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ORIGINAL PAPER

Effects of Exposure Conditions on Antioxidant Depletion, Tensile Strength, and Creep Modulus of Corrugated HDPE Pipes Made With or Without Recycled Resins

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Abstract

The service lifetime of corrugated high-density polyethylene (HDPE) pipes used in transportation infrastructure applications has been the subject of several studies over the years. How factors specific to North American climate conditions—such as freeze/thaw cycles, deicing salts, and abrasion—affect pipe service life have not been addressed. In this study, laboratory accelerated-aging tests were conducted to examine the thermo-oxidative degradation as well as tensile strength and creep modulus of HDPE pipes made with or without recycled resins. Specimens were immersed in saline solution (NaCl), acidic solution (H_2SO_4), alkaline solution (NaOH) at 60 °C, or in water and exposed to freeze/thaw cycles ranging from − 40 to + 40 °C. Measuring the oxidative induction time (OIT) at various time intervals quantified the amounts of antioxidants present in the pipes, revealing that the rate of antioxidant depletion depended on the surrounding environment. The depletion rate was faster in the saline solution and slower in the freeze/thaw cycles. For specimens exposed to freeze/thaw cycles, the depletion was 1.12 to 1.91 lower than in the acidic solution, 1.25 to 2.64 times lower than in the alkaline solution, and 1.43 to 3.25 times lower than in saline solution. Nevertheless, the HDPE pipes all met the OIT requirements of a minimum of 20 min. Furthermore, the one-way analysis of variance (ANOVA) and Tukey's HSD test indicate that the tensile strength and creep modulus values of the recycled and virgin HDPE pipes were unchanged before and after immersion in the different solutions.

Keywords Corrugated high-density polyethylene (HDPE) pipe · Recycled resins · Exposure conditions · Oxidative induction time (OIT) · Antioxidant depletion · Tensile strength · Creep modulus · ANOVA · Tukey's HSD test

Introduction

Corrugated high-density polyethylene (HDPE) pipes used for transportation infrastructure applications are usually buried underground. Many studies [1–4] have shown that the properties of polymer materials can change over time and due to exposure conditions (e.g., sunlight, temperature, thermal cycles, and oxygen). Understanding long-term performance is important in estimating a pipe's lifetime. In general, the long-term service lifetime of HDPE follows three aging phases [5, 6]. In the first phase (I), the pipes mainly fail in a ductile manner under significant loads (normally > 30% of yield). In the second phase (II), pipes are subjected to intermediate loads, which initiates microcracks at the junction of the liner and corrugated part, leading to brittle failures. Lastly, in the third phase (III), failures are generally brittle under lower stresses when the antioxidants have been consumed and the pipe undergoes oxidation.

Three main factors control the lifetime of a plastic pipe: material (resins, additives), environment (chemical agents, UV radiation, microbiological effects), and loading (static and dynamic loading) [7]. In stark contrast to pressure pipes, it can be assumed that stress cracking is not a major problem for buried pipes. They could be in a state of relaxing

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bending stress under an occasional load or a dynamic load of vehicles [8]. In fact, the choice of resin type, additives, or manufacturing process can be planned early on. In contrast, environmental factors are a major issue with buried pipes. The water in culverts and sewer systems contains heavy materials, synthetic chemicals, petroleum, or gas. Moreover, the surrounding soil might contain significant amounts of transition metal and moisture that cause pipe degradation [5]. Oxidation occurs once the antioxidant (AO) content has been exhausted. Oxidation can decrease of the molecular weight of the polymer due to chain scission, making the pipe brittle over time [9]. Therefore, oxidative degradation is an important issue affecting pipe degradation. Antioxidants (AOs) and stabilizers incorporated into HDPE pipes retard or halt the oxidation process [10]. Primary and secondary antioxidants are two common types for antioxidant packages [11]. A primary antioxidant's function is to act as free radical (ROO·, RO·, OH·) scavengers and form ROOH, ROH, and H_2O. Secondary antioxidants convert carboxylic acids (ROOH) to inactive alcohol (ROH) [6, 12]. In general, the oxidative degradation process, which can be evaluated by measuring OIT values, occurs in three stages, as shown in Fig. 1 [6, 13, 14]. The total loss of antioxidants takes place in stage t_1. Stage t_2 is the induction time to the onset of material degradation. Stage t_3 is the process of material degradation with a reduction of properties. The time taken for a material property to drop to a specified value (typically 50% of its initial value) is defined as the material's lifetime (t_{life}). For HDPE pipes, this is the combined duration of the three stages ($t_1 + t_2 + t_3$).

Other studies have evaluated the antioxidant depletion and mechanical properties of pipes with accelerated aging tests to simulate long-term exposure and estimate pipe durability. Gedde et al. [15] studied the long-term behavior of polyolefin pipes used in hot-water environments. Thomas [16] studied the long-term performance of corrugated pipe manufactured with recycled polyethylene content. Most of the work done in the past focused on HDPE water pipes. Hoang and Lowe [17] studied the depletion of antioxidants and initiation of thermooxidative degradation of PE100 blue water pipes. They had been exposed to hydrostatic pressure at 20 °C, 40 °C, 60 °C, or 80 °C. The results indicated that the antioxidant content decreased by 95% after 2500 h of immersion at 80 °C. Whelton et al. [18] evaluated the impact of the aging of HDPE water pipes by exposing them to chlorinated water. The results demonstrated that after 141 days, the OIT value decreased by 69% during the first 90 days and remained relatively unchanged afterwards. They also reported unchanged tensile strength. In addition, Hsuan and Wong [14] evaluated the oxidation degradation of corrugated HDPE pipe. The tested specimens contained 2.5 wt% of carbon black (CB) and an antioxidant package that consisted of 0.05% Irganox 1010 and 0.1% Irgafox 168. Specimens were incubated in a forced-air oven at temperatures of 65 °C, 75 °C, and 85 °C and in a water bath at 85 °C. The results also indicated that the OIT value decreased with incubation time. Krushelnitzky and Branchman's work pointed out that the antioxidant depletion rate of HDPE pipes was faster when the pipes were immersed in a synthetic municipal solid-waste leachate rather than in air [8]. Recent works have pointed out that antioxidant depletion rates were faster for HDPE geomembranes exposed to leachate than that exposed to water or air [10, 19]. Jeon et al.'s work also indicated that the antioxidant depletion time of HDPE geomembranes was longer in an acidic solution and shorter in an alkaline solution [20]. Furthermore, the effects of different exposure conditions on the mechanical properties (tensile strength, creep modulus) have also been investigated. Rowe and Rimal [21] reported that tensile strength at yield and break was almost unchanged after 35 months of incubation in synthetic leachates at 85 °C for an HDPE geomembrane in a composite liner. Rowe et al. [22, 23] had similar results for the unchanged tensile strength of an HDPE geomembrane under landfill conditions. HDPE geomembranes, however, are different than HDPE pipes because of their constituents and conditions of use. The literature contains little about the effect of exposure conditions on antioxidant depletion, tensile strength, and creep modulus of corrugated HDPE pipe for transportation infrastructure applications. In contrast, no research has evaluated the antioxidant depletion rate, or the mechanical properties of corrugated HDPE pipes made with virgin and recycled resins exposed to North American climate conditions.

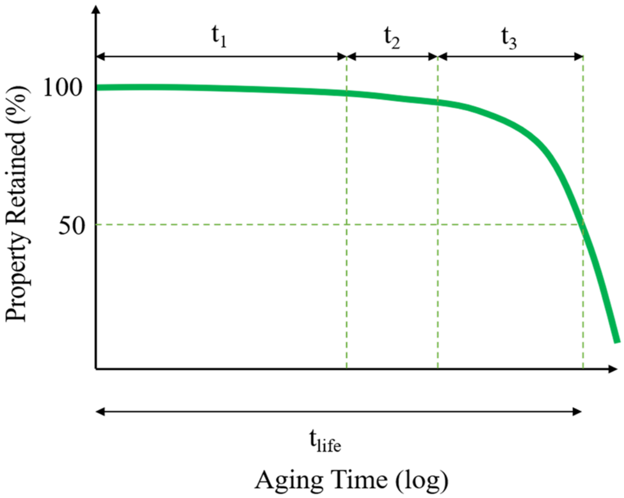

Fig. 1 Three-stage conceptual model of chemical aging of HDPE (adopted from Hsuan and Koerne [6])

The exposure conditions for HDPE pipe in past studies do not reflect the actual conditions in North America, including their durability characteristics related to freeze/thaw cycles, deicing salts, and abrasion conditions. In our study, the failure of corrugated HDPE pipe in phase III (oxidation) was assessed by determining the depletion rate of antioxidants under different aging conditions (freeze/thaw cycles; saline, acidic, and alkaline solutions). The study aimed at comparing the antioxidant depletion rate between these different aging durations. Moreover, the effects of different solutions on the tensile strength and creep modulus of recycled and virgin HDPE pipes were identified.

Fig. 2 HDPE pipes under investigation [24]

Materials and Methods

Materials

The material used has been described elsewhere [24, 25]. It consisted of six corrugated HDPE pipes, two (A-R, D-R) manufactured with post-consumer recycled resins (household waste), one (B-R) with post-industrial recycled (pipe scrap), and three with virgin resins (A-V, B-V, and C-V) from four different North American manufacturers (Fig. 2). These pipes, used for transportation infrastructure applications (e.g., storm drainage and storm sewers), were manufactured according to BNQ 3624-120, Class A [26]. Their density was greater than 0.945 g/cm^3 [27] and they were stabilized with 2–4% carbon black and antioxidants. The letters A, B, C, and D designate the manufacturers, R stands for recycled resin, and V for virgin resin. Each pipe had an external diameter of 900 mm and length of 3000 mm. As shown in Table 1, the detailed properties and parameters used for this study were summarized in a past study [25].

Laboratory-Accelerated Aging Tests

Polyethylene (PE) is well-known for its low chemical reactivity with polar fluids and solvents—such as water, acids, and bases—due to its hydrophobicity and chemical structure [33, 34]. HDPE pipes are therefore resistant to chemical corrosion [35]. The available literature related to short-term applications does not address road infrastructure applications. In addition, current environments—including exposure to detergents, leachates, hot water, or chlorinated

Table 1 Properties of investigated HDPE pipes

Property	Method (ASTM)	Specimen					
		A-R	A-V	B-R	B-V	C-V	D-R
Density[a] (g/cm^3)	D792-13 [28]	0.988	0.977	0.976	0.987	0.976	0.964
Density[b] (g/cm^3)	–	0.975	0.964	0.963	0.976	0.965	0.950
MFI (g/10 min)	D1238-13 [29]	0.103	0.077	0.066	0.061	0.115	0.058
10^{-3} M_n molecular weight (gmol^{-1})	–	17.8	14.7	17.0	15.3	16.0	19.0
10^{-3} M_w molecular weight (gmol^{-1})	–	150	153	169	163	146	151
CB (%) content	D5805-00-19 [30]	3.0	2.8	3.0	2.5	2.5	3.2
Hardness (HD)	–	56	55	55	60	65	61
Softening temperature by DMA	–	54	52	45	45	52	58
Softening temperature by TMA	–	51	50	45	45	45	54
Mass loss (°C)	E2550-17 [31]	390	375	415	420	413	413
Tensile strength at yield (MPa)	D638-14 [32]	25.12	23.58	18.83	17.44	19.81	22.56
Elastic modulus (MPa)	–	525	393	236	219	424	390

MFI melt flow index, *CB* carbon black, *DMA* dynamic mechanical analysis, *TMA* thermomechanical analysis, *ASTM* American Society for Testing and Materials

[a] Compound density

[b] Resin density

water—do not simulate Northern conditions. Our study focused on the impact of Canada's winter (e.g., low temperature, variations in daytime and nighttime temperatures, and freeze/thaw cycles) on antioxidant depletion, tensile strength, and creep modulus. Historical weather records for Quebec indicate that the average highest and lowest temperatures annually are around 40 °C and – 40 °C, respectively. Weather characteristics with long, cold winters, and short summers, as well as the impact of sunlight at certain times (installation, repair, etc.), are among the factors that affect the long-term performance of pipelines. To simulate the Northern climate, specimens taken from the pipe liner (Fig. 3) were conditioned in different chemical solutions. Specimens consisting of $40 \times 10 \times 2$ mm^3 plates were used to investigate antioxidant depletion and creep modulus. Dog-bone specimens were used to evaluate the tensile strength. The following sections provide details on the dimensions of the test specimens. Four exposure conditions were applied: freeze/thaw cycles and exposure to saline, acidic, or alkaline solutions. It is well known that freeze/thaw cycles are one of the problems facing transportation infrastructure in North America. These conditions could cause internal cracks by inducing additional internal stresses (shrinkage and expansion). To simulate freeze/thaw cycles, specimens were immersed in water for 625 cycles at temperatures between – 40 and 40 °C. Each cycle consisted of 8 h of freezing and 6 h of thawing. To simulate deicing-salt, acidic, and alkaline service conditions, specimens were immersed in either a saline solution (NaCl 1 M), sulfuric acid (H_2SO_4, pH of 2), or sodium hydroxide (NaOH, pH of 10). The immersion tests were performed at 60 °C. It should be noted that acceleration tests are valid when only the temperature affects the rate of degradation [10, 19, 36, 37]. The selected temperature for accelerated aging tests is not so high that it modifies the nature of the material and consumes the antioxidants. Hence, it is recommended that the upper temperature limit for laboratory accelerated-aging tests of HDPE pipes not exceed the softening point at 60 °C. Specimens were retrieved at various time intervals, including 4, 8, and 12 months. The oxidative induction time was monitored to evaluate the rate of antioxidant depletion. Unfortunately, it is impossible to predict the service life of pipes under real-life conditions with the AO depletion rate determined in our study. The AO depletion rate is not proportional to temperature. It is important to remember that the elevated incubation temperature steps (50 °C, 60 °C, 70 °C, etc.…) are required to extrapolate the master curve at the service temperature. Due to time limitations, our study focused on the depletion rate of oxidants in different media at 60 °C. Tensile properties were also investigated. Due to the availability of the conditioning chamber, specimens for creep modulus were investigated after 6.5 months.

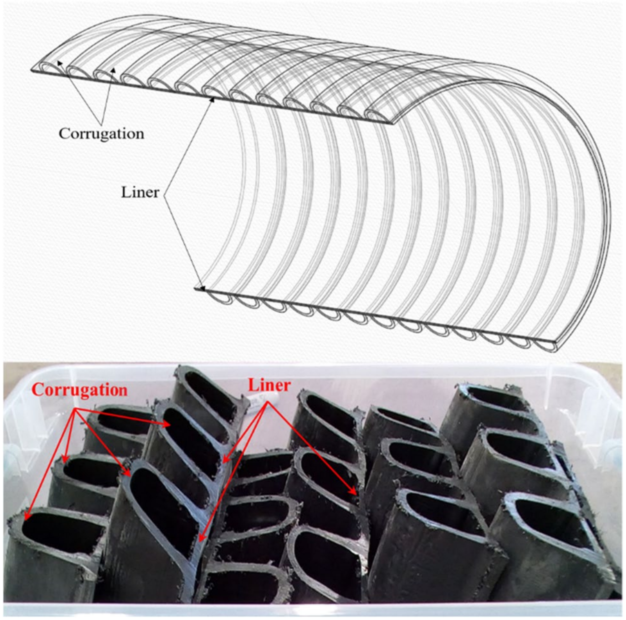

Fig. 3 Sectional view of the HDPE pipe showing the location of corrugation and liner

Oxidative Induction Time

Oxidative induction time is a standardized test to determine the amount of AO in the pipes. This test was conducted with differential scanning calorimetry (DSC 6000 from PerkinElmer) in accordance with ASTM D3895-19 [38]. Specimens weighing 5–10 mg were heated under nitrogen from room temperature to 200 °C at 30 °C/min. Once the higher temperature was reached, oxygen was introduced and the value of OIT was taken as the onset of the exothermic peak appearing when the sample oxidized. Figure 4 provides a typical evaluation of OIT from the recorded time-based thermal curve.

Tensile Tests

The tensile properties of the HDPE pipes before and after immersion in the different solutions were investigated according to ASTM D638 [32] with type IV dog-bone specimens. The specimens were punched directly from the pipe liner by a stainless-steel die with respect to the direction of extrusion. The specimen geometry and dimension are described in ASTM D638, as illustrated in Fig. 5. The thickness of test specimens cut from pipes A-R, A-V, B-R, B-V, C-V, and D-R was 4.80, 4.40, 7.50, 7.60, 2.70, and 3.80 mm, respectively. The test was performed at 23 °C on an MTS (Material Testing Systems) universal testing machine with a constant crosshead speed of 50 mm/min. The specimen extension was measured with a 634.12F-24 extensometer. At

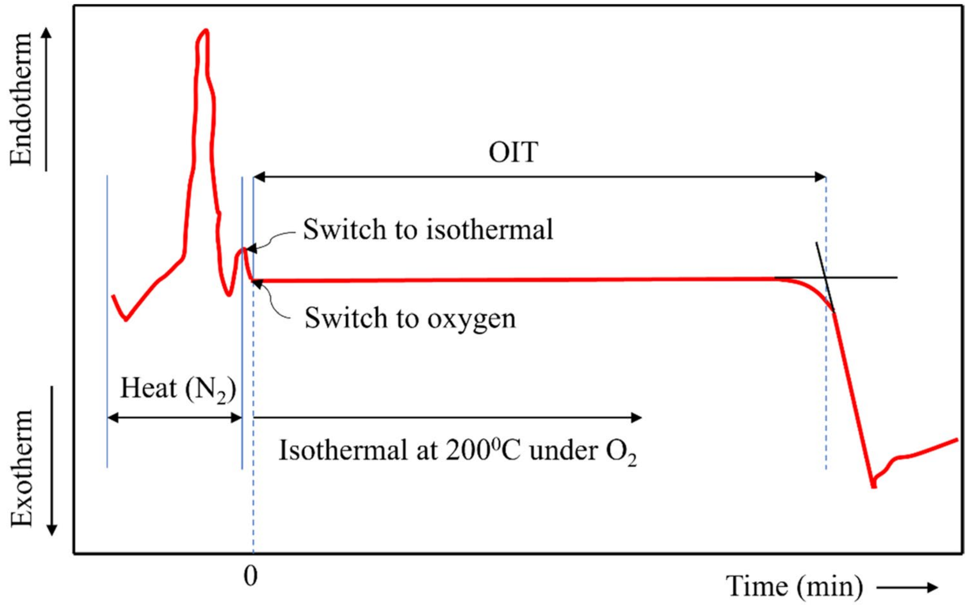

Fig. 4 Evaluation of OIT from the recorded time-based thermal curve

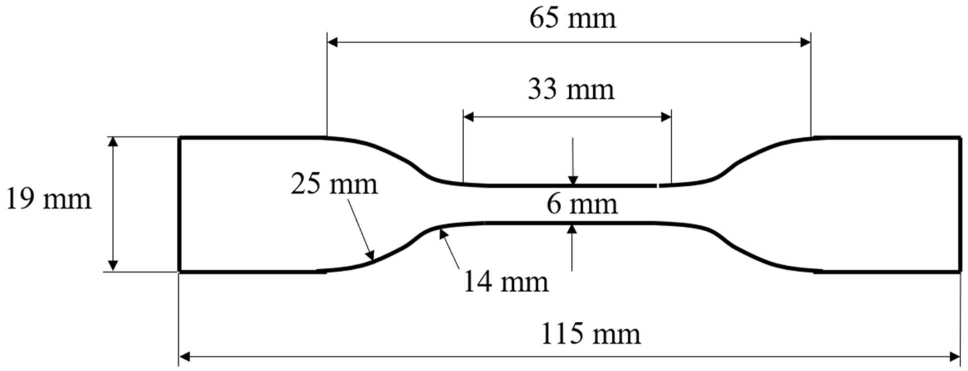

Fig. 5 Typical dog-bone specimens for evaluating tensile strength

least four replicates were performed to evaluate the tensile strength before and after immersion.

Time–Temperature Superposition (TTS)/DMA Tests

Time–temperature superposition (TTS) is the acceleration test most often used to investigate the viscoelastic behavior and creep modulus with elevated temperature. The relaxation curves obtained at high frequency and temperature steps can be shifted to generate a master curve at the reference temperature. The master curve is then extrapolated to predict the creep modulus of materials. In the current study, the tests were carried out using a DMA (DMA 8000 from PerkinElmer). The specimens—measuring $40 \times 10 \times 2$ mm^3—were tested using the single-cantilever mode with a span length of 17.5 mm. The temperature ranged from 5 to 100 °C, with 5 °C steps. Frequencies from 0.01 to 100 Hz were applied. A heating rate of 5 °C/min and an isotherm of 3 min for each temperature step were applied. The strain applied was 0.05%. At least three replicates were performed with a test duration of 10 h.

Statistical Analysis

Variations between the values measured before and after specimens have been immersed in solutions are normal. Therefore, a one-way analysis of variance (ANOVA) was used to evaluate whether the differences between measurements were significant. This method indicates no significant difference between measurements (treatments) if the F-value is less than the F-critical value or the p-value is greater than the alpha level (0.05). The F-value is determined as the ratio of the variance of the group means (means square between) to the mean of the group variance (mean squared error). The F-critical value is determined by the numerator (df1) and denominator (df2) degrees of freedom with a specific significance level (alpha level) from the F-distribution table [39]. The p-value is the probability value describing how likely it is that the data could have occurred under the null hypothesis

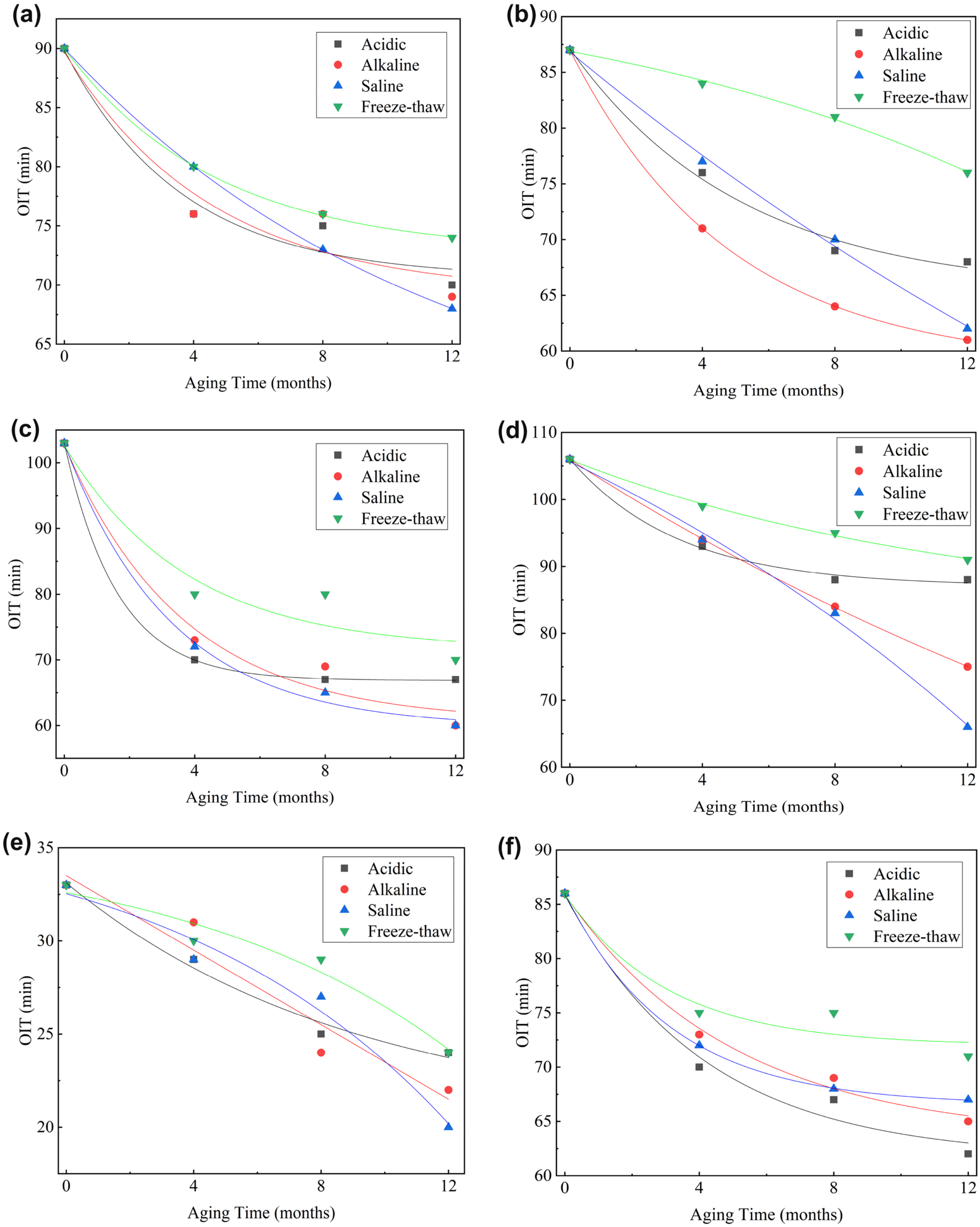

Fig. 6 OIT value of HDPE pipes with aging time under different exposure conditions **a** A-R **b** A-V **c** B-R **d** B-V **e** C-V **f** D-R

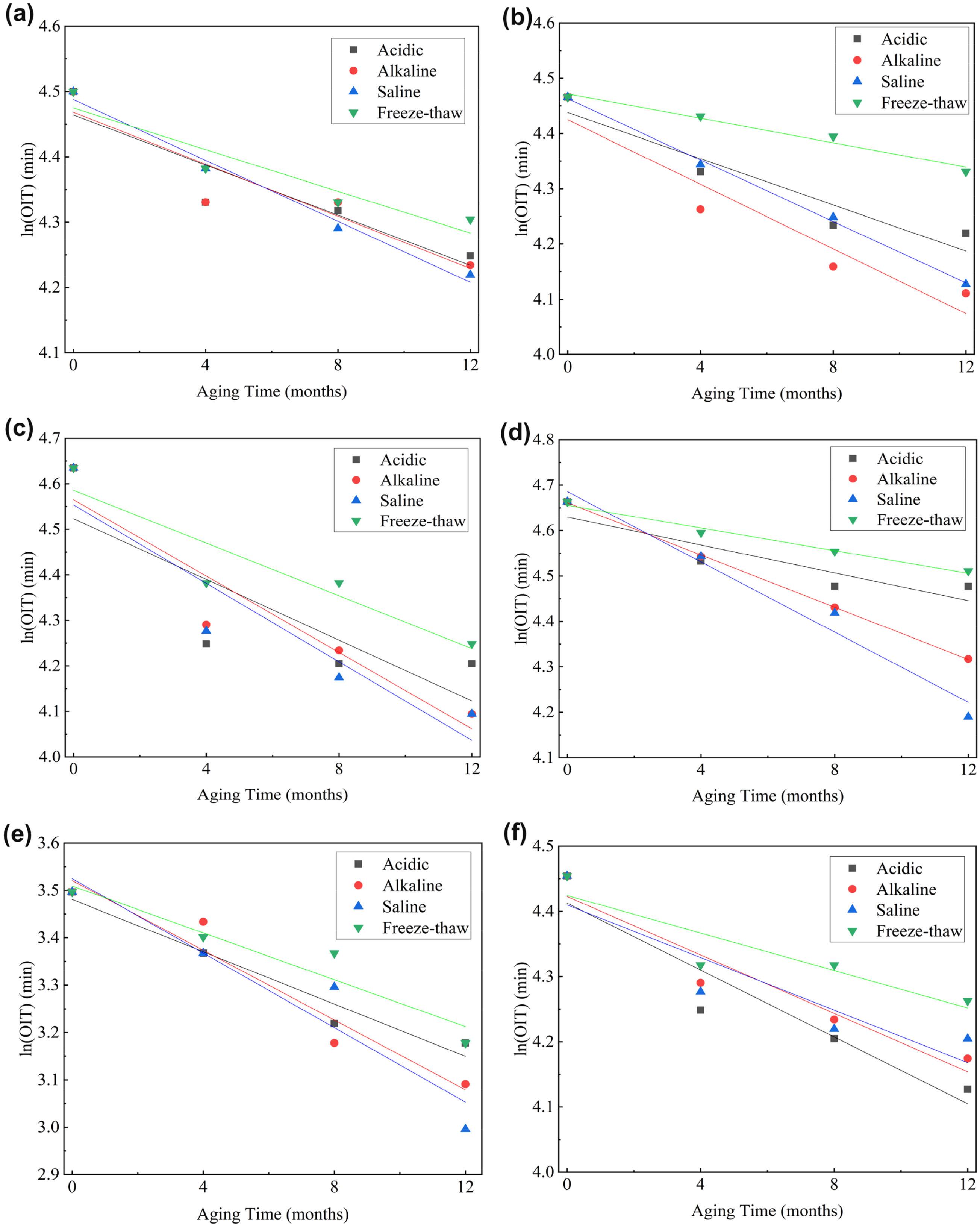

Fig. 7 Plot of ln(OIT) with aging time under different exposure conditions **a** A-R **b** A-V **c** B-R **d** B-V **e** C-V **f** D-R

Table 2 AO depletion rates of HDPE pipes under different exposure conditions

Sample	Exposure conditions (month^{-1})				Comparison		
	Acidic	Alkaline	Saline	Freeze/thaw	Acidic to FT	Alkaline to FT	Saline to FT
A-R	0.019 ± 0.0034	0.020 ± 0.0007	0.023 ± 0.0014	0.016 ± 0.0008	1.19	1.25	1.44
A-V	0.021 ± 0.0003	0.029 ± 0.0014	0.028 ± 0.0002	0.011 ± 0.0002	1.91	2.64	2.55
B-R	0.033 ± 0.0029	0.042 ± 0.0031	0.043 ± 0.0014	0.029 ± 0.0023	1.14	1.45	1.48
B-V	0.015 ± 0.0004	0.029 ± 0.0023	0.039 ± 0.0031	0.012 ± 0.0005	1.25	2.42	3.25
C-V	0.028 ± 0.0010	0.037 ± 0.0023	0.039 ± 0.0019	0.025 ± 0.0013	1.12	1.48	1.56
D-R	0.026 ± 0.0028	0.022 ± 0.0057	0.020 ± 0.0006	0.014 ± 0.0014	1.86	1.57	1.43

FT freeze/thaw

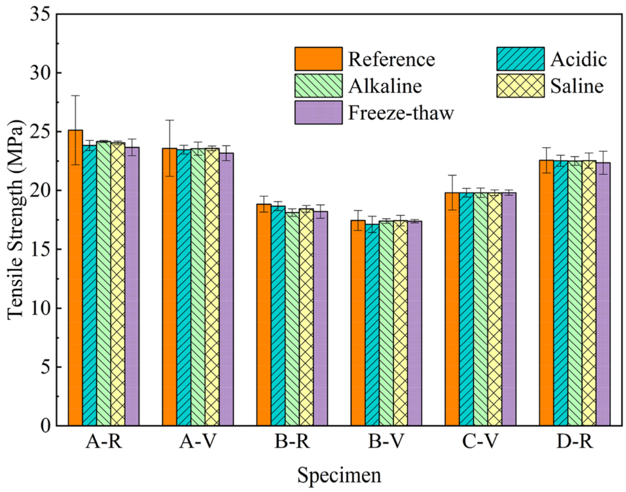

Fig. 8 Tensile strength of HDPE pipes before and after exposure to different conditions

(no significant difference between measurements). In contrast, if one or more treatments are significantly different, post-hoc tests are then applied to clarify which pairs of treatments are significantly different. In this study, Tukey's HSD test was applied as a post-hoc test. The first issue is to determine the critical value of Tukey's HSD (q-critical) based on the number of treatments and degree of freedom for the error term and for the alpha level (0.05) from the tables of the inverse Studentized Range Distribution [40]. Next, Tukey's HSD q-statistic is calculated by taking the absolute value of the difference between pairs of means and dividing it by the standard error of the mean, as shown in Eqs. 1 and 2.

$$q_{i,j} = \frac{\left|x_i - x_j\right|}{SE} \tag{1}$$

$$SE = \sqrt{\frac{1}{2}MS_w\left(\frac{1}{n_i} + \frac{1}{n_j}\right)} \tag{2}$$

where $q_{i,j}$ is Tukey's HSD q-statistic; $x_i - x_j$ is the difference between pairs of means; SE is the standard error of a group mean; MS_w is the mean square within group; n is the group; and i,j are the columns labeled i and j.

For Tukey's post-hoc test, if the q-statistic value is less than q-critical, there is no significant difference between the pair of treatments and vice versa.

Results and Discussions

Rates of Antioxidant Depletion

Although OIT measurements cannot identify the types of antioxidants, the relative concentration of antioxidant retained in the aging specimen can be determined. A higher OIT value indicates a greater amount of AO [41]. In other words, the OIT is proportional to the antioxidant concentration. In our investigation, the AO depletion rate followed a first-order decay model, which can be expressed as Eqs. 3 and 4 [6, 13].

$$OIT_t = OIT_0.e^{-st} \tag{3}$$

or taking the logarithm on both sides

$$\ln\left(OIT_t\right) = -st + \ln\left(OIT_0\right) \tag{4}$$

where OIT_t is the OIT at time t (in min), OIT_0 the initial OIT value (in min), s the rate of AO depletion (in month^{-1}), and t the aging time (in months).

Figure 6a–f show the OIT value as a function of time and exposure conditions for six corrugated HDPE pipes. The OIT was measured at various time intervals with three replicate specimens for each pipe. The exponential curve fitting was used to fit experimental data. The results indicate that, after 12 months of aging in different solutions, the OIT percent retained was above 55%. Therefore, all the pipes met the OIT minimum of 20 min according to ASTM D3895-19 [38]. Hsuan and Koerner [6] observed that OIT decreased with time and different exposure conditions. In

Table 3 One-way ANOVA of tensile strength before and after immersion

Specimen	Description	Sum of squares	df	Mean squares	F-value	P-value	F-critical
A-R	Between groups	6.1	4	1.5	0.665	0.625	3.007
	Within groups	36.8	16	2.3	–	–	–
	Total	42.9	20	–	–	–	–
A-V	Between groups	0.5	4	0.1	0.076	0.988	3.007
	Within groups	25.5	16	1.6	–	–	–
	Total	26.0	20	–	–	–	–
B-R	Between groups	1.6	4	0.4	1.684	0.203	3.007
	Within groups	3.8	16	0.2	–	–	–
	Total	5.4	20	–	–	–	–
B-V	Between groups	0.3	4	0.1	0.243	0.910	3.007
	Within groups	5.1	16	0.3	–	–	–
	Total	5.4	20	–	–	–	–
C-V	Between groups	0.0004	4	9E-05	0.0002	1.000	3.007
	Within groups	9.9519	16	0.6	–	–	–
	Total	9.9523	20	–	–	–	–
D-R	Between groups	0.1	4	0.03	0.043	0.996	3.007
	Within groups	9.9	16	0.6	–	–	–
	Total	10	20	–	–	–	–

other words, antioxidant depletion is dependent on chemical reactions and leaching. Reaction with oxygen and oxidizers as well as diffusion/extraction processes cause the consumption of AO [42]. Figure 7a–f present the typical plot of ln(OIT) with aging time for pipes immersed in saline, acidic, and alkaline solutions and subjected to freeze/thaw cycles. With a linear regression of data (Eq. 3), the AO depletion rate corresponds to the slopes of the graph. Table 2 presents the rate of antioxidant depletion values determined for these conditions. The results show that the lowest depletion rates were observed with the freeze/thaw cycles, while the highest depletion rates were obtained with the saline solution. The test temperature in saline solution was higher than that in freeze/thaw cycles, which accelerated the diffusion of antioxidants within the sample. A comparison between the AO depletion rate after immersion in the acidic, alkaline, and saline solutions with freeze/thaw cycles indicates that the depletion rate in saline solution varied from 1.43 to 3.25 times the depletion rate after freeze/thaw cycles (Table 2). It should be highlighted that the initial OIT value differed from one pipe to the next because of the composition of their antioxidant and stabilizer package, which is exclusive and proprietary for each manufacturer. These pipes might have been stabilized with phenolic (Irganox 1010) and phosphate (Irgafos 168) antioxidants, which would probably make them more susceptible to chemical solutions than to freeze/thaw cycles. In addition, freeze/thaw cycles are the physical process and their effect on the AO depletion rate differs from the acidic, alkaline, and saline solutions. The AO depletion rate was slightly faster in the chemical solutions rather than in freeze/thaw cycles, possibly due to the higher solubility of antioxidants. This is related to their polar structure as well as the elevated temperatures in chemical solutions, which accelerate the diffusion/extraction from the material surface. The thickness of the pipe wall could also be a factor influencing antioxidant depletion [8, 43, 44]. In our study, the specimens were all cut from pipe liners with the same thickness (2 mm). For the OIT measurements, the 5–10 mg specimens were taken from the skin of aged specimens. Therefore, the pipe-wall thickness factor is not mentioned herein. In addition, both sides of the investigated specimens were in contact with the solutions. It should be noted that polyethylene (PE) is well-known for its low chemical reactivity with polar fluids and solvents. Exposing both sides of

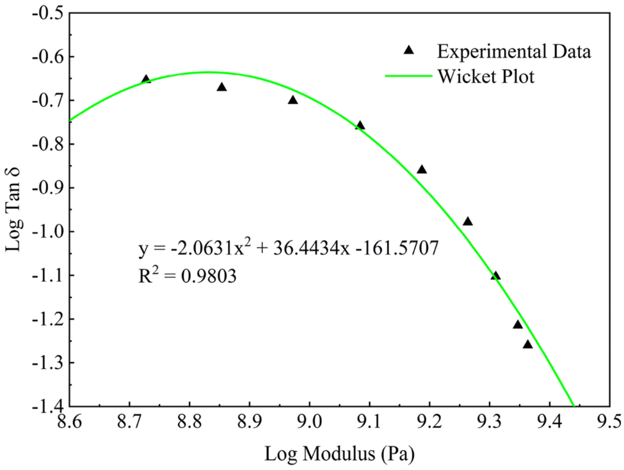

Fig. 9 Theoretical wicket plot

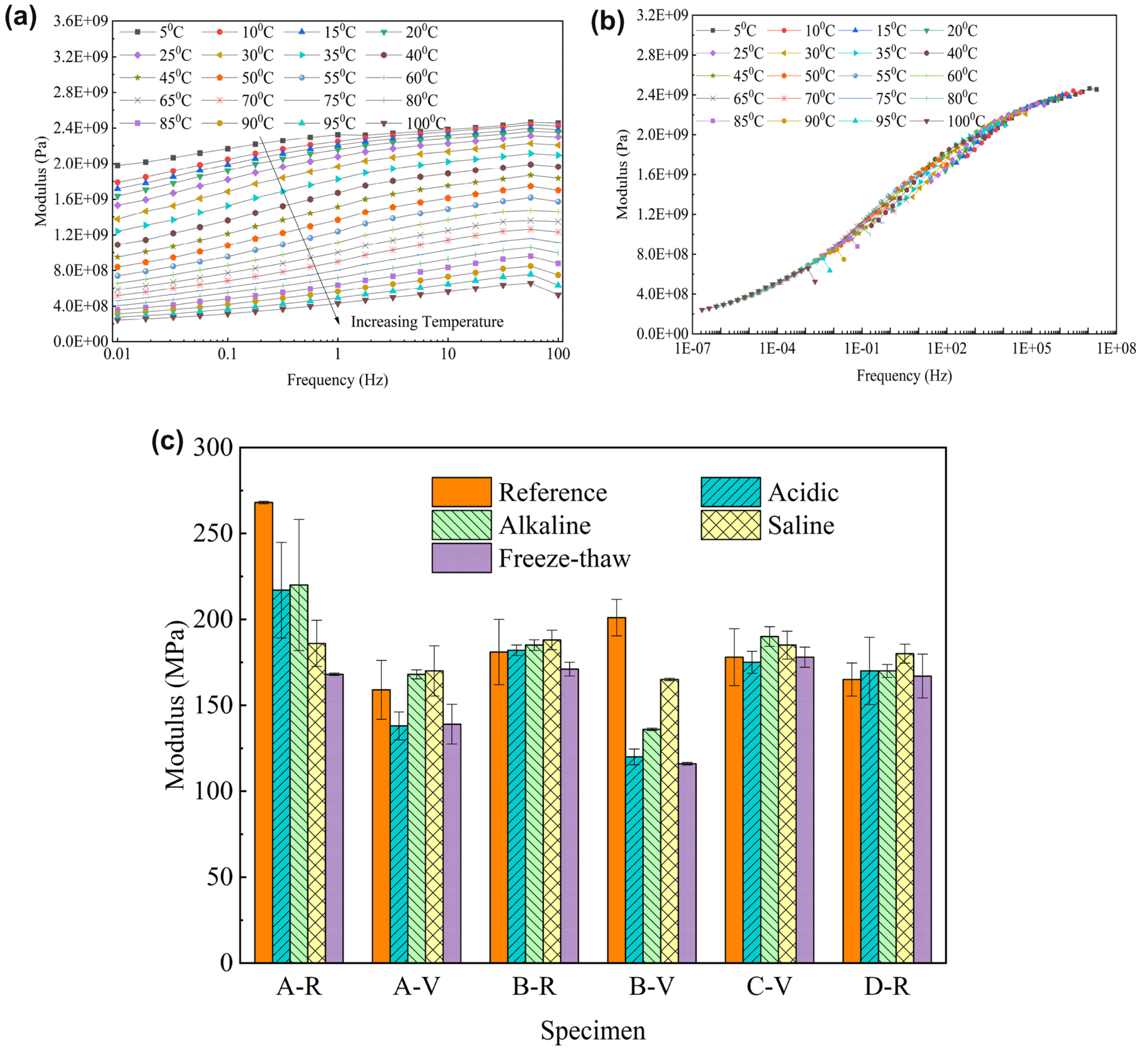

Fig. 10 TTS/DMA analysis **a** Temperature–frequency sweep **b** A typical of master curve for A-R pipe **c** Creep modulus of HDPE pipes before and after exposure to different conditions

specimens, therefore, helps to accelerate the degradation of the material. In fact, the actual antioxidant depletion rate will be significantly lower since only one side of the pipe is exposed to the medium. Lastly, the AO depletion rates in recycled and virgin pipes cannot be compared because they contain different and unknown antioxidants, which do not react in the same way with their environments.

Tensile Strength

Figure 8 shows the average tensile strength of original and aged HDPE pipes for 12 months. The standard deviation is represented by vertical bars. The stress–strain behavior of specimens is similar to that in another study [24] in which three regions were observed: elastic, neck propagation, and plastic. The tensile strength of the specimens before immersion was considered the reference. There was no significant degradation in pipe tensile strength before and after immersion in the different solutions. There was, however, a small variation between the measurements. Therefore, a one-way analysis of variance (ANOVA) was used to clarify if there are significant differences between measurements. Table 3 shows that the F-value was less than the F-critical value and that the p-value was greater than 0.05 for tensile strength.

Table 4 One-way ANOVA of creep modulus before and after immersion

Specimen	Description	Sum of squares	df	Mean squares	F-value	P-value	F-critical
A-R	Between groups	11,802.1	4	2950.5	4.445	0.042	4.120
	Within groups	4646.2	7	663.7	–	–	–
	Total	16,448.3	11	–	–	–	–
A-V	Between groups	2778.3	4	694.6	4.848	0.020	3.478
	Within groups	1432.7	10	143.3	–	–	–
	Total	4211.0	14	–	–	–	–
B-R	Between groups	496.4	4	124.1	1.448	0.289	3.478
	Within groups	857.3	10	85.7	–	–	–
	Total	1353.7	14	–	–	–	–
B-V	Between groups	13,824.8	4	3456.2	90.217	4E-06	4.120
	Within groups	268.2	7	38.3	–	–	–
	Total	14,093.0	11	–	–	–	–
C-V	Between groups	451.6	4	112.9	1.261	0.347	3.478
	Within groups	895.3	10	89.5	–	–	–
	Total	1346.9	14	–	–	–	–
D-R	Between groups	386.9	4	96.7	0.709	0.604	3.478
	Within groups	1364.7	10	136.5	–	–	–
	Total	1751.6	14	–	–	–	–

Therefore, under laboratory conditions including conditioning in saline, acidic, or alkaline solutions as well as freeze/thaw cycles, it can be concluded that the tensile strength was not significantly affected. Furthermore, the tensile strength values for the recycled pipes exposed to the different solutions were relatively unchanged compared to the reference values.

Creep Modulus

Time–temperature superposition (TTS) is an extrapolation technique widely applied to predict the creep modulus of materials. The theory of this technique is based on the empirical Williams–Landel–Ferry (WLF) relationship [45]. Although this technique was initially developed for amorphous polymers, Nielsen and Landel [46] recommended that it could also be employed for semicrystalline polymers (HDPE). The wicket plot [47, 48] was used to confirm that the materials in this study were suitable for analysis with the WLF relationship. A specimen measuring $40\times10\times2$ mm^3 was heated from 0 to 100 °C at a heating rate of 5 °C/min at 1 Hz using the single-cantilever mode. As shown in Fig. 9, the wicket plot is almost symmetrical, which is suitable for applying the principle of TTS.

As discussed above, the effects of temperature and frequency variations are interchangeable according to the TTS principle. Therefore, data obtained over a limited frequency range can be extended by applying a shift factor to data obtained over a range of temperatures. First, a specimen was heated from 5 to 100 °C at steps of 5 °C along a range of frequencies between 0.01 and 100 Hz (Fig. 10a). The curves obtained were then shifted to generate a master curve (Fig. 10b) using a reference temperature and shift factor as illustrated in the WLF equation (Eq. 5). A reference temperature of 50 °C was used, which is just below the softening temperature of the materials. With the TTS principle, temperatures below the reference temperature are shifted to higher frequencies, while temperatures above are shifted to lower frequencies.

$$\log(aT) = \frac{-C_1(T - T_r)}{C_2 + T - T_r} \tag{5}$$

where a_T is the shift factor, C_1 and C_2 the WLF coefficients (K), T the temperature (K), and T_r the reference temperature (K).

Figure 10c shows the creep modulus of the HDPE pipes before and after immersion in the different solutions for 6.5 months. A higher variation in the creep modulus values was observed. Therefore, a one-way analysis of variance (ANOVA) was used to determine if there were a significant difference between the measurements before and after immersion. Table 4 indicates that the F-value is higher than the F-critical value for specimens A-R, A-V, and B-V, which suggests that one or more pairs of measurements were significantly different. In contrast, the creep modulus values for specimens B-R, C-V, and D-R were relatively unchanged before and after immersion. Tukey's HSD test was then applied to confirm whether the pairs of measurements were indeed significantly different. Table 5 shows no significant difference between the pair of measurements for specimens A-R and A-V as the q-statistic value is less than the q-critical

Table 5 Post-hoc Tukey's HSD test

Specimen	Treatment pair	Tukey's HSD q-statistic	Tukey's HSD q-critical	Tukey's HSD inference
A-R	Ref. vs. Acidic	3.0367	5.0615	Insignificant
	Ref. vs. Alkaline	2.8362	5.0615	Insignificant
	Ref. vs. Saline	4.5012	5.0615	Insignificant
	Ref. vs. freeze/thaw	5.4893	5.0615	Significant
	Acidic vs. alkaline	0.2241	5.0615	Insignificant
	Acidic vs. saline	1.8942	5.0615	Insignificant
	Acidic vs. freeze/thaw	2.9765	5.0615	Insignificant
	Alkaline vs. saline	2.0946	5.0615	Insignificant
	Alkaline vs. freeze/thaw	3.1770	5.0615	Insignificant
	Saline vs. freeze/thaw	0.9881	5.0615	Insignificant
A-V	Ref vs. acidic	3.0388	4.6552	Insignificant
	Ref vs. alkaline	1.2541	4.6552	Insignificant
	Ref vs. saline	1.4953	4.6552	Insignificant
	Ref vs. freeze/thaw	2.8941	4.6552	Insignificant
	Acidic vs. alkaline	4.2930	4.6552	Insignificant
	Acidic vs. saline	4.5341	4.6552	Insignificant
	Acidic vs. freeze/thaw	0.1447	4.6552	Insignificant
	Alkaline vs. saline	0.2412	4.6552	Insignificant
	Alkaline vs. freeze/thaw	4.1483	4.6552	Insignificant
	Saline vs. freeze/thaw	4.3894	4.6552	Insignificant
B-V	Ref vs. acidic	22.7602	5.0615	Significant
	Ref vs. alkaline	16.4778	5.0615	Significant
	Ref vs. saline	9.2192	5.0615	Significant
	Ref vs. freeze/thaw	21.4837	5.0615	Significant
	Acidic vs. alkaline	3.8796	5.0615	Insignificant
	Acidic vs. saline	11.1381	5.0615	Significant
	Acidic vs. freeze/thaw	1.1263	5.0615	Insignificant
	Alkaline vs. saline	6.6261	5.0615	Significant
	Alkaline vs. freeze/thaw	4.5697	5.0615	Insignificant
	Saline vs. freeze/thaw	11.1959	5.0615	Significant

value. The difference was mainly observed with specimen B-V before and after immersion in the different solutions. Moreover, the average molecular weight of HDPE pipes before and after immersion in saline was investigated, as shown in Fig. 11. As mentioned earlier, the saline solution was a more severe environment than the other solutions. The average molecular weight was therefore investigated only after immersion in saline. Figure 11 shows no significant difference in molecular weight before or after immersion in saline, except for the B-V specimens. Therefore, the molecular weight could be the cause of the variation in creep modulus values of the B-V specimens before and after immersion in the various media. Many parameters of pipe manufacturing (e.g., composition, grade of resin) could explain this change. They are, however, protected by trade secrets and therefore were not available to the authors. Moreover, further studies need to be conducted to better explain this change. Based on the results, it can be concluded that the creep modulus of the HDPE pipes might not be significantly affected by the saline, acidic, or alkaline solutions or the freeze/thaw cycles.

Conclusion

The antioxidant depletion rate of six HDPE pipes was evaluated with accelerated aging in acidic, alkaline, and saline solutions and under freeze/thaw cycles. The results show that the OIT value decreased with aging time in the solution. The OIT percent retained was above 55% for all solutions. After the 12 months of aging had been completed, the antioxidant depletion rate was: saline > alkaline > acidic > freeze/thaw. For specimens subjected to freeze/thaw cycles, the depletion was about 1.12 to 1.91 lower than that in the acidic solution, 1.25 to 2.64 times lower than that in the alkaline solution, and 1.43–3.25 times lower than in the saline solution. The

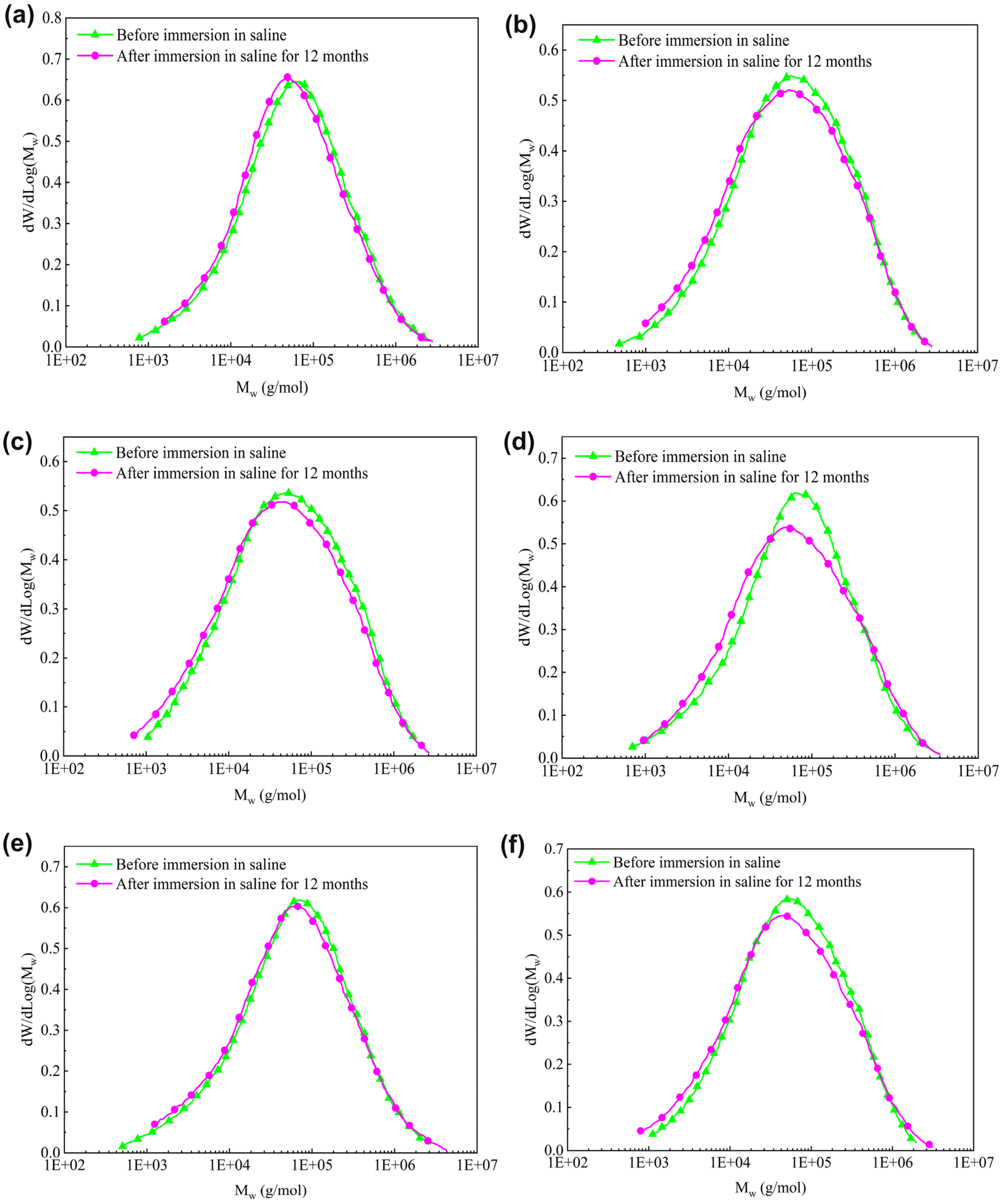

Fig. 11 Molecular-weight distribution curves of the HDPE pipes before and after immersion in saline: **a** A-R **b** A-V **c** B-R **d** B-V **e** C-V **f** D-R

antioxidant depletion rate herein refers only to the first stage (t_1) of the degradation process and does not consider stages t_2 and t_3, corresponding to the degradation with a reduction in properties. The HDPE pipes all met the OIT minimum of 20 min according to ASTM D3895-19, regardless of the manufacturing process and antioxidant package. Moreover, it must be pointed out that the laboratory conditionings were more corrosive than the actual environment of the pipes in the field. Based on one-way analysis of variance (ANOVA) and Tukey's HSD tests, the specimens exhibited no significant changes in tensile strength or creep modulus after immersion in the different solutions. Whether produced with recycled or virgin resins, the pipes maintained good mechanical properties after laboratory aging. The findings from this study again confirm the appropriateness of using pipes made with recycled HDPE for transportation infrastructure applications. Recycled pipes can deliver short- and long-term performance when they contain appropriate antioxidants.

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Data Availability Some or all data, models, or code that support the findings of this study are available from the corresponding author upon reasonable request.

Declarations

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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