

# The effect of stress on geosynthetic durability

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**ABSTRACT:** The durability of geosynthetic materials is defined in terms of resistance to physical (eg. abrasion), tensile (eg. creep and stress cracking), mechanical (eg. puncture resistance), oxidative (eg. thermal or UV light) or chemical (eg. immersion) agencies as well as combinations such as weathering. The effect of tensile stress on chemical agency attack of some geopolymers is well-documented but is not considered within the current test methodologies for durability to such agencies. Evidence is presented here which relates the creep sensitivity of polypropylene geotextile tapes to the stress-sensitivity of their thermal oxidation.

## 1 INTRODUCTION

During installation and in use, it is generally accepted that geosynthetic materials are subjected to a range of potentially degrading agencies, usually acting in combinations which vary within themselves and with regard to relative intensity and contribution as functions of time. Typical agencies have been listed and discussed recently (Horrocks and D'Souza, 1989) both to illustrate their complexity and to demonstrate our generally poor understanding of multiagency degradation. Consequently, laboratory ageing tests designed to simulate real exposure to defined agencies are concerned with selection of single agencies at any one time. For instance, exposure to tensile loads enables creep-resistance to be determined. Use of a defined chemical condition may be used to quantify the durability of a specified geosynthetic to a single or mixture of chemicals (eg EPA Method 9090, 1985; ASTM D5322-92) and resistance to normal atmospheric oxidation at ambient temperature may be simulated by elevated-temperature ageing techniques.

This paper does not intend to review current laboratory ageing test methodologies, although the general aspects of geosynthetic durability have been discussed elsewhere (Horrocks and D'Souza, 1992). It will highlight the inadequacies of current test methodologies which demand only single

Table 1. Chemically-degrading agencies and particular geosynthetic material sensitivities.

Degrading agency	Geopolymer*	Sensitising factor
HEAT: Thermal degradation and oxidation	Melt-extruded/processed polymers: PET, N66, PP, PE and PVC	PET, PE & PP; catalyst residues PET & N66; metallic residues & additives
LIGHT: Photodegradation Photo-oxidation	Aramids Polyamides Polyolefins (PE, PP)	Impurities; Delustrant & therm. deg. products impurities;
WATER: Acidic, alkaline or neutral hydrolysis	Cellulosics** Polyamides Polyester	pH, catalyst residues, therm. deg. products
CHEMICAL: Oxidative	Cellulosics Polyamide Polyolefins (PE, PP)	pH, Heavy metals; pH, Therm. deg products,
IONISING RADIATION	All Geosynthetics	
MICRO-ORGANISM	Cellulosics Polyamides	pH, moisture

Notes: \* PET = polyester, N66 = nylon 6.6, PP = polypropylene, PE = polyethylene (LDPE & HDPE);

\*\* Cellulosics = regenerated and natural cellulose fibres.

agency testing and, in particular, focus on the influence of stress on chemical degradation and hence the durability of geopolymers. Table 1 briefly summarises the main chemically-driven degrading agencies and highlights particular geosynthetic sensitivities and sensitising agencies. Many of these sensitising species are associated with the presence of impurity or similar species, or are the products of degradation that have originated during some previous treatment or exposure history. Because of the heterophasic, polycrystalline character of many geopolymers, impurity sites and degraded polymer chain segments will concentrate in the non-crystalline regions and at crystallite surfaces and defect zones. When the polymer is subjected to tensile load, these centres will concentrate stress. Since they are also the points of future chemical activity, it follows that an average stress field across the polymer matrix will be distorted into high concentration zones at these impurities. This has two principal effects, one is that stress may enhance rates of chemical attack (eg. by encouraging chain scission, see Section 3.1 below) and the other is that they will act as microcrack propagation sites once degradation is underway.

Typical of these are residual products of thermal oxidation introduced during melt or heat processing which then sensitise subsequent UV degradation by virtue of their ability to absorb near-UV (290-400nm) radiation to which the polymer itself is transparent. In fact, the UV and hence weathering sensitivity of polyolefins, typified by polypropylene in geotextile fabrics and high density polyethylene in geomembranes, is a consequence of their previous melt-processed histories. Thus thermal stabilisers may be introduced during such process stages which not only reduce the thermal degradative damage but also reduce the potential UV sensitivity of the thermally-stable polymer.

## 2. EFFECT OF TEMPERATURE

All chemical reactions are thermally activated which means that normal chemical degradations have higher rates at higher temperatures; at much higher temperatures, usually above 250°C for the common geopolymers, thermal degradation will occur which, in the presence of atmospheric oxygen, is more usually thermal oxidative in character. During outdoor weathering in tropical

regions, ambient temperatures may be quite high and over time scales of several months may cause thermal oxidative degradation in addition to photo-oxidation. Experiments by Schneider (Schneider, 1990) suggested that in carbon black-filled geosynthetics, the black body heat-absorbing characteristics were great enough to cause very high subsurface temperatures (>100°C) to the extent that observed deterioration during outdoor exposure was a consequence of thermal and not photodegradation. In this case, it may be argued that applied stress might influence both UV and thermally-initiated degradations.

### 2.1 The Arrhenius Law

The temperature sensitivity of all chemical reactions, if they are assumed to be homogeneous in character, are determined by the Arrhenius Law:

$$k = A e^{-E/RT} \dots\dots\dots(1)$$

where k is the reaction rate constant (s<sup>-1</sup>) at temperature T (K), A is the pre-exponential factor (often called the collisional factor), E is the activation energy (J mol<sup>-1</sup>) and R is the gas constant (= 8.3136 J mol<sup>-1</sup> K<sup>-1</sup>). Use of this equation enables accelerated ageing experiments carried out at elevated temperatures to yield predictive lifetime data for the same materials used under ambient conditions (Horrocks, 1992).

The validity of this equation not only assumes that degradation reactions occur in homogeneous environments but also that during the temperature interval between ambient (the "real" condition) and the elevated temperature (the "accelerated" condition), the polymer does not undergo a phase change.

### 2.2 Second order transition temperatures

For many of the commonly used geopolymers, while the melting points are too high to worry about during accelerated testing, their glass or second order temperatures, T<sub>g</sub>, may fall within the range. Examples of second order temperatures are listed in Table 2 from which it is seen that of the common fibre-forming polymers, polypropylene, nylons 6 and 6.6 and polyester have T<sub>g</sub> values between -15 to 75°C. This range, which spans the extremes of terrestrial temperatures, including

black body absorption temperatures in hotter regions.

When a polymer is above is heated above  $T_g$  (and note that in Table 2, LDPE, HDPE, polypropylene and wet polyamides are above respective  $T_g$  values at normal ambient temperatures of 10-20°C), it shows a slight change in its physical properties as a consequence of the increased mobility of non-crystalline polymeric chain segments. In addition this same increased segmental freedom enables greater rates of access by degrading agencies. For example, the polymer structure now becomes especially more accessible to smaller attacking species such as oxygen and water molecules and hydrogen, hydroxide and heavy metal ions (see Table 1). Thus rates of chemical attack increase by more than the Arrhenius equation might predict because of an effective change in the reaction "constants" A and/or E in equation (1) above. However, for polymers which are above their  $T_g$  values at ambient temperatures, it is highly likely that elevated temperature ageing will be less affected by such polymer transitions unless a higher temperature recrystallisation transition occurs.

The implications of  $T_g$  and applied stress are twofold in that a stress concentration zone present when the ambient temperature is below the second order temperature may relax when heated above it (eg. during accelerated ageing). Secondly, if E is stress-dependent (see Section 3.1) then once the polymer is heated above  $T_g$ , E may change because of this and stress relaxation.

### 3. STRESS AND DURABILITY

The consequences of applied tensile load are most commonly evident as creep which has received

Table 2. Second order transition temperatures of the common geopolymers.

Polymer	$T_g$ , deg C	
Low density PE	-100	
High density PE	-100	
Polypropylene	-15 to 10	
Polyamide, Nylons 6 &	Dry	100
	65% rh	45
	Wet	0
Polyester	65% rh	75
	wet	60
Polyaramid	340	

considerable attention within geosynthetic structures for a number of years (Greenwood and Myles, 1986). Its determination is formalised in both national (BS 6906 Pt 5: 1990; ASTM D5262-92) and international standards ( draft prEN) and is usually a required parameter by which most geosynthetic materials must be partially characterised. While it leads to gradual and significant changes in dimensions with time in the first instance, it also gives rise to premature failure as the material flows under stress, thereby creating stress concentrations which ultimately exceed the tensile stress. This reduction in time-to-failure is usually considered to be a consequence of physical flow of mobile polymer chains and may be defined by an equation similar to the Arrhenius equation (1) above where E is the activation energy of polymer chain segmental flow; this represents the energy barrier which must be surmounted if stressed, adjacent chains are to flow past each other.

In support of this mechanism is the observation that creep coefficients decrease as second order transition temperatures increase for the common fibre-forming geotextile fibres, ie

LDPE,HDPE < PP < PET < Polyaramid  
 .....  $T_g$  .....>

<.....Creep coefficient (Horrocks, 1992)....>

However, as will be shown below, considerable evidence exists for the creep process in polyolefins particularly to be chemical degradation driven and related.

#### 3.1 Chemical effects of stress

The application of stress to a polymer can significantly change the rate of chemical attack by degrading agencies. Considerable research has been undertaken in Russia during the last fifteen years or so and this has been extensively reviewed for a range of polymers (Popov, Rapoport and Zaikov, 1991) and more recently, for geosynthetic materials (Horrocks and D'Souza, 1992). While this phenomenon has been widely ignored by the geosynthetic community, it was noted and quantified for polypropylene geotextile yarns in 1982 (Wisse and Birkenfeld, 1982) and has been considerably researched since 1989 by the current author (Horrocks and D'Souza, 1990, 1994) for

the thermal oxidative durabilities of polypropylene tapes. This recent research demonstrated that:

- (i) low stresses have negligible effects on the thermal oxidation of tapes;
- (ii) moderate stresses (upto 10% of ultimate breaking stress) possibly have a slight stabilising effect;
- (iii) higher stresses (above 10% of ultimate) significantly enhance both physical and chemical degradation; and
- (iv) presence of carbon black may reduce the effects of stress upto and including values of 10% of ultimate but may enhance stress-related effects above this level.

These results agree with the observed low/moderate stress-stabilisation of polypropylene to UV degradation (Peeva and Evtimova, 1984) and to thermal oxidation (Czerny, 1972) which gave rise to the concept of a "safe stress" by the latter author. These and related results (Popov, Rapoport and Zaikov, 1991) have been explained in terms of a stress-sensitive competition between reduced oxygen diffusion and favoured polymer chain oxidation at low-to-moderate stresses and enhanced defect formation and growth and favoured chain scission reactions at high stresses. Our research also showed that failure at stresses above 10% of ultimate was accompanied by transverse crack formation which was not evident at lower stresses. These results support the comments made above in Section 1 regarding the possible effect of tensile load on degradation centres in heterogeneous geopolymers.

More importantly and especially to the field of geosynthetic materials, Krisyuk and coworkers (Krisyuk et al., 1983) have demonstrated that the mechanism of creep is driven by the same chain scission reactions that determine failure by thermally-initiated degradation reactions. They showed that the processes of free radical formation, durability of the polymer and rate of creep in polypropylene are determined by the following rate formulae:

$$\text{Radical formation} = A_r \exp(-E_{0,r} - \beta_r \delta / RT) \dots\dots\dots(2)$$

$$\text{Durability} = A_t \exp((E_{0,t} - \beta_t \delta) / RT) \dots\dots\dots(3)$$

$$\text{Creep rate} = A_{cr} \exp(-E_{0,cr} - \beta_{cr} \delta / RT) \dots\dots(4)$$

where A and E are the Arrhenius factors and activation energies for radical formation (r),

durability(t) (defined as the time, t for a given sample to embrittle or fail when subjected to a defined temperature, T and stress,  $\delta$  environment and creep(cr). Respective  $\beta$  values are the stress sensitivity parameters. Experiment showed that within experimental error, A and E values of the three different processes were the same. Therefore, creep is suggested to be a consequence of chemical chain scission and not physical chain movement. One consequence of this observation is that ruptured radical chain ends may act as sensitisers for subsequent oxidative degradation.

### 3.2 Creep and thermal oxidative sensitivities for polypropylene.

Recent studies in our own laboratories confirm the above findings in that a relationship exists between the resistance of a polypropylene tape to creep and its degradative tendency under load.

Reconsideration of the previously published results (Horrocks and D'Souza, 1994) within this context, produces the following analysis. Reference to this research shows that the polypropylene polymers in Table 3 were converted into high tenacity, geotextile quality tapes for both creep (at ambient temperatures) and oven-ageing (at 130°C) as previously described (Horrocks and D'Souza, 1990, 1994).

Polymers P1 to P4 differ in their MFI values and in the case of P4, the antioxidant formulation which gives it a significantly higher thermal stability. Tapes comprising polymer PC contained P2 polymer and 2.5% carbon black and were

Table 3. Polymer grades converted to tapes.

Polymer*	Tape MFI**	Stabiliser
P1	3	Irganox 1010 + Irgafos 168
P2	3.8	Irganox 1010 + Irgafos 168
P3	7.5	Irganox 1010 + Irgafos 168
P4	12.4	Irganox 1330 + Irgafos 168
PC	3.6	Irganox 1010 + Irgafos 168

Notes: \* Polymer codes relate to previously published codes; \*\* Melt flow index units are g per 10 min at 2.16 kg, 230°C.

commercial samples from Scott and Fyfe Ltd., UK. Creep experiments carried out on P2, P4 and PC tapes suggested that P2 and P4 had similar stress-dependent creep tendencies but that the presence of carbon black enhanced the creep resistance of PC tapes. The creep versus time curves (Horrocks and D'Souza, 1994) were approximately linear upto 50 days from which the creep rates (expressed as percentage increase in length per second, % s<sup>-1</sup>) as a function of applied stress are listed in Table 4.

Table 4. Creep rates during the first 50 days of stress application.

Tape	Stress, %	Creep rate, Cr	
		% per sec. x 9 orders*	ln Cr
P2	5	1.16	-20.57
	10	3.82	-19.38
	15	6.25	-18.89
P4	5	0.46	-21.5
	10	1.16	-20.57
	15	3.24	-19.55
PC	5	0.93	-20.8
	10	1.85	-20.11
	15	3.47	-19.48

Note: \* Creep rates are multiplied by 10<sup>9</sup>

The results in Table 4 may be plotted as ln Cr versus percentage (p) of ultimate stress,  $\delta_u$  where stress,  $\delta = p/100 \cdot \delta_u$  according to equation (4) above.

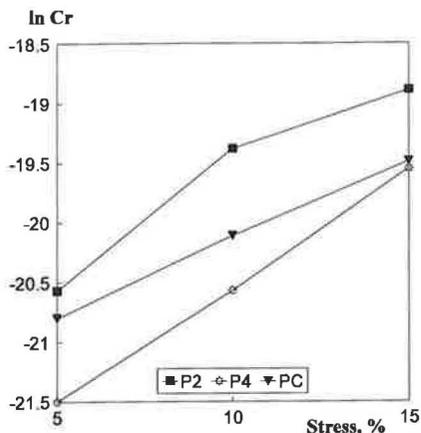


Figure 1. Plots of ln Cr versus stress,  $\delta$  for P2, P4 and PC tapes.

The plots are shown in Figure 1. In spite of there being only three points on each plot, the trends are apparently quite linear as predicted by equation (4); linear regression gives the respective slopes ( $\beta_{cr} \delta_u / RT$ ) and intercepts ( $\ln A_{cr} - E_{0,cr} / RT$ ). Values for these are listed in Table 5 along with respective  $\delta_u$  and calculated  $\beta_{cr}$  parameters (assuming that the ambient temperature is 290 K). It is not possible to calculate the A and E factors, but the calculated  $\beta_{cr}$  values indicate the relative creep sensitivities of each tape from which it is seen that the order of increased creep tendency is:

$$PC < P2 < P4$$

...creep tendency....>

This shows that higher polymer molecular weight (ie. lower MFI) and the presence of carbon black increase the creep resistance of polypropylene tapes.

Table 5. Derived intercepts, slopes and stress sensitivity parameters from plots in Figure 1.

Tape	$\ln A_{cr} - E_{0,cr} / RT$	$\beta_{cr} \delta_u / RT$	$\delta_u$ , GN/m <sup>2</sup>	$\beta_{cr}$ , m <sup>3</sup> /mol x 9 orders*
P2	21.3	0.168	0.506	809
P4	22.5	0.195	0.45	1056
PC	21.4	0.131	0.467	683

Note: \* $\beta_{cr}$  values are multiplied by 10<sup>9</sup>

If creep is a chain-scission related process, then the formation of free radical-ended polymer chain fragments may be considered as initiators for subsequent oxidative attack. From this it might be suggested that a relationship between creep tendency and stress-sensitivity of thermal oxidation could exist. To test this hypothesis, reanalysis of earlier results (Horrocks and D'Souza, 1994) in the light of the commonality of equations (2)-(4) may be undertaken. From our earlier studies, it was shown that of all the measured polypropylene properties, tape breaking strain and its retention during thermal exposure was the most sensitive indicator of degradation and, more importantly, its stress-dependence of thermal oxidation.

Percentage retention of breaking strain, e may be related to the relative increase in polymer chain scissions during degradation and so the rate of loss of breaking strain, -de/dt at any exposure time, t will be proportional to the rate of radical formation as defined by equation (2). If at any time and applied stress value,  $\delta$  the rate of loss in breaking

strain is  $-(de/dt)_\delta$  then the ratio of loss on increasing the stress from zero to  $\delta$  will be  $(de/dt)_\delta / (de/dt)_{\delta=0} = (dr/dt)_\delta / (dr/dt)_{\delta=0}$

where  $(dr/dt)$  is the rate of radical formation. From equation (2):

$$\begin{aligned} (de/dt)_\delta / (de/dt)_{\delta=0} &= \frac{\exp(-E_{0,r} - \beta_r \delta / RT)}{\exp -E_{0,r}} \\ &= \exp -(\beta_r \delta / RT) \dots \dots \dots (5) \end{aligned}$$

Thus a plot of  $\ln (de/dt)_\delta / (de/dt)_{\delta=0}$  versus  $\delta$  or % stress,  $p$  ( since  $\delta = p/100 \cdot \delta_{100}$  ) will have a slope related to the stress sensitivity factor  $\beta_r$ . Results of percentage breaking strains for the tapes in Table 3 oven-exposed in air at 130°C under varying  $p$  values from the previously cited research (Horrocks and D'Souza, 1994) are tabulated in Table 6.

Unfortunately, the data does not allow straightforward analysis because increases in breaking strain occur after short exposure times as a consequence of chain scission-initiated

Table 6. Percentage breaking strains of oven-exposed polypropylene tapes.

Polymer/days exposure	Applied stress, % of ultimate				
	0	5	10	12.5*/15	
P1	1	116.2	89.2	83.8	94.6
	4	83.8	108.1	(3)**83.8	f
	8	135.1	100	(6)**110.8	f
P2	1	62.2	51	44.9	37.8
	3	62.2	72.4	52	36.7
	6	62.2	59.2	42.9	f
	10	56.1	58.2	f	f
P3	1	140.7	108.5	69.5	48.3*
	3	132.2	118.6	74.3	42.4*
	6	116.9	98.3	59.3	40.7*
	10	148	98.3	f	f
P4	1	100	97.6	85.7	57.1*
	10				57.1*
	15	11.9	123.8	92.9	f
	29	95.2	116.7	88.1	f
	45	109.5	114.3	f	f
	60	81	150	f	f
PC	1	102.5	91.1	88.6	f
	3	105.1	94.9	81	f
	6	108.9	97.5	77.2	f
	10	102.5	102.5	79.7	f

Notes: \* denotes 12.5% stress; \*\* denotes different exposure time; f denotes tape failure

reordering of the polymer structure. However, a measure of the relative stress sensitivities may be obtained by considering the relative change in  $e$  as a consequence of applied stress for each tape exposed at each defined time, ie  $(e_\delta/e_{\delta=0})_t$ . This ratio is calculated from the  $e$  values in Table 6 and displayed in Table 7.

Comparison of  $(e_\delta/e_{\delta=0})_t$  data sets for a given tape/stress exposure experiment shows that while a considerable spread occurs, the mean of each set decreases for a given tape as applied stress increases. This effect is more evident in Figure 2 where average values of  $(e_\delta/e_{\delta=0})_t$  data sets are plotted against applied stress for all tapes. In Figure 2 it is seen that the relative stress-sensitivities of each tape to thermal oxidation may be compared. Three stress ranges may be identified, 0 - 5%, 5 - 10% and >10% ultimate stress. In the first stress zone, stress sensitivities are generally less than at higher values; in fact, for P4 (which has the highest MFI and is most highly stabilised), stresses upto 5% may be considered to have a stabilising effect. Within the 5 - 10% stress

Table 7. Relative effect of applied stress on relative reductions in exposed tape breaking strains  $(e_\delta/e_{\delta=0})_t$ .

Polymer/days exposure	Breaking strain ratio; $e(\delta)/e(\delta=0)$			
	5% stress	10% stress	12.5*/15% stress	
P1	1	0.768	0.721	0.814
	4	1.29	(3)** 1	
	8	0.74	(6)** 0.82	
P2	1	0.82	0.722	0.608
	3	1.164	0.836	0.59
	6	0.952	0.69	
	10	1.037		
P3	1	0.771	0.494	0.343*
	3	0.987	0.564	0.321*
	6	0.841	0.507	0.348*
	10	0.664		
P4	1	0.976	0.857	0.51*
	10			0.51*
	15	1.106	0.83	
	29	1.226	0.925	
	45	1.044		
	60	1.85		
PC	1	0.889	0.864	
	3	0.903	0.771	
	6	0.895	0.709	
	10	1	0.778	

Note: \* denotes 12.5% stress; \*\* indicates days' exposure

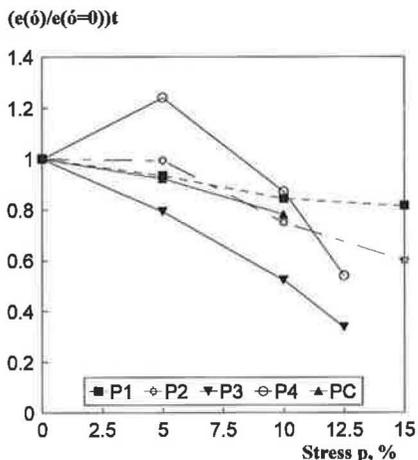


Figure 2. Plots of averaged  $(e_{\delta}/e_{\delta=0})_t$  data sets from Table 7 for each tape/stress condition versus applied stress

range, the following order of increasing sensitivity of thermal oxidation to stress is evident if slopes in this range are compared:

$$P1 < PC < P2 < P3 < P4$$

.....stress sensitivity.....>

Above 10% stress, however, the following order is apparent determined either by relative slope or by failure stress:

$$P1 < P2 < P3 < P4 < PC$$

.....stress sensitivity.....>

These results show that relative sensitivities of thermal oxidation to stress are themselves stress-dependent. Over both these stress ranges, the stress-sensitivity of the clear tapes increases with polymer MFI; in other words, lower molecular weight polypropylene chains are more sensitive to oxidation under applied stress than longer chains. Of special note is the relative stress-insensitivity of the carbon black-filled tape, PC below 10% stress and its poor performance above this level.

#### CORRELATION OF CREEP AND THERMAL OXIDATIVE SENSITIVITIES

The order for P2, P4 and PC tapes in the 5 - 10% stress range reflect the order of increased creep

tendency shown in Section 3.2 above, ie:

$$PC < P2 < P4$$

.....creep tendency.....>

.....stress sensitivity.....>

(5 -10%)

This similarity of trends, therefore confirms the above suggestion that, for the same geopolymer, creep tendency and stress sensitivity to thermal oxidation are related through the common influence of the relative ease with which polymer chain scission occurs. These results are the first, to the author's knowledge, of a relationship between the physical and chemical consequences of stress. In addition, lower molecular weight polypropylenes are more sensitive to creep and thermal oxidation under applied stress.

Clearly the importance of this to the design of geosynthetic materials and selection of appropriate materials requires further attention. Furthermore, the need to include application of stress to geopolymers during chemical durability testing protocols merits serious consideration.

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