

Article

High-Temperature Characteristics of Polyphosphoric Acid-Modified Asphalt and High-Temperature Performance Prediction Analysis of Its Mixtures

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Abstract: To promote the application of economical and sustainable polyphosphoric acid (PPA)-modified asphalt in road engineering, styrene-butadiene block copolymer (SBS), styrene-butadiene rubber (SBR), and PPA were used to prepare PPA/SBS and PPA/SBR composite-modified asphalts, which were tested and the data analyzed. Fourier transform infrared spectroscopy (FTIR) tests and thermogravimetric analysis (TG) tests were carried out to study the modification mechanisms of the composite-modified asphalts, and the high-temperature performance of the PPA-modified asphalt and asphalt mixtures was analyzed by dynamic shear rheology (DSR) tests and wheel tracking tests. A gray correlation analysis and a back-propagation (BP) neural network were utilized to construct a prediction model of the high-temperature performance of the asphalt and asphalt mixtures. The test results indicate that PPA chemically interacts with the base asphalt and physically integrates with SBS and SBR. The PPA-modified asphalt has a higher decomposition temperature than the base asphalt, indicating superior thermal stability. As the PPA dosage increases, the $G^*/\sin\delta$ value of the PPA-modified asphalt also increases. In particular, when 0.6% PPA is combined with 2% SBS/SBR, it surpasses the high-temperature performance achieved with 4% SBS/SBR, suggesting that PPA may be a good alternative for polymer modifiers. In addition, the creep recovery of PPA-modified asphalt is influenced by the stress level, and as the stress increases, the R-value decreases, resulting in reduced elastic deformation. Furthermore, the BP neural network model achieved a fit of 0.991 in predicting dynamic stability, with a mean percentage of relative error (MAPE) of 6.15% between measured and predicted values. This underscores the feasibility of using BP neural networks in predictive dynamic stability models.

Keywords: polyphosphoric acid; high-temperature performance; multiple stress creep recovery; back propagation neural network; sustainability



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1. Introduction

Asphalt pavement is widely used in high-grade highways due to its advantages of having a smooth surface, no joints, and low noise. Asphalt has good plasticity and viscosity, which binds aggregate particles together to form a strong pavement structure. However, with the increase of pavement traffic volume and axle load, as well as the influence of external environmental factors, asphalt will flow at high temperatures, which will lead to the destabilization of the aggregate skeleton, resulting in rutting, pushing, and other diseases, which will seriously affect the service life and durability of the pavement.

Therefore, improving the performance and durability of asphalt binders is crucial to the sustainable development of asphalt pavements.

Compared to traditional asphalt, modified asphalt can significantly enhance the high-temperature performance of asphalt pavements and reduce rutting [1,2]. Therefore, researchers have proposed the incorporation of modifiers into asphalts to improve their performance. Commonly utilized modifiers include polymers, waxes, and acids [3–8]. Among the polymers that have been identified as effective in improving the properties of asphalt are styrene-butadiene block copolymer (SBS) and styrene-butadiene rubber (SBR). These polymers have been shown to enhance the elastic recovery, low-temperature ductility, and adhesion properties of asphalt. Nevertheless, the disparities in internal structure and solubility parameters between SBS, SBR, and asphalt result in suboptimal storage stability of the blends. Additionally, the cost of polymer-modified asphalt is considerably higher than that of matrix asphalt [4,9,10]. To solve these problems, it is necessary to improve the compatibility of polymer-modified asphalt and reduce its cost, which can be achieved by combining it with other modifiers, either by chemical reaction or physical mixing [4].

Polyphosphoric acid (PPA) is an acid modifier with excellent anti-rutting ability, anti-aging ability, and outstanding storage stability, and its use in road engineering materials has been increasing year by year [11–14]. Previous research has suggested that the inclusion of PPA modifiers can enhance the high-temperature performance of both asphalt and asphalt mixtures. Baldino and Zhang et al. studied the transition of PPA-modified asphalt from a viscoelastic state to viscous fluid at elevated temperatures [15,16]. Their results indicated that the introduction of PPA improved the performance of the asphalt at high temperatures. Zhang et al. used a dynamic shear rheometer (DSR) to scan the frequency of PPA/SBS composite-modified asphalt at a fixed temperature [17]. They evaluated the rutting resistance and found that introducing PPA improved the interaction between asphalt components, which effectively improved the performance of SBS asphalt before and after aging at high temperatures. Liu et al. and Wei et al. performed an investigation of the rheological properties of PPA-modified asphalt [18,19]. The results demonstrated that the rheological properties were affected by the PPA content, and there exists a positive correlation between the PPA content and the enhanced high-temperature performance of the asphalt. Song et al. investigated the behavior of asphalt mixtures at high temperatures based on the high-temperature rutting test and the Hamburg wheel tracking test [20]. The findings indicated that the PPA composite-modified asphalt mixture had better high-temperature behavior than the SBS-modified asphalt mixture. Ma et al. investigated the high-temperature performance of PPA/SBS composite-modified asphalt mixtures through the wheel tracking test [21]. The data indicated that PPA can be partially replaced by the SBS modifier without negatively impacting the high-temperature rutting performance of the asphalt mixture. The incorporation of PPA into the mixtures was shown to reduce the required SBS content and improve resistance to high-temperature rutting. Liang et al. evaluated the compatibility and rheological properties of asphalt after adding PPA to SBR-modified asphalt [4]. The results showed that the incorporation of PPA significantly increased the adhesion, elasticity, and high-temperature behavior of SBR-modified asphalt. In conclusion, the addition of PPA to asphalt enhances the high-temperature performance capabilities of modified asphalt. In this sense, PPA might be a good alternative for polymer modifiers [15,22,23]. The enhancement impact is contingent upon the quantity of PPA, temperature, asphalt property, and volumetric index of the asphalt mixture. Nevertheless, in previous studies, the high-temperature performance of PPA-modified asphalts or mixtures has been assessed individually without modeling or predicting their relationship.

A neural network is a biologically derived computing model that learns and processes information by mimicking the behavioral characteristics of animal neural networks [24]. Many scholars have applied neural network algorithms to road engineering research, particularly pavement performance prediction [25–28]. In pavement performance prediction, traditional prediction methods, which can only consider a single influencing factor, fail to reflect the pavement performance comprehensively [29]. Neural network algorithms are

highly applicable due to their ability to handle the diversity, uncertainty, and complexity of the influencing factors [30]. Tan et al. developed a prediction model for asphalt mixture characteristics, such as low-temperature performance, asphalt property, and volume parameters, using a back-propagation (BP) neural network [31]. Yin et al. utilized a BP neural network algorithm to establish a non-linear prediction model of the factors influencing water stability and the TSR (freeze–thaw splitting strength ratio) of asphalt mixtures [32]. The model achieved high accuracy by minimizing the deviation between predicted and actual values within the engineering precision boundary. Yin et al. developed a BP neural network model to quantify the effects of recycled asphalt content, temperature, stress level, and other factors on the fatigue performance of hot mix recycling asphalt pavement [33]. The model accurately predicted the results, demonstrating its reliability. Lu et al. developed a BP neural network prediction model using MATLAB software to forecast the stability of asphalt mixtures under different wet and dry cycling conditions [34]. Previous studies have shown that BP neural networks are capable of predicting the road performance of asphalt mixtures. However, the correlation between the input and output values was not analyzed before the establishment of the prediction model. The inclusion of factors with low correlation in the input values increased the prediction error.

Given the time-consuming and resource-intensive nature of the high-temperature wheel tracking test for asphalt mixtures and the lack of an accurate prediction model for the relationship between asphalt high-temperature performance and asphalt mixture high-temperature stability, it is evident that further research is required in this area. In this paper, the microscopic properties of PPA were initially investigated through the use of FTIR and TG. The high-temperature performance of PPA-modified asphalt was analyzed by temperature scanning and multiple stress creep recovery (MSCR) tests using DSR. Influencing factors were selected from two aspects, i.e., asphalt creep property and Marshall test index. The correlation between each influencing factor and dynamic stability (DS) was analyzed by gray relational analysis. On this basis, a DS prediction model was established using the BP neural network to train on, learn, and verify asphalt mixture DS test data. The related test protocols are illustrated in Figure 1. In the figure, DH represents Donghai-70# produced by Sinopec, KL represents Kunlun-90# produced by Qinhuaodao PetroChina Fuel Asphalt Company, 2S represents an SBS content of 2%, and 2R represents an SBR content of 2%.

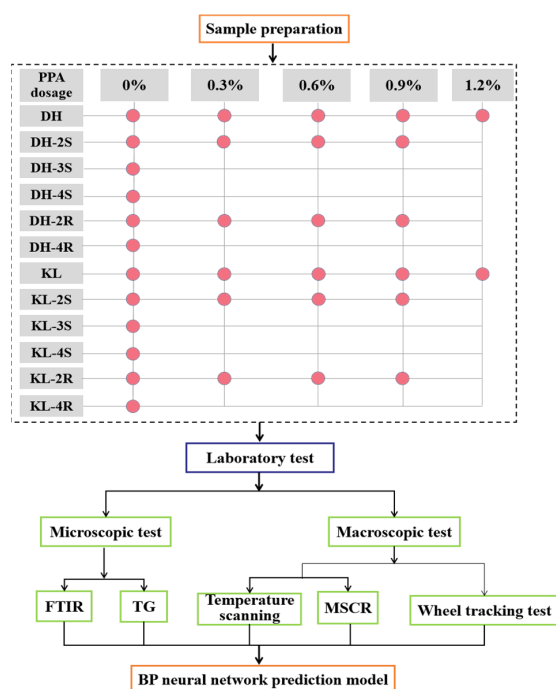


Figure 1. Flow chart of the research protocol.

2. Materials and Methods

2.1. Materials

2.1.1. Asphalt and Modifier

The tested base asphalts were DH-70# (Sinopec, Beijing, China) and KL-90# (Qinhuangdao, China). The technical parameters of the asphalts were measured by the “Standard Test Methods for Bitumen and Bituminous Mixtures for Highway Engineering” (JTG E20-2011). Table 1 presents the data from these tests. The polymer modifiers were SBS1401 (Sinopec, China) and SBR (Beijing, China). The PPA selected was industrial-grade polyphosphoric acid with a relative mass fraction of 110% from Nanjing Chemical Reagent Co., Ltd., (Nanjing, China) and technical features are listed in Table 2. Combined with the preparation method of PPA-modified asphalt used by scholars at home and abroad and the previous research results of the research group [35,36], the preparation method of PPA composite-modified asphalt was determined as follows. Firstly, the asphalt was heated to 150 °C and 2.0% SBS/SBR was added after stirring for 30 min at 4500 rpm. Then, 0.6% PPA was poured and sheared at 4500 rpm for 30 min. The resulting mixture was then subjected to swelling for one hour at a temperature of 180 °C in an oven. For ease of analysis, KL added with 0.3% PPA is represented as KL-0.3P, KL added with 2% SBS is represented as KL-2S, and KL added with 2% SBS and 0.3% PPA is designated as KL-2S-0.3P. This method is also used to represent other modified asphalts.

Table 1. Technical indexes of asphalts.

Test Project	Results		Test Method (JTG E2011)
	DH	KL	
Softening point (°C)	53.1	49.5	T 0606
Ductility (10 °C, cm)	39.4	>100	T 0605
Penetration (25 °C, 0.1 mm)	64.2	93.5	T 0604

Table 2. Main technical indicators of modifiers.

Type of Modifier	Parameter	Unit	Value
SBS1401	Block ratio, S/B	-	40/60
	Volatiles	%	≤0.7
	Ash content	%	≤0.2
	Tensile strength	MPa	24.0
	Elongation at break	%	730
	Shore hardness	-	85
SBR	Particle size	Mesh	10~80
	Bound styrene content	%	10~50
	300% stretching stress	MPa	15
	Tensile strength	MPa	≥20
PPA-110	P ₂ O ₅ concentration	%	82.0
	Chloride (Cl)	%	≤0.001
	Heavy metal (Pb)	%	≤0.003
	Iron (Fe)	%	≤0.002

2.1.2. Asphalt Mixture

The asphalt mixture selected was crushed limestone as coarse and fine aggregate with fine ground limestone mineral powder as filler, and the mineral grading was AC-13 type, as shown in Figure 2. The asphalt was modified asphalt compounded with DH, KL base asphalt, and a PPA/SBS/SBR modifier. The Marshall test method was used to determine the Optimum Asphalt Content (OAC) by determining the Relative Bulk Density of Asphalt Mixture (γ), Voids in Mineral Aggregate (VMA), Volume of Air Voids (VV), Marshall Stability (MS), Flow Value (FL), and Voids Filled with Asphalt (VFA). The specific indicator data are shown in Table 3. According to the rutting test in the specification (JTG E20-2011),

the dynamic stability of different modified asphalt mixtures was tested at 60 °C and their high-temperature stability was evaluated. The test results are presented in Table 3.

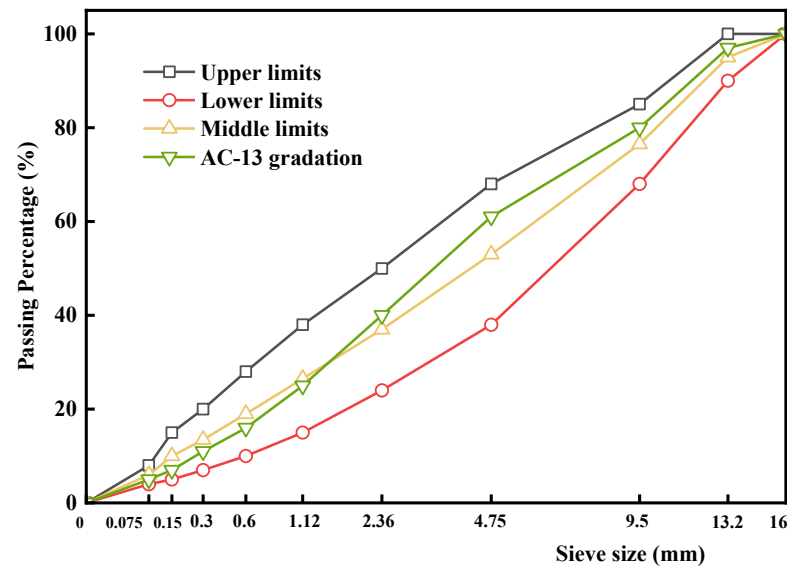


Figure 2. The grading curve of the AC-13.

Table 3. Results of Marshall testing and dynamic stability.

Asphalt Mixtures Sample Number	Asphalt Binder Type	OAC /%	γ /g·cm ^{−3}	MS /kN	VMA /%	VV /%	FL /mm	VFA /%	DS (Times/mm)
1	DH	5.11	2.400	11.00	13.81	4.01	3.03	70.96	2952
2	DH-0.3P	4.54	2.384	11.32	14.07	4.70	3.41	66.65	3036
3	DH-0.6P	4.83	2.331	13.83	16.38	4.28	4.07	74.04	3966
4	DH-0.9P	5.38	2.390	11.06	14.12	4.27	2.74	69.76	3828
5	DH-1.2P	5.47	2.384	12.90	14.29	4.56	3.53	68.16	4326
6	DH-2S-0.3P	5.22	2.380	8.78	14.08	4.34	1.98	68.76	5322
7	DH-2S-0.6P	5.42	2.380	9.91	14.81	4.59	2.08	69.01	5658
8	DH-2S-0.9P	5.47	2.382	9.95	14.58	4.63	2.11	69.20	5724
9	DH-2S	5.31	2.380	10.78	14.55	4.32	2.31	70.31	4926
10	DH-3S	4.76	2.401	12.81	14.15	4.27	3.83	70.17	4794
11	DH-4S	4.61	2.384	10.71	14.25	4.64	3.79	67.95	5772
12	DH-2R	4.85	2.410	10.69	13.92	4.58	2.62	67.10	4716
13	DH-2R-0.3P	4.82	2.385	10.36	13.94	4.65	2.60	67.30	5166
14	DH-2R-0.6P	4.93	2.390	10.39	13.98	4.71	2.56	66.31	5250
15	DH-2R-0.9P	4.50	2.501	14.82	13.31	3.85	3.76	71.11	4494
16	DH-4R	4.72	2.394	9.82	14.18	4.36	3.52	70.19	4332
17	KL	5.10	2.380	9.83	14.02	3.98	3.10	71.61	2580
18	KL-0.3P	4.67	2.342	10.23	15.36	4.12	3.64	73.26	3606
19	KL-0.6P	4.83	2.331	13.83	16.38	4.28	4.07	74.04	3966
20	KL-0.9P	5.39	2.410	9.96	14.17	4.24	2.88	70.08	3222
21	KL-1.2P	4.76	2.430	11.60	13.81	4.03	3.26	70.80	4170
22	KL-2S-0.3P	4.46	2.402	11.23	14.64	4.00	2.35	66.74	4146
23	KL-2S-0.6P	5.37	2.380	10.26	14.77	4.48	2.14	69.67	4386
24	KL-2S-0.9P	4.84	2.390	13.60	13.72	4.04	2.18	70.60	6006
25	KL-2S	5.28	2.400	10.41	14.64	4.37	2.46	70.15	3894
26	KL-3S	4.50	2.386	11.14	14.11	4.59	3.77	67.50	3438
27	KL-4S	4.68	2.385	14.50	14.29	4.59	3.98	68.20	4938
28	KL-2R	4.83	2.390	9.88	13.87	4.56	2.65	67.12	3780
29	KL-2R-0.3P	4.48	2.386	11.96	14.62	4.00	2.15	67.26	3636
30	KL-2R-0.6P	4.95	2.380	10.19	13.95	4.66	2.35	66.59	4224
31	KL-2R-0.9P	4.52	2.390	11.76	14.77	4.00	2.26	67.30	3930
32	KL-4R	4.75	2.396	11.36	14.67	4.00	2.29	66.50	3888

2.2. Methods

2.2.1. FTIR

To analyze the modification mechanisms of PPA/SBS/SBR modifiers added to base asphalt, a Nicolet iS50 Fourier infrared spectrometer was used to conduct the tests and collect the specimens' spectra. The wavenumber area was between 400 cm^{-1} and 4000 cm^{-1} , the number of scans was 32, and the frequency resolution was 4 cm^{-1} . The spectra were analyzed using OMNIC 7.3 software.

2.2.2. TG

To analyze the curve changes to evaluate the thermal stability of different modified asphalts, TG tests were performed using a NETZSCH STA449F5 TGA. The temperature range consisted of $25\text{ }^{\circ}\text{C}$ to $800\text{ }^{\circ}\text{C}$ and the heat-up frequency was $15\text{ }^{\circ}\text{C}$ per minute, the test environment was a nitrogen atmosphere, and the nitrogen flow rate was 50 mL per minute.

2.2.3. Temperature Sweep Experiment

A Discovery HR-1 DSR, manufactured by TA, was used for the temperature sweep experiment. The temperature was $58\text{--}76\text{ }^{\circ}\text{C}$ (temperature interval of $6\text{ }^{\circ}\text{C}$). The rotational speed was 10 radians per second, with an applied strain of 12.0% magnitude. The parallel plates were spaced 1.0 mm apart and 25.0 mm in diameter. The complex shear modulus (G^*) and phase angle (δ) of different modified asphalts were tested and the rutting factor ($G^*/\sin\delta$) was calculated to analyze the high-temperature rheological properties of the asphalts.

2.2.4. MSCR

The MSCR testing used a dynamic shear rheometer to control the stress loading and simulate the viscoelastic properties of asphalt under various stress conditions [37,38]. In this paper, the same instrument and parallel plate as the temperature scanning testing were used, with the test temperature set at $60\text{ }^{\circ}\text{C}$.

2.2.5. Wheel Tracking Test

To evaluate the rutting resistance of the asphalt mixture at high temperatures, the testing was conducted following T0719 of JTG E20-2011. The index DS was obtained to evaluate the high-temperature stability of the asphalt mixtures. A rutting specimen of $30\text{ cm} \times 30\text{ cm} \times 5\text{ cm}$ size was placed in a constant temperature of $60 \pm 1\text{ }^{\circ}\text{C}$ in the thermostat. The holding time of the specimen was not less than 5 h and not more than 12 h. The test wheel pressure was 0.7 MPa. The deformation curve and specimen temperature were recorded during the test and the DS was calculated.

2.3. BP Neural Network

A BP neural network was created using a back-propagation algorithm that continuously adjusts the weights and thresholds of the network based on the function of gradient descent. The main objective is to minimize the total squared error of the network [39]. Figure 3 displays the flowchart of the algorithm. BP neural networks, widely employed in different fields, have gained popularity due to the utilization of MATLAB R2022a software [40]. They consist of a hidden layer, an input layer, and an output layer, each containing multiple nodes. Weights and bias govern the interconnection between input and output values for each node.

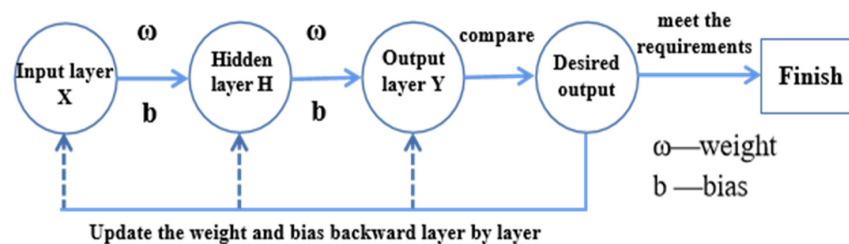


Figure 3. BP neural network algorithm flowchart.

3. Results and Discussion

3.1. FTIR Test Analysis

To analyze the changes in the molecular structure and functional group properties of asphalt, DH and KL asphalt and PPA-SBS/SBR single/composite-modified asphalt were selected for infrared spectroscopy testing. The corresponding test results are given in Figures 4 and 5.

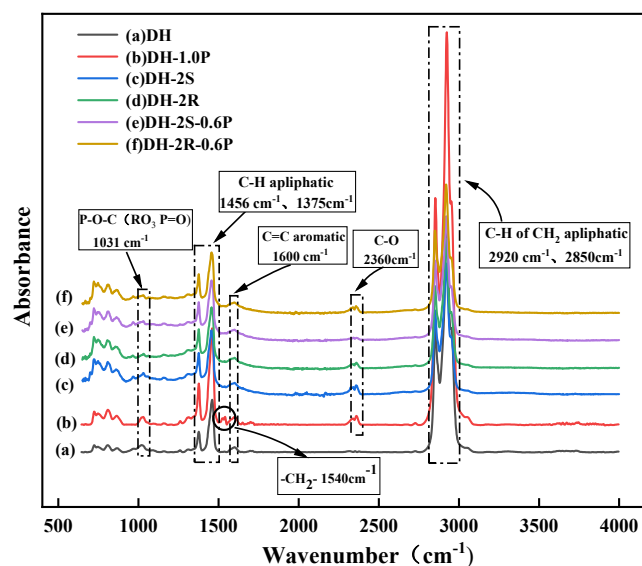


Figure 4. Infrared spectra of DH–PPA single/composite-modified asphalt.

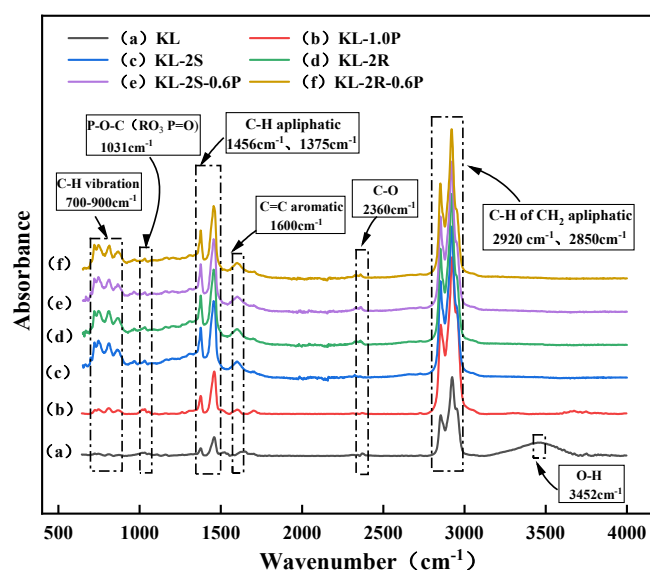


Figure 5. Infrared spectra of KL–PPA single/composite-modified asphalt.

As can be seen in Figures 4 and 5, the DH-PPA-modified asphalt with the addition of PPA showed a new characteristic peak at 2360 cm^{-1} , which corresponds to the absorption peak of the C-O bond stretching vibration. The KL-PPA-modified asphalt displays a new absorption peak near 1031 cm^{-1} , which corresponds to the P-O-C antisymmetric stretching vibration of the phosphate ester $(\text{RO}_3)\text{P}=\text{O}$ group. The absorption peak of KL-PPA-modified asphalt disappears at 3452 cm^{-1} , where the absorption peak is $-\text{OH}$ stretching vibration. The creation and disappearance of the three peaks indicate that PPA chemically reacts with the base asphalt to produce new substances.

Further analysis is shown in Figure 4; after the addition of PPA/SBS and PPA/SBR to the DH base asphalt, the infrared absorption of the modified asphalt changes compared to the base asphalt, and the absorption peaks at 2360 cm^{-1} change at approximately the same location, except that the peak intensities change slightly, with a peak height of 2.17 for DH-2S, a peak height of 0.54 for DH-2S-0.6P, a peak height of 1.46 for DH-2R, and a peak height of 1.34 for DH-2R-0.6P. As illustrated in Figure 5, after the addition of PPA to KL/SBS- and KL/SBR-modified asphalt, no significant new characteristic peaks appear in the functional group region ($1333\text{--}4000\text{ cm}^{-1}$). In contrast, a significant change in wave intensity occurs in the range of $800\text{--}1000\text{ cm}^{-1}$, as exemplified by KL-2S-0.6P at 810 cm^{-1} , where the peak height increases from 0.004 to 3.05 for base asphalt. This may be because SBS, SBR, and PPA do not react chemically, but are just a physical superposition of the infrared spectra of polystyrene and polybutadiene, such that the position of the absorption peaks remains unchanged and the intensity changes. This shows that the PPA is physically blended with the SBS and SBR.

3.2. TG Test Analysis

To research the thermal stability of PPA-modified asphalt, PPA-110 was added to DH base asphalt at dosages of 0.5%, 1.0%, and 1.5% to prepare PPA-modified asphalt samples, which were then subjected to TG analysis. Table 4 and Figure 6 show the experimental results.

Table 4. Pyrolysis parameters of PPA-modified asphalts at different dosages.

Type of Asphalt	T_m ¹ (°C)	T_n ² (°C)	$T_{5\%}$ ³ (°C)	$T_{10\%}$ ⁴ (°C)	$T_{50\%}$ ⁵ (°C)	Weight Percent (%)
DH	359.5	555.7	373.4	384.1	463.5	99.1
DH-0.5P	351.3	579.5	372.7	383.4	478.4	97.8
DH-1.0P	373.4	570.4	384.1	389.7	458.9	99.1
DH-1.5P	325.5	581.4	351.8	380.9	473.1	99.9

¹ Temperature at initiation of decomposition. ² Temperature at termination of decomposition. ³ Temperature at 5% weight lost. ⁴ Temperature at 10% weight lost. ⁵ Temperature at 50% weight lost.

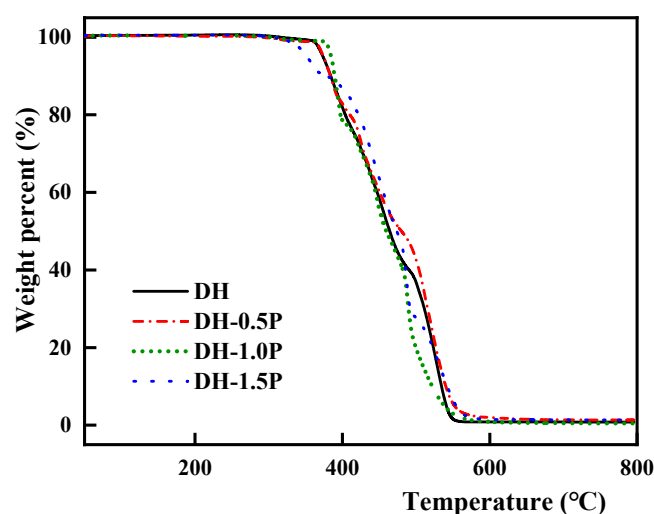


Figure 6. Comparison of TG curves of PPA-modified asphalts at different dosages.

The trend of the TG curve of the PPA-modified asphalt at various contents is similar to that shown for the base asphalt, as illustrated in Table 4, and Figure 6. The pyrolysis reaction mainly occurs between 350 °C and 580 °C, and there are apparent sharp peaks. The base asphalt and modified asphalt curves show pronounced sharp peaks in this temperature range. The decomposition initiation temperature is more prominent compared to the base asphalt at a PPA dosage of 1.0%, indicating that a certain amount of PPA added to the base asphalt would contribute to thermal stability improvement. The TG curve plateaus around 550–580 °C, where the decomposition is completed. The decomposition termination temperature T_n of PPA-modified asphalt exhibits an increase with rising PPA dosage, demonstrating that adding PPA could enhance the thermal stability of asphalt. In further analysis, when the temperature is below 300 °C, the results for a different dosage of PPA-modified asphalt mass loss are unchanged, and the actual service environment temperature of asphalt pavement is within 300 °C, indicating that the addition of PPA does not affect the thermal stability of asphalt under the actual service temperature.

3.3. Temperature Sweep Test Analysis

The rutting factor is the quotient of the complex shear modulus G^* and the sine of the phase angle δ , characterizing the anti-deformation ability of asphalt under high temperatures. A more significant rutting factor represents a better high-temperature performance of asphalt [41]. According to the Superpave Binder Performance Specification, the temperature at which unaged asphalt has a rutting factor of 1.0 kPa is the high-temperature failure temperature of the asphalt. Temperature sweep tests were conducted on PPA single/composite-modified asphalt at 58 °C to 76 °C (an interval of 6 °C). The loading frequency was set as ten radians per second, while the strain control rate was 12.0%. The experimental findings obtained are illustrated in Figures 7 and 8.

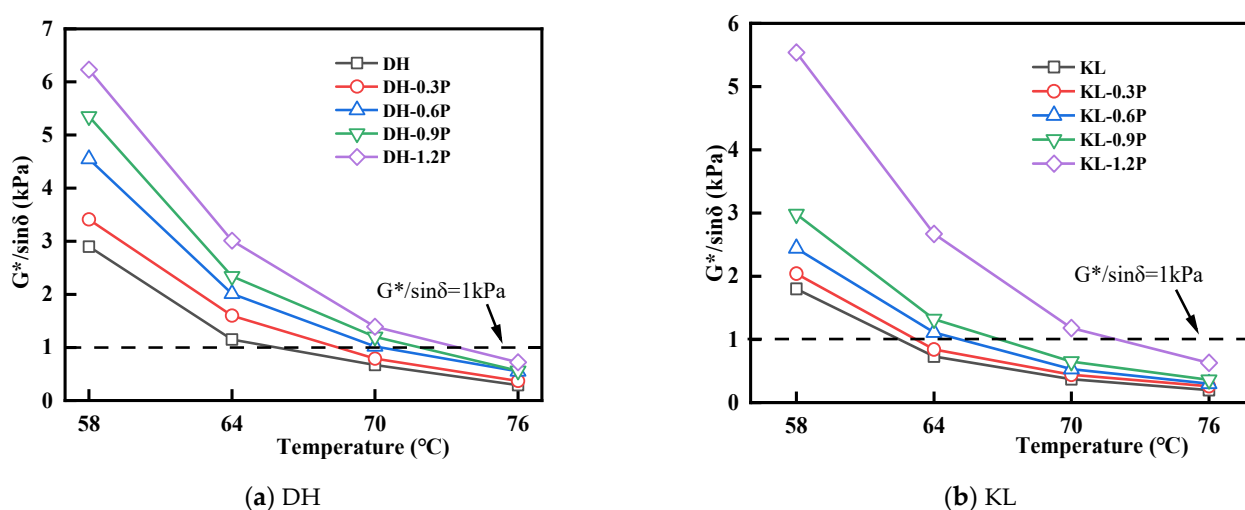


Figure 7. The relationship between $G^*/\sin\delta$ and temperature for PPA single-modified asphalts.

Figure 7 illustrates the curves of $G^*/\sin\delta$ versus temperature for PPA single-modified asphalt with different base asphalts and different PPA blends. At the same temperature, the $G^*/\sin\delta$ values of the PPA-modified asphalt are all greater than those of the base asphalt, and the high-temperature performance is better. Further analysis shows that the high-temperature failure temperature of asphalt increases with increasing PPA content. The DH asphalt increases from 65.6 °C of the original sample to 74 °C at 12% PPA content, and the KL asphalt increases from 62.3 °C of the original sample to 71.7 °C at 12% PPA content. This shows that the asphalt can be used at higher temperatures after PPA is added to the asphalt. The primary reason is that PPA converts the colloidal component in asphalt to hard asphaltenes, which increases the asphalt's resistance to deformation and therefore improves its high-temperature stability [42].

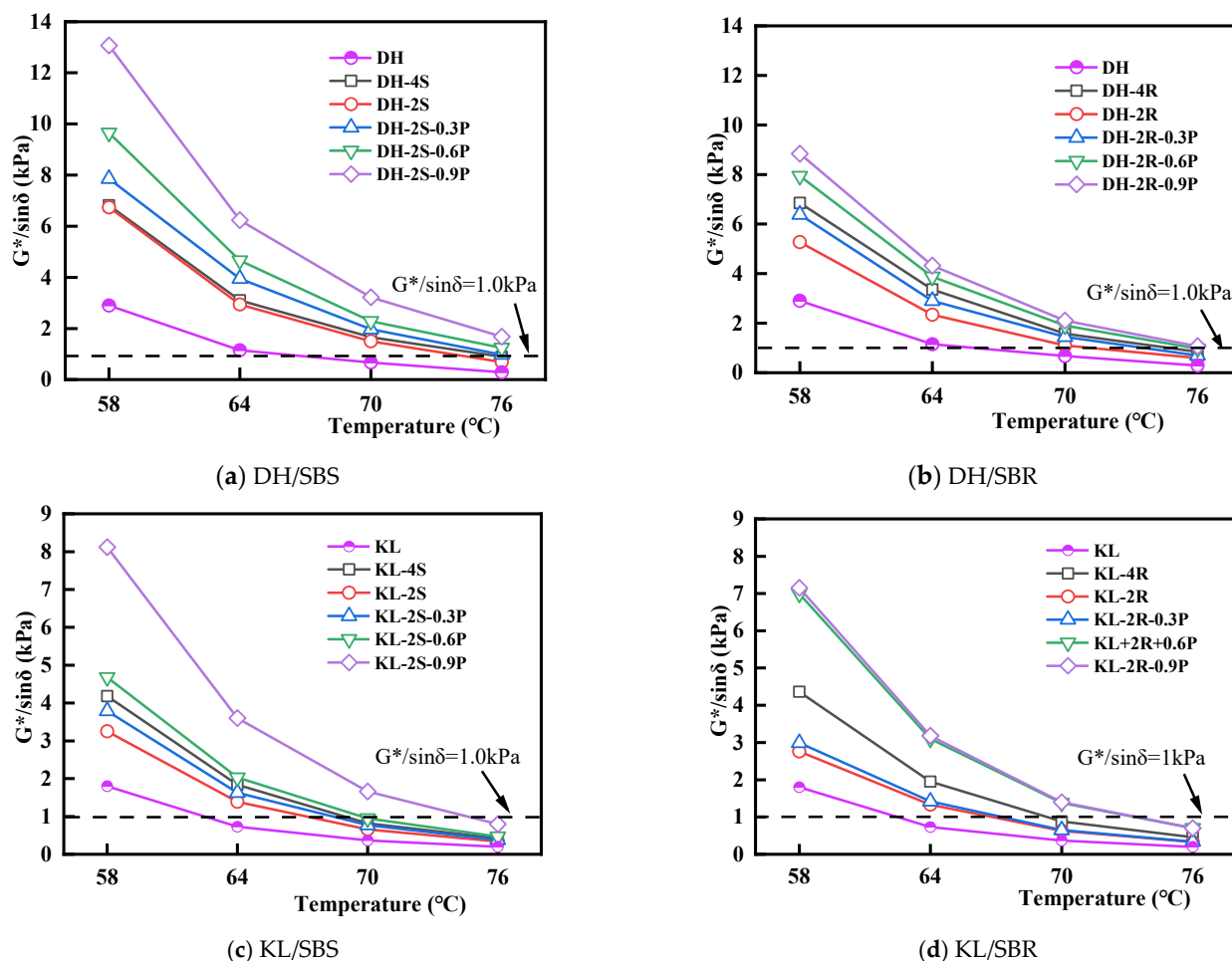
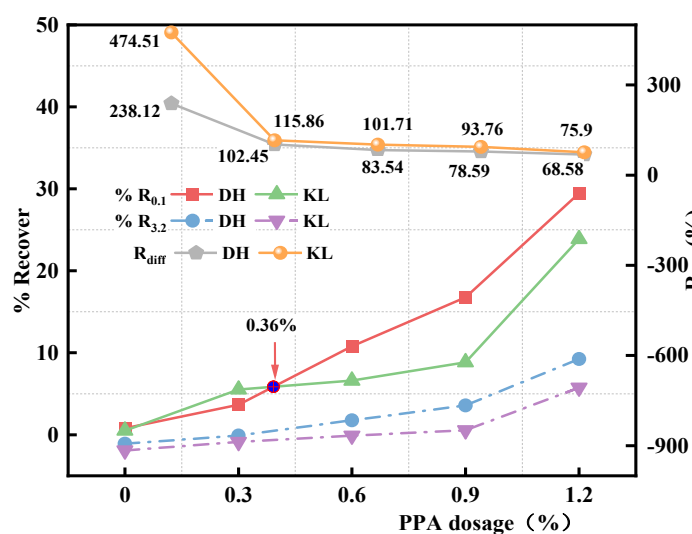


Figure 8. The relationship between $G^*/\sin\delta$ and temperature for PPA/polymer modifier composite-modified asphalts.

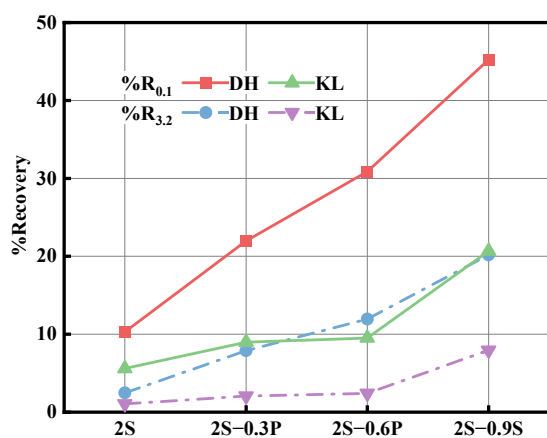
Figure 8 presents the $G^*/\sin\delta$ versus temperature curves for PPA and polymer modifier composite-modified asphalt. For DH base asphalt, the addition of different PPA/SBS/SBR modifiers increased $G^*/\sin\delta$. SBS had the most significant modifying effect. For KL base asphalt, the increase in $G^*/\sin\delta$ values of the different modified asphalt was less than that of the DH base asphalt. The increase in $G^*/\sin\delta$ value was twice as large for 0.9% PPA compared to 0.6% PPA for the KL base asphalt when the SBS dosage was 2%. At 58 °C, the $G^*/\sin\delta$ of KL-2S-0.6P is 1.60 times that of KL, while that of KL-2S-0.9P is 3.51 times. Further analysis of Figure 8 shows that as the temperature increases, the $G^*/\sin\delta$ value of different PPA-modified asphalts decreases. This indicates that with increasing temperature, the high-temperature performance of asphalt deteriorates and rutting resistance decreases. At the same temperature, PPA-modified asphalt has a greater $G^*/\sin\delta$ value than SBS/SBR-modified asphalt. This shows that with the increase of PPA content, the high-temperature performance of the asphalt binder is enhanced. When 0.6% PPA is compounded with 2% SBR, the high-temperature stability exceeds that of modified asphalt containing 4% SBR, indicating that PPA might be a good alternative for polymer modifiers. This viewpoint is also corroborated by the elevated temperature at which the asphalt fails. When the PPA content is 0.6%, the high-temperature failure temperature of 2% SBR/PPA composite asphalt is higher than that of 4% SBR-modified asphalt. The primary reason for this is the absence of a chemical reaction between PPA and the SBS or SBR composite, which results in the dispersion of the latter into fine particles. This enhances the role of dissolution, improves the SBS and SBR modification effect, and thereby improves the high-temperature performance [43].

3.4. MSCR Result Analysis

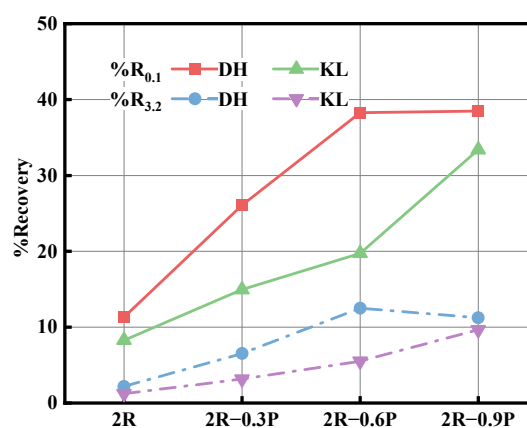
The MSCR method can simulate the viscoelastic properties of asphalt at various loading conditions and provide the asphalt's creep and recovery curves at varying loading levels and temperatures. The testing was first conducted at 0.1 kPa for 20 consecutive cycles and then at a pressure of 3.2 kPa for ten cycles. Every cycle was separated into a 1 s loading creep section and a 9 s unloading recovery period. The entire MSCR test lasted 300 s. Calculating the collected strains, the percent recovery (%R) and the non-recoverable creep compliance (J_{nr}) of the asphalt were determined. The %R is the proportion of the recoverable strain in the total creep strain. In contrast, the J_{nr} is the quotient of the unrecoverable strain value and loaded stress. In this study, MSCR tests were carried out on PPA-modified asphalt, both single and composite, with PPA dosages ranging from 0.3% to 1.2%, utilizing a DSR. Results are presented in Figures 9 and 10, which illustrate the %R and J_{nr} of the PPA-modified asphalt.



(a) Base asphalt



(b) PPA/SBS



(c) PPA/SBR

Figure 9. %R of PPA single/composite—modified asphalt.

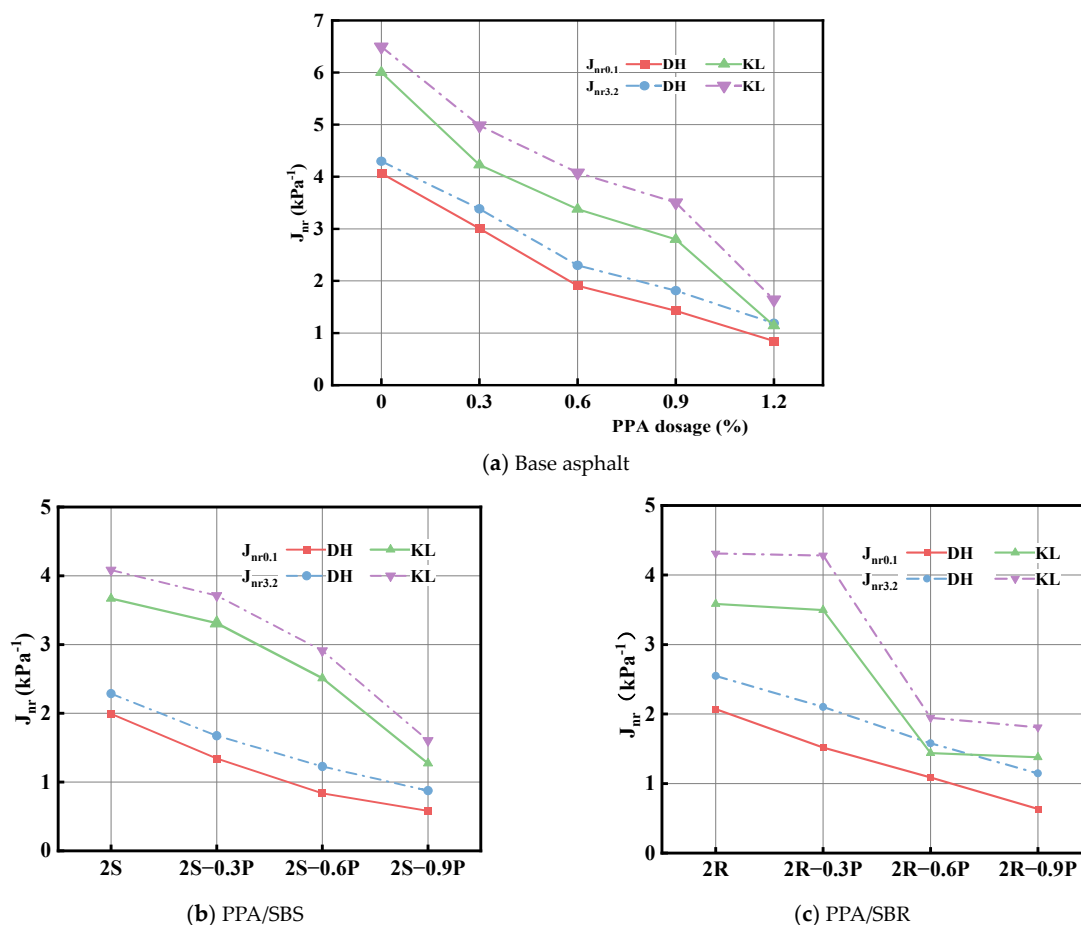


Figure 10. Non-recoverable creep compliance J_{nr} of PPA single/composite-modified asphalts.

3.4.1. Percent Recovery

Figure 9 is the %R of modified asphalt under different strains and PPA contents. The %R indicates the elastic properties of the asphalt. The higher the %R, the greater the elastic recovery.

According to Figure 9a, it is evident that the %R of both kinds of asphalt increases as the PPA dosage increases. Improvements in percent recovery were observed in both types of asphalt after the introduction of PPA; this is because adding PPA to the asphalt promotes the conversion of colloidal components into asphaltenes, thus increasing the asphaltene content. Macroscopically, the asphalt's ability to withstand high temperatures becomes enhanced with rising asphaltene content. At the same loading level, the modified DH asphalt exhibits a larger %R than the modified KL asphalt. At 0.1 kPa, modified DH asphalt exhibits a contrast when the PPA content reaches 0.36%. The magnitude of %R variation at 0.1 kPa is larger than that at 3.2 kPa, mainly because asphalt belongs to the category of viscoelastic material and its mechanical behavior is affected by stress. In the early stage (20-cycle test at 0.1 kPa), the asphalt undergoes elastic deformation that is not immediately recovered but gradually recovers with time. The percent recovery at the later stage (10-cycle test at 3.2 kPa) is affected by the cumulation of delayed elastic recovery. As a result, the accumulated elastic deformation recovered at the later stage is comparable to the residual deformation. This is manifested by a reduction in residual deformation at the last stage, an increase in recoverable deformation, and a larger increase in the %R. The base asphalt has a greater R_{diff} than PPA-modified asphalt. As the amount of PPA increases, the ability of the asphalt to recover from creep under load decreases. Generally, the R_{diff} of the KL asphalt exceeds that of the DH asphalt, implying that the KL asphalt is more sensitive to stress changes.

PPA added to asphalt is decomposed to H_2PO_4^- and H^+ , inducing protonation reactions that result in the loss of hydrogen bonds in the asphaltene and asphaltene disaggregation. After disaggregation, the asphaltene becomes polar and prone to cross-linking with H_2PO_4^- to form covalent compounds. This is evidenced by a visible increase in asphaltene content [44], which strengthens the asphalt's capacity to endure high temperatures and regain its shape after deformation. KL asphalt is a softer grade and is more sensitive to stress response at high temperatures.

As shown in Figure 9b,c, there is an upward trend in the %R of PPA/SBS, and PPA/SBR composite-modified asphalts with increasing PPA dosage, which suggests that the inclusion of PPA improves the percent recovery and high-temperature performance of the modified asphalts. This finding aligns with the temperature sweep results. Additionally, the change in the %R of the modified asphalt is not influenced by the stress level, showing that the PPA dosage has a more substantial effect on the creep recovery of modified asphalt. For the same type of base asphalt, the %R decreases as the stress level is elevated from 0.1 kPa to 3.2 kPa; this is because, at higher stress levels, the deformation is less recoverable and the elastic deformation amount is smaller.

3.4.2. Non-Recoverable Creep Compliance

Figure 10 displays the non-recoverable creep compliance J_{nr} of PPA single/composite-modified asphalts. A reduced J_{nr} implies less deformation of the asphalt under identical loads and improved resistance to deformation.

Figure 10a shows that the J_{nr} of DH and KL asphalts declines with increasing PPA dosage at the same stress level, with a larger variation amplitude at 0.1 kPa than that at 3.2 kPa. This shows that using PPA increases the deformation strength of asphalts and decreases their irreversible deformation at high temperatures. The J_{nr} value of PPA/DH is smaller than that of PPA/KL, indicating that the unrecoverable deformation of PPA/DH is smaller than that of PPA/KL, which may be due to the fact that adding PPA to DH asphalt induces the transformation of light components into asphaltenes. This boosts the asphaltene content, improves the high-temperature performance of asphalt, and reduces unrecoverable asphalt deformation. Within the same kind of asphalt, the J_{nr} of the DH asphalt rises as the load increases, while the J_{nr} of KL asphalt exhibits a pattern of initial decline and subsequent rise as the stress level increases.

Figure 10b,c show that after creep recovery at different stress conditions, the J_{nr} of PPA/SBS and PPA/SBR composite-modified asphalts shows a reducing trend with the augmentation of PPA dosage. It is shown that the inclusion of PPA reduces the non-recoverable deformation of asphalt under load while enhancing its elastic deformation, thereby improving its resistance to high-temperature deformation. There are differences in the variation of creep compliance of different asphalts. For DH asphalt, the J_{nr} of PPA/SBS and PPA/SBR composite-modified asphalt has little change. However, the change is more obvious for KL asphalt. When the content of PPA is 0.3–0.6%, the J_{nr} of PPA/SBR is substantially lower than the one of PPA/SBS, indicating that the resistance of PPA/SBR to high-temperature deformation is more robust than that of PPA/SBS.

3.5. High-Temperature Performance Prediction for PPA-Modified Asphalt Mixtures

3.5.1. Determination of Input and Output Values

Many factors affect the DS of asphalt mixtures, among which the asphalt creep recovery characteristic and Marshall test index are the two most important aspects. The relational degree of the influencing factors, such as %R, J_{nr} , $G^*/\sin\delta$, VV, VFA, OAC, γ , VMA, MS, and FL with the DS of the asphalt mixture was analyzed using MATLAB R2022a software. A larger correlation coefficient indicates a stronger correlation between these factors and the DS. The gray correlation coefficients are given in Figure 11.

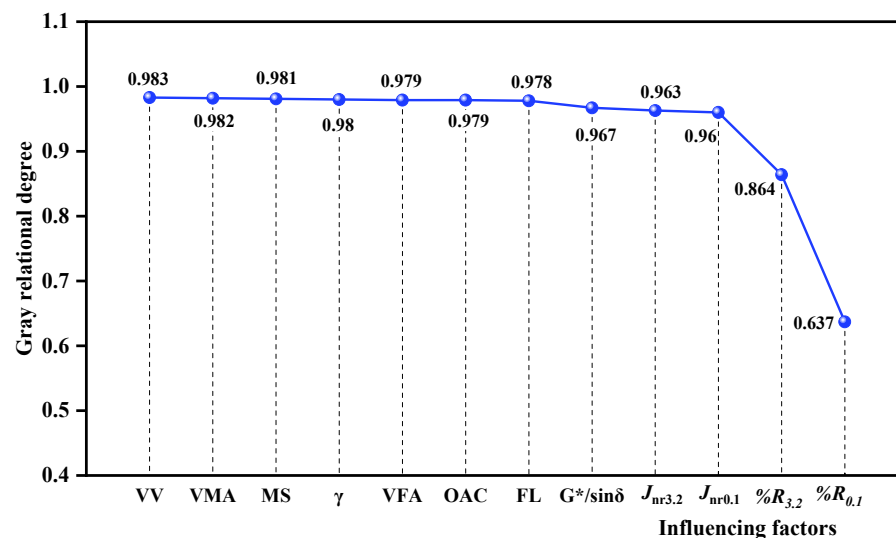


Figure 11. The gray correlation degree between each influencing factor and dynamic stability.

Based on the above analysis, the correlation degree between each factor and dynamic stability is large and the correlation degree of the ten factors is more than 0.9. The higher the correlation value, the stronger the correlation with the dynamic stability of the asphalt mixture. Therefore, the ten influencing factors above were selected as the input indices of the BP neural network model. The output variable is dynamic stability.

To enable the high-temperature performance of PPA-modified asphalt and its mixtures to be predicted, the proposed model follows a neural network structure with a single hidden layer of 10–20–1, as shown in Figure 12. Research indicates that the amount of hidden layer nodes is typically more than twice the number of input layers by one layer. Nodes in the hidden layer are twice the number of nodes in the input layers, and then plus one [32]. The original amount of hidden layer nodes in this study was set to 21. After training and error analysis, the optimal number of hidden layer nodes was finally determined to be 20.

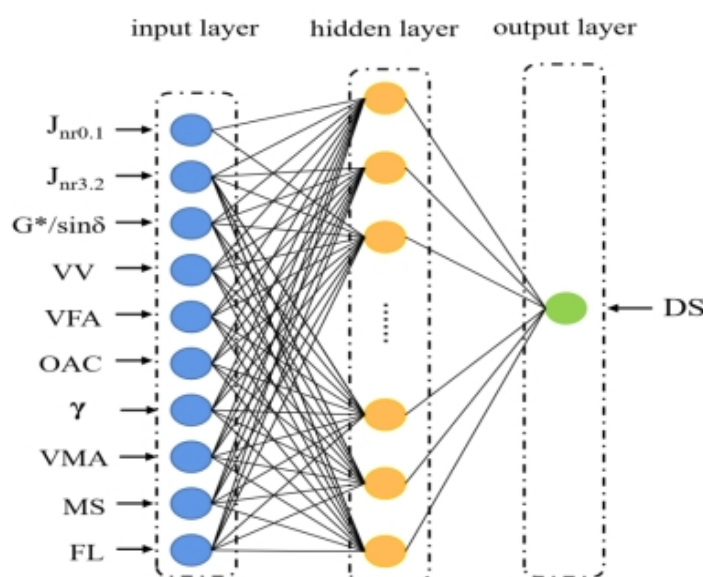


Figure 12. Structure diagram of asphalt mixture DS prediction.

3.5.2. Model Prediction Results

In this paper, 32 sets of sample data were tested, with the data randomly divided into training sets, verification sets, and test sets.

The MATLAB R2022a software was utilized to build the neural network, with the Mapminmax function employed to normalize the input and output values within the [0,1] range, thereby enhancing the sensitivity of the network weights and thresholds to the changes in values of each factor. The transfer function connecting the input layer and hidden layer is determined by an S-type tangent function Tansig. The linear excitation transfer function Purelin is used for the output layer, and the training function is Trainlm using the Levenberg–Marquardt algorithm. The output data after training must be denormalized based on the pre-calculated maximum and minimum values to obtain the simulation results. After 34 training iterations, the fitting degree and prediction effectiveness of the BP neural network model were obtained, as shown in Figures 13 and 14, respectively.

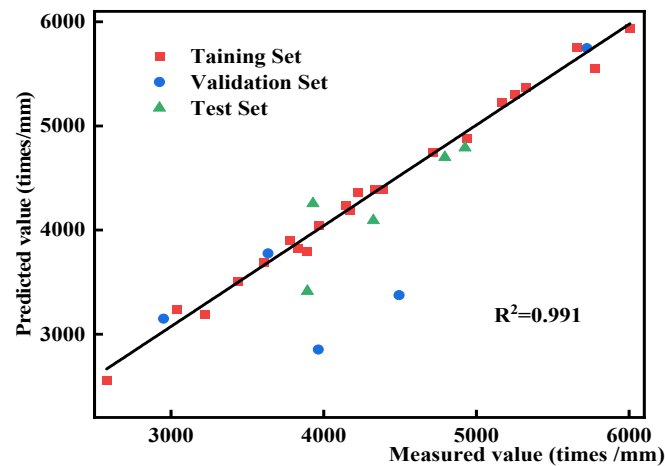


Figure 13. Fitting degree of the neural network model.

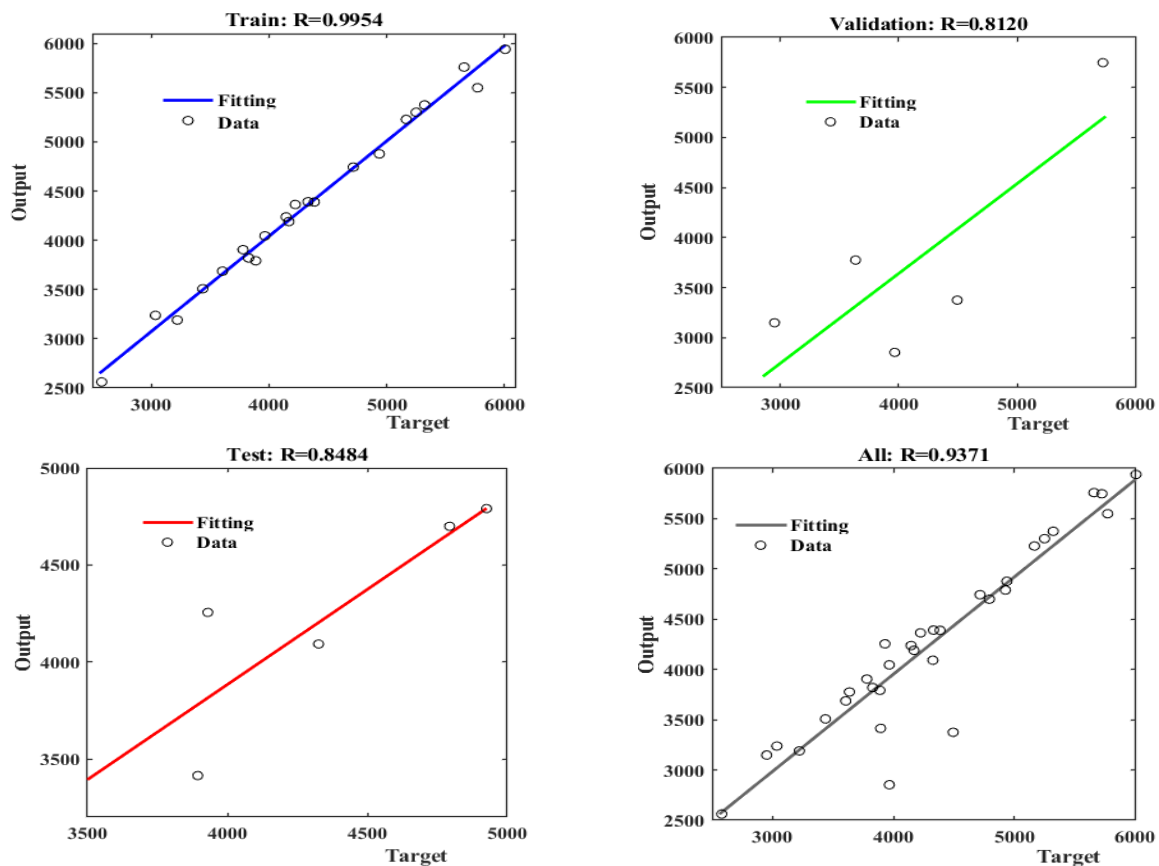


Figure 14. The relationship between predicted value and measured values of the neural network.

It can be seen from Figures 13 and 14 that the correlation coefficients between the measured values and the predicted values of the training set, validation set, test set, and the whole model are 0.9954, 0.8120, 0.8484, and 0.9371, respectively. The model fitting degree R^2 is 0.991, which indicates that the BP neural network model can predict the dynamic stability of asphalt mixtures very well.

3.5.3. Error Analysis

The Mean Absolute Percentage of Error (MAPE) is used to quantify the prediction accuracy of the neural network; the lower the MAPE, the higher the prediction accuracy. Equation (1) shows the calculation formula [45].

$$MAPE = \frac{\sum_{i=1}^M \left| \frac{A_i - \tilde{A}_i}{A_i} \right|}{M} \times 100\% \quad (1)$$

where M is the number of samples in the test set, A_i is the measured value of DS in the test set, and $\tilde{A}_i$ is the predicted value of DS from the prediction model.

As shown in Figure 15, the maximum residual variance between the measured and predicted values is 480 and the maximum relative error is 12.33%. The minimum relative error is as low as 1.98% and the error between the measured and predicted values is small. In further analysis, the MAPE value is 6.15% and the prediction accuracy of the prediction model is high, which indicates that the prediction of high-temperature performance of asphalt mixtures using the BP neural network has high feasibility.

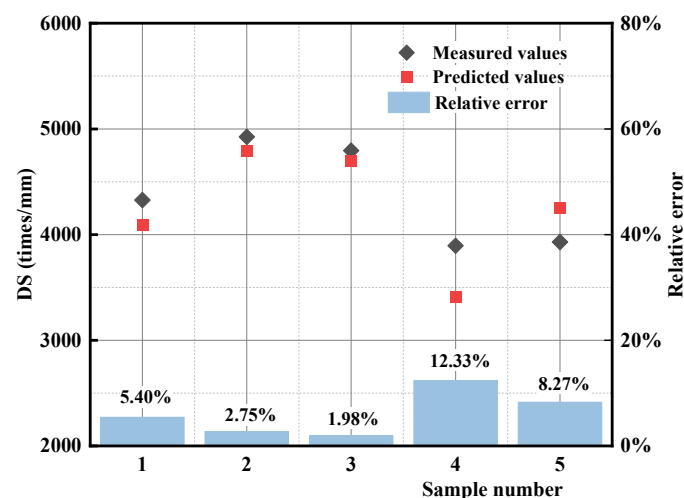


Figure 15. Prediction effectiveness of the neural network.

4. Conclusions

This paper examined the high-temperature performance of PPA-modified asphalt and asphalt mixtures. The factors influencing the high-temperature performance of asphalt mixtures were analyzed and a prediction model for the DS of asphalt mixtures was established based on a BP neural network. The findings of this paper are as follows:

- (1) When PPA is added to asphalt, it undergoes a chemical reaction with the base asphalt while physically blending with SBS or SBR. The thermal decomposition reaction of PPA-modified asphalt occurs primarily between 350 °C and 580 °C. The termination temperature of asphalt decomposing due to PPA modifications at varying dosages exceeds that of the base asphalt. This represents heightened thermal stability in the modified asphalt.
- (2) The $G^*/\sin\delta$ values of the PPA-modified asphalt were found to increase with increasing PPA loading. When PPA at a concentration of 0.6% was compounded with 2%

- SBS/SBR, the high-temperature stability exceeded that of the 4% SBS/SBR-modified asphalt blends, indicating that PPA might be a good alternative for polymer modifiers.
- (3) PPA was incorporated into the asphalt, increasing the percent recovery and the high-temperature performance of the modified asphalt. At the same stress level, the stiffness of the base asphalt decreases as the PPA content increases. The range of change is greater at the 0.1 kPa pressure level than at 3.2 kPa. At varying stress levels, both PPA/SBS and PPA/SBR composite-modified asphalt exhibit a decrease in J_{nr} with increasing PPA dosage.
 - (4) By combining MATLAB R2022a software and a BP neural network, we constructed a model to predict the high-temperature performance and high-temperature stability of asphalt mixtures. The fitting degree of the model is 0.991, the correlation coefficient between the predicted and measured values is 0.8484, and the *MAPE* value is 6.15%. These results demonstrate the feasibility of using the neural network to predict the high-temperature performance of asphalt mixtures.

This study established a BP neural network model that can better predict the high-temperature stability of PPA asphalt mixtures. Next, further work will be done mainly from two aspects: First, considering that the modified asphalt greatly influences the aggregate structure, subsequent research will increase the aggregate gradation types and the base asphalt types studied. Second, the BP neural network will be used to construct other performance-prediction models of PPA asphalt and asphalt mixtures. For example, the long-term aging performance of asphalt mixtures is predicted by the aging performance of PPA-modified asphalt, and the long-term performance of modified asphalt is quickly judged, which provides a basis for selecting pavement materials.

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