priority areas as discussed below.

- Long-term degradation of the reinforcing fibres or stitching of the two types of geotextile-encased GCLs.
- Long-term degradation of the encasing geotextiles themselves in the three types of geotextile-encased GCLs.
- Study of the potential effects of future leachates to the long-term attenuation and permeability characteristics of the bentonite in GCLs.

Ideally, a programme of long term integrity tests would form the central core of a research study into the long-term degradation of both reinforcing fibres / stitching and the encasing geotextiles. However, such tests are themselves long-term. For example, GRI is conducting some tests of this type in a five-year research programme. There are a number of research and university facilities in the UK where such tests could be carried out. An alternative may be to collaborate with a research organisation already conducting similar tests, e.g. GRI or BAM but there may be difficulty in agreeing project objectives, scope of testing, the schedule of the availability of results for publication, etc.

In view of the complex nature of a laboratory test programme, a scope of testing has not been proposed as this would require careful consideration and agreement, but if the Environment Agency is, in principle, prepared to fund a long-term laboratory test programme, then a detailed project scope could be prepared for further consideration.

Whether or not a laboratory programme is commissioned, a literature study should first be conducted into the current "state of the art" on durability of both GCL reinforcing fibres / stitching and the encasing geotextiles. While there is limited literature currently available, it is a subject receiving increasing attention and, at the least, the outcome of the literature search would be the early preparation of interim guidance on GCL durability for use in risk assessments and for regulatory guidance. Effects to be examined would include:

- oxidative and UV degradation of GCL reinforcement fibres and stitching, and of encasing geotextiles;
- long-term loss of confinement and disintegration of encasing geotextiles;
- long-term creep and reduction in internal shear strength.

The literature sources identified in Section 9.2.2 would provide the latest information augmented where possible by direct contact with GRI (through the Environment Agency membership), BAM, ERI and other centres conducting relevant research.

A number of researchers have addressed the issue of GCL leachate compatibility, and the permeability and performance (including attenuation) of GCLs in contact with various permeants that could occur at the base of a landfill. A review of the literature on this subject would result in a report on the "state of the art" on the subject. To extend the value of the literature study, a laboratory study of GCL permeability and performance using synthetic leachate to model current and projected future UK leachates would be beneficial.

A1.2.5 Material standards

In this project, the issue of material standards for geomembrane liners, GCLs and protection geotextiles has arisen. In Germany, BAM sets particular material specifications which have to be

met (and certified) before the materials can be used in landfills. GRI in the USA has published or is due to publish generic material specifications on various geomembranes, protection geotextiles, drainage geocomposites, GCLs and geogrids (available on the GRI website). The GRI standards are being increasingly adopted in the USA as reasonable, well-founded material standards.

In the UK, no such national specifications exist. While the dangers of over-prescriptive regulation should be avoided, it would be beneficial to the industry and regulators alike for there to be a set of consistent minimum Environment Agency specifications for the primary materials used in landfill liner construction. Currently, designers specify the materials on behalf of the operators and there can be a wide spread of material properties specified, not always being appropriate or reasonable. This presents difficulties to the Environment Agency officers in deciding if a specification is inadequate, excessive or reasonable. Standard minimum material specifications would remove this uncertainty and would establish consistent requirements, benefiting regulators, operators, designers and suppliers. This should also lead to improved standards.

It is recommended that a project be commissioned to develop Environment Agency minimum material specifications for HDPE and LLDPE geomembranes, GCLs and protection geotextiles. It is expected that these would be developed from a knowledgeable review of the background of the GRI, BAM and specifications from other respected sources, and discussions with material suppliers.

A1.3 Dense asphaltic concrete

A1.3.1 Background

Dense asphaltic concrete (DAC) is a densely graded combination of stone and sand aggregate, filler and bitumen. The critical parameter to ensure an essentially impermeable material is an air voids ratio < 3% by volume. The materials are pre-mixed hot in batches, then laid and compacted in layers. DAC has been used in dam construction and more recently for lining of lagoons and some landfills. Its use is concentrated in Switzerland and parts of Europe, with two UK landfills having DAC linings.

DAC can be used to form basal liners and manufacturers have developed techniques to use DAC in lining steep side slopes (up to 1H:1.2V) and vertically as part of steepwall lining systems. DAC installation is relatively plant-intensive compared to traditional mineral lining systems. Paving machines, track-mounted finishers, special steering equipment and compactors are required. Slopes require special variants of these machines as well as powerful winching equipment. Less compactive effort is generally delivered on slopes. On slopes, the mix stability must be increased to avoid creep, and this stability adequately demonstrated by testing.

Mix design considerations include permeability, workability, stability (physical and thermal), ensuring adequate bond between layers and avoidance of blister formation. Mix designs will vary dependent on the particle size distribution and angularity of the stone and sand aggregate. The preliminary laboratory and field tests lead to a design which is specified in the contract in terms of an air voids ratio < 3% by volume and a corresponding mass density, proven on site by the number of blows on each side of a Marshall specimen.

A1.3.2 Literature review

There is a relatively recent body of literature, most of it in German, on the use of DAC in landfill applications. A proportion of this derives from work undertaken by manufacturers and installers, however much of it has come from seminars and research undertaken in Germany over the last decade. Key bibliography and sources include the following.

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In terms of volume of data, the major aspects in relation to the use of DAC appear to have been well investigated. These data are however largely inaccessible to anglophone designers and regulators because they are predominantly in German. There is a clear need for a comprehensive review of the key German language sources of data on DAC material, its use in landfill applications and its durability.

A1.3.3 Hydraulic conductivity

The performance of DAC as a hydraulic barrier is in some senses analogous to that of a geomembrane. There is effectively no interconnection between voids in DAC with an air voids content < 3%. This factor means that Darcy's law is not strictly applicable for determining the hydraulic conductivity of DAC. Contaminant migration can occur due to advective flow through holes in DAC and by diffusion through intact DAC.

A1.3.4 Design and installation guidance

The long history of using bituminous, asphaltic membranes in hydraulic engineering and more recent experience of transferring the technology to the differing requirements of landfill liners means that most of the groundwork for a detailed understanding of these materials is already in place. However, much of the knowledge is contained within academic papers and foreign language sources. There is a need by regulators and designers in the UK for a detailed review with the production of a guidance document, independent from those produced by manufacturers and installers.

A review would have several objectives including the following.

- Impartial assessment of the technical merits of DAC as a landfill liner.
- Determination of the appropriate design methodology and input parameters in the context of stability, integrity and assessment of suitable QC and QA test methods.
- Review on current knowledge on long-term durability and degradation of DAC in landfill environments.

There is a need for greater transparency of design methodology and to assist regulatory review of the suitability of proposed solutions for particular sites and available source materials.

The CQA regime required for DAC liners is unlike that for typical mineral liners in terms of the material test requirements, placement specification and qualifications of CQA inspector. Much work has been undertaken already in Germany by the DIBt (German Institute for Construction Techniques) on defining appropriate CQA to ensure that design objectives are achieved. It is recommended these guidelines be reviewed for their applicability to the UK regulatory context.

A1.3.5 Degradation mechanisms

Physical integrity

DAC lining systems are offered for side slopes (up to 1V:1.2H) and even for vertical lining systems, the latter on the basis of the lining system being supported by two granular shoulders, a variant of standard DAC dam core formation techniques. One of the advantages of DAC, namely that it can accommodate some differential settlement without a reduction in performance, means that DAC liners are also susceptible to creep. The most critical physical stresses are considered to arise from settlement. Angular distortion of up to 1:10 is reported to be accommodated in hydraulic structures and greater settlements may be acceptable in the higher temperatures in landfills but the elevated temperatures may lead to excessive creep.

The relatively simple “Van Asbeck” test (Van Asbeck, 1959) is used to determine qualitatively the stability of a DAC batch. Empirical based tests are ideal for rapid site control. However, these should be complemented by appropriate tests of engineering values such that stability analyses can be determined for the full range of loading scenarios (construction, initial waste placement, full height waste and long term loading) to determine the relevant factors of safety.

Biological attack

There are a substantial number of publications referring to biological attack on thin films of bitumen in aerobic conditions. However, there appears to be little reported research on thick layers of asphaltic concrete in the anaerobic conditions of a landfill and basic laboratory research would be needed to investigate the resistance of DAC liners to biological attack.

Chemical integrity

Organic compounds have the potential to cause damage to DAC liners owing to their swelling and solvent actions. In landfills, the low concentrations of the more aggressive organic compounds (non-polar solvents and vegetable and animal oils and fats) and the exposure conditions make it unlikely that these compounds will pose a serious threat to the barrier function of the DAC.

DAC degrades with time, dependent upon the exposure medium (oxygen, leachate, water, etc.), ambient temperature and UV exposure. Ageing of bitumen is linked to thermo-distillative effects (loss of lighter fraction, especially between 30° and 80°C, leading to loss of mass and increase in viscosity), thermo-oxidative reactions and structural changes. Some ageing mechanisms are permanent and some, including structural changes, are reversible. Ageing generally leads to mass loss, although some reactions with atmospheric oxygen (oxidation, polymerisation and polycondensation) can lead to mass gain. Asphalts have different propensities to hardening during the ageing process dependent upon the provenance of their base oil. This is defined by the asphalt ageing index [add reference] (typical range 0.018 for Californian asphalt to 0.071 for Venezuelan asphalt). Although there is coverage in the literature of these ageing issues, the service life of DAC in a landfill context does not appear to have been adequately assessed.

A1.3.6 Recommendations

From published literature, new fundamental research and testing is not warranted at this stage. Although biological attack in a landfill environment appears poorly researched at present, this is not likely to be a critical degradation mechanism. Hence, fundamental laboratory research in this subject is not considered a priority. Further work is required however in two areas:

- the provision, in English, of guidance on the design, CQA and construction aspects of DAC liners;
- assessing the long-term durability / service life of DAC liners from a literature review.

These are described in greater detail below.

Guidance on DAC liners

An independent review of the body of available literature on DAC is required and practical guidance on the design and construction of DAC liners prepared for UK designers and regulators. The guidance should also place DAC in an appropriate UK regulatory context. The review should examine the range of German language sources and it is anticipated that the project would benefit from liaison with the German Institute for Construction Technology (DIBt), German Asphalt Institute (DAI), and the Federal Institute for Materials Testing and Research (BAM).

In particular:

- the review should include an impartial assessment of the technical merits and limitations of DAC as a mineral landfill liner;
- guidance should be developed on design, technical specifications, construction techniques and CQA including suitable QC and QA test methods;
 - a more transparent approach is necessary for input to standard liner risk assessment tools. It is recommended DAC be characterised in terms of hydraulic conductivity and diffusion coefficients for the likely range of contaminants and concentrations;
 - empirical tests, which exist for rapid site control of stability on slopes need to be complemented by appropriate tests of engineering values. Thus, stability analyses can be determined for the full range of loading scenarios (construction, initial waste placement, full height waste and long-term loading) to evaluate relevant factors of safety.

It is recommended that the outcome of this review would form the basis of EA guidance on the use of DAC in landfill engineering (noting that the EA may wish to delay publication until after the work described below is completed, or combine the two efforts).

Long term durability of DAC liners

To assess the potential long term risks to groundwater where DAC is used as a landfill liner, it is necessary to review the likely long-term durability of DAC. This review should consider the various degradation (including physical, chemical and biological) and other failure mechanisms of DAC.

This review would follow on naturally from the review stage described above. Case histories from the UK and abroad should be examined, with reference made to dam construction and other hydraulic engineering case histories where lessons can be drawn from previous failures. The use of DAC in a landfill environment with exposure to the range of typical current and projected future leachates should be examined. Consideration should be given to current changes in legislation likely to affect waste composition and leachate quality and their impacts on DAC liners. DAC as a basal liner should be examined and consideration given to its suitability for use in side slopes and steep-wall systems.

The review should also consider whether new research, including long-term laboratory tests, is necessary.