



# Antioxidant depletion patterns of high-density polyethylene geomembranes in landfills under different exposure conditions



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## ABSTRACT

The service life of high-density polyethylene (HDPE) geomembranes is directly determined by the landfill environment, and the antioxidant depletion stage is the first and most important stage of the HDPE geomembrane aging process. In this study, the antioxidant depletion stage was chosen to investigate the effects of different exposure environments on the HDPE geomembrane lifespan. The antioxidant depletion rate (ADR) and the antioxidant depletion time (ADT) of HDPE geomembranes under various exposure conditions were calculated based on the aging parameters obtained by fitting the collected reported data with the Arrhenius model. Also, the influence of exposure conditions on the HDPE geomembrane performance degradation was analyzed. The results showed that the aging method had the greatest effect on the antioxidant depletion period, while the HDPE geomembrane thickness had the least effect. The ADR sensitivity to brand/material, aging method, leachate components, and exposure medium decreased with increasing temperature; only the sensitivity to thickness showed a slight increasing tendency with increasing temperature. The ADTs of HDPE geomembranes under different exposure conditions ranged from 6 years to 900 years, indicating that the HDPE geomembranes can complete the service time of landfills under reasonable exposure conditions. This study provides a reliable methodological basis for the risk control and life prediction of HDPE geomembranes.

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## 1. Introduction

Landfilling is one of the main methods of solid waste disposal, especially among developing countries. Because of its advantages such as low cost and low technical barriers, it is used as the preferred method of solid waste disposal and management (Grugnaletti et al., 2016). High-density polyethylene (HDPE) geomembrane is widely used in leachate containment systems, rainwater containment systems, and diversion systems in landfills. Therefore, it is a key material for leachate control and containment of hazardous constituents. The continuous deterioration of the HDPE geomembrane performance will eventually lead to a gradual loss of the landfill ability to trap solid waste and its toxic and hazardous components and a significant increase in the risk of environmental contamination. (Xu et al., 2019; Gallagher et al., 2016).

According to the deterioration mechanism, there are two main causes of HDPE geomembrane deterioration: (i) physical factors, such as the effects of sharp objects (roots or stones) and machinery

(laying machinery and landfill machinery) (De Donno and Cardarelli, 2016) and (ii) aging oxidation reactions, which are the main factor responsible for the aging of HDPE geomembranes (Abdelaal et al., 2014; Rowe and Sangam, 2002). In these reactions, polyethylene undergoes chain breaks, crosslinking, changes in molecular chain structure, and changes in side-groups (Ewais and Rowe, 2014). The mechanism is essentially the same as the oxidation reaction mechanism of small hydrocarbon molecules, that is, the free radical chain reaction of spontaneous catalytic oxidation (Gulminea et al., 2003; Klein et al., 2001). In general, to inhibit the HDPE geomembrane photooxidation process and increase lightfastness, a certain percentage of carbon black is usually added to the membrane (Kyrikou et al., 2011). Other antioxidants are also added to slow down the oxidation aging process of the membrane (Wu et al., 2010). Considering the role of antioxidants in preventing oxidation of HDPE geomembrane, Hsuan and Koerner, (2011) proposed a 3-STAGE model to characterize the HDPE geomembrane deterioration. Base on this model and the aging parameters obtained from accelerated aging experiments, scholars have analyzed the HDPE geomembrane durability under different aging conditions, and some results show that the life expectancy of HDPE geomembranes is about 100–500 years at a service temperature of

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**Nomenclature**

HDPE	High-density polyethylene	T	Experimental temperature, K
ADR	Antioxidant depletion rate	A <sub>1</sub>	Preexponential factor
ADT	Antioxidant depletion time	OIT <sub>t</sub>	Antioxidant residues at t moment, min
Std-OIT	Standard oxidation induction time	OIT <sub>0</sub>	Antioxidant residues at the initial moment, min
HP-OIT	High-pressure oxidation induction time	t	Time, month
s <sub>1</sub>	Antioxidant depletion rate, month <sup>-1</sup>		
E <sub>a1</sub>	Activation energy during antioxidant depletion, J/mol		
R	Gas constant, 8.314 J/mol/K		

20 °C to 35 °C and decreases with increasing temperature, and the life expectancy is less than 20 years at 60 °C (Collins, 1993). Besides under extreme solar radiation conditions, the life expectancy of HDPE geomembranes exposed to subtropical conditions is only 30 years (Sun et al., 2019).

In term of the factors and mechanism that affect HDPE geomembrane life, a multitude of studies have reported the influence of different factors, such as different geomembrane thicknesses and exposure media (Rowe et al., 2016; Rowe and Islam, 2009), on the oxidation and aging process of HDPE geomembrane, but they are all limited to the influence law of a single factor without comparative analysis of the different effects between different factors. One case for example is the research on the antioxidant depletion period. The antioxidant depletion period is the first stage in the aging process and is the most important period in the HDPE geomembrane life cycle because this period can indirectly affect the HDPE geomembrane service life by influencing the service temperatures of the oxidation induction period and the performance degradation period (Bian and Liu, 2014). Rowe et al., 2009 studied the effects of different media (air, pure water, and leachate) on the first stage aging rate and time; Islam, 2009 studied the effects of different thicknesses on the first stage aging rate and time. However, the comparative study of aging rate difference caused by medium and thickness is still lacking. The same situation also exists in other factors affecting the aging characteristics of HDPE film, such as HDPE film brand, experimental conditions, etc.

The current study aims to provide theoretical support for the life prediction of HDPE geomembranes and support the medium and long-term scientific and technological planning for the risk management of landfills. This study used reported data are combined with the first stage of the aging process, that is, the antioxidant depletion period, to investigate the effects of various factors, such as the aging method, brand/material, exposure medium, leachate components, and geomembrane thickness, on the antioxidants. Also, the characteristics of the depletion are investigated, and the depletion pattern is summarized based on the Arrhenius equation fitting to obtain the antioxidant depletion rate (ADR) and the antioxidant depletion time (ADT) under different exposure conditions. On this basis, the influence of each factor on the antioxidant depletion period under different temperatures was analyzed, and the time of the antioxidant depletion period was predicted by simulation.

**2. Materials and methods**

*2.1. Materials and exposure conditions*

To investigate the deterioration patterns of the antioxidant depletion periods of different types of HDPE geomembranes under different exposure conditions, five different HDPE geomembranes of four thicknesses (1.0, 1.5, 2.0, and 2.5 mm) were selected from two brands (GSE, Solmax International). Although some samples

were of the same brand or thickness, the initial values of the standard oxidation induction time (Std-OIT) and the high-pressure oxidation induction time (HP-OIT) have significant differences and are thus classified into different types (Abdelaal et al., 2014; Rowe and Sangam, 2002; Rowe and Rimal, 2008). Under three kinds of simulated aging conditions (A. immersion test, B. simulated liner test, and C. simulated stress load test) and different exposure media (air, pure water, leachate with different components), the simulated aging experiments of HDPE geomembranes were performed at different temperatures (temperature selection is described in Section 2.3 and Supplementary Material).

The Method A immersion test is the earliest and most widely used method to simulate the HDPE geomembrane aging in landfills. Here, the HDPE geomembrane is directly immersed in the simulated leachate environment, and both sides are in contact with the leachate without any stress. Method B simulates the liner experiment, which is an improvement of Method A. Only one side is in contact with the leachate, but the stress load is still not considered. Method C is further improved by adding a stress load. Due to the experimental conditions and the limitations of human and material resources, it cannot be guaranteed that each type of HDPE geomembrane can complete the experiments under each exposure condition. The specific experimental conditions are presented in Table 1 and the HDPE geomembranes characteristics are listed in Table 2.

*2.2. Data processing and model fitting method*

The antioxidant depletion process of the HDPE geomembrane is a simple chemical reaction; only oxidation occurs to reduce the antioxidants; therefore, the process follows a first-order reaction and also conforms to the Arrhenius equation (Ounas et al., 2011). To comprehensively consider various factors in the HDPE geomembrane aging, this study employs the Arrhenius equation fitting of the temperature and ADR to obtain the corresponding expression.

**Table 1**  
HDPE geomembrane materials, aging test methods, and medium types.

Variable	Value	Remarks	
HDPE geomembrane brands	G1	GSE	–
	G2	GSE	–
	S1	Solmax International	–
	G3	GSE	–
	S2	Solmax International	–
Aging test	A. Immersion test	Double-sided immersion	
	B. Simulated liner test	Single-sided immersion	
	C. Simulated stress load test	Single-sided immersion with stress	
Medium types	Air	–	
	Pure water	–	
	Leachate	–	

**Table 2**  
Basic characteristics and experimental parameters of HDPE geomembrane.

No.	Thickness/mm	Aging method	Medium <sup>1</sup>	Leachate	Properties of HDPE geomembrane			Brand	Data source
					Std-OIT/min	HP-OIT/min	Crystallinity/%		
HGM1a	1.5	A	L	L <sub>a</sub>	135	660	49.0	G2	Rowe and Rimal, 2008
HGM1b	1.5	B	L	L <sub>a</sub>	135	660	–	G2	
HGM2a	2.0	A	L	L <sub>a</sub>	133	380	–	G1	Rowe et al., 2009
HGM2b	2.0	A	P	–	133	380	–	G1	
HGM2c	2.0	A	A	–	133	380	–	G1	
HGM3a	1.5	A	L	L <sub>a</sub>	135	244	47.6	S1	Islam, 2009
HGM3b	2.0	A	L	L <sub>a</sub>	150	265	50.3	S1	
HGM3c	2.5	A	L	L <sub>a</sub>	136	235	46.6	S1	
HGM3d	1.5	C	L	L <sub>a</sub>	(135)115	(244)241	47.6	S1	Rowe and Rimal, 2008
HGM4a	1.5	A	L	L <sub>a</sub>	174	903	37.7	G3	
HGM4b	1.5	A	L	L <sub>b</sub>	174	903	37.7	G3	Abdelaal et al., 2014
HGM4c	1.5	A	L	L <sub>c</sub>	174	903	37.7	G3	
HGM4d	1.5	A	L	L <sub>d</sub>	174	903	37.7	G3	
HGM5a	1.0	A	L	L <sub>a</sub>	175	960	53.6	S2	Rowe and Ewais, 2014
HGM5b	1.5	A	L	L <sub>a</sub>	175	960	48.0	S2	
HGM5c	2.0	A	L	L <sub>a</sub>	175	960	46.7	S2	
HGM5d	2.5	A	L	L <sub>a</sub>	175	960	48.4	S2	

Note: 1. medium: A is air, P is pure water, L is leachate.

Based on this expression, the ADR under the same temperature and different exposure conditions was calculated. The ADR sensitivity to various factors was analyzed, and the key influencing factors were identified. According to the Arrhenius equation, the relationship between the ADR  $s_1$  and experimental temperature  $T$  is as follows (Rath and Staudinger, 2001):

$$s_1 = A_1 \exp\left(\frac{-E_{a1}}{RT}\right) \quad (1)$$

It was then indexed to:

$$\ln(s_1) = \ln(A_1) - \left(\frac{E_{a1}}{R} \frac{1}{T}\right) \quad (2)$$

where  $s_1$  is the ADR, month<sup>-1</sup>;  $E_{a1}$  is activation energy during antioxidant depletion, J/mol;  $R$  is gas constant, 8.314 J/mol/K;  $T$  is the experimental temperature, K;  $A_1$  is the preexponential factor.

Since both  $\ln(A_1)$  and  $(E_{a1}/R)$  are constant, a linear relationship exists between the reciprocal of temperature ( $1/T$ ) and the natural logarithm of the ADR,  $\ln(s_1)$ . This relationship can be fitted by three or more temperatures and the corresponding ADR to obtain the activation energy  $E_{a1}$  and the preexponential factor  $A_1$ . After the ADR  $s_1$  is obtained, the OIT<sub>t</sub> of the residual antioxidant amount of the HDPE geomembrane at any time  $t$  can be calculated by the following equation (Hsuan and Koerner, 2001):

$$OIT_t = OIT_0 \cdot e^{-s_1 t} \quad (3)$$

where  $OIT_t$  is the antioxidant residues at moment  $t$ , min;  $OIT_0$  is the antioxidant residue at the initial moment, min; and  $t$  is time, month.

Assuming that the end of antioxidant depletion ( $t_e$ ) is completed when the antioxidant residue is 1% of the initial value, i.e., when  $OIT_{t_e} = 1\%OIT_0$ , then (3) can be rewritten as follows:

$$\frac{1}{100} = e^{-s_1 t_e} \quad (4)$$

Thus, the antioxidant depletion period  $t_e$  can be obtained as follows:

$$t_e = \frac{\ln(0.01)}{-s_1} = \frac{4.61}{s_1} \quad (5)$$

### 2.3. Temperature selection

Temperatures and basic information on HDPE geomembranes in landfills around the world were collected by reviewing reference

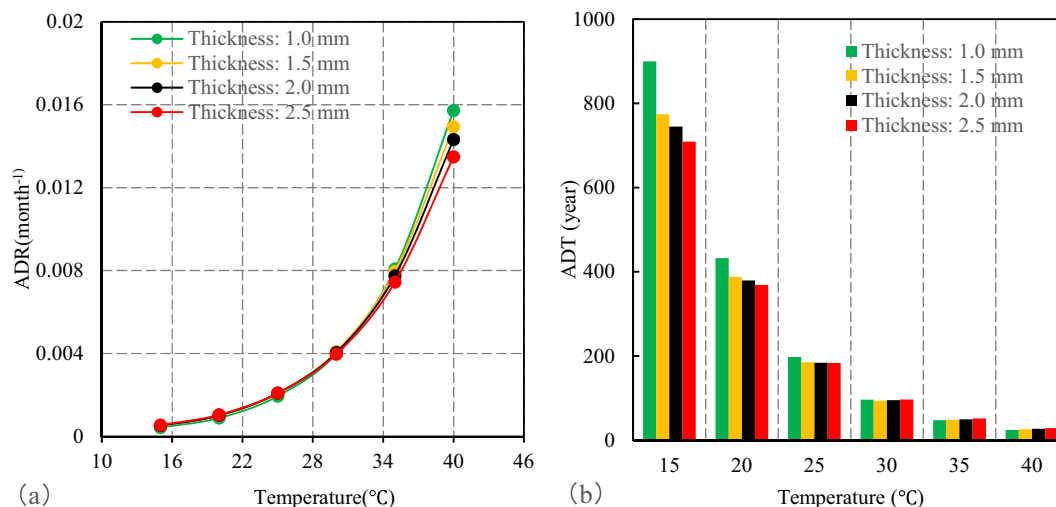
material, as detailed in Supplementary Material. According to Supplementary Material, most of the highest temperatures of HDPE geomembrane are around 30 °C, and combined with the annual average temperature of major cities in China (20–35 °C), six temperature levels were selected: 15, 20, 25, 30, 35 and 40 °C. The methods given in Section 2.2 were used to calculate the ADR and ADT under different exposure conditions and to analyze the influence of each factor and the ADR sensitivity.

## 3. Results and discussion

### 3.1. Effect of HDPE geomembrane thickness on antioxidant depletion period

HDPE geomembranes made of S2 used in the experiment of Rowe and Ewais (2014), designated HGM5a, HGM5b, HGM5c, and HGM5d (Table 2), were selected to analyze the effect of the geomembrane thickness on the antioxidant depletion period under the same aging method A and the same leachate. Fig. 1(a) and (b) display the ADR and ADT of the HDPE geomembrane, respectively. Fig. 1(a) shows that the ADR increased with the HDPE geomembrane thickness when the temperature was greater than 30 °C. For example, at 40 °C, the ADRs of HDPE geomembranes with thicknesses of 1.0, 1.5, 2.0, and 2.5 mm were 0.0157, 0.0149, 0.0143, and 0.0135 month<sup>-1</sup>, respectively, corresponding to ADTs of 24.4, 25.7, 26.8 and 28.4 years. Rowe and Ewais (2014) also found that the oxidation aging rate of polymers decreased with increasing thickness. Thicker polymers had a longer induction time for oxidation because oxidation is related to the number of oxygen molecules available to attack the polymer chain, and increasing the HDPE geomembrane thickness decreases the potential of oxygen to attack the polymer. In addition, the HDPE geomembrane thickness may also affect the rate of the outward diffusion of the antioxidant, thereby reducing the depletion rate.

However, as the temperature decreased, the difference between the ADRs of HDPE geomembranes of different thicknesses gradually decreased; for example, under 40 °C, the difference between the ADRs of the 1.0 and 2.5 mm HDPE geomembranes was 16.6%; at 35 °C, the difference was 8.4%; and at 30 °C, the ADR of the 1 mm HDPE geomembrane was only 0.4% less than that of the 2.5 mm geomembrane. When the temperature was 25 °C or even lower, the depletion rate of the 2.5 mm HDPE geomembrane was higher than that of 1.0 mm geomembrane. Since the ADR is as low as  $2 \times 10^{-3}$  to  $4 \times 10^{-5}$  month<sup>-1</sup> that the requirements for test



Material: S2; aging method: A (double-sided immersion); exposure medium: leachate

Fig. 1. ADRs and ADTs under different HDPE geomembrane thickness.

refinement significantly increase to the point where test errors may result; however, from an engineering viewpoint, this rate fully meets the expected requirements of the project life.

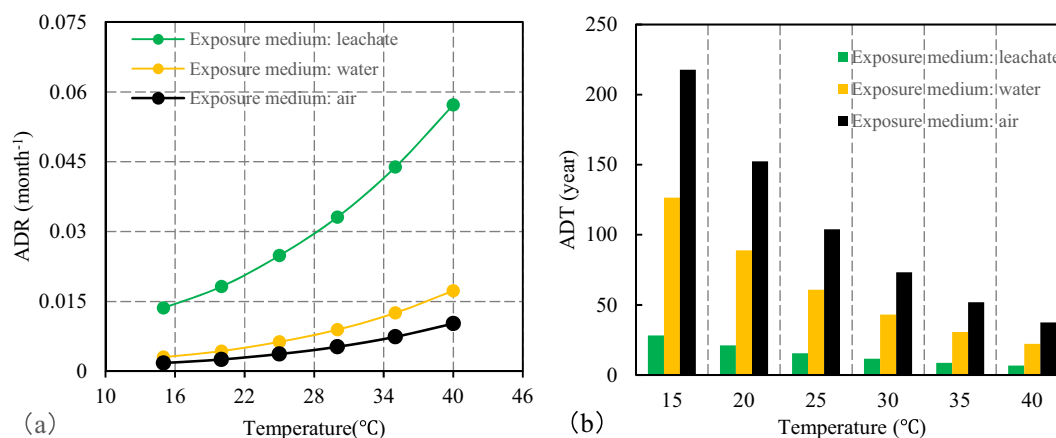
### 3.2. Effect of different exposure medium on antioxidant depletion period

Furthermore, 2.0 mm HDPE geomembranes of material G1 used in the experiment of Rowe, Rimal, and Sangam (2009), designated HGM2a, HGM2b, and HGM2c (Table 2), were selected to analyze the effect of different exposure media (air, pure water, and leachate) on the antioxidant depletion period under the same aging method A. Fig. 2(a) and (b) show the ADR and ADT of the HDPE geomembrane, respectively. Fig. 2 shows that the ADR of the HDPE geomembrane immersed in the liquid was significantly higher than that of the sample exposed to air. Under the temperature condition of 15 °C–40 °C, the ADR of the HDPE geomembrane exposed to pure water was 1.69–1.72 times of the geomembrane in air, while the aging rate of the HDPE geomembrane exposed in leachate was 3.3–4.5 times of that of the geomembrane in pure water. The corresponding ADT of the HDPE geomembrane in air

was 37–218 years, that of the geomembrane in pure water was 22–127 years, and that of the geomembrane in leachate was only 6.7–21.1 years. The higher ADR in the liquid phase than in the air phase may be because the antioxidants in the HDPE geomembranes enter the liquid phase via extraction, whereas extraction does not occur in the air phase.

Moreover, the ADR in pure water was significantly lower than that in leachate. One possible explanation is that the transition metals (e.g., Co, Mn, Cu, Al, and Fe) in the leachate decompose hydroperoxides through redox reaction and generate additional free radicals (Rowe and Sangam, 2002); these radicals can diffuse into the HDPE geomembrane and directly react with the antioxidants or catalyze or accelerate the reaction (Koerner and Koerner, 2006). These transition metals are usually from residual catalysts used in polymerization resins, but they have also been found in leachate (Sarmiento et al., 2019), especially for landfills. Therefore, they are the focus of the research on the long-term durability of HDPE geomembranes.

Also, from Fig. 2(a), different from the effect of the thickness on the ADR, the effect of the exposure medium on the ADR increases as the temperature decreases. This may be because at lower tem-



Material: G1; aging method: A (double-sided immersion); thickness: 2.0 mm

Fig. 2. ADRs and ADTs under different exposure medium.

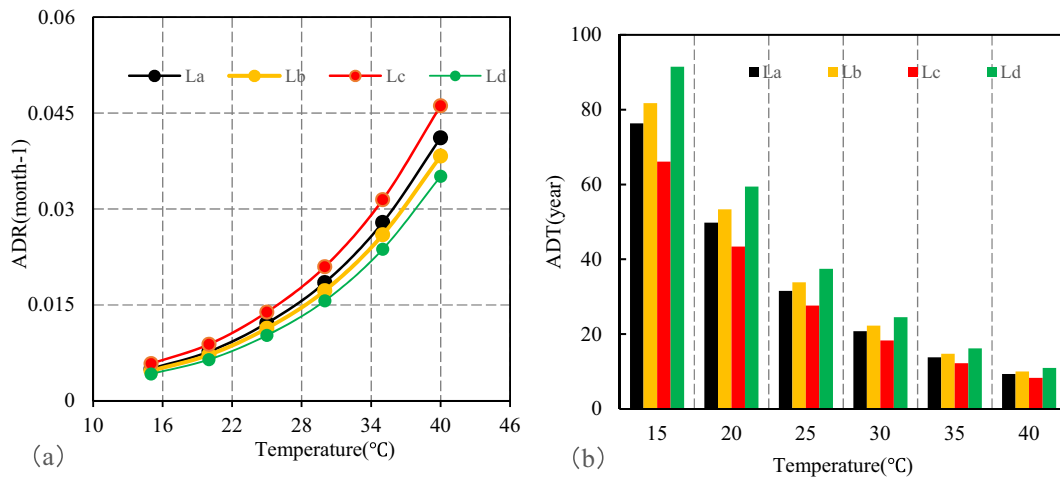


Fig. 3. ADRs and ADTs under different leachates.

peratures, the antioxidant can sufficiently contact the polymer to increase the ADR. For example, at 40 °C, the ADR in the leachate was 3.7 times that in pure water, but at 35 °C, 30 °C, 25 °C, and 10 °C, the ADRs were 3.9, 4.2, and 4.5 times those in pure water, respectively.

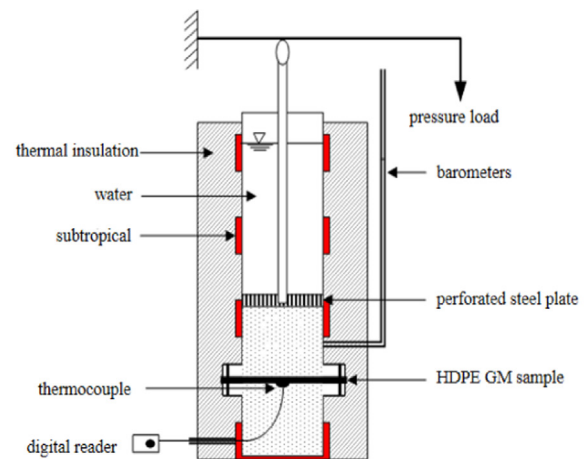
In summary, the ADR of the HDPE geomembrane decreased under the exposure media as follows: leachate > pure water > air. The difference in the ADR under different exposure medium conditions increased with temperature increase. At 40 °C, the maximum difference in the ADR was a factor of 4.8 (between air and leachate), which led to a 36-year antioxidant depletion period difference (44 years and 8 years). At 15 °C, the maximum difference in the ADT was 190 years (218 and 28 years).

3.3. Effect of different leachate component on antioxidant depletion period

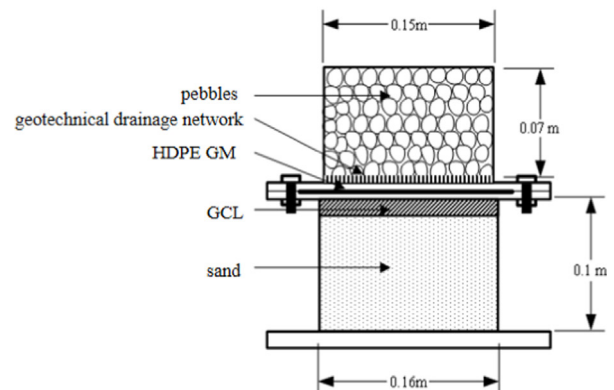
A significant difference existed between the ADRs of HDPE geomembranes exposed to pure water and leachate under the same conditions. The components in the leachate played a key role in this difference. The HDPE geomembranes of material G3 used in the experiment of Abdelaal, Rowe, and Islam (2014), HGM4a, HGM4b, HGM4c, HGM4d (Table 2), were selected to analyze the effect of different leachate components on the antioxidant depletion period. In Fig. 3,  $L_a$  represents ordinary leachate,  $L_b$  is leachate with volatile fatty acids and salts,  $L_c$  is leachate with salts, and  $L_d$  is leachate with volatile fatty acids. Fig. 3(a) and (b) show the ADR and ADT of the HDPE geomembrane, respectively. Under 40 °C, the depletion rates of HDPE membrane antioxidants in the different leachates were of the following order:  $L_c > L_a > L_b > L_d$  and the rates were 0.046, 0.041, 0.038 and 0.035 month<sup>-1</sup>, respectively, with the maximum difference of a factor of 0.31. As the temperature decreased the difference increased; for example, under 15 °C, the difference was a factor of 0.38. The results suggest that salts can accelerate the antioxidant depletion, while volatile fatty acids can alleviate the effect of salts. This is probably because the salts react with the antioxidants, increasing the ADR. The volatile fatty acids can consume some of the salts and therefore control ADR (Abdelaal et al., 2014).

Since the leachate components of actual landfills are more complex, the effect of leachate components on the HDPE geomembrane aging will be mainly considered in the subsequent work. In summary, at the same temperature, a high salt content of the leachate

component can accelerate the ADR, while a high content of volatile fatty acids can slow down the ADR. Moreover, the lower the temperature, the greater the difference in the ADR.



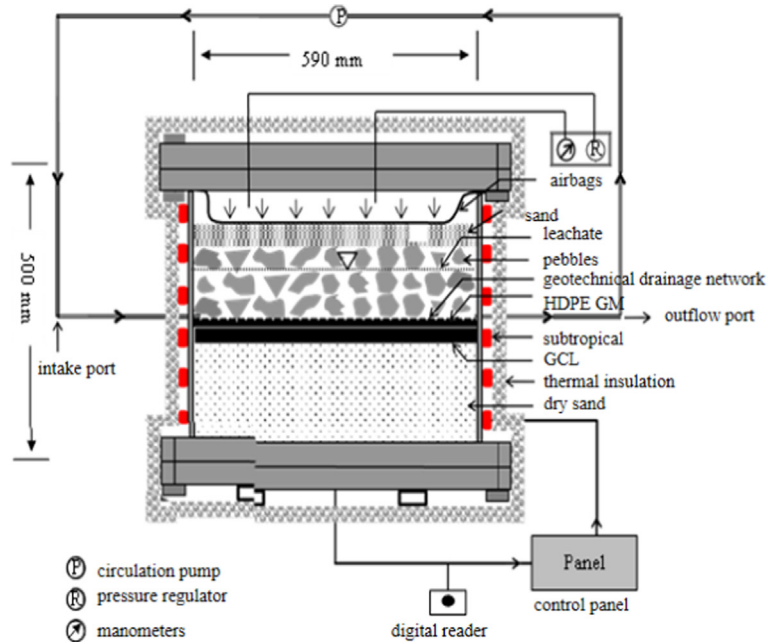
A. Double-sided immersion aging simulation device



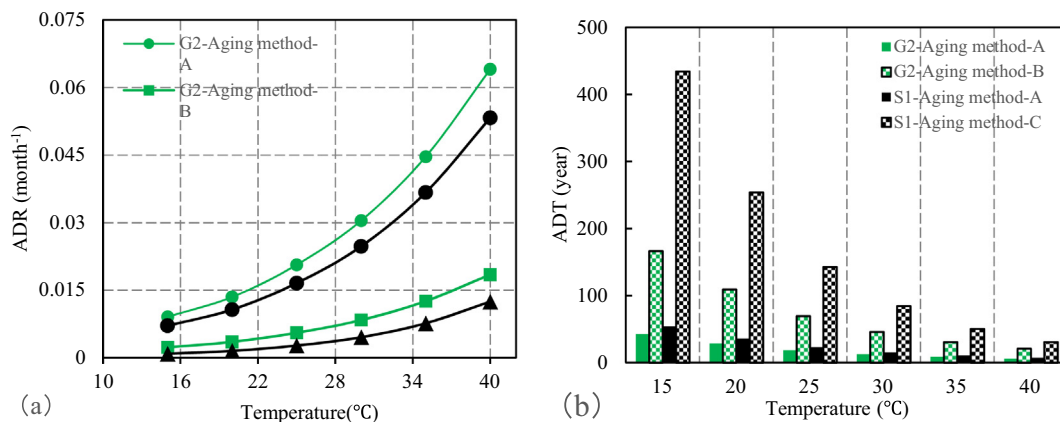
B. Single-sided immersion aging simulation device

Fig. 4. ADRs and ADTs under different aging methods.





C. Device for aging simulation via single-sided immersion with stress



Material: G2, S1; exposure medium: leachate; thickness: 1.5 mm

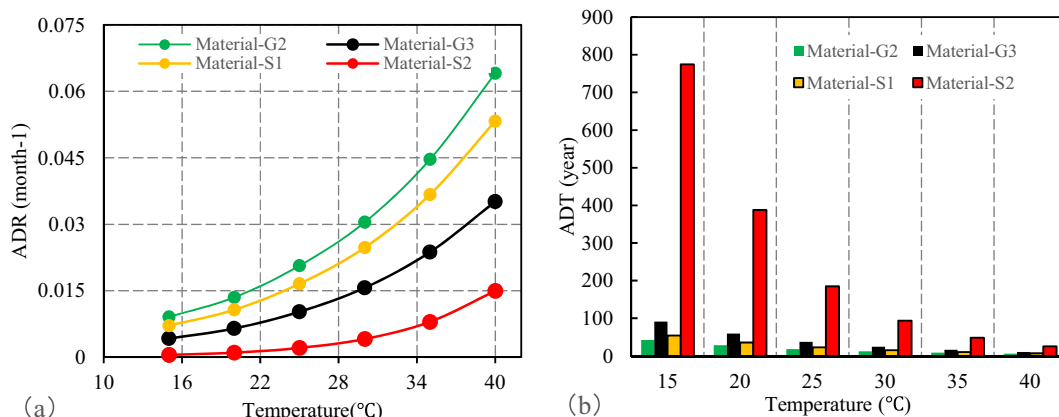
Fig. 4 (continued)

### 3.4. Effect of aging method on antioxidant depletion period

Furthermore, 1.5 mm HDPE geomembranes of material G2 and S1 used in the experiment of Rowe and Rimal (2008) and Islam (2009), designated HGM1a, HGM1b, HGM3a, and HGM3c (Table 2), were selected to analyze the effects of different aging methods on the antioxidant depletion period in the same leachate. The diagrams of the aging modes are displayed in Fig. 4. Fig. 4A and B show the aging modes of HDPE geomembranes made of G2, and Fig. 4A and C show the aging modes of the geomembranes made of S1. Fig. 4(a) and (b) illustrate the ADR and ADT of the HDPE geomembrane, respectively. Fig. 4(a) shows that under different exposure conditions, that is, aging method A and C, the ADRs of S1 were 0.007–0.0053 and 0.0009–0.012, respectively; the difference could reach a factor of 3.3–7.0. Also, under aging method A and B, the ADRs of G2 were 0.009–0.064 and 0.002–0.018, respectively; the difference could reach a factor of 2.8–3.5. Therefore, the

ADR between C and A was not much different from that between B and A, indicating that the ADR does not increase significantly despite the stress load addition in C; this indirectly indicates that the load in the landfill environment (around 26 kPa) does not exacerbate the oxidation aging process of HDPE geomembranes.

Hsuan, (2002) showed that there is a safe threshold of stress for polymers below which an increase in stress does not affect the oxidation degradation; however, the applied stress above this threshold causes a significant acceleration of polymer embrittlement. Moreover, a study on the aging performance of HDPE pipelines for natural gas transportation (Rowe and Sangam, 2002) showed that this threshold is typically above 7000 kPa. For the HDPE geomembrane at the landfill bottom, assuming a landfill height of 30 m and a density of 2 g/cm<sup>3</sup>, the equivalent pressure load is about 588 kPa, which is much lower than the stress load threshold. Therefore, under this stress loading condition, the effect of stress on the oxidation process can be completely neglected.



Aging method: A (double-sided immersion); exposure medium: leachate; thickness: 1.5 mm

Fig. 5. ADRs and ADTs under different HDPE geomembrane brand/material.

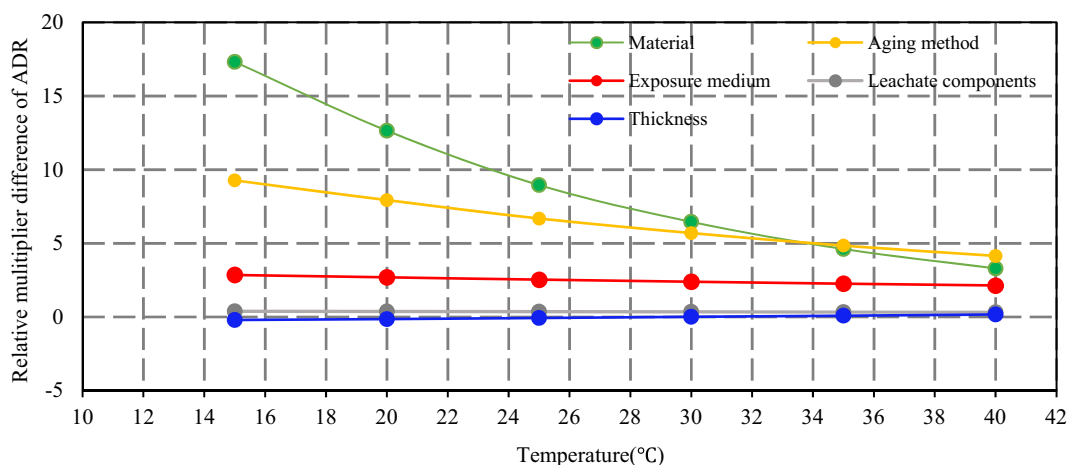


Fig. 6. Variation of the ADR sensitivity to each factor with temperature.

In addition to the above analysis, the normal stress loading of the landfill does not affect the antioxidant depletion period. The different exposure methods (double-sided immersion and single-sided immersion) have a significant effect on the antioxidant depletion rate and duration up to a maximum of a factor of 7.0.

### 3.5. Effect of HDPE geomembrane brand/material on antioxidant depletion period

Furthermore, 1.5 mm HDPE geomembranes of the G2, G3, S1, and S2 materials used in the experiment of Rowe and Rimal (2008), Abdelaal, Rowe, and Islam (2014), Islam (2009), and Rowe and Ewais (2014) (designated HGM1a, HGM4a, HGM3a, and HGM5c in Table 2) were selected to analyze the effect of different brands and materials on the antioxidant depletion period of membranes under the same leachate and aging method A. Materials G2 and G3 were of GSE brand, and S1 and S2 belonged to Solmax International. Fig. 5(a) and (b) show the ADR and ADT of the HDPE geomembrane, respectively. Fig. 5(a) shows that there are not only differences between the ADRs of the HDPE geomembranes of different brands, but also large differences between the ADRs of geomembranes of different materials but the same brand. Under

40 °C, the ADR of the GSE G2 sample was 4.3 times that of the Solmax S2 sample, and the difference increased as the temperature decreased. At 15 °C, the ADR of G2 was 17.3 times that of S2. Also, for the same GSE, the ADR of G2 was 1.8 and 2.2 times those of G3 at 40 °C and at 15 °C, respectively.

In general, the ADRs of HDPE geomembranes of different brands were different, and the ADRs of geopolymers of the same brand but different materials were also different. The ADRs of HDPE geomembranes of different brands differ by a factor of 3.3–17.3 times (40 °C ~ 15 °C), and the ADRs of HDPE geomembranes of the same brand and material also differed by a factor of 0.8–1.2 (40 °C ~ 15 °C).

### 3.6. Parameter sensitivity analysis of antioxidant depletion

According to the analysis of the above results, the ADR sensitivity to each of the influencing factors is of the following order: aging method > brand/material > exposure medium > leachate components > geomembrane thickness, under the temperature of 40 °C, which can lead to ADR differences of factors of 4.1, 3.3, 2.12, 0.31 and 0.17, corresponding to 23, 19.7, 30.8, 2.6 and 4 years of the difference in ADT, respectively.

A further comparison of the changes in the ADR sensitivity to the above factors with increasing temperature (Fig. 6) showed that the ADR sensitivity to brand/material, aging method, and exposure medium decreased with increasing temperature; while the ADR sensitivity to the leachate component changed less with temperature but also decreased with increasing temperature; only the ADR sensitivity to thickness showed a slight tendency to increase with increasing temperature. The ADTs of HDPE geomembrane ranged from 6 (HGM1a, 40 °C) to 900 years (HGM5a, 15 °C) under different exposure conditions.

#### 4. Conclusion

Through reference data collection and theoretical analysis, this study investigated the pattern of the antioxidant depletion period of HDPE geomembranes in landfills under different exposure conditions. Moreover, a method for ADR determination and a prediction model for the ADT is established. Based on the data and analysis, the following conclusions can be drawn. First, the antioxidant depletion was mainly due to the attack of oxidation on polymer chains and the extraction process in the liquid phase. The aging method had the greatest influence on the antioxidant depletion period of HDPE geomembranes, while the geomembrane thickness had the least influence. The factors influenced the antioxidant depletion period in the following order: aging method > brand/material > exposure medium > leachate components > geomembrane thickness. Second, the simulation results showed that the antioxidant depletion phase of the HDPE geomembrane was strongly influenced by temperature. Variation in temperature affects the ADR sensitivity to individual exposure factors. The ADTs of HDPE geomembranes ranged from 6 (HGM1a, 40 °C) to 900 years (HGM5a, 15 °C) under different exposure conditions. This indicates that the HDPE geomembranes can complete the service life of landfills under reasonable exposure conditions.

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#### Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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