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Aging Performance and an Improved Evaluation Method for PE80 and PE100 Pipelines for Urban Gas

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Abstract: Polyethylene (PE) pipes are widely used in urban gas transportation due to their good toughness and corrosion resistance. Currently, the designed service life of a PE pipeline is 50 years, and some urban gas PE pipelines are approaching their service life. However, research on the aging assessment of PE pipelines is not complete, and it is impossible to effectively predict their aging status in service. Once urban gas PE pipelines are damaged, serious accidents may be caused. PE80 and PE100 pipelines are commonly used for urban gas, and improved accelerated aging tests were conducted considering different conditions of pressure, pipe diameter, and temperature. According to the experimental results, an aging life prediction method for PE pipes was constructed based on the Arrhenius formula considering multiple effect factors.

Keywords: polyethylene (PE) pipeline; antioxidant; thermal oxidative aging; life prediction

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1. Introduction

As a kind of polymer material, PE pipes inevitably age during the service process [1–4]. There are three main types of aging: photo-oxidative, thermo-oxidative, and stress cracking (Table 1). For buried PE pipes, the thermo-oxidative process is the main process, which also includes stress crack expansions. The aging process depends on the material, the environment, and the working conditions [5].

There are two main methods for research on the aging life prediction of PE pipes. Some research is focused on the properties of the PE material [2,6,7]. In this research, the PE specimens were required to be made into plate-like samples for aging experiments. For example, based on cyclic cracked bar (CRB) tests on four types of pipes, including PE80, PE80-MD, PE100, and PE100-RC, a life prediction method was proposed by Frank et al. [8], combined with linear elastic fracture mechanics. According to thermal oxidative aging experimental results, the relationships between oxidation induction time (OIT) and mechanical properties were explored by Byrne et al. [9], including PE63, PE80B, PE80, and PE100 pipes. However, the aging characteristics of the pipes cannot be actually reflected according to these kinds of specimens. Another piece of research was about integral PE pipes aging under experimental conditions that were more similar to actual aging conditions. According to thermal oxygen aging tests, a life prediction method for PE80 considering the inner pressure was proposed by Wang Yang [10,11]. Based on the thermal oxidative aging tests of PE100 pipes under cyclic pressure loading, a method for service life predicting of PE100 pipes was established by Chen Guohua [12,13]. However, research about the aging status of PE pipes with different diameters is not enough. An effective method for aging status evaluating of PE pipes has not yet been established. Based on the common thermal oxygen aging method, the experimental equipment was improved by being combined with the operating characteristics of urban gas PE pipes in this paper. Accelerated aging tests of PE pipes for various pressures, materials, and diameters were undertaken. Based on these experimental results, a prediction model for the aging status of PE pipes was proposed.

Table 1. Aging types for polyethylene pipelines.

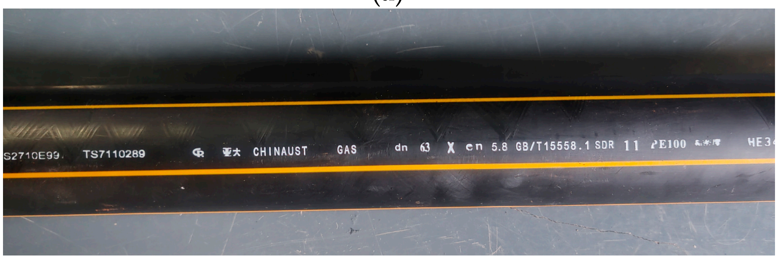
Aging Type	Mechanism	Description
Photo-oxidative	Photodegradation	The polymer chains will break down under prolonged exposure to ultraviolet (UV) light, causing surface embrittlement and discoloration and impacting resistance reduction.
Thermo-oxidative	Chain Scission and Crosslinking	The polymer chains will break or crosslink in an environment with oxygen, which reduces the flexibility and strength of the PE pipe.
Stress Cracking	Crack Initiation and Propagation	Micro-cracks on a PE pipe may occur in service, which will grow under mechanical stress.

2. Accelerated Aging Experiment

2.1. Sample Tube Preparation

At present, PE pipes are mainly used for medium- and low-pressure gas transport [14–17]. Commonly, the design pressure of these PE pipes is 0.4 MPa and the operating pressure is 0.2 MPa. SDR11 DN63 PE80, SDR11 DN110 PE80, and SDR11 DN63 PE100 pipes produced by the same company were used in the thermal oxygen accelerated aging experiments mentioned in this paper (Figure 1). In the aging experiments, the DN63 pipes should be longer than 320 mm and the DN110 pipes should be longer than 490 mm.

(**a**)



(**b**)

(**c**)

Figure 1. PE pipes used for aging test: (**a**) PE80 SDR11 DN63 (melt mass flow rate (190 °C/5 kg): 0.83 g/10 min); (**b**) PE100 SDR11 DN63 (melt mass flow rate (190 °C/5 kg): 0.23 g/10 min); and (**c**) PE80 SDR11 DN110 (melt mass flow rate (190 °C/5 kg): 0.83 g/10 min).

2.2. Aging Experimental Process

The improved aging experimental platform is shown in Figure 2. In order to prevent the oxidation of the inner wall during the accelerating aging experiment, nitrogen gas was used for the inner pressure control. The experimental pressure can be automatically controlled to be constant through the pressure regulator and solenoid valve. During the experiment, the experimental pressure was controlled constantly by the pressure-raising and pressure-reducing solenoid valves. Considering the melting point of the materials, the design pressure, and the operating pressure of the pipes, the experimental conditions were designed as shown in Table 2.

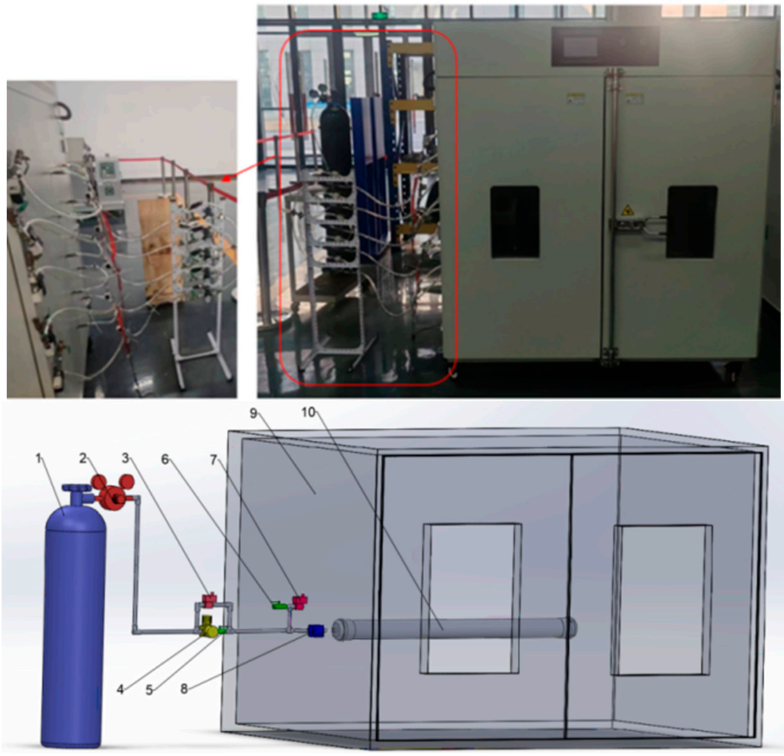


Figure 2. Aging experimental platform: 1—nitrogen cylinder; 2—cylinder regulator; 3—pressure-raising solenoid valve; 4—regulator; 5—pressure-raising ball valve; 6—pressure-reducing ball valve; 7—pressure-reducing solenoid valve; 8—electronic barometer; 9—thermo-oxidative aging chamber; and 10—polyethylene pipe.

Table 2. Aging conditions for polyethylene pipelines.

Temp/°C	Pressure/MPa	Aging Times				
		1st/h	2nd/h	3rd/h	4th/h	5th/h
80	0	0	144	288	576	864
	0.2	0	144	288	576	864
	0.4	0	144	288	576	864
90	0	0	96	192	288	384
	0.2	0	96	192	288	384
	0.4	0	96	192	288	384
100	0	0	24	48	96	192
	0.2	0	24	48	96	192
	0.4	0	24	48	96	192
110	0	0	8	16	32	72
	0.2	0	8	16	32	72
	0.4	0	8	16	32	72

3. The Aging Status of PE Pipes

An antioxidant is usually added to improve the anti-aging ability of PE pipes during their production. An OIT test is usually used to determine the consumption of the antioxidant and the aging status of the PE pipes. A differential scanning calorimetry (DSC) was used to detect changes in the OIT during polyethylene pipe aging by Hoang et al. [18], who characterized the content of the residual antioxidant in the PE pipes. Its effectiveness was verified by the method of iodine titration. The properties of PE gas pipelines were tested by Bachir Bey et al. [19], with the pipes having been in service in Algeria for 30 years. Even though the mechanical properties still met the manufacturer's requirements, the density and OIT values had significantly decreased. According to the relevant standards [20], PE pipes should be replaced when the degradation of the outer surface exceeds 10% of the wall thickness. Therefore, 9% of the wall thickness from the outer surface of the PE pipe was taken as the testing sample for the OIT testing in the aging experiments [5].

3.1. OIT Tests

The OIT was measured through a DSC (NETZSCH DSC 200 F3 Maia®, NETZSCH-Gerätebau GmbH, Selby, Germany) (Figure 3). The specimens were tested according to ISO 11357-6 [21]. The thickness of the test specimen obtained from the pipe was about 9% of the wall thickness and its weight was 15.0 ± 0.5 mg. The temperature was raised at a rate of 20 °C/min to 200 °C under a nitrogen flow rate of 50 cm^3/min, and the thermogram was recorded once per minute. Then, the nitrogen was switched to oxygen at a flow rate of 50 cm^3/min and this point on the thermogram was marked. The thermogram continued to be recorded until the exotherm descended to its lowest point.

Figure 3. Differential scanning calorimeter.

3.2. Results and Comparison

The minimum OIT value was obtained from the three experimental results as the final test result. The results are shown in Table 3, and the confidence coefficient of the experimental results was higher than 95%. An aging analysis of PE pipes can be conducted according to the experimental data below.

Table 3. Oxidation induction periods under various experimental conditions.

Temp/°C	Time/h	OIT/min								
		PE80 DN63			PE100 N63			PE80 DN110		
		0 MPa	0.2 MPa	0.4 MPa	0 MPa	0.2 MPa	0.4 MPa	0 MPa	0.2 MPa	0.4 MPa
80	0	88.5	88.5	88.5	87.3	87.3	87.3	88.5	88.5	88.5
	144	82.3	79.9	78.4	82	79.8	79.2	84.4	84	83.9
	288	78.1	76.7	74.6	79.7	78.2	77.8	82.3	82.1	82
	576	76	74	71.2	78.2	75.8	74.6	80.1	80	78.1
	864	73.8	70.1	65.4	76.6	74	71	78.9	76.8	75.4
90	0	88.5	88.5	88.5	87.3	87.3	87.3	88.5	88.5	88.5
	96	81.2	80	79	83.2	82.7	82.6	85.1	84.4	83.8
	192	80.3	78.1	75.7	82.3	78.6	77.8	83	82.5	82.2
	288	77.9	76.7	74.8	80.5	77.4	75.8	81.4	80.2	79.5
	384	76.7	74.7	73.3	77.1	74.8	72	79.4	78	76.5
100	0	88.5	88.5	88.5	87.3	87.3	87.3	88.5	88.5	88.5
	24	86.1	84.3	83.3	84.8	82.8	81.9	85	84.5	83.7
	48	83.3	82.4	79.9	82	79.2	80.3	83.8	83	82.5
	96	80.2	77.2	76.3	79.8	77.2	77	83.2	82.3	80.5
	192	78.5	76.3	73.4	77.8	75	73.8	81	79.4	78
110	0	88.5	88.5	88.5	87.3	87.3	87.3	88.5	88.5	88.5
	8	85.8	85.6	85.4	84.6	83.9	83.6	85.6	84.7	83.9
	16	85.6	83.1	82	83.1	81.5	80.1	85.2	84.2	83.3
	32	83.5	81.9	78.6	82.2	79.8	78.5	83.5	83.4	82.3
	72	80.6	79.1	76.3	80.1	77.8	74.5	82	80.1	78.3

4. Aging Rate Calculation Model

4.1. Aging Rate Calculation Results

The OIT data of aged SDR11 DN63 PE80, SDR11 DN110 PE80, and SDR11 DN63 PE100 gas pipes were obtained from the experiments above. According to the Arrhenius equation, the aging rates of PE pipes can be calculated as follows [22].

$$Q = f(q) = A\exp(-Kt) \quad (1)$$

where Q, $f(q)$ is the rate constant (F/F_0); F is the property of materials at failure; F_0 is the current property; t is the aging time; K is the reaction rate constant; and A is the material constant.

Equation (1) can be changed into Equation (2).

$$F = AF_0\exp(-Kt) \quad (2)$$

The relationship between K and the temperature follows the Arrhenius equation (Equation (3)):

$$K = Ze^{\frac{-E}{RT}} \quad (3)$$

where T is the absolute temperature (K); E is the apparent activation energy (J·mol^{-1}); Z is the frequency factor (d^{-1}); and R is the gas constant (8.31 J·K^{-1}·mol^{-1}).

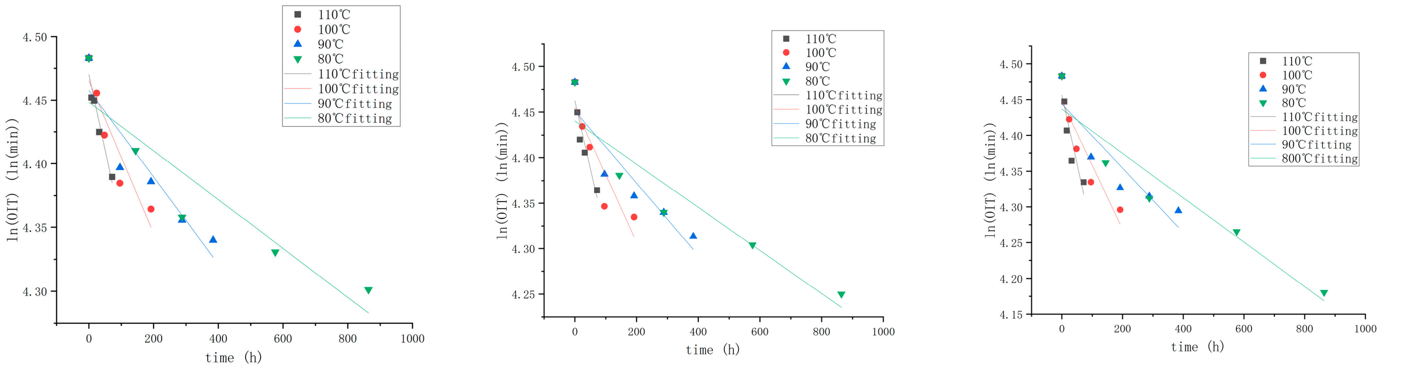
Let $X = t$, $Y = \ln F$, $a = \ln AF_0$, and $b = -K$; then, Equation (2) can be changed into a linear equation.

$$Y = a + bX \quad (4)$$

Let $X_1 = 1/T$, $Y_1 = \ln K$, $a_1 = \ln Z$, and $b_1 = (-E)/R$; then, Equation (3) can be changed as follows:

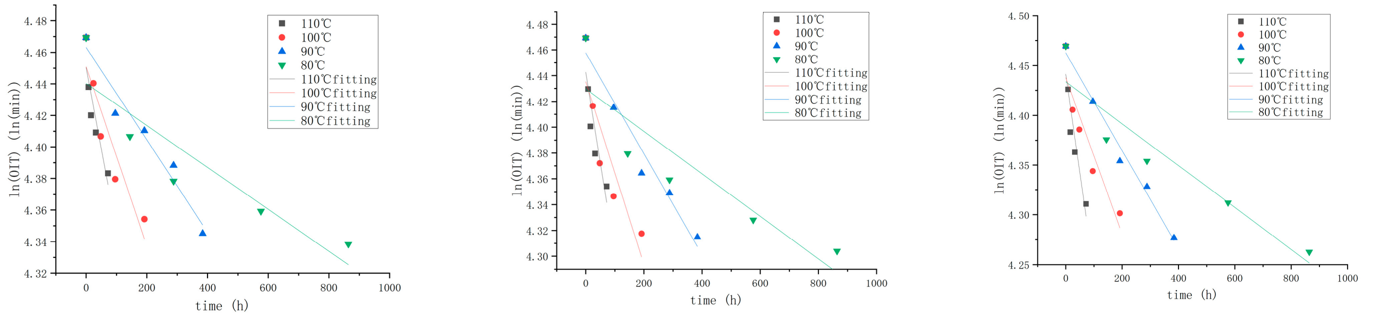
$$Y_1 = a_1 + b_1X_1 \quad (5)$$

The relationship between time and ln(OIT) was plotted to determine the reaction rate *K* (Figures 4–6,Table 4).



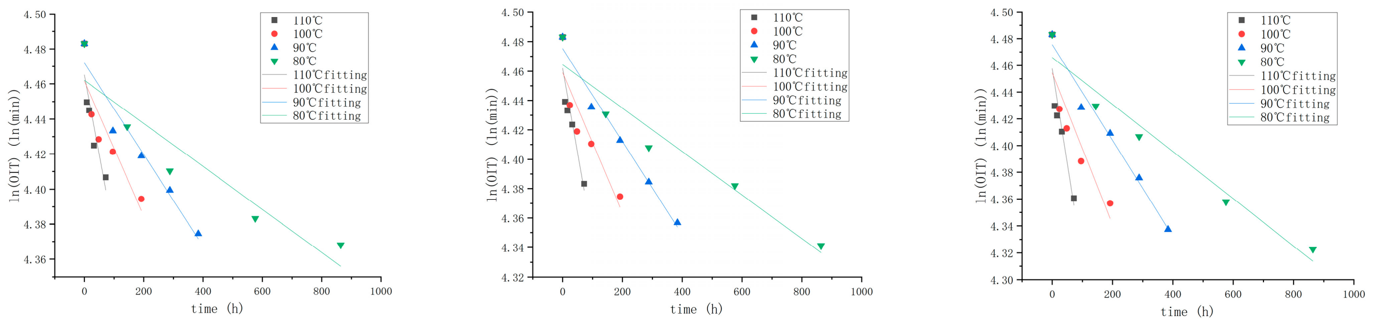
0 MPa 0.2 MPa 0.4 MPa

Figure 4. Relationship between time and ln(OIT) of PE80 DN63.



0 MPa 0.2 MPa 0.4 MPa

Figure 5. Relationship between time and ln(OIT) of PE100 DN63.



0 MPa 0.2 MPa 0.4 MPa

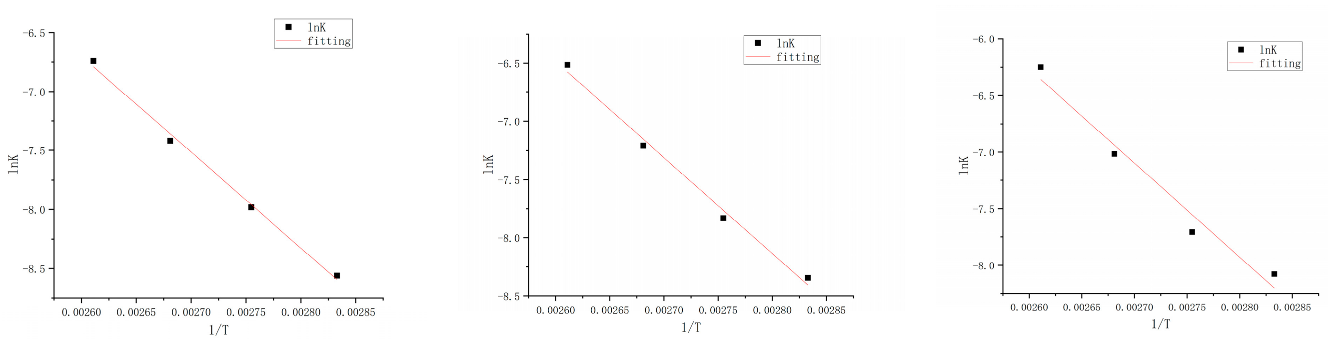
Figure 6. Relationship between time and ln(OIT) of PE80 DN110.

According to the fitting results of ln(OIT), the reaction rate constant *K* obtained under different conditions is shown in Table 4.

Table 4. *K* of different materials under experimental conditions.

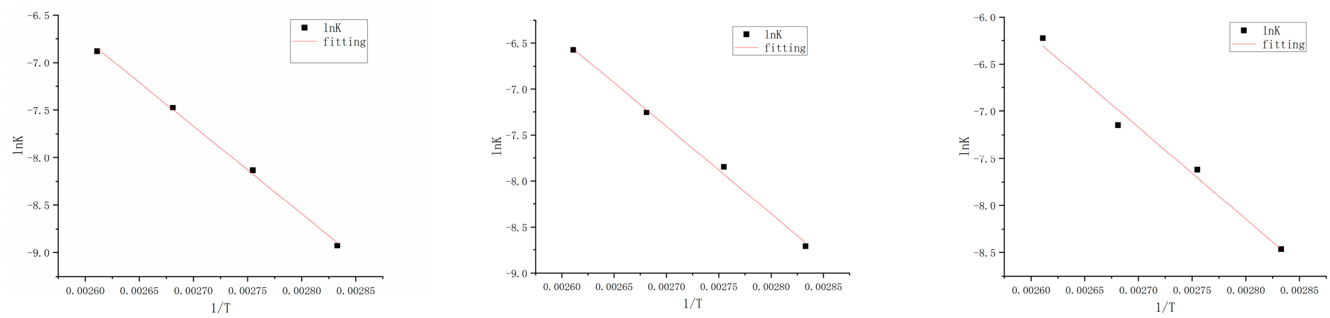
Pressure/MPa	Temp/°C	*K*		
		PE80 DN63	PE100 DN63	PE80 DN110
0	110	0.00118	0.00103	0.000911
	100	0.0006	0.000568	0.000385
	90	0.000341	0.000293	0.000261
	80	0.000191	0.000133	0.000123
0.2	110	0.0012	0.0014	0.00116
	100	0.000739	0.000707	0.000478
	90	0.000369	0.000391	0.000316
	80	0.000212	0.000165	0.000148
0.4	110	0.00193	0.00198	0.00142
	100	0.000896	0.000786	0.000567
	90	0.000449	0.000491	0.000358
	80	0.00031	0.000211	0.000176

The relationships between ln*K* and 1/*T* (temperature) are shown in Figures 7–9. The reaction rate constant *K* of different type of PE pipes at room temperature (23 °C) can be obtained.



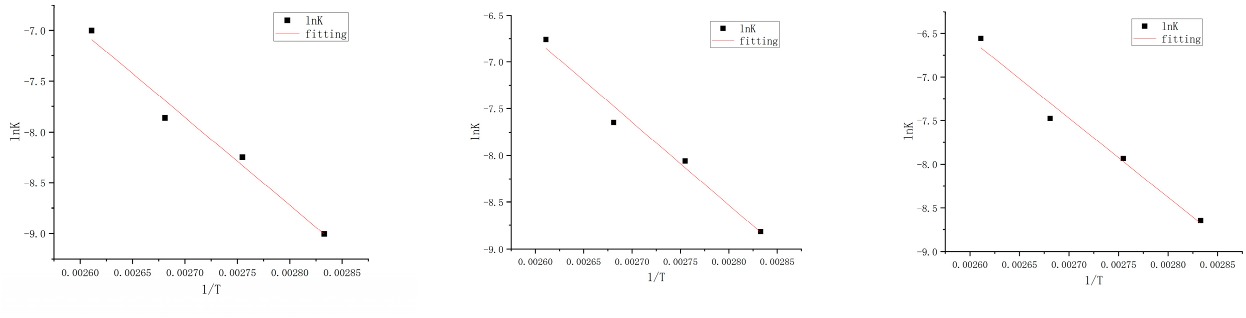
0 MPa 0.2 MPa 0.4 MPa

Figure 7. Arrhenius plot of ln*K* and 1/*T* of PE80 DN63.



0 MPa 0.2 MPa 0.4 MPa

Figure 8. Arrhenius plot of ln*K* and 1/*T* of PE100 DN63.



0 MPa 0.2 MPa 0.4 MPa

Figure 9. Arrhenius plot of ln*K* and 1/*T* of PE80 DN110.

Table 5 shows the reaction rate constant K obtained under different PE pipes at room temperature (23 °C).

Table 5. *K* of different PE pipes at room temperature (23 °C).

Pressure/MPa	*K*		
	PE80 DN63	PE100 DN63	PE80 DN110
0	2.19655×10^6	9.02178×10^7	1.10495×10^6
0.2	2.48867×10^6	9.68853×10^7	1.15063×10^6
0.4	2.93855×10^6	1.05662×10^6	1.21285×10^6

When the inner pressure is high, the diffusion and consumption of antioxidants in PE pipes will be accelerated and the aging rate is higher. The reaction rates of PE100 are lower than those of PE80 under the same service conditions. The antioxidant capacity of PE100 is better. When the SDRs are the same, the aging rate for the same type of PE pipe decreases as the diameter increases. According to the SDR, the relationship between the diameter and the reaction rate can be characterized by changes in the wall thickness.

4.2. The Modified Calculation Model

There is a relation between the activation energy, pressure, and frequency factor in the Arrhenius equation [12]. By substituting it into Equation (3), Equation (6) can be finally obtained, considering the effects of pressure and temperature.

$$K = Bexp\left[-\frac{E_0}{RT} - \frac{\alpha\frac{P_g}{P_f}}{T} + \beta\frac{P_g}{P_f}\right] \tag{6}$$

Based on the aging test results, the constant values for the life calculation of the PE80 DN63 and PE80 DN110 pipes are shown in Table 6.

Table 6. Wall thickness and calculation constants of PE80 DN110 and PE80 DN63.

	PE80 DN63	PE80 DN110
e_n/mm	5.8	10
A	0.970515076	0.981791678
E_0/J·mol^{-1}	67,611.62464	71,869.73828
B	1,889,022.391	5,392,124.386
α	46.2374	107.85093
β	0.22897	0.38765

As the wall thickness of the pipes increases, the material constant A and activation energy E_0 of PE80 DN110 and PE80 DN63 at standard pressure are basically unchanged. However, the values of B, α, and β change significantly as the wall thickness changes. Due to the influence of the wall thickness on the reaction rate K, it is necessary to consider the wall thickness while calculating the aging rate. In Equation (6), B, α, and β can be expressed as functions of the wall thickness e. Combined with Equation (2), an improved life prediction equation can be established as Equation (7), which includes the effects of temperature, pressure, and wall thickness. According to the calculation constants in Table 5, the functions of different wall thicknesses can be simplified. In subsequent research, more experimental data should be supplemented to improve the accuracy of the life prediction method.

$$t = \frac{\ln \frac{AF_0}{F}}{B(e)\exp\left[-\frac{E_0}{RT} - \frac{\alpha(e)\frac{P_g}{P_f}}{T} + \beta(e)\frac{P_g}{P_f}\right]} \tag{7}$$

5. Conclusions

According to the characteristic of aging for gas PE pipes, simulating aging experiments were established in this paper based on actual gas transmission conditions. Based on the experimental results, relevant research has been conducted combined with the current aging testing and life predicting methods. The conclusions are as follows:

1. The aging experimental results of PE pipes are closer to the actual performance than those obtained from specimens. The aging process can be simulated with different pipe types, diameters, and pressures according to this method. The aging properties of PE pipes can be well characterized by the OIT value of the materials.
2. In the aging experiments mentioned in this paper, the effects of pressure, temperature, diameter, and material types on the aging of PE pipes are considered. Based on the Arrhenius formula, the aging reaction rate at room temperature (service temperature) can be calculated under different conditions.
3. Comparing the reaction rate coefficients, the diffusion ability of antioxidants in PE pipes enhances as the pressure increases. The antioxidant content is higher in PE pipes with a bigger diameter and wall thickness (when the SRD is constant) and the anti-aging ability is stronger. When the wall thickness doubles, the aging rate decreases about half of the rate under no pressure conditions.
4. The anti-aging ability of PE100 is stronger than PE80. The aging life of PE100 is twice as much that of PE80 at room temperature.
5. Based on the Arrhenius formula, the evaluation of the aging status can be extended. Combined with the aging experimental results of PE pipes, an improved aging evaluating method considering wall thickness is proposed in this paper, which can be a support for subsequent model improvements.

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