# Study on the thermal aging kinetics of EPDM rubber

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**Abstract:** Heat aging stability is a hallmark of EPDM rubber and a requirement for the application of rubber hoses, sealing strips, roofing membranes, and other rubber products. This paper derives a model for predicting the service life of products at actual application temperatures by testing the tensile properties of EPDM vulcanized rubber after aging at temperatures of 100°C, 120°C, 130°C, and 150°C. The test results indicate that peroxide-vulcanized EPDM rubber exhibits higher strength and lower elongation at break compared to sulfur-vulcanized samples. After aging, the tensile strength initially shows a sharp decrease, and as the aging time increases, the decrease gradually flattens out. The elongation at break shows a higher and continuous loss, and the higher the aging temperature, the more severe the loss of strength and elongation at break. Both vulcanized rubber samples exhibit the same pattern of physical property loss. Based on the established aging model, it is inferred that a product using sulfur-vulcanized rubber can last for 17 years, while one using peroxide-vulcanized rubber can last for 44 years, providing strong evidence for the practical application of the product.

**Key words:** EPDM;fractional strain energy; rubber aging rate constant

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## 0 Introduction

Ethylene propylene diene monomer (EPDM) rubber itself possesses excellent aging resistance. Due to its stable and saturated polymer backbone structure, EPDM exhibits superior heat resistance, oxidation resistance, ozone resistance, and weathering resistance. EPDM, based on the copolymerization of ethylene and propylene, introduces non-conjugated dienes as a third monomer. Its molecular chain still maintains its original saturation, while the side chains contain a small amount of double bonds introduced by the addition of the third monomer. Therefore, EPDM not only retains the excellent heat resistance of the original saturated rubber but also enables the rubber compound to be vulcanized using various common vulcanization systems such as sulfur-accelator systems, thus achieving broader applications. The combination of ethylene and propylene monomers forms this chemically saturated and stable polymer backbone, and the third diene monomer, typically ethylnorbornene (ENB), dicyclopentadiene (DCPD), 1,4-hexadiene (HD), and other monomers, copolymerizes in a controlled manner to maintain a saturated main chain and place unsaturated side chains on the side chains that can be used for vulcanization or polymer modification chemistry. Since the production of EPDM began in 1960 , more than sixty years have passed, and many scholars have continuously improved and innovated its properties. Many scholars have studied EPDM rubber formulations . EPDM vulcanized rubber is widely used in automotive engine hood hoses and gaskets, door panel seals, heat-resistant hose conveyor belts, roof waterproof and thermal insulation layers, etc. ExxonMobil's P.S. Ravishankar specifically described the development history of EPDM. The high performance and long-term heat and weathering resistance of EPDM vulcanized rubber products are key requirements for many rubber applications.

**Biography:** Jiang Huabo (1981-), male, is an intermediate engineer, mainly engaged in the research and development of transmission belts and other rubber products.

Many scholars have conducted research on its aging properties . The rubber industry heavily relies on accelerated aging measurements, especially thermal aging, as quality assurance tests and specifications. Gao Yue from Shenyang University of Technology briefly mentioned using compression heat generation to predict the service life of rubber. However, compression heat generation testing is cumbersome and the cycle is long. To ensure product performance and guide product development, proposing a simple aging model to predict its lifespan is of great significance.

The process by which raw rubber, vulcanized rubber, and rubber products undergo damage to their composition and structure due to the combined effects of internal and external factors during storage, transportation, processing, and use, resulting in a gradual loss of their original excellent properties and even their loss of use value, is called aging. The aging process is divided into physical aging and chemical aging based on whether the reactions and changes are reversible. Generally, rubber aging refers to irreversible chemical aging, which is an inherent characteristic of rubber materials and one of the important reasons for rubber failure. This article studies the changes in the tensile properties of two types of EPDM rubber, which are vulcanized with sulfur and peroxide, after different aging times at different aging temperatures. A linear regression equation is derived for the change in fractional strain energy with aging time, and the aging rate constants at different temperatures are determined. Furthermore, the reaction activation energy is calculated, and a simple empirical model is established to predict the lifespan of the product under use and storage conditions, guiding its practical application.

## 1 Experimental part

### 1.1 Main raw materials

EPDM3250, sourced from Lanxess, Germany. Antioxidants, vulcanizing agents, sulfur, DCP, stearic acid, zinc oxide, co-crosslinking agents, paraffin wax, carbon black, and other additives are all commercially available.

### 1.2 Product formula

1#refers to sulfur-cured EPDM rubber; 2#refers to peroxide-cured EPDM rubber.

### 1.3 Main instruments and equipment

XSM-1/lO-120 type internal mixer, product of Shanghai Kechuang Rubber & Plastics Machinery Equipment Co., Ltd.; XK-160 type two-roll open mill, product of Shanghai Shuangyi Rubber & Plastics Machinery Co., Ltd.; MV2000 type Mooney viscometer and MDR3000 type curing press, products of Alpha Technology Co., Ltd. of the United States; XLB-D 600×600 type flat vulcanizing press, product of Zhejiang Huzhou Dongfang Machinery Co., Ltd.; AI-3000 type rubber tensile testing machine, product of High-Speed Rail Testing Instrument (Dongguan) Co., Ltd.

### 1.4 Sample preparation

The mixing of rubber compound is carried out in two stages. The first stage is conducted in a mixer, with the mixing process as follows: mix raw rubber for 30 seconds, add 2/3 of the carbon black, mix for 40 seconds, add the remaining 1/3 of the carbon black and small materials, mix for 50 seconds, clean, add oil, and discharge the rubber compound when the cumulative time reaches 205 seconds. The second stage is conducted on an open mill, with the mixing process as follows: mix the first stage rubber compound for 35 seconds, add a vulcanizing accelerator, mix for 45 seconds, clean, and discharge the sheet when the cumulative time reaches 90 seconds. After being placed at room temperature for 8 hours, the sample is prepared for vulcanization.

### 1.5 Performance testing

The performance testing of rubber compound is conducted in accordance with the corresponding national standards. Reference is made to the evaluation method for ethylene-propylene-diene rubber (EPDM) specified in GB/T42268-2022. The Mooney viscosity test is conducted in accordance with the provisions of GB 1232 standard, and the Mooney viscosity is expressed as $M_L$(1+4) at 125℃. The tensile strength and elongation at break properties are tested using a universal material testing machine, with samples cut according to Type I specifications in the national standard GB/T 528-2009.

### 1.6 Aging test

After cutting the vulcanized rubber sheet into Type I dumbbell-shaped test specimens (115 mm×6 mm×2 mm) according to the standard GB/T528-2009, hang it freely in a hot air aging oven. After aging at the specified temperature and time, remove it and let it stand at the standard test temperature (23±1) ℃ for 24 hours. Then, conduct the tensile property test

after aging.

## 2 Results and discussion

### 2.1 Rubber compound processing performance

The Mooney viscosity and vulcanization characteristics of the rubber compound are shown in Table 1.

**Table 1 Vulcanization characteristics of rubber compound**

| Project | Formula Number | |
|---|---|---|
| | 1# | 2# |
| $M_L$(1+4)125 ℃ | 56.2 | 60.6 |
| $t_{10}$/min | 3 | 2 |
| $t_{90}$/min | 21 | 18 |

### 2.2 Aging performance standards for rubber compound

Thermo-oxidative aging is the primary cause of rubber damage. Rubber undergoes thermo-oxidative aging below 200 ℃, with oxygen being the main factor causing aging, and heat playing a role in activating oxidation and accelerating the process. Aging occurs through free radical reactions, where bonds in the polymer break, forming carbon radicals, which may lead to the formation of new cross-linking bonds. The generation of new cross-linking bonds causes the polymer to harden and become brittle. For EPDM, changes in elongation at break are a sensitive indicator of aging.

Conveyor belts are frequently utilized in high-temperature environments such as chemical and metallurgical industries, necessitating their maintenance of superior mechanical properties under such conditions. According to the specifications outlined in GB/T20021-2005 for heat-resistant canvas core conveyor belts, these belts are categorized into four grades based on their level of heat resistance. Specifically, Grade T3 is suitable for use in environments with temperatures up to 150℃, while Grade T4 is suitable for temperatures up to 175℃. Generally, heat-resistant conveyor belts of Grades T3 and T4 incorporate EPR or EPDM in combination with other heat-resistant materials, along with additional measures, to enhance their heat resistance.

The automotive and roofing industries widely use accelerated thermal aging methods for quality assurance testing of elastomeric vulcanizates, typically specifying minimum values after aging. Standards ASTM D 2000 and SAE J200 specify test conditions for many automotive applications. Typical EPDM aging temperatures include 70°C, 100°C, 125°C, 150°C, and 175°C. ASTM D 4637 specifies requirements for black EPDM roofing membranes, which undergo oven aging at 116°C for 28 days or 7 days, with tensile strength after aging not less than 8.3 MPa and elongation at break not less than 200%.

This study tested the tensile properties of a kind of EPDM vulcanized rubber with two formulations after aging at temperatures of 100°C, 120°C, 130°C, and 150°C, and established a simple model for predicting the thermal aging properties of EPDM rubber at different temperatures and durations.

### 2.3 Change in tensile properties after aging

Tables 2~5 and Figure 1 show the changes in tensile strength of 1# and 2# ethylene propylene diene monomer (EPDM) vulcanizate samples after undergoing different aging times at 100 ℃, 120 ℃, 130 ℃, and 150 ℃, respectively. From the figures and tables, it can be observed that the peroxide-vulcanized 2# vulcanizate has higher strength and lower elongation at break compared to the sulfur-vulcanized 1# vulcanizate. After aging, both vulcanizate samples exhibit the same pattern of physical property loss, with the tensile strength initially showing a sharp decrease, followed by a gradual decrease as the aging time increases, while the elongation at break continues to decrease significantly. Furthermore, the higher the aging temperature, the more severe the loss in strength and elongation at break. From the figure, it is also possible to estimate the corresponding time at various aging temperatures assuming a certain level of strength and elongation at break after aging. These results indicate that high-temperature short-time aging can be used to infer the application at low temperatures.

**Table 2 Performance of the sample before and after aging, with an aging temperature of 100 ℃**

| Aging time /h | 1# | | 2# | |
|---|---|---|---|---|
| | tensile strength $T_b$ | Elongation at break $E_b$ | tensile strength $T_b$ | Elongation at break $E_b$ |
| 0 | 12 | 532 | 14.4 | 472 |
| 168 | 12.1 | 301 | 14.9 | 321 |
| 336 | 12 | 266 | 14.9 | 296 |
| 672 | 11.9 | 254 | 14.6 | 243 |
| 1008 | 11.2 | 216 | 13.8 | 230 |
| 1680 | 10.7 | 160 | 13 | 196 |
| 4224 | 8.8 | 92 | 11.8 | 118 |
| 8760 | 8.4 | 73 | 11.7 | 35 |

**Table 3 Properties of the sample before and after aging, aging temperature 120 ℃**

| Aging time /h | 1# | | 2# | |
|---|---|---|---|---|
| | tensile strength $T_b$ | Elongation at break $E_b$ | tensile strength $T_b$ | Elongation at break $E_b$ |
| 0 | 12 | 532 | 14.4 | 472 |
| 168 | 11.8 | 276 | 14 | 283 |
| 336 | 12.1 | 253 | 14.4 | 276 |
| 672 | 10.8 | 196 | 13.3 | 206 |
| 1008 | 10 | 162 | 13.1 | 192 |
| 2520 | 8.7 | 103 | 10.9 | 110 |
| 4032 | 8.4 | 23 | 9.4 | 63 |

**Table 4 Properties of the sample before and after aging, with an aging temperature of 130 ℃**

| Aging time /h | 1# | 2# | 1# | 2# |
|---|---|---|---|---|
| | tensile strength $T_b$ | Elongation at break $E_b$ | tensile strength $T_b$ | Elongation at break $E_b$ |
| 0 | 12 | 532 | 14.4 | 472 |
| 24 | 12.4 | 326 | 14.3 | 336 |
| 72 | 11.4 | 271 | 14.4 | 303 |
| 168 | 11 | 232 | 13.9 | 257 |
| 336 | 10.1 | 191 | 12.7 | 214 |
| 672 | 9.6 | 141 | 11.7 | 152 |
| 1008 | 8.8 | 112 | 10.4 | 123 |
| 1680 | 7 | 62 | 9.6 | 36 |

**Table 5 Properties of the sample before and after aging, with an aging temperature of 150 ℃**

| Aging time/ h | 1# | | 2# | |
|---|---|---|---|---|
| | tensile strength $T_b$ | Elongation at break $E_b$ | tensile strength $T_b$ | Elongation at break $E_b$ |
| 0 | 12 | 532 | 14.4 | 472 |
| 8 | 11.7 | 366 | 14.4 | 352 |
| 24 | 11.3 | 327 | 14.3 | 311 |
| 48 | 11.1 | 276 | 13.6 | 292 |
| 72 | 10.8 | 246 | 13.3 | 246 |
| 168 | 9.6 | 142 | 13.3 | 138 |

## 2.4 Change in fractional strain energy after aging

The loss of tensile strength and elongation can be described by the fractional strain energy factor at fracture, as expressed by Bergstrom $(T_E)_{fO}$ $(T_E)_f$ is defined as the ratio of the product of the tensile strength after aging and the elongation at fracture to that before aging. As shown in Equation

$$(TE)_f = \frac{(T_b \times E_b)_{\text{before aging}}}{(T_b \times E_b)_{\text{after aging}}} \quad (1)$$

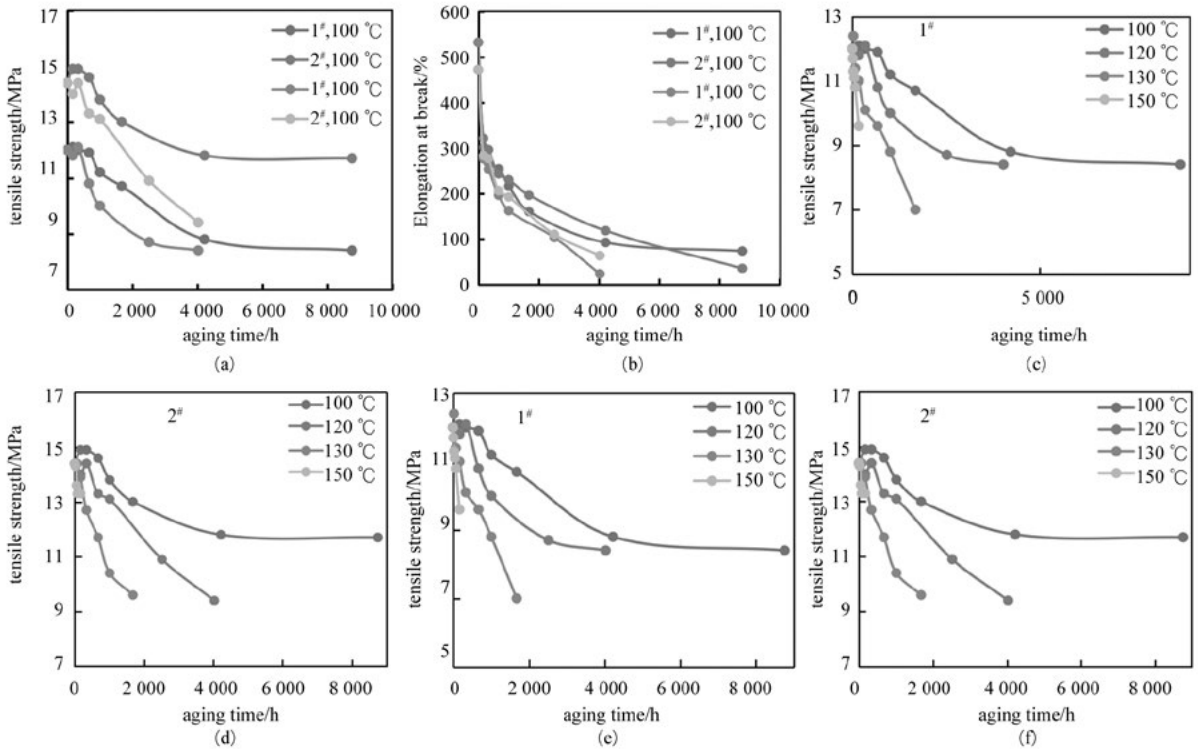


**Figure 1 Changes in tensile strength and elongation at break with aging time at different temperatures**

In the formula, $T_b$ and $E_b$ represent the tensile strength and elongation at break, respectively. The fractional strain energy factor at break is a dimensionless parameter that allows for the comparison of materials with different properties. In other studies, the fractional strain energy factor at break has been variously used to characterize performance degradation.

Based on the fractional strain energy factor at fracture, an empirical expression is established to quantitatively describe the observed aging properties, as shown in Equation (2):

$$\left[1/(TE)_f - 1/a^{'}\right] = k^{'}t \quad (2)$$

In the formula, $(TE)_f$ represents the fractional strain energy at time t; $k'$ is the effective rate coefficient at temperature T; a is the intercept factor. This rate k refers to the overall performance degradation rate of a specific vulcanized rubber sample. The value of k can be determined by regression analysis of experimental data using the slope over time at a specific temperature. As shown in Tables 2~5, the tensile properties of 1# and 2# EPDM vulcanizates at four aging temperatures are presented. The fractional strain energy was calculated based on the test results in Tables 2~5, From the figure 2 and Table 6, it can be seen that the correlation coefficients of the regression equations for 1# and 2# samples at different temperatures are above 0.986, indicating a good correlation, especially in the initial stage, which suggests that the responses are generally consistent for most testing times. At the same aging temperature, the rate constant of 1# sample is higher than that of 2# sample, indicating that sulfur-vulcanized samples have poorer heat resistance than peroxide-vulcanized samples. For the same sample, the higher the aging temperature, the larger the rate constant, with the reaction rate increasing by 2~3 times for every 10°C increase. In this study, the empirical rule of rate is not a simple constant value. When the aging temperature increases by 10°C, the rate constant of 1# sample increases by 1.5~2 times, and that of 2#sample increases by about 2 times, indicating an intensified aging degree. The relationship expressed by Equation (2) can conveniently describe the aging behavior of EPDM compounds in this study over a linear time period. As components requiring heat resistance aging, the use of low-sulfur and peroxide curing systems, as well as the selection of composite components, can improve the heat aging performance of EPDM compounds.

**Table 6 Relation equation between reciprocal of strain energy and aging time under different aging temperatures for test samples**

| Temperature/ ℃ | 1# | | | 2# | | |
|---|---|---|---|---|---|---|
| | Trend equation | Rate constant k' | $R_2$ | Trend equation | Rate constant k' | $R_2$ |
| 100 | y=0.001564x+1.210 | 0.001564 | 0.992 | y=0.000875x+1.213 | 0.000875 | 0.993 |
| 120 | y=0.002338x+1.354 | 0.002338 | 0.989 | y=0.001770x+1.158 | 0.001770 | 0.987 |
| 130 | y=0.005007x+1.462 | 0.005007 | 0.989 | y=0.004045x+1.184 | 0.004045 | 0.994 |
| 150 | y=0.020697x+1.128 | 0.020697 | 0.986 | y=0.0153x+1.077 | 0.01530 | 0.988 |

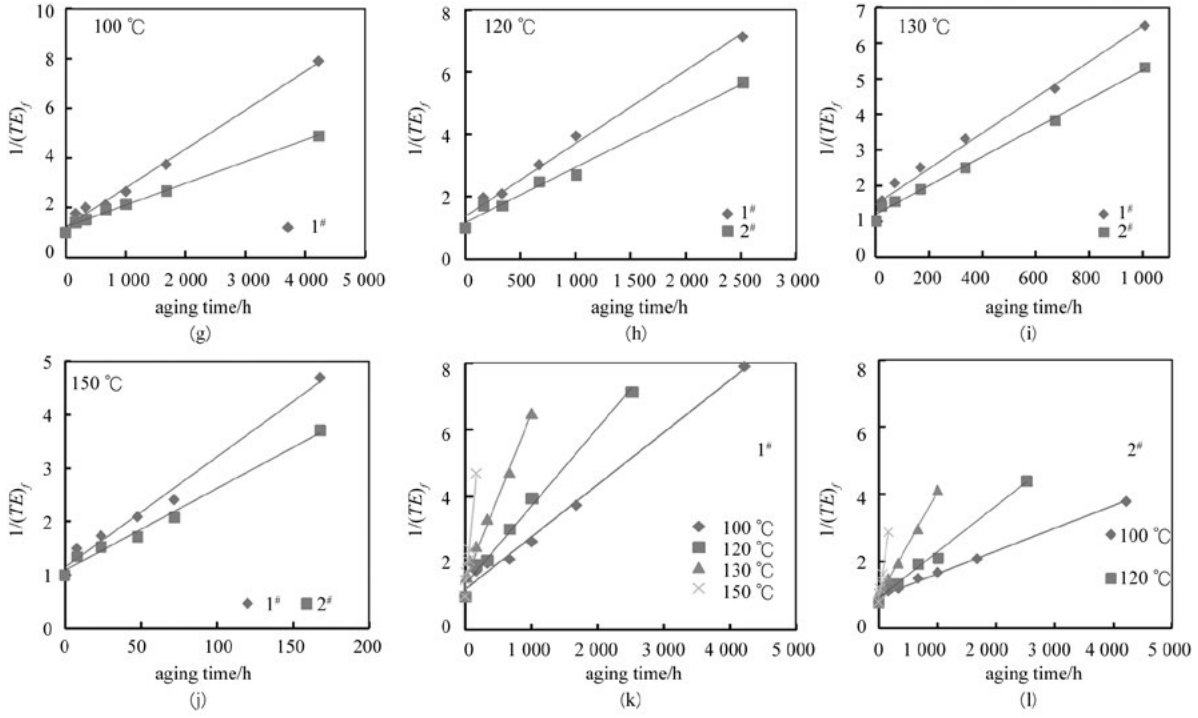


**Figure 2 Change in reciprocal of strain energy of the sample with aging time**

## 2.5 The influence of temperature on degradation rate

There are precedents for the application of the Arrhenius relationship in elastomers and plastic materials. According to the Arrhenius equation, as shown in equation (3):

$$\ln k = -(E_a / RT) + \ln A \quad (3)$$

In the formula, $k$ represents the reaction rate, $E_a$ denotes the activation energy, which is affected by temperature but the impact is generally negligible and often ignored. $R$ stands for the gas constant, and $T$ represents absolute temperature. Given the reaction rate $k$ at different temperatures, the activation energy $Ea$ can be determined by the slope of lnk versus 1/$T$.

After aging, the stress-strain properties of EPDM vulcanized rubber decrease, and the Arrhenius relationship can be used to predict the changes at different temperatures and durations. The activation energy ($E_a$) value is determined by logarithmic linear regression of k against 1/$T$. The correlation coefficient $R^2$ is 0.911~0.911. The relationship between ln k and 1/$T$ is shown in Figure 3 and Table 7. The apparent activation energy ($E_a$) of performance changes caused by thermal aging in EPDM films 1# and 2# is 68.7~75.8 kJ/mol in the range of 100℃~150℃.

**Table 7 Relation equation between reciprocal of strain energy and aging time of samples at different aging temperatures**

| recipe | Trend equation | $R_2$ | Slope-$E_a$/R | Activation Energy $E_a$ (kJ/mol) | LnA' |
|---|---|---|---|---|---|
| 1# | Y=−8263.6x+15.398 | 0.911 | −8263.6 | 68.7 | 15.398 |
| 2# | Y=−9120.8x+17.188 | 0.958 | −9120.8 | 75.8 | 17.188 |

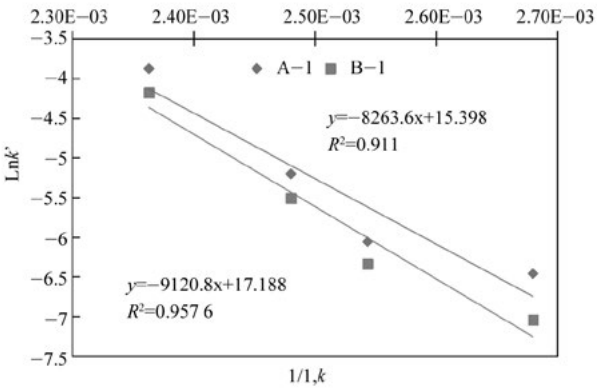


**Figure 3 Relationship between the logarithm of rate constant and the reciprocal of aging temperature**

## 2.6 Estimation of service life

As mentioned earlier, within the temperature range of 100°C to 150°C, there is a linear relationship between EPDM vulcanized rubber 1/$(TE)_f$ and aging time, and the aging rate constant follows Arrhenius' law. Therefore, the service life of rubber at different usage temperatures can be inferred. The time t' required to reach a specific comparison value or reference value ($(TE)_f$ f at a set temperature can be estimated using equations (2) and (5), which can be expressed as:

$$\left[1/(TE)_f - 1/a'\right] = k't \quad (4)$$

$$\ln k' = -(E_a' / RT) + \ln A' \quad (5)$$

$$t' = \left[1/(TE)_f' - 1/a\right] \div A' \exp(-E_a' / RT) \quad (6)$$

Alternatively, one can simply estimate based on the graph of 1/$(TE)_f$ versus time.

The extrapolation of accelerated thermal aging to actual usage conditions and lifespan is of interest to most applications. For example, in practical use, a product's tensile strength should not be less than 13 MPa, and its elongation at break should not be less than 200%. Assuming an actual average usage temperature of about 50°C, and using vulcanizates of formulas 1# and 2# respectively, calculations based on a 100°C aging model indicate that the vulcanizate using formula 1# can last for 17 years, while the one using formula 2# can last for 44 years. Depending on the customer's product design requirements, the appropriate formula can be selected or adjusted accordingly. However, such predictions may encounter issues, as dynamic tensile fatigue, changes in temperature and humidity, ultraviolet radiation, ozone, rainwater, and other environmental conditions may occur during the product's actual usage. These factors can affect its performance and aging, and they cannot be easily simulated and predicted in the laboratory. Nevertheless, if thermal aging is the primary mechanism of deterioration in actual use, the performance retention rate can be estimated from the results of accelerated thermal aging in the laboratory.

## 3 Conclusion

Based on the Arrhenius equation, a simple empirical

thermal aging model based on fractional strain energy was established to predict the service life of EPDM vulcanized rubber. By testing the tensile properties of samples after aging at different temperatures and durations, the fractional strain energy was calculated, and a linear regression equation between fractional strain energy and aging time was derived to determine the effective rate constant and apparent activation energy of the aging reaction. The results confirmed that sulfur and peroxide vulcanized EPDM rubber, as commonly used materials for heat-aging applications such as tapes, hoses, automotive parts, and building roofing membranes, are characterized by stress-strain properties that prove to be a predictable method for characterizing thermal aging behavior. Based on this model, the service life of the formulation under actual application environmental temperatures can be predicted, providing a reference for product design.