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Research Article

Lifetime Analysis of Rubber Gasket Composed of Methyl Vinyl Silicone Rubber with Low-Temperature Resistance

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Most machines and instruments constantly require elastomeric materials like rubber for the purposes of shock absorption, noise attenuation, and sealing. The material properties and accurate lifetime prediction of rubber are closely related to the quality of machines, especially their durability and reliability. The properties of rubber-like elastomers are influenced by ambient conditions, such as temperature, environment, and mechanical load. Moreover, the initial properties of rubber gaskets must be sustained under working conditions to satisfy their required function. Because of its technical merits, as well as its low cost, the highly accelerated life test (HALT) is used by many researchers to predict the long-term lifetime of rubber materials. Methyl vinyl silicone rubber (VMQ) has recently been adopted to improve the lifetime of automobile radiator gaskets. A four-parameter method of determining the recovery ability of the gaskets was recently published, and two revised methods of obtaining the recovery were proposed for polyacrylate (ACM) rubber. The recovery rate curves for VMQ were acquired using the successive zooming genetic algorithm (SZGA). The gasket lifetime for the target recovery (60%) of a compressed gasket was computed somewhat differently depending on the selected regression model.

1. Introduction

Most machines and instruments constantly require elastomeric materials like rubber for the purposes of shock absorption, noise attenuation, and sealing [1]. The rubber elastomer is classified into three types: natural rubber (NR), synthetic rubber (SR), and NR + SR blended at a given ratio. SR exhibits many excellent properties in terms of mechanical performance. NR is often inferior to certain SRs, especially with respect to thermal stability and compatibility with petroleum products [2]. The SR ethylene propylene diene monomer (EPDM) rubber, which has the characteristic of high-temperature resistance, has been mainly adopted for a radiator gasket of an automobile until now. However, methyl vinyl silicone rubber (VMQ) has recently begun to be used as a radiator gasket material compatible with an extreme temperature range and low temperatures, according to SAE J200, because previous gasket design criteria stated that low-temperature applications for automobiles reached temperatures in the range of –70°C to –55°C by major automotive companies. The VMQ specimen used in this study is made from the final master batch of Burim FMB Co. in ROK, which has been made from the silicone base of Dow Corning Co. by adding 1 PHR of curing agent and 0.5 PHR of pigment.

In this study, we predict the lifetime of a VMQ radiator gasket recently developed by a local company using the method proposed in 2014 [3]. Generally three methods are used for the lifetime prediction of a rubber gasket. The most practical method with mathematical concepts is the highly accelerated life test (HALT), which applies temperatures higher than the service temperature over a short period. Using this method, the long-term lifetime of a gasket at lower temperatures can be predicted by extrapolating the data [4]. The second lifetime prediction method under service conditions is economically disadvantageous because of its long testing time, high cost, and labor requirements. The third method is to rely on an experienced engineer specializing in rubber materials, which is less reliable and does not yield objective results.

The HALT is a test methodology that accelerates the degradation of material properties using several specimens, and it has been used by many researchers during the material development stage and design process. This test is also commonly applied to rubber materials for gaskets and dampers and facilitates the identification and resolution of weaknesses in new product designs. The methodology diminishes the probability of in-service failures; that is, it increases product quality by virtue of reliability and decreases the development cost and time [5]. The HALT for VMQ was performed at temperatures of 150–200°C under a compression rate of 30%, which is the actual compression rate under service conditions for the radiator gasket. Additionally, a low-temperature test at –70°C was performed under the same compression rate.

In this method of lifetime prediction, the Arrhenius model [6] is simpler and more effective for most cases than the Eyring model [7] and uses experimental data. The lifetime of the gasket is defined as the time when the recovery rate meets 60% of the target value after undergoing a 30% compression rate, which depends on the service temperature, whereas ISO 11346 [8] stipulates that the failure time of chemical materials is the point where its initial properties are reduced to 50%.

According to most references investigating a lifetime evaluation adopting the linear Arrhenius equation [9] for the $\ln(t) - (1/T)$ relationship, where t is the lifetime and T is the temperature, small errors in the lifetime at high temperatures from the HALT evaluation may lead to large errors in the predicted lifetime at low temperatures. Unlike most papers, which do not consider the recovery~$\ln(t)$ curve, one study made use of four parameters instead of two parameters in the Arrhenius plot to accurately draw the recovery~$\ln(t)$ curve and correctly determine the long-term lifetime.

With accurate lifetime predictions at high temperatures, the linear Arrhenius model in the $\ln(t)-(1/T)$ plane can yield correct quantitative analysis of the lifetime of VMQ at a low working temperature.

2. Successive Zooming Genetic Algorithm Method for Optimum Parameters

The successive zooming genetic algorithm (SZGA) method is used to achieve a smart reduction of the search space around the candidate optimum point [10, 11]. Although this method can also be applied to a general genetic algorithm (GA), it was applied to a micro-genetic algorithm (MGA). The computing procedure of the SZGA is as follows. First, the initial population is generated and an MGA is applied. Subsequently, after every 100 generations, the optimum point with the highest fitness is identified. Second, the search domain is reduced to $(X_{k\text{opt}} - \alpha^k/2,\ X_{k\text{opt}} + \alpha^k/2)$, and the optimization procedure continues based on the reduced domain; that is, a new initial population is generated within the new boundaries. This reduction of the search domain increases the resolution of the solution, and the procedure is repeated until the identified solution is satisfactory (δ is the error ratio, $X_{k\text{opt}}$ is the optimum point after $(100 \times k)$th generation, α is the zooming factor, and N_{zoom} is the number of zooming operations). δ is the relative ratio of $(F^k_{\text{opt}} - F^{k-1}_{\text{opt}})/F^k_{\text{opt}}$, and F^k_{opt} and F^{k-1}_{opt} are the kth and $(k-1)$th optimum function values. The critical ratio δ_0 is 1×10^{-6}. To fit the recovery rate curve of the polyacrylate (ACM) rubber gasket, the optimal parameters of the smallest mean squared error (MSE) [12] were obtained using this SZGA method. Figure 1 shows the flowchart and the schematics of the SZGA:

$$\text{Mean squared error (MSE)} = \frac{1}{n}\sum_{i=1}^{n}\left(f\left(\mathbf{k}, x_i\right) - D_i\right)^2, \quad (1)$$

where $\mathbf{k}$: unknown parameters.

3. Methods of Predicting the Quantitative Lifetime of a VMQ Gasket

Methods of mathematically predicting the quantitative lifetime are introduced in this section. To obtain an Arrhenius plot of the long-term lifetime, we first needed to fit the recovery rate curve for a given temperature to obtain the lifetime corresponding to a recovery rate of 60%. Two methods of fitting curves for the ACM were adopted here for the VMQ [3]. The recovery rate of a rubber gasket was assumed to be two exponential functions represented by four parameters. The recovery rate curves were fit using the four optimized parameters, and the lifetimes were solved from the obtained functions. Before we explain the methods of obtaining lifetimes at each given temperature by adopting recovery models, let us first explain the prediction of long-term lifetime using the Arrhenius plot.

3.1. Arrhenius Equation and Plot. An Arrhenius equation presents the kinetic rate K as a function of the reciprocal of the temperature T in Kelvin [5]. This model is used widely to estimate the reciprocal effect of temperature as

$$K = Ae^{-E_a/RT}. \quad (2)$$

For a single rate-limited thermally activated process, an Arrhenius plot gives a straight line as a function of the activation energy and temperature as

$$\ln(K) = \ln(A) - \frac{E_a}{R}\left(\frac{1}{T}\right), \quad (3)$$

where K is the rate constant, A is the preexponential factor, R is the gas constant, T is the absolute temperature (°K), and E_a is the activation energy.

Equation (3) can be rearranged to give a time-temperature relation by applying $t \propto 1/K$, as

$$\ln(t) = \ln\left(\frac{1}{A}\right) + \frac{E_a}{R}\left(\frac{1}{T}\right). \quad (4)$$

The lifetimes for higher temperatures are plotted in Arrhenius form, and the long-term lifetimes may be predicted by linearly extrapolating the given data in the semilog

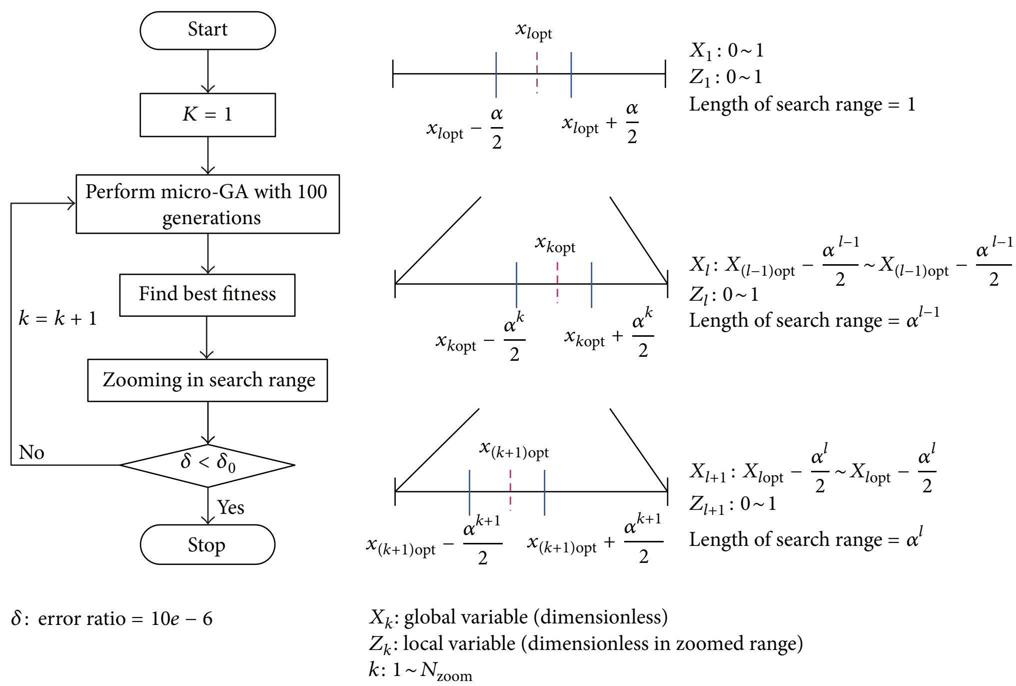


Figure 1: Flowchart and schematics of SZGA.

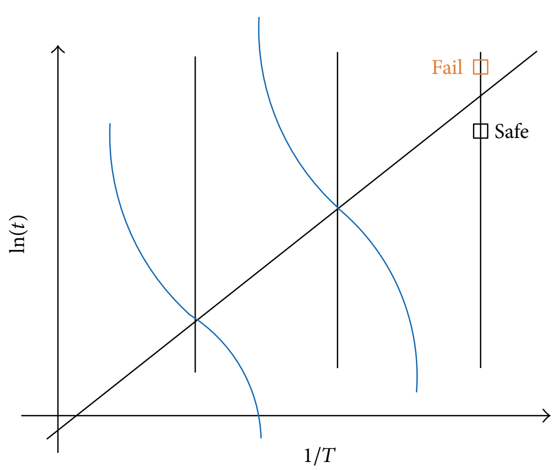


Figure 2: Arrhenius plot used to predict lifetime.

plane of $\ln(t) - (1/T)$. Figure 2 shows a plot of the long-term lifetime prediction method using the Arrhenius plot. If the part is used for more time (upper square) than the allowable lifetime (dot) at a given temperature, it will fail; if it is used for less time (lower square) than the lifetime, it will be safe.

3.2. Four-Parameter Method 1. Four-parameter (t_0, f_0, k_1, k_2) method 1 is composed of two exponential functions, and when the time is less than the reference time t_0, one function is used, as in (5). When the lifetime t is zero in (5) [13], the equation can be rewritten as (6), from which f_c is confirmed with the other three parameters f_0, k_1, and t_0:

$$f(t) = f_c - (f_c - f_0)\, e^{(t-t_0)k_1}: \quad (t < t_0) \tag{5}$$

$$100\,(\%) = f_c \left(1 - e^{(-t_0 \cdot k_1)}\right) + f_0 e^{(-t_0 \cdot k_1)} \tag{6}$$

$$f_c = \frac{100 - f_0 e^{(-t_0 \cdot k_1)}}{1 - e^{(-t_0 \cdot k_1)}}. \tag{7}$$

When the time is greater than the reference time t_0, the recovery equation is represented as in (8). Figure 3 schematically shows the recovery rate curve using four-parameter method 1:

$$f(t) = f_0 e^{-(t-t_0)k_2}: \quad (t > t_0). \tag{8}$$

3.3. Four-Parameter Method 2. In four-parameter (t_0, f_0, k_1, k_2) method 2, f_c is a constant and not dependent on f_0, k_1, and t_0 [14]. When the lifetime t is $-\infty$, the recovery is assumed to be 100% to make the recovery curve symmetric, as in (9). Therefore, four-parameter method 2 is actually the same as method 1, except that $f_c = 100$ instead of the definition in (7) (Figure 4):

$$\begin{aligned} f(t) &= 100 - (100 - f_0)\, e^{(t-t_0)k_1}: \quad (t < t_0) \\ f(t) &= f_0 e^{-(t-t_0)k_2}: \quad (t > t_0). \end{aligned} \tag{9}$$

4. Experiments

Before the HALT test of the VMQ, a rubber material property test was performed by the Korea Testing and Research

Table 1: Material properties of VMQ.

Material properties	Exp. value	Test standard
Basic properties		
Hardness (IRHD)	70	
Tensile strength (MPa)	7.8	ASTM D412
Ultimate elongation (%)	150	
Heat resistance		
Change in hardness (%)	0	
Change in tensile strength (%)	−19.9	ASTM D573
Change in elongation (%)	−7.6	
Compression set (%)	9.2	ASTM D395, method B; 22 h, 150°C, plied
Compression set (%)	33.6	ASTM D395, method B; 1000 h, 150°C, plied
Fluid resistance		
Change in hardness (IRHD)	−5	
Change in tensile strength (%)	−10.1	ASTM D471, ASTM oil number 1; 70 h, 150°C
Change in elongation (%)	−3.3	
Change in volume (%)	3.4	
Fluid resistance		
Change in hardness (IRHD)	−10	ASTM D471, ASTM oil number 3; 70 h, 150°C
Change in volume (%)	17.5	
Low temp. brittleness	No cracking	ASTM D2137, method A; −55°C, 3 min

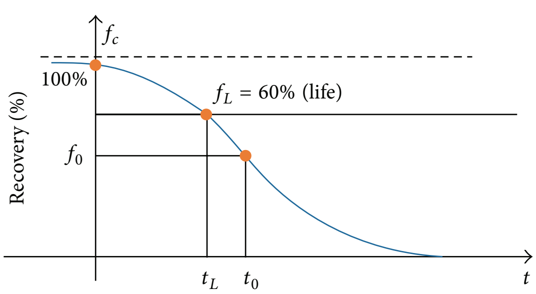


Figure 3: Recovery rate curve using four-parameter method 1.

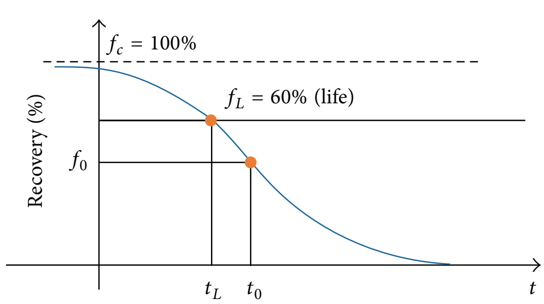


Figure 4: Recovery rate curve using four-parameter method 2. f_c: critical recovery, f_L: life recovery, t_L: lifetime, f_0: reference recovery, and t_0: reference time.

Institute (KTR) and the Korea Polymer Testing & Research Institute (KOPTRI) according to ASTM standards. The test results are given in Table 1.

The ability of a rubber to return to its original thickness after prolonged compression is measured by a compression set test at high and low temperatures. The compression set tests in this study were carried out under a compression rate of 30% with VMQ silicon rubber gaskets. For the lifetime prediction at a high temperature, the compression set test was performed with components that were heat-aged in an oven at temperatures of 150, 180, and 200°C for periods ranging from 20 to 500 h, and a cold resistance test was performed at a temperature of −70°C for periods ranging from 48 to 120 h. The dimensions of the specimen (diameter = 29 mm and thickness = 12.5 mm) and the compression set were determined according to ISO 815-1 (Figure 5) [15]. The compression set and recovery rate were calculated using

$$\mathrm{CS}\ (\%) = \frac{(l_0 - l_2)}{(l_0 - l_1)} \times 100$$
$$\text{Recovery}\ (\%) = 100 - \mathrm{CS}, \tag{10}$$

where CS = compression set, l_0 = thickness of the specimen, l_1 = thickness in the compressed state, and l_2 = thickness after removal of the load.

Table 2 lists the experimental data for each temperature. First, the experiments were performed at 150°C according to method B in the ASTM D395. The higher temperature of 180°C was adopted for the HALT as in [3]. The temperature of 200°C seems rather high. The experimental data, with the exception of a couple of erroneous data points, were selected to optimize the four parameters in four-parameter methods 1 and 2 by SZGA.

5. Results

Representative automobile companies require the compression set rates of engine head rubber gaskets to be less than

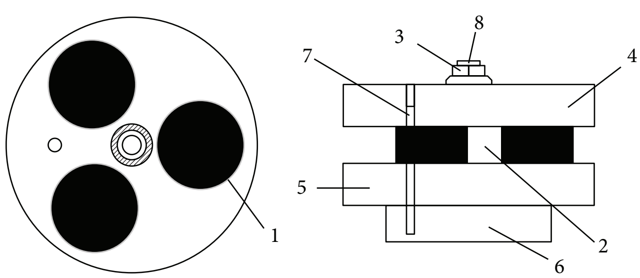
TABLE 2: Results of the compression set test with a compression rate of 30% at (a) high temperatures and (b) a low temperature.

(a)

Temp. (°C)	Time (h)	CS (%)	Recov. (%)	Temp. (°C)	Time (h)	CS (%)	Recov. (%)	Temp. (°C)	Time (h)	CS (%)	Recov. (%)
150	22	9.20	90.80	180	48	10.33	89.67	200	20	10.67	89.33
	1000	33.60	66.40		96	25.67	74.33		30	18.33	81.67
					120	31.00	69.00		40	19.67	80.33
					196	32.67	67.33		50	30.00	70.00
					240	34.33	65.67		200	83.00	17.00
									300	86.00	14.00
									400	92.00	8.00
									500	97.00	3.00

(b)

Temp. (°C)	Time (h)	CS (%)	Recov. (%)
−70	48	1.93	98.07
	72	2.73	97.27
	96	3.34	96.66
	120	3.78	96.22



(1) Rubber sample
(2) Spacer
(3) Nut
(4) Upper plate
(5) Lower plate
(6) Vice grip
(7) Position control pin
(8) Bolt

FIGURE 5: Jig for measuring the compression set.

40%. In other applications, the lifetimes of a gasket are defined as the time until its recovery rate is 60%. Both heat and cold resistance tests were performed. Experimental data at each temperature were obtained from the compression set test, and the recovery rate curves were fit using the SZGA method to find the optimal parameters of the smallest MSE between the best-fit function and the experimental data. Subsequently, the lifetime of the recovery rate at 60% was obtained from the best-fit recovery rate curve using a bisection method to solve the nonlinear equation. Finally, a linear regression model was fit by superimposing the recovery rate curve on the Arrhenius plot to obtain the long-term lifetime at the working temperature.

5.1. Lifetime of VMQ Gaskets under High Temperatures. The lifetime evaluations have been made on the two differently regressed curves of methods 1 and 2.

TABLE 3: MSEs of the four-parameter methods at a compression rate of 30%.

Method	MSE 150°C	MSE 180°C	MSE 200°C	Total MSE
Four-parameter method 1	0	10.432	3.998	14.430
Four-parameter method 2	0	6.712	3.988	10.700

5.1.1. Recovery Rate Curves from Four-Parameter Methods 1 and 2. The recovery rate curves were acquired using four-parameter methods 1 and 2. The SZGA method was used to optimize the four parameters, and the recovery rate curves of the two methods were fit using these four parameters. The recovery rate curves were compared with the experimental data. The results showed that the recovery rate curves could be fit properly using the four parameters. Figures 6 and 7 [14, 16] show the recovery rate curves at different temperatures and a compression rate of 30%.

5.1.2. Mean Squared Error of Four-Parameter Methods 1 and 2. The MSE [12] can be calculated to gauge the extent to which the data points vary from the recovery rate curves. Table 3 compares the MSEs of four-parameter methods 1 and 2. For the life prediction of the rubber gasket, either method can be used to find the least MSE:

$$\text{Mean squared error (MSE)} = \frac{1}{n}\sum_{i=1}^{n}\left(y_i - \hat{y}_i\right)^2. \qquad (11)$$

One can see that four-parameter method 2 yields a smaller MSE than method 1 for a compression rate of 30%.

5.1.3. Results of Quantitative Lifetime Prediction. Compression set rates less than 40% are required by major automobile

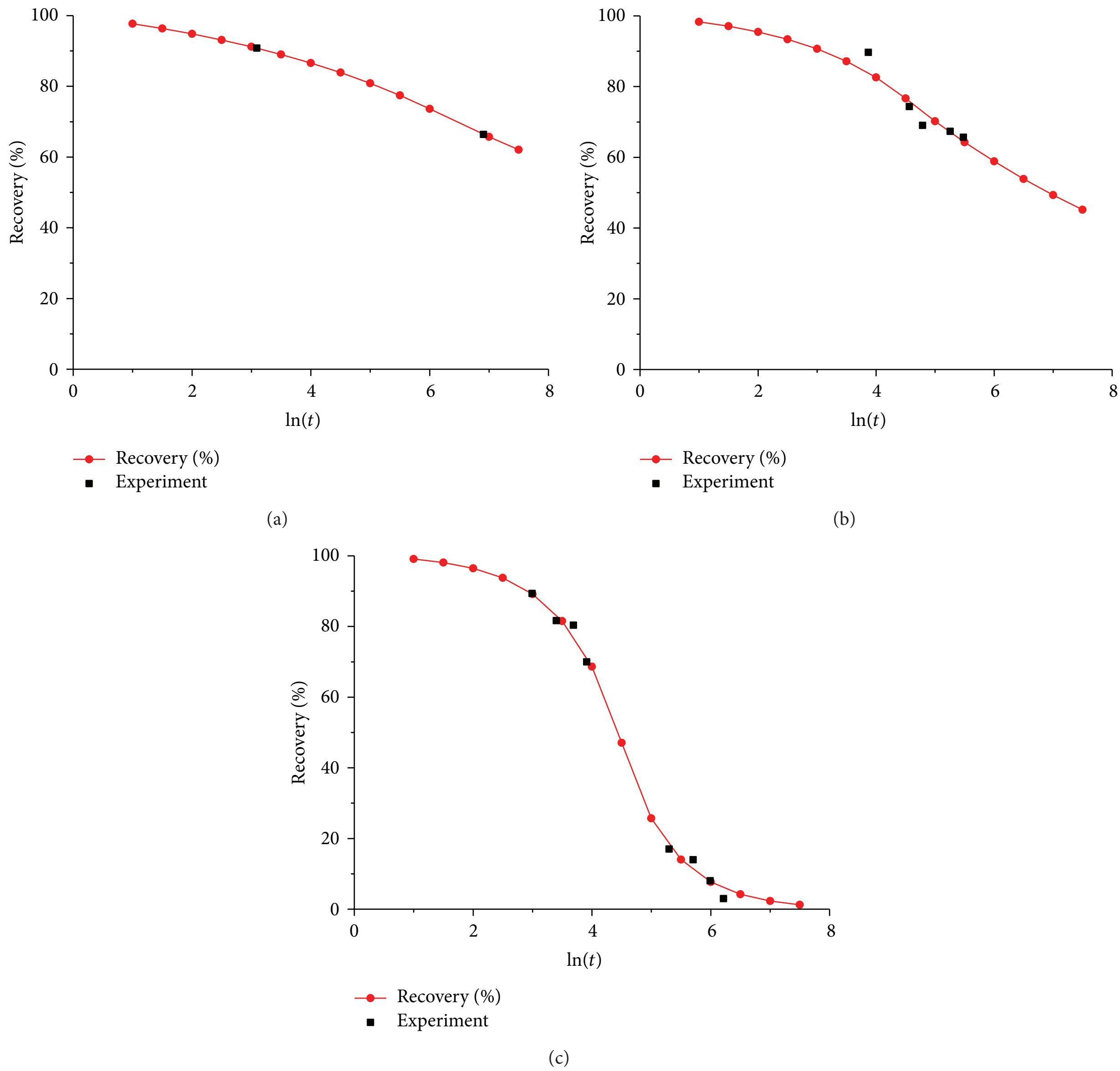


Figure 6: Recovery rate curves using four-parameter method 1 with a compression rate of 30% at temperatures of (a) 150°C, (b) 180°C, and (c) 200°C.

companies. The precise lifetime corresponding to a recovery rate of 60% can be determined using the bisection method from the four-parameter equations. The method with the minimum MSE is the best choice to obtain the lifetime with a 60% recovery rate. The lifetime data acquired from each recovery rate curve were plotted using linear regression, and a linear equation was derived using the Origin Pro system (Figure 8). This equation can be used to estimate the lifetime at a specific temperature from the Arrhenius equation. Subsequently, the lifetime and lifetime mileage were calculated by

$$\begin{aligned} \text{Time (hour)} &= \exp\{\ln(t)\} \\ \text{Life mile} &= 30\,\text{mph (mile/hour)} \times \text{time (hour)}. \end{aligned} \tag{12}$$

Under operating conditions of 30 mph (mile/hour) at 100°C, the lifetime mileage values of the rubber gasket predicted by four-parameter methods 1 and 2 are 6,836,220 and 7,805,780 mi, respectively, as shown in Table 4. Because the operating time is assumed to be an average of 3 h/day, the predicted quantitative lifetimes of the rubber gasket calculated using four-parameter methods 1 and 2 are 208 and 273 years, respectively. Thus, the lifetime of the VMQ silicon rubber gasket predicted at a working temperature of 100°C meets the performance requirements of 100,000 mi and 10 years.

5.2. Lifetime of VMQ Gaskets under Low Temperatures. A closer look at the experimental data at a low temperature

Table 4: Lifetime mileage determined using the Arrhenius equation.

Compression rate 30%			
Four-parameter method 1		Four-parameter method 2	
$\ln(t) = 14.06674 \times \{(1/(T\,(^\circ\mathrm{C}) + 273)) \times 1000\} - 26.37589$		$\ln(t) = 14.39015 \times \{(1/(T\,(^\circ\mathrm{C}) + 273)) \times 1000\} - 26.11032$	
Temperature (°C)	Lifetime mileage (10^3 miles)	Temperature (°C)	Lifetime mileage (10^3 miles)
80	57,911	80	69,453
100	6,836	100	7,805
120	1,003	120	1,095
140	177	140	186
160	36	160	37
180	9	180	9

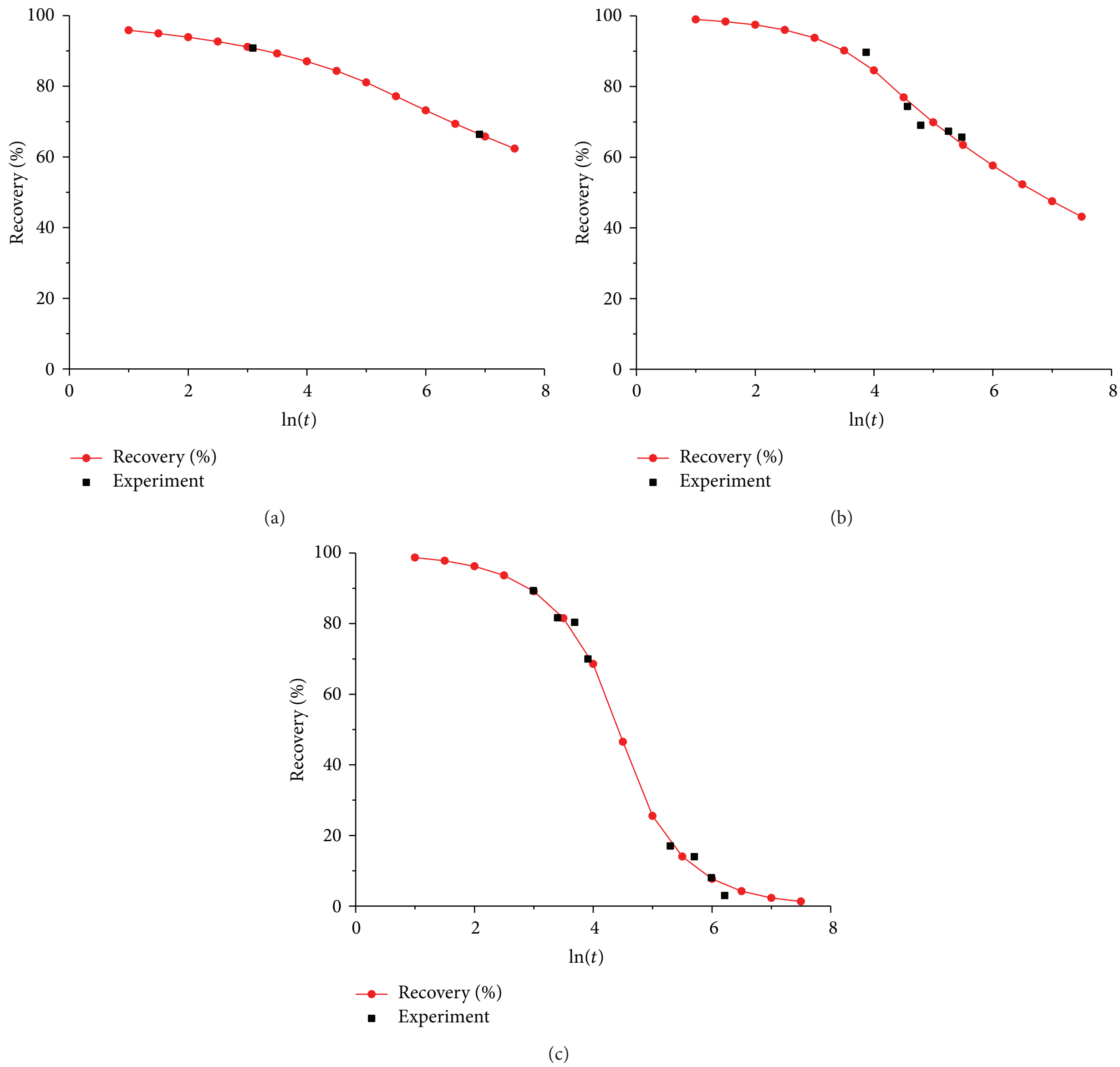


Figure 7: Recovery rate curves using four-parameter method 2 with a compression rate of 30% at temperatures of (a) 150°C, (b) 180°C, and (c) 200°C.

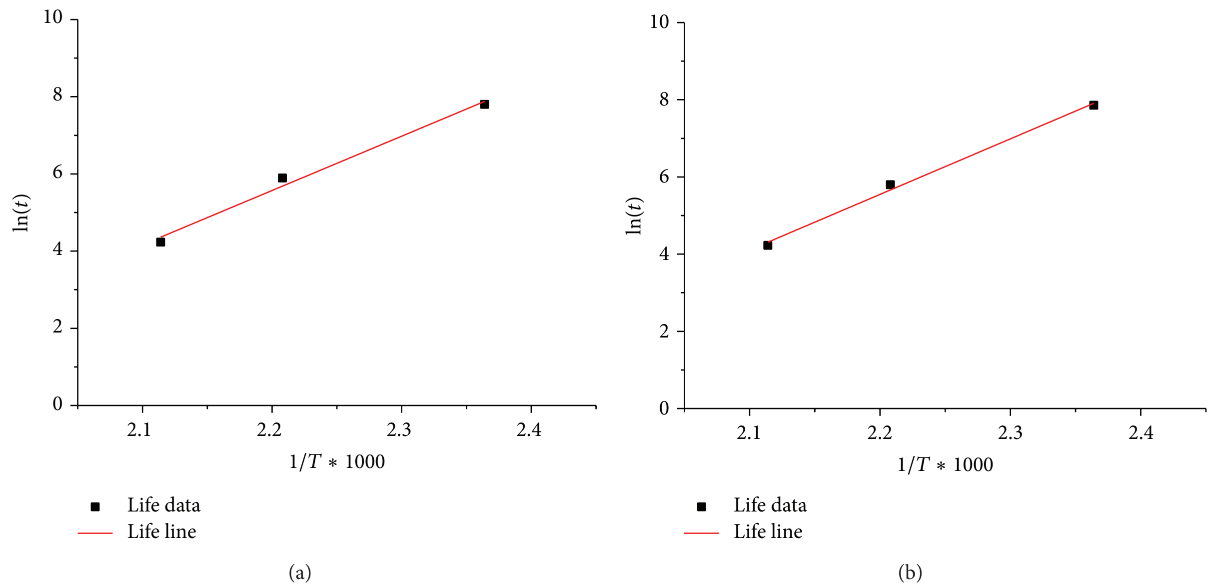


Figure 8: Arrhenius plots for four-parameter methods (a) 1 and (b) 2.

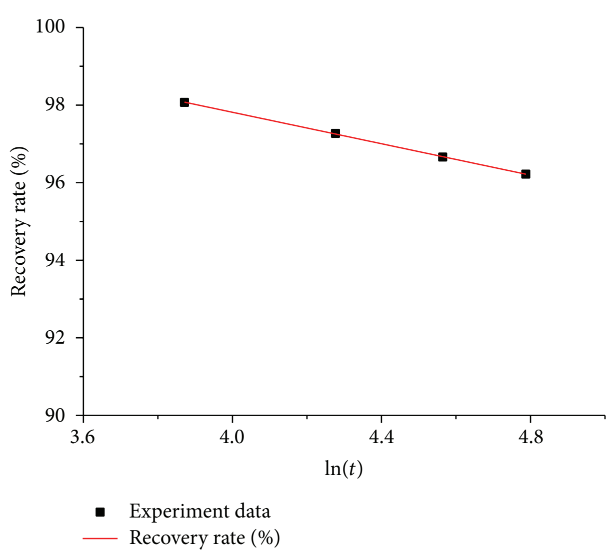


Figure 9: Recovery rate curve at low temperature (–70°C).

(Table 2(b)) indicates that the relationship between the recovery rate and lifetime can be represented as a straight line. Thus, a linear regression was performed using the experimental data from the compression set test at a low temperature, and the intercept and slope of the linear equation were obtained. This means that only two parameters were needed to predict the lifetime, unlike in the four-parameter methods. Figure 9 shows the regression line from the linear relation between the recovery rate and lifetime. In cold regions, like the Antarctic, where temperatures can reach –70°C, shrinkage and shrinkage leaking in the rubber may occur because of the low temperatures, and these problems can reduce the recovery rate of VMQ. Thus, the standard required recovery rate of over 60% should be applied to predict the lifetime in this case. Table 5 shows the lifetime of the rubber gasket obtained from the best-fit line at different recovery rates ranging from 70 to 90%. After establishing the standard recovery rate, depending on the method and purpose of use, the lifetime of VMQ can be predicted for each case. For example, according to the linear equation, the lifetime of the VMQ rubber gasket corresponding to a recovery rate of 80% was 357,960 h. Assuming that the rubber gasket in an automobile radiator is continuously exposed to temperatures of –70°C, its lifetime would be 41 years. This result leads to the conclusion that the VMQ rubber gasket sufficiently satisfies the lifetime requirement of 10 years.

Table 5: Lifetimes from the fitted line.

Recovery rate (%) = $-2.02719 \times \ln(t) + 105.92382$			
Time (h)	$\ln(t)$	Compressed set (%)	Recovery rate (%)
2,579	7.8552	10	90
30,384	10.3217	15	85
357,960	12.7882	20	80
4,217,197	15.2547	25	75
49,683,641	17.7212	30	70

6. Conclusion

A compression set test was carried out on developed VMQ gaskets at a compression rate of 30%. The SZGA method was applied to determine the optimal four parameters for the two four-parameter methods used in this study and calculate the recovery rate curves. The MSEs of the regression functions from different models and the experimental data were compared. By comparing the results of both methods, it was determined that either method can be used to accurately predict gasket lifetime because they showed only small differences in their results. We obtained the target lifetime for a recovery rate of 60% (80% for −70°C) through the fitted recovery rate curve using the bisection method at each temperature.

Referring to the data points of the 60% (80% for −70°C) recovery found from the recovery rate curves, a linear Arrhenius plot in the $\ln(t) - (1/T)$ plane was constructed to determine the quantitative lifetime at any given temperature.

The results are summarized as follows.

(1) A procedure using four-parameter methods 1 and 2 to predict the long-term lifetimes of rubber gaskets was suggested.

(2) Using four-parameter methods 1 and 2, the quantitative lifetime of a rubber gasket could be accurately predicted at any given temperature.

(3) The lifetime mileage of VMQ was predicted to be 6,836,220 and 7,805,780 mi using four-parameter methods 1 and 2, respectively, at a working temperature of 100°C.

(4) The lifetime of the VMQ rubber is 41 years at an ambient temperature of −70°C based on the standard recovery rate of 80%.

Conflict of Interests

The authors declare that there is no conflict of interests regarding the publication of this paper.

Acknowledgments

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