

Research Article

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Optimizing the performance of TPCB/SCA composite-modified asphalt using improved response surface methodology

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Abstract: A composite-modified asphalt was developed by incorporating tire pyrolytic carbon black (TPCB) with a silane coupling agent (SCA) to enhance its ductility and storage stability. The functional binding between asphalt and TPCB was improved by the addition of SCA. The gray correlation comprehensive evaluation method is incorporated into the response surface methodology (RSM), leading to an improved RSM. The improved RSM was employed to optimize the content of the modifier and the preparation process to enhance overall performance. A central composite design-based experiment was conducted to optimize the penetration, softening point, 10°C ductility, Brookfield rotational viscosity, and storage stability indicators of modified asphalt. The response surface of the gray comprehensive evaluation value of the evaluation indicators was calculated using the gray correlation analysis method, and a second-order regression model was established between independent variables such as TPCB, SCA content, and shear time factor and the gray comprehensive evaluation value. This model analyzed the impact of a single variable and variable interaction on the performance of the TPCB/SCA composite-modified asphalt. The optimized model results showed that the preparation parameters that optimize the expected

comprehensive performance of the composite-modified asphalt are 9.2% TPCB content, 2.1% SCA content, and a shear time of 56.3 min. Finally, the TPCB/SCA composite-modified asphalt prepared with the improved RSM exhibited the highest gray comprehensive evaluation value and the best overall performance, indicating the feasibility of the optimized preparation parameters with the improved RSM.

Keywords: road engineering, modified asphalt, tire pyrolytic carbon black, silane coupling agent, improved response surface methodology

1 Introduction

Due to the rapid growth of the transportation industry and the increase in the number of vehicles, waste tires have become one of the largest types of waste worldwide. As a result, recycling and reuse of waste tires have become urgent issues that need to be addressed [1]. There are two main ways to recycle waste tires: direct utilization, such as reuse and rubber product remanufacturing, and indirect utilization, such as powder utilization, desulfurization regeneration, material modification, pyrolysis distillation, and alternative fuels [1–3]. Pyrolysis is a process that degrades the organic components in rubber powder; generates volatile substances and solid coke; and produces carbon black, pyrolysis oil, combustible gas, and steel wire from waste tires under anaerobic conditions [4]. This process is mainly divided into two parts: natural rubber pyrolysis and synthetic rubber pyrolysis and has the advantage of large processing capacity, no secondary pollution, and multiple resource recycling and utilization [1,5]. Carbon black is an important byproduct of the pyrolysis method for treating tires, with a production yield of usually 30–40% [6]. Since the emergence of waste tire pyrolysis technology in the 1980s, the application of waste tire pyrolysis carbon black (TPCB) has gained significant attention. In 1995, Lesueur *et al.* found that with increasing amounts of TPCB, the high-temperature performance of asphalt gradually improved, and TPCB had

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rheological properties similar to those of mineral powder [7]. In 2016, Feng *et al.* found that TPCB improved the high-temperature performance of asphalt and significantly improved the thermal, light, and oxygen aging of matrix asphalt [8]. Kumar *et al.* studied the storage stability performance of tire pyrolysis carbon (TPC)-modified adhesives for waste tires with and without the addition of crosslinking agents. The TPC-modified binder exhibits good storage stability in sulfur (optimal dosage of 0.3%) and PPA (optimal dosage of 1%). In addition, waste plastic pyrolysis carbon-modified sulfur-containing binder can provide a stable storage formula [9,10]. In 2018, Li *et al.* confirmed that TPCB improved the anti-rutting performance of asphalt mixtures [11]. In 2021, Feng *et al.* found that TPCB weakened the interfacial interaction force between aggregate and asphalt, leading to poor low-temperature crack resistance, and the characteristics of TPCB insoluble in asphalt and organic solvents resulted in its poor storage stability [12]. In 2022, Li *et al.* proposed a process for preparing asphalt mixtures modified by TPCB through dry mixing [1]. Chen *et al.* revealed the modification mechanism of TPCB/TPO-MB. They showed that using TPCB and TPO composite materials to modify asphalt is feasible [13]. TPCB-modified asphalt significantly improves the high-temperature performance of asphalt and asphalt mixtures; enhances resistance to heat, light, and oxygen aging; and provides a new choice for green road construction materials with good economic and environmental performance. TPCB-modified asphalt has attracted significant attention and has been applied in real engineering projects in China [14].

Silane coupling agent (SCA) is an organic compound that simultaneously possesses a hydrophilic inorganic group and a hydrophobic organic group [15]. It is of great significance for bonding materials of different types and properties, which can improve the interfacial conditions and combine with various materials, such as glass fiber, adhesive, coating, textile, and filler. It improves the interaction between matrix materials and functionalized silicon compounds by forming “molecular bridges.” Some road researchers used SCA as intermediate materials to surface-treat the existing road materials. In 2009, Guo *et al.* [16] used SCA to pretreat oil shale waste powder and then remade composite-modified asphalt. It was found that the addition of SCA significantly improved the high-temperature rheological properties and storage stability of the asphalt and effectively improved the temperature sensitivity of the asphalt. In 2012, Chen and Tang [17] used SCA to surface treat asphalt flame retardants, and the two underwent chemical reactions to form new chemical bonds, improving the dispersibility and compatibility of the flame retardants in the asphalt and improving the thermal stability of the asphalt flame retardants. In 2012, Liang *et al.* [18] found that increasing the coupling agent content gradually

increased the contact angle of the asphalt, indicating that the surface tension of the asphalt decreased, the hydrophobicity increased, and the surface activity increased. Compared with the matrix asphalt, the bonding properties between modified asphalt and aggregates are excellent. In 2020, Hui [19] used SCA to treat coal liquefaction residue-modified asphalt, which effectively improved the low-temperature performance of the coal liquefaction residue-modified asphalt. In 2021, Lv *et al.* [20] studied the composite-modified asphalt of fish scale powder and KH-560 SCA and found that the addition of coupling agents could increase the storage modulus and loss modulus of asphalt binders. It was confirmed that there was a chemical reaction between fish scale powder, SCA, and asphalt. In 2022, Hao *et al.* studied the effect of SCA on a new thermosetting polyurethane asphalt binder (PUAB). The study showed that SCA can improve the thermal and storage stability of PUAB [21]. Numerous road researchers have attempted to use coupling agents to link different road materials, thereby improving the physical and chemical properties and contact effects of asphalt, aggregates, and modifiers, and thus improving the high- and low-temperature properties and water stability of asphalt mixtures.

In order to scientifically determine the optimal modification formula and preparation process of composite-modified asphalt, it is necessary to study the comprehensive effects of multi-level and multiple factors on multi-objective and use response surface methodology (RSM) to model and analyze the problems under the influence of multiple factors by combining mathematical and statistical methods. RSM is an experimental design methodology that combines mathematical methods with statistical analysis. It is commonly used to optimize experimental processes, predict experimental results, and quantitatively describe the functional relationship between multiple independent variables and response variables. Due to its small experimental size and ability to accurately reflect significant effects of interaction between multiple independent variables, RSM has been increasingly introduced into road engineering experimental design. Tang *et al.* used RSM central composite design (CCD) to study the effect of areca fiber content, shear time, and oil-to-stone ratio on the water stability of areca fiber-modified asphalt mixture [22]. Gong *et al.* used the Box–Behnken design method in RSM to conduct a three-factor three-level design, studying the response variables of Marshall stability, flow value, VFA, VMA, and other multiple indicators of the mixture, and verified the optimal nano-TiO₂/CaCO₃, basalt fiber composite content, and optimal oil-to-stone ratio through experiments, with maximum and minimum errors of 6.96 and 0.096%, respectively [23]. Li *et al.* used RSM based on CCD to conduct experiments on the formed Marshall specimens, studying

and analyzing the effects of asphalt content and TPCB content on the volumetric parameters, strength, and high-temperature road performance of the asphalt mixture [24]. Bala *et al.* used CCD to analyze the interaction effects of two independent variable factors, nanosilica and binder content, on nanocomposite strength, volumetric, and performance properties and developed a model to fit experimental results to predict the response [25]. Adnan *et al.* used RSM to study the influence of graphite oxide (GO) and asphalt content on the volume and strength characteristics of asphalt mixture and the interaction effect of temperature, GO content, and loading frequency on the rutting performance of GO-modified asphalt and established a prediction mathematical model [26,27]. Yuan *et al.* adopted a comparison matrix method to evaluate the comprehensive performance of NAM, BAM, RAM, and RABAM [28]. The results of the above research demonstrate that RSM can be used to optimize the content of road materials and process parameters with good prediction accuracy. In this study, the RSM is introduced into the optimization design of composite modified asphalt, which can comprehensively investigate the interaction between different modifier dosages and preparation processes and balance its multiple evaluation indicators to prepare the composite modified asphalt with the best comprehensive performance.

Since Professor Deng proposed the gray correlation model, it has gradually become one of the important theoretical methods for studying uncertain systems. The basic idea of gray correlation analysis theory is based on the geometric similarity of behavioral factor sequences. It converts the discrete points of observation sequences into continuous piecewise lines and constructs a gray correlation model by extracting the geometric characteristic differences of the lines [29]. Gray correlation degree comprehensive evaluation is one of the important branches of gray system theory. It is an analytical evaluation method proposed to measure the degree of influence between factors or the contribution of factors to the main behavior. Compared with traditional mathematical and statistical analysis methods, it is better at dealing with data with the characteristics of “limited effective data and incomplete information.” In 2015, Tian used the gray correlation analysis evaluation method to obtain the optimal preparation parameters for preparing carbon black-modified asphalt [30]. In 2021, Zhou found the relationship between the length of glass fiber and the enhancement effect on the road performance of asphalt mixture through a comprehensive gray correlation evaluation analysis and recommended the optimal length of glass fiber for application in the mixture [31]. The gray correlation degree comprehensive evaluation method can assign values to various

evaluation indicators of the evaluation object and thus make a comprehensive judgment on the evaluation object under the joint action of multiple independent variable factors.

In summary, the incorporation of TPCB into asphalt has been found to be effective in improving its high-temperature shear deformation resistance and aging resistance. However, TPCB does not improve the low-temperature performance and resistance to water damage of asphalt and may even reduce them. To address this issue, the use of SCA has been proposed to improve the dispersibility and adhesion of TPCB in asphalt. This is expected to prevent TPCB agglomeration and low-temperature cracking in asphalt and enhance the adhesion between asphalt and stone, effectively improving the low-temperature performance and water stability of asphalt and asphalt mixtures. However, research studies on TPCB and SCA as composite modifiers for asphalt are relatively rare. Therefore, this article aims to investigate the composite modification of asphalt using TPCB and SCA. It is expected that TPCB-modified asphalt will have better high-temperature performance and anti-aging performance, while its low-temperature performance and storage stability will also be improved. The independent variables are TPCB and SCA content, and the key process parameter for preparing composite-modified asphalt is shear time. Other process parameters, such as temperature, shear rate, stirring time, and static development time, will be set according to Guo *et al.* [16]. The evaluation of composite asphalt's performance is crucial in assessing its quality. To improve the high and low-temperature performance of asphalt while considering other properties, various performance evaluation indicators such as penetration, softening point, 10°C ductility, Brookfield viscosity, and storage stability are employed. In this study, the gray correlation comprehensive evaluation method is introduced to the response surface design to evaluate the five performance parameters for each group of design experiments. The gray comprehensive evaluation value is calculated based on this method, and an improved RSM is developed. A quadratic regression model is established using the independent variables of TPCB, SCA content, and shear time, and the gray comprehensive evaluation value. This model helps to determine the optimal TPCB content, SCA content, and shear time for composite modified asphalt, leading to the best comprehensive performance and maximum gray comprehensive evaluation value. Ultimately, this study aims to identify the optimal preparation parameters for TPCB/SCA composite asphalt to improve its comprehensive performance and enhance the pavement performance of the TPCB-modified asphalt mixture.

Table 1: Technical indicators of matrix asphalt

Detect items	Unit	Technical requirement	Test result	Test method
Penetration (25°C, 100 g, 5 s)	0.1 mm	60–80	67	T0604-2011
Softening point	°C	≥46	48.4	T0606-2011
Ductility (15°C)	cm	≥100	> 100	T0605-2011
Dynamic viscosity (60°C)	Pa·s	≥180	215.2	T0620-2000
RTFOT				
Mass change	%	±0.8	+0.4	T0610-2011
Penetration ratio	%	≥61	69.8	T0604-2011
Residual ductility	cm	≥15	18.5	T0605-2011

2 Experimental materials and methods

2.1 Asphalt

The matrix asphalt used in this study is Grade A 70# road petroleum asphalt. The performance indicators of the asphalt were tested according to the methods outlined in “Test Methods of Bitumen and Bituminous Mixtures for Highway Engineering JTG E20-2011.” The test results are shown in Table 1.

2.2 TPCB from waste tire pyrolysis

Domestically produced TPCB was selected for this study. TPCB is a mixture of various industrial carbon blacks with varying sizes and aggregates. The particle size distribution of the TPCB used in the experiment is shown in Figure 1 [32].

As shown in Figure 1, the particle size of TPCB mainly ranges from 0 to 10,000 nm in diameter. The D10, D50, and

D90 of TPCB are 197, 265, and 358 nm, respectively, with an average particle size of 307.3 nm. TPCB was tested according to the specifications outlined in “Additives for Asphalt Mixtures – Part 7: Waste Tire Pyrolysis Carbon Black JT/T 860.7-2017,” and the results are shown in Table 2, which meets the technical requirements of the standard.

2.3 SCA

The SCA used in this study is KH550, with a chemical name of γ -amino propyl trimethoxy silane. It can be used in the surface treatment of inorganic fillers such as carbon black, silica, and clay to enhance the wettability and dispersibility of the fillers in the polymer. The main physical and chemical properties of KH550 are shown in Table 3.

2.4 Improved RSM

To establish a quadratic response surface equation for composite-modified asphalt, a sequential methodology

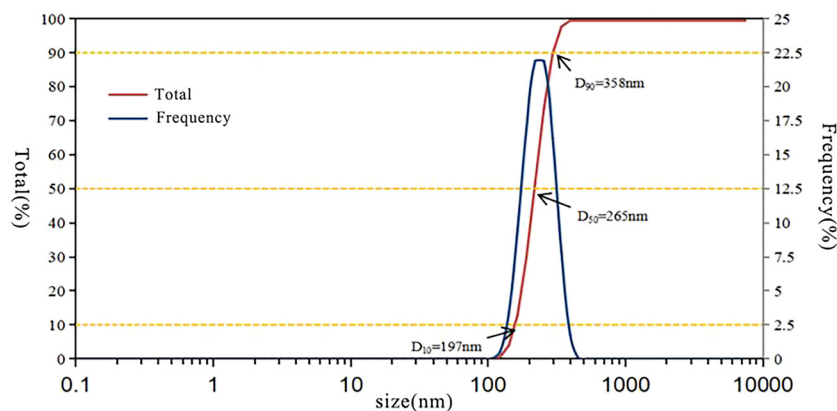
**Figure 1:** TPCB particle size distribution diagram.

Table 2: Technical indicators of TPCB

Detect items	Unit	Technical requirement	Test result	Test method
Appearance	—	Black powder	Black powder	Visual inspection
Ash content	%	≤18.5	15.02	JTG E20/T 0614
Iodine absorption	mg·g ⁻¹	≥80	85.1	GB/T 3780.1
Water content	%	≤2.0	1.5	JTG E20/T 0103
Oil absorption	10 ⁻² mL·g ⁻¹	≥70	87.1	GB/T 3780.2
Hydrogen ion concentration	—	≥6	7.68	GB/T 3780.7
Granularity (the rate of passed 0.045 mm)	%	100	100	JTG E42/T 0302

Table 3: Physicochemical properties of SCA

Items	Physical and chemical properties
Chemical formula	NH ₂ (CH ₂) ₃ Si (OCH ₂ CH ₃) ₃
Relative molecular mass	221.4
Appearance	Colorless, transparent liquid
Refractive index	1.421
Flash point (°C)	96
25°C Density (g·cm ⁻³)	0.946
Boiling point (°C)	217
Solubility	It can be miscible with organic solvents such as ethanol, ether, toluene, and xylene, react with CCl ₄ , and hydrolyze into silicanols in the presence of water, presenting an alkaline state
Matters needing attention	Strictly seal and store in a dry, cool, and dark room

was employed in the response surface design. Each factor that affects the response variable was analyzed separately, followed by multi-factor and multi-level experiments conducted using the CCD method. The optimal solution was found using mathematical methods. By using this method, the optimal values of different asphalt performance evaluation indicators can be predicted under different influencing factors. However, the optimal solution for composite-modified asphalt varies for different evaluation indicators. Therefore, it is crucial to assign weights to multiple evaluation indicators based on different application conditions to determine the total evaluation value characterizing the comprehensive performance of the composite-modified asphalt.

To achieve this, the gray correlation analysis comprehensive evaluation method was introduced into the response surface design. This method assigns an evaluation value to each evaluation indicator of the evaluated object based on the given application conditions, enabling a comprehensive evaluation of the evaluated object under the interaction of multiple factors. In this study, qualitative and quantitative analyses were combined to calculate the total gray comprehensive evaluation value for the comprehensive evaluation of the

researched object. An improved response surface design method was developed by establishing a quadratic response surface equation between the total gray comprehensive evaluation value and each influencing factor to determine the optimal content of each modifier and shearing time for composite-modified asphalt with the best comprehensive performance. The main calculation steps of the gray correlation comprehensive evaluation method are as follows:

(1) Determine the reference sequence X_0 and the comparison sequence X_i , and perform non-dimensional processing.

Reference sequence (also known as the parent sequence):

$$X_0 = \{X_0(1), X_0(2), \dots, X_0(k)\}.$$

Comparison sequence (also known as the sub-sequence):

$$X_i = \{X_i(1), X_i(2), \dots, X_i(k)\}, \text{ where } i = 0, 1, 2, \dots, m; \\ k = 1, 2, \dots, n.$$

(2) Calculate the correlation coefficient $\zeta_i(k)$

$$\zeta_i(k) = \frac{\min_i \min_k |x_0(k) - x_i(k)| + \rho \max_i \max_k |x_0(k) - x_i(k)|}{|x_0(k) - x_i(k)| + \rho \max_i \max_k |x_0(k) - x_i(k)|}, \quad (1)$$

where $|x_0(k) - x_i(k)|$ is the absolute difference between x_0 sequence and x_i sequence at point k :

$\min_i \min_k |x_0(k) - x_i(k)|$ is the minimum difference between the two levels.

$\max_i \max_k |x_0(k) - x_i(k)|$ is the maximum difference between the two levels.

Let $\Delta_i(k) = |x_0(k) - x_i(k)|$, where $i = 0, 1, 2, \dots, m$; $k = 1, 2, \dots, n$, then

$$\zeta_i(k) = \frac{\min_i \min_k \Delta_i(k) + \rho \max_i \max_k \Delta_i(k)}{\Delta_i(k) + \rho \max_i \max_k \Delta_i(k)}. \quad (2)$$

$\rho \in (0, \infty)$ is called the resolution coefficient. The smaller the ρ , the higher the resolution. The general ρ value range is (0,1), and the actual value can vary depending on the situation. When $\rho \leq 0.5463$, the resolution is best, and ρ is usually taken as 0.5.

(3) Calculate the correlation degree R_i

Since the correlation coefficient is a measure of the degree of association between the comparison sequence and the reference sequence at each preparation parameter condition (*i.e.*, each point on the curve), it has multiple values, and the information is too dispersed to facilitate overall comparison. Therefore, it is necessary to consolidate the correlation coefficients of each preparation parameter condition (*i.e.*, each point on the curve) into one value, namely, the average value, as a quantitative representation of the degree of correlation between the comparison sequence and the reference sequence, called the correlation degree R_i . The correlation degree R_i is calculated according to formula (3):

$$R_i = \frac{1}{n} \sum_{k=1}^n \zeta_i(k), \quad (3)$$

$i = 1, 2, \dots, m$; $k = 1, 2, \dots, n$,

where R_i is the correlation degree of the i th group of experiments, $\zeta_i(k)$ is the correlation coefficient; k is the evaluation index, n is the number of experimental groups, and m is the number of evaluation indexes.

(4) Calculate the proportion P_i and the gray comprehensive evaluation value $GC(k)$ for each evaluation index.

$$P_i = R_i / \sum_{i=1}^m R_i, \quad (4)$$

where P_i is the proportion of evaluation index, $i = 0, 1, 2, \dots, m$

$$GC(k) = \sum_{i=1}^m P_i \zeta_i(k), \quad (5)$$

where $GC(k)$ is the gray comprehensive evaluation value, $i = 1, 2, \dots, m$; $k = 1, 2, \dots, n$.

2.5 Preparation process of the composite-modified asphalt

To ensure the quality of the composite-modified asphalt, it is necessary to comprehensively consider the characteristics of each modifier and determine the appropriate feeding sequence and processing conditions. The surface pre-treatment method can make SCA fully contact with inorganic materials, resulting in better results. Based on relevant research [16], the following preparation process steps are determined:

- 1) *Hydrolysis*. Before surface modification, a certain proportion of water and ethanol solution is added, and the pH of the solution is adjusted to 4–6 with a small amount of cold acetic acid. Then, SCA is added to the prepared hydrolysis solution and mixed for 30 min to achieve sufficient hydrolysis. The hydrolyzed SCA solution is then immersed in the surface of TPCB and stirred evenly in a constant temperature water bath at 65°C. The hydroxyl groups formed by the hydrolysis reaction of the silanol groups first form hydrogen bonds on the surface of TPCB and then form a coating film on the surface of TPCB through a dehydration reaction. The degree of hydrolysis of SCA affects the surface modification of TPCB. According to relevant research [15], the ratio of SCA to water and ethanol is 5:45:50.
- 2) *Curing*. The surface of TPCB treated with SCA solution also needs to undergo a curing reaction, *i.e.*, thermal drying. After the curing reaction, some hydrogen bonds form covalent bonds due to dehydration, and SCA will be in a solid or fixed state on the surface of TPCB. When TPCB is coated on the substrate, a chemical reaction occurs between SCA and TPCB, making their bonding tighter, thus improving the adhesion performance of the coating. The curing time and curing temperature affect the bonding strength. The purpose of thermal drying is mainly to remove water and ethanol solution during the hydrolysis process. According to the actual preheating requirements, the curing temperature is selected as 105°C and the curing time is 8 h to prepare SCA surface-modified TPCB.
- 3) *Shear mixing*. After the curing process, TPCB treated with SCA should be shear mixed with the matrix asphalt to create TPCB/SCA composite-modified asphalt. First, preheat the matrix asphalt to 150°C in an oven, then add the SCA-treated TPCB according to the desired mixing ratio. Stir the mixture by hand for 2 min and then shear it using an asphalt shear apparatus at a speed of 4,000 rpm for a shearing time of 30–90 min. After shearing, let the asphalt stand at 150°C for 30 min to develop, and the TPCB/SCA

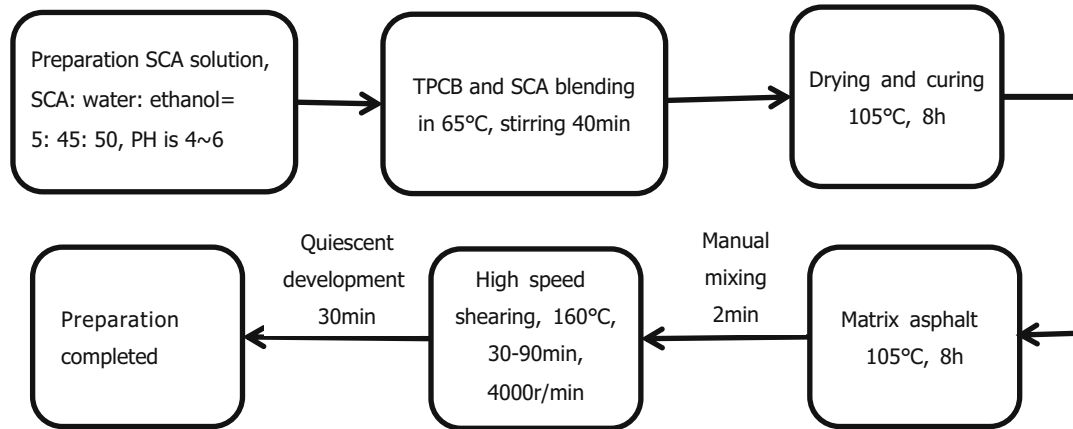


Figure 2: Preparation process of the TPCB/SCA composite-modified asphalt.

composite modified asphalt is obtained. Figure 2 shows the process flow for preparing TPCB/SCA composite-modified asphalt.

2.6 Experimental design and results

Based on the relevant literature [8,11,12,24], this study proposed comprehensive evaluation indices and expected targets for composite-modified asphalt. Penetration, softening point, 10°C ductility, Brookfield viscosity, and storage stability were used as evaluation indices for analysis. The penetration was used as the consistency index, as it reflects the consistency and grading of the asphalt. The expected penetration value should be moderate to ensure proper construction and ductility performance. The softening point was used as the high-temperature index, as it has a relatively small parallel error, and the experimental steps are relatively simple. A higher softening point indicates better high-temperature performance of the asphalt. The 10°C ductility test was used as the low-temperature index, as it measures the ductility of the asphalt at low temperatures and reflects its low-temperature performance. A higher ductility value indicates better low-temperature ductility. The 135°C Brookfield viscosity was used as the construction and workability index, as it characterizes the construction and workability of asphalt. The

storage stability index, softening point difference, was used to measure the degree of separation of modified asphalt under heat and the quality stability of modified asphalt in actual use.

According to previous studies [4,7,8,12,14,19,24,30], the optimal TPCB content is 15%, and the shear time is 60 min. When the TPCB content is less than 10%, it has excellent storage stability, but when it is greater than 15%, its low-temperature performance will be affected. In order to ensure the rigor of this experiment and test the performance of composite modified asphalt, the TPCB content of 0–20% and the shear time of 30–90 min were selected. According to Lv *et al.*'s research and previous experiments, the optimal dosage of SCA for modification is 1% [20,33]. To meet the requirements of this experiment, a dosage of

Table 4: Corresponding values of each factor code

Factor code	−1.682	−1	0	1	1.682
TPCB content (%)	0	4.1	10	15.9	20
SCA content (%)	1	1.4	2	2.6	3
Shearing time (min ^{−1})	30	42	60	78	90

Table 5: Results of the segregation test

Number	Lower softening point	Upper softening point	Softening point difference
0	54	53.7	0.3
1	46	45.3	0.7
2	48.4	45.5	2.9
3	52	50.8	1.2
4	48.6	46.2	2.4
5	52.5	51.9	0.6
6	49	46.2	2.8
7	54	53.5	0.5
8	49.2	47.2	2
9	46.3	46	0.3
10	53.5	50.2	3.3
11	53.9	51.5	2.4
12	50.8	49.7	1.1
13	51	49.6	1.4
14	50.2	48.9	1.3
15–20	51.6 ± 0.5	50.5 ± 0.5	1.1 ± 0.5

Table 6: Response surface test and data analysis results

Serial number	A	B	C	25°C Penetration/0.1 mm		Softening point (°C)		10°C Ductility (cm)		135°C Brookfield rotational viscosity (Pa-s)		Storage stability (°C)		Gray comprehensive evaluation value (%)
				Measured value	Correlation coefficient ζ_i	Measured value	Correlation coefficient ζ_i	Measured value	Correlation coefficient ζ_i	Measured value	Correlation coefficient ζ_i	Measured value	Correlation coefficient ζ_i	
0	Reference performance			57.3	1	53.7	1	23.4	1	0.219	1	0.3	1	100
1	-1	-1	-1	60.8	0.78	46.1	0.46	22.3	0.73	0.329	0.60	0.7	0.47	62.55
2	1	-1	-1	54.9	0.70	48.4	0.32	13.8	0.39	0.446	0.50	2.9	0.36	47.03
3	-1	1	-1	52.1	0.52	52.4	0.69	23.4	1.00	0.332	0.59	1.2	0.40	65.28
4	1	1	-1	55.0	0.71	48.5	0.41	15.9	0.41	0.45	0.49	2.4	0.36	49.43
5	-1	-1	1	59.6	0.71	52.7	0.62	21	0.67	0.321	0.61	0.6	0.50	63.38
6	1	-1	1	54.5	0.67	48.9	0.42	13.6	0.34	0.438	0.50	2.8	0.36	47.13
7	-1	1	1	60.2	0.66	53.3	0.66	23.2	0.87	0.368	0.55	0.5	0.56	66.92
8	1	1	1	54.3	0.66	49	0.37	15.7	0.52	0.432	0.50	2	0.37	49.85
9	-1.682	0	0	68.7	0.33	46.1	0.62	23.2	0.87	0.219	1.00	0.3	1.00	63.00
10	1.682	0	0	53.9	0.63	53.5	0.68	12.1	0.31	0.428	0.51	3.3	0.35	50.36
11	0	-1.682	0	56.4	0.86	53.7	1.00	19.8	0.42	0.399	0.53	2.4	0.36	65.16
12	0	1.682	0	57.1	0.97	50.7	0.42	23.4	1.00	0.411	0.52	1.1	0.41	69.76
13	0	0	-1.682	58.4	0.84	50.9	0.38	22.1	0.76	0.413	0.52	1.4	0.39	60.43
14	0	0	1.682	56.9	0.93	50.4	0.29	20.8	0.69	0.423	0.51	1.3	0.39	59.47
15-20	0	0	0	56.7 ± 1.8	0.90	51.4 ± 0.6	0.95	21.4 ± 1.6	0.92	0.402 ± 0.012	0.52	1.1 ± 0.5	0.41	76.76
Relevance				0.73		0.55		0.63		0.56		0.45		—
Proportion of evaluation indicators				0.25		0.19		0.22		0.19		0.15		—

Note: A – TPCB content; B – SCA content; and C – shearing time.

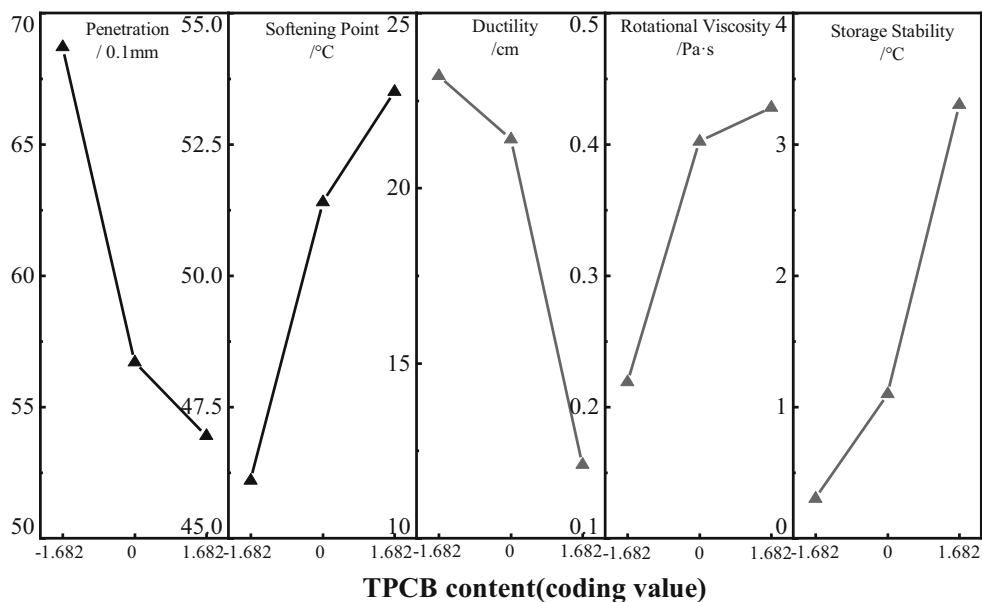
Table 7: Dynamic shear rheological test results

Type	Rutting factor (kPa)							Critical temperature (°C)
	54°C	58°C	62°C	66°C	70°C	74°C	78°C	
BA-70	6.25	4.37	3.28	2.43	1.19	0.86	0.44	71.2
TMA	24.78	14.58	7.82	4.11	2.27	1.16	0.56	75.1
TSMA	27.02	16.36	9.04	4.84	2.32	1.19	0.57	75.4

1–3% was selected for SCA. To comprehensively consider the interaction between different influencing factors and effectively estimate the primary and secondary terms, CCD was adopted in this study as a 3-factor, 5-level experimental plan. The corresponding values of each factor code are shown in Table 4. In order to examine the comprehensive performance of the test experiments, the optimal value of the evaluation index was assumed to be the “ideal performance” as the reference performance X_0 , and the various performance indicators of each test group constituted a comparative sequence X_{ij} . The gray system analysis method was used to calculate the gray comprehensive evaluation value (GC) of each group of tests and rank the comprehensive characteristics of each group of experiments according to the size of the GC. The preparation parameters corresponding to the highest gray comprehensive evaluation value are optimal. Excel 2016 was used for data processing. The results of the segregation test are shown in Table 5. The experimental results, the correlation coefficient of the experimental group, and the gray comprehensive evaluation values are shown in Table 6.

In order to test the rheological properties of TPCB/SCA composite-modified asphalt, this study used dynamic shear rheological tests to compare 70# matrix asphalt (BA-70), TPCB-modified asphalt (TMA), and their short-term thermal oxidative aging samples (TSMA), and verify the rheological properties and applicability of TPCB/SCA composite-modified asphalt. The test results are shown in Table 7.

According to Table 7, as the temperature increases, the $G^*/\sin \delta$ of BA-70, TMA, and TSMA gradually decrease. Asphalt is a temperature-sensitive material, so its ability to resist permanent deformation will decrease with increasing temperature. The critical temperatures of BA-70, TMA, and TSMA asphalt are 71.2, 75.1, and 75.4°C. The addition of TPCB significantly increased the rutting factor of asphalt, resulting in a critical temperature increase of 3.9 and 4.2°C for TMA and TSMA compared to BA-70. With the continuous increase of temperature, the difference in high-temperature performance of these three types of asphalt gradually narrows. At 58°C, their $G^*/\sin \delta$ difference is more obvious, and after 70°C, the difference between the three types of asphalt is no longer significant. Therefore, the addition of pyrolysis carbon black

**Figure 3:** Effect of TPCB content on asphalt performance.

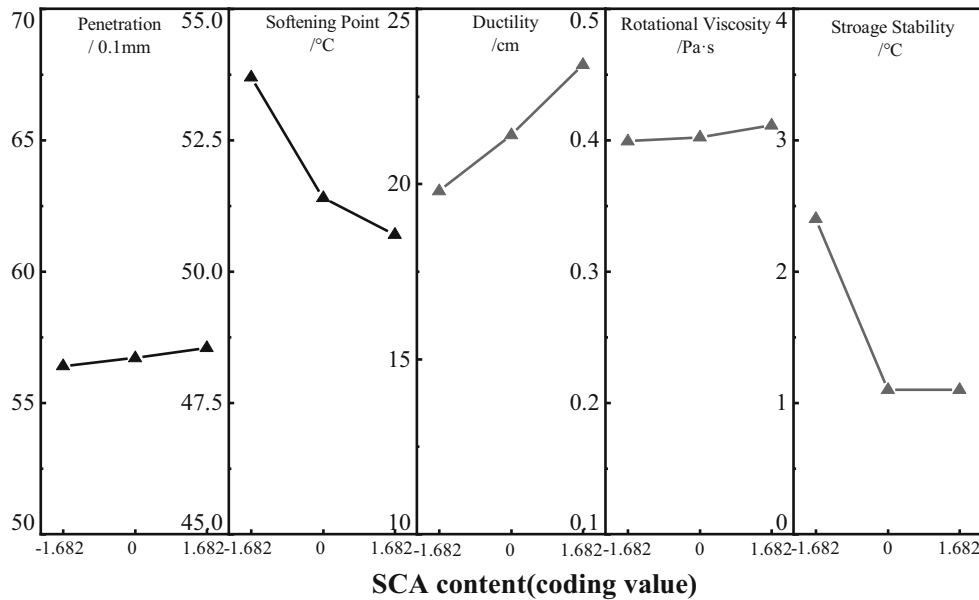


Figure 4: Effect of SCA content on asphalt performance.

from waste tires can effectively improve the rheological properties of asphalt.

3 Data processing and analysis

3.1 Analysis of single-factor effects

By analyzing the experimental results of eight test points (namely, 9 (−1.682, 0, 0), 15–20 (0, 0, 0), and 10 (1.682, 0, 0)), we can obtain the variation pattern of asphalt performance with TPCB content. To simplify the analysis, the experimental results of points 15–20 are averaged for calculation. Figure 3 shows that as the TPCB content increases from code −1.682 to code 0, the penetration value of asphalt decreases from 68.7 to 56.7. If the asphalt is categorized by grade number, the modified asphalt has changed from 70# to 50#. The decreasing trend of penetration value slows down as the TPCB content continues to increase. The changes in softening point and rotational viscosity are strongly correlated with the changes in the penetration value. When the TPCB content increases from code −1.682 to code 0, the softening point and viscosity increase significantly by 11.5 and 83.6%, respectively. However, as the TPCB content continues to increase, the increments of softening point and viscosity decrease to 4.1 and 6.5%, respectively.

At a low TPCB content level, its influence on the ductility and storage stability of asphalt is not significant, but at

a higher level, the ductility of asphalt decreases significantly. At the same time, the storage stability of asphalt decreases significantly, and the difference in softening point reaches 3.3°C, which exceeds the maximum value of 2.5°C specified in the “Technical Specifications for Construction of Highway Asphalt Pavements.” TPCB has a larger specific surface area and a developed pore structure, which leads to a certain adsorption effect on the light components in asphalt, increasing the viscosity, consistency, and stiffness of asphalt. The reduction of light components also weakens the extensibility of asphalt and reduces its ductility. Moreover, excessive TPCB will cause agglomeration in asphalt, making it prone to settling and segregation. Therefore, it is essential to optimize the TPCB content to minimize its negative effects on the viscosity, ductility, and storage stability of asphalt.

Similarly, by analyzing the results of the eight test points, namely 11 (0, −1.682, 0), 15–20 (0, 0, 0), and 12 (0, 1.682, 0), under the condition of other factors being constant, the influence curve of SCA content on asphalt performance can be obtained, as shown in Figure 4. It can be seen that compared with the TPCB content, the influence of SCA content on asphalt performance is significantly weaker. Changes in the SCA content have almost no effect on asphalt penetration and rotational viscosity. When SCA content increases from code −1.682 to code 1.682, the softening point of asphalt decreases by 3°C, and the elongation increases by 3.6 cm. This indicates that SCA has a certain improvement effect on the low-temperature performance of asphalt. This can be attributed to the lower molecular weight of SCA, which has a certain regulatory effect on the components

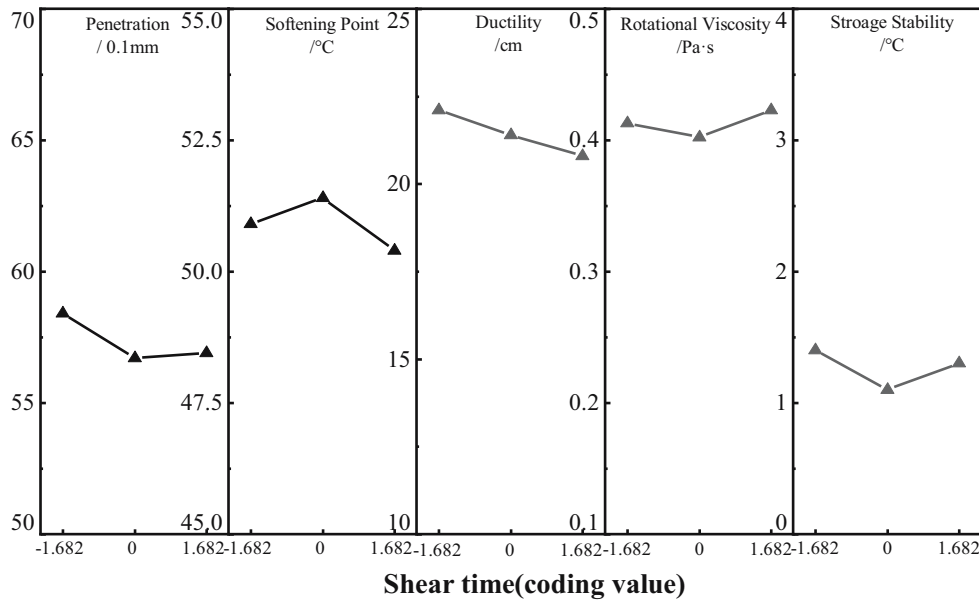


Figure 5: Effect of shearing time on asphalt performance.

of asphalt. However, compared with the enhancement effect of TPCB, this effect is very limited. In addition, as SCA content increases from code -1.682 to code 0 , the softening point difference of asphalt decreases from 2.4 to 1.1°C , with a decrease of 54.2% . This indicates that SCA plays a good bridging role between TPCB and asphalt and can significantly improve the compatibility of TPCB-modified asphalt. However, when the SCA content continues to increase to code 1.682 , the softening point difference of asphalt does not continue to decrease but remains at 1.1°C . This indicates that

there is an optimal SCA content to fully ensure the compatibility between TPCB and asphalt. To avoid wasting resources, the optimal SCA content should be further determined through a predictive equation.

By analyzing the results of eight testing points, namely 13 $(0, 0, -1.682)$, 15–20 $(0, 0, 0)$, and 14 $(0, 0, 1.682)$, under other constant conditions, the curve of the influence of shear time on the performance of asphalt can be obtained, as shown in Figure 5. Compared with the changes in TPCB and SCA contents, the influence of shear time on the performance of asphalt is less significant. As the shear time increases from the coding of -1.682 to 0 and finally to 1.682 , there is a turning point in the penetration and softening point of asphalt. This may be because asphalt undergoes

Table 8: Analysis of variance of the fitting equation

Source	Degree of freedom	Adj SS	Adj MS	F value	P value
Model	9	1032.78	114.753	5.04	0.045
Linear	3	567.81	189.268	8.31	0.022
A	1	540.88	540.881	23.75	0.005
B	1	26.79	26.786	1.18	0.328
C	1	0.14	0.138	0.01	0.941
Square	3	464.17	154.724	6.79	0.033
A × A	1	402.62	402.618	17.68	0.008
B × B	1	114.27	114.275	5.02	0.035
C × C	1	296.58	296.577	13.02	0.015
Two-factor interaction	3	0.8	0.267	0.01	0.998
A × B	1	0.17	0.165	0.01	0.935
A × C	1	0.48	0.475	0.02	0.891
B × C	1	0.16	0.160	0.01	0.937
Error	5	113.88	22.777	—	—
Total	14	—	—	—	—

Table 9: Quadratic polynomial coefficients and their significance analysis

Term	Coefficient	Coefficient standard deviation	T value	P value
Constant	78.969	0.874	90.4	0
A	1.905	0.238	8.01	0
B	0.828	0.238	3.48	0.008
C	0.319	0.238	1.34	0.238
A × A	-15.661	0.357	-43.85	0
B × B	-8.3	0.357	-23.24	0
C × C	-2.586	0.357	-7.24	0.001
A × B	2.097	0.311	6.75	0.001
A × C	0.087	0.311	0.28	0.789
B × C	-1.257	0.311	-4.05	0.010

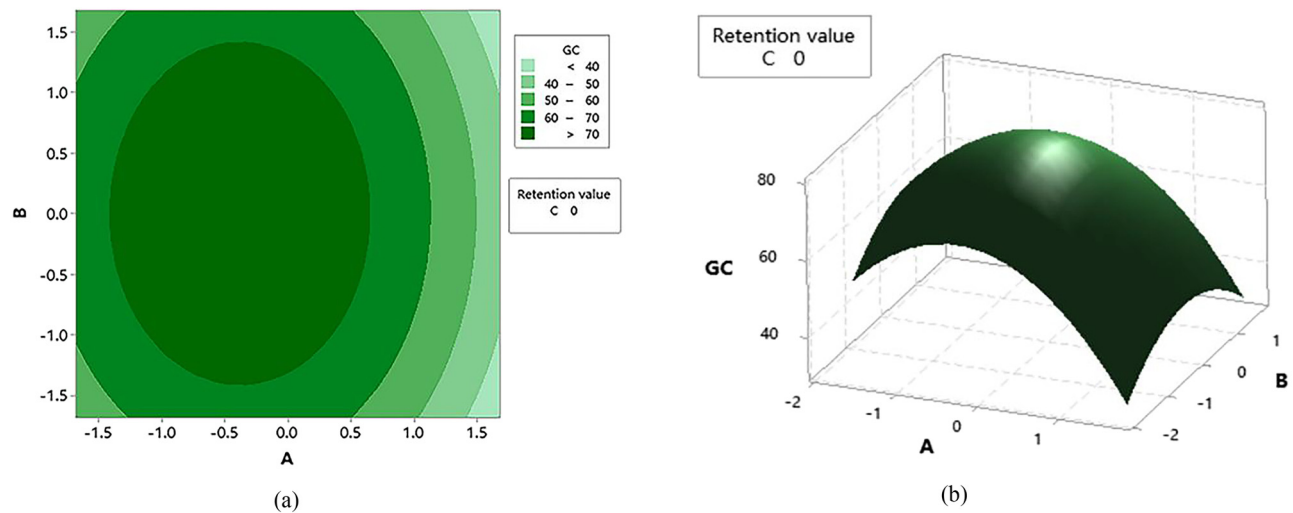


Figure 6: Contour and response surface diagrams of A and B. (a) Contour map and (b) response surface graph.

some aging during the long-term shearing process, resulting in increased viscosity and high-temperature performance. As the shear continues to deepen, the crosslinking degree of SCA and TPCB increases and the reinforcing effect of TPCB is partially neutralized by the plasticizing effect of SCA, resulting in the recovery of asphalt performance. However, this process does not significantly affect the ductility of asphalt. Dominated by the aging of asphalt, the ductility of asphalt continues to decrease slowly with the prolongation of shear time. The changes in rotational viscosity and storage stability are not significant, and their numerical fluctuations remain within the range of experimental errors. Therefore, it can be considered that shear time does not significantly affect the viscosity and storage performance of asphalt.

3.2 Interaction analysis

In order to quantitatively analyze the effects of TPCB content, SCA content, and shear time on the gray comprehensive evaluation value and to determine the optimal ratio, a quadratic polynomial equation was fitted to the experimental data using MINITAB19 software. A quadratic function of A, B, and C was established to describe the relationship between the gray comprehensive evaluation value GC and A, B, and C. The regression equation is shown in Eq. (6):

$$\begin{aligned} GC = & 77.52 - 6.29A + 1.40B + 0.10C - 8.15A \times A \\ & - 4.34B \times B - 7.00C \times C - 0.14A \times B \\ & - 0.24A \times C + 0.14B \times C. \end{aligned} \quad (6)$$

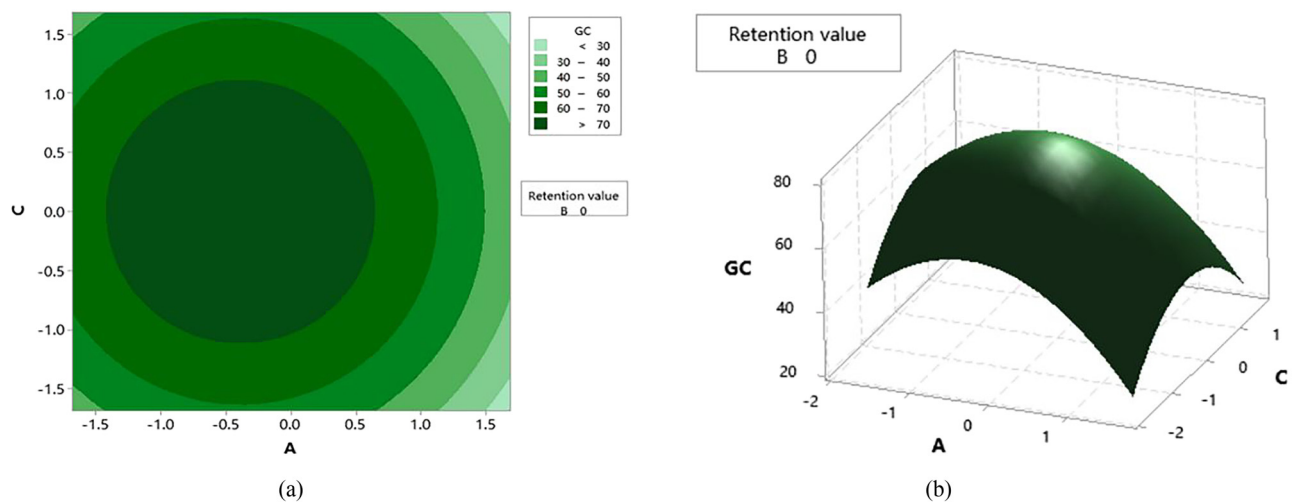


Figure 7: Contour and response surface diagrams of A and C. (a) Contour map and (b) response surface graph.

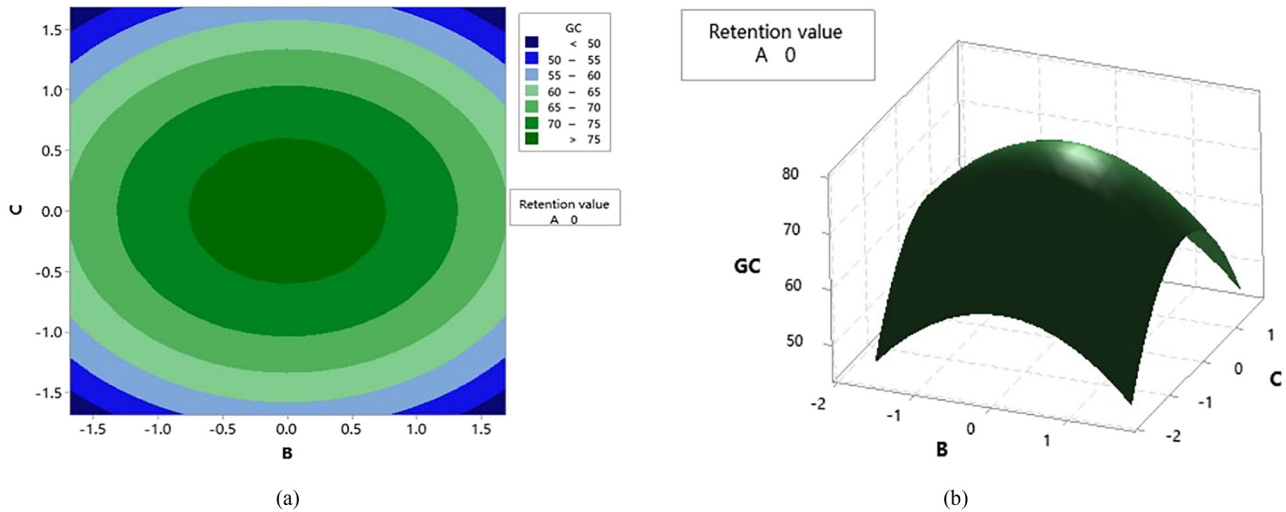


Figure 8: Contour and response surface diagrams of B and C . (a) Contour map and (b) response surface graph.

Checking the reliability of the model is an important part of data analysis. If the fit is insufficient, the model function will give incorrect responses. In order to evaluate the significance of the model, a variance test analysis was performed, and the results are listed in Table 8.

According to the variance analysis data in Table 8, the F -value of the fitted variance model is 5.04. After removing five items of B , C , $A \times B$, $A \times C$, and $B \times C$ from the model, all other items have $P \leq 0.05$. According to the relevant research results [34,35], the smaller the P value, the better the fitting effect and the more credible the regression equation. Therefore, the fitting degree of the model in this article is high, and it can accurately reflect the relationship between the gray comprehensive evaluation value and the various influencing factors. On the basis of the overall significance test of this model, the significance test was conducted on each sub coefficient of the model to analyze the effects of three influencing factors, as shown in Table 9.

It can be seen from Table 9 that the coefficients of some primary, secondary, and interaction terms of the three influencing factors A , B , and C are all greater than 1, which indicates that there is a strong interaction between the influencing factors, and also verifies the applicability of the improved RSM for optimizing the preparation of modified asphalt. To more intuitively analyze the degree of interaction between various factors and the influence on the performance of asphalt, a two-dimensional contour plot and a three-dimensional response surface plot of the factors A , B , and C , and the comprehensive evaluation value were generated based on the regression model (6) established according to the experimental data in Table 6. The contour plot is a projection of the response surface plot on the horizontal plane for more intuitive observation and analysis of the relationship

between the comprehensive evaluation value and various factors, as shown in Figures 6–8.

Figure 6 shows the contour map and response surface map of the influence of A and B on the gray comprehensive evaluation value, which indicates the interactive effect of TPCB content and SCA content on the gray comprehensive evaluation value of asphalt when C is at the central level, i.e., the shearing time is 60 min. From the contour map formed by TPCB content and shearing time in Figure 6(a), the graph shows an elliptical shape, indicating that the interaction between TPCB content and shearing time is more significant than other factors. In Figure 6(b), when the encoding value of A is around -0.2 and the content of B is around 0.1 , the response variable can reach the maximum value. This indirectly proves that a small amount of SCA can undergo a branching reaction with TPCB, making TPCB stable and evenly distributed in the asphalt and promoting the modification effect of TPCB. However, excessive SCA has no significant effect on the asphalt and may even have a negative effect, leading to a decrease in the gray comprehensive evaluation value of the asphalt.

Figure 7 shows the contour map and response surface map of the influence of A and C on the comprehensive evaluation value. From the contour map in Figure 7(a), the shape of the graph is circular. Combined with Figure

Table 10: Indicators of model adequacy

Signal-to-noise ratio (SN)	R^2	R_A^2 (adjusted)	R_B^2 (predicted)
8.73	82.71%	87.65%	83.47%

7(b), it can be seen that irrespective of the value set for factor *A*, the response variable can reach a maximum value when factor *B* takes the encoding value of 0. Similarly, when factor *B* takes the encoding value of -0.4 , the response variable can reach a relative maximum value within the process, regardless of the specific value of factor *A*. This indicates that the interaction between *A* and *C* is not significant, and when *A* and *C*, respectively, take the optimal solutions under single-factor action, the response variable, *i.e.*, the optimal value in the global response process, is also obtained. The model does not have the characteristic of a quadratic bending response.

Figure 8 shows the interaction effect of SCA content and shearing time on the gray comprehensive evaluation value of asphalt when *A* is at the central level, *i.e.*, TPCB content is 10%. From the contour map in Figure 8(a), it can be seen that the interaction between *B* and *C* is relatively significant, and when the encoding value of *B* is -0.1 , and the encoding value of *C* is -0.1 , *i.e.*, SCA content is 1.9% and shearing time is 58.2 min, the comprehensive evaluation value of asphalt is the maximum. The main reason may be that when the shearing time is too short, TPCB/SCA is not uniformly mixed in the asphalt, affecting its modification effect; however, when the shearing time is too long, the asphalt will undergo thermal oxidation aging, leading to a decrease in its overall performance.

3.3 Determination of optimal parameters

In Table 7, a p -value < 0.05 indicates the significance of the model and parameters in evaluating the response variable. However, some unimportant interactions were analyzed for each formula model with variance (p -value > 0.01). From the response surface plots of the interaction terms, it can also be seen that the interaction effects of the factors achieve the optimal value when the values of each factor are at their optimal level during the single-factor variation process. Therefore, the interaction terms in the model are not significant enough to predict the accuracy of the model. To improve the model and optimize the results, these unimportant terms can be eliminated from the model. The optimized model is shown as follows:

$$GC = 77.52 - 10.58A - 23.07A \times A - 12.29B \times B - 19.80C \times C. \quad (7)$$

Eq. (7) shows that there is quadratic bending in the gray comprehensive evaluation value with the three influencing factors *A*, *B*, and *C*. According to the variance analysis data in Table 7, the F -value of the fitted variance

Table 11: Test data for optimal content and process

Optimal formula and process	25°C Penetration/0.1 mm	Softening point (°C)	10°C ductility (cm)	135°C Brookfield viscosity (Pa·s)	Storage stability (°C)	Gray comprehensive evaluation value (%)
9.2% TPCB + 2.1% SCA, Cutting time 56.3 min	56.9	51.4	21.4	0.413	0.6	78.92
Test value range	52.1–68.7	46.1–53.7	12.1–23.4	0.219–0.450	0.3–2.9	—
Target expectations	Median	Bigger	Bigger	Smaller	Smaller	—

model is 5.04. After removing five items of B , C , $A \times B$, $A \times C$, and $B \times C$ from the model, all other items have $P \leq 0.05$. Therefore, the fitting degree of the model in this article is high, and it can accurately reflect the relationship between the gray comprehensive evaluation value and the various influencing factors. The adjusted R^2 , predicted R^2 , and signal-to-noise ratio (SN) after adjustment through variance analysis using MINITAB19 are shown in Table 10. The signal-to-noise ratio represents the ratio of signal to noise and is generally ideal when it is greater than 4. The fitness of the model is evaluated by the value of R^2 . Models with $R^2 > 0.8$ are considered to be well-fitting [36]. The higher correlation coefficients R^2 and adjusted R_A^2 indicate good consistency between the predicted values and actual values. The correlation coefficient R^2 is 82.71%, and the adjusted R_A^2 (adjusted) is 87.65%, indicating that 82.71% of the reliability of the comprehensive evaluation value can be explained by the three influencing factors A , B , and C . This suggests that the comprehensive asphalt performance prediction method established in this article can describe its variation law well with the three influencing factors, and the quadratic equation is suitable for this regression model. The R_B^2 (prediction) is used to judge the predictive ability of the regression model. R_B^2 (prediction) is 83.47%, which is close to R^2 and greater than 0.8, indicating that the prediction of this model is highly reliable.

Based on the analysis of response surface experimental data and variance significance, it is known that the established mathematical model has a high degree of fitting, and on this basis, predictive research has been carried out, which can effectively approach the actual situation. The response value optimization was carried out using MINITAB19 software, and the expected value of the gray comprehensive evaluation value was set to the maximum value. The results showed that the optimal proportional code values of the pyrolysis carbon black content (A), SCA content (B), and shearing time (C) were -0.14, 0.19, and -0.21, respectively, and the corresponding data were 9.2%, 2.1%, and 56.3 min. The predicted comprehensive evaluation value was 77.14%. Then, TPCB/SCA composite-modified asphalt was prepared according to the optimal formula and preferred process for related tests to verify the optimization results and the accuracy of the response, and the experimental results are shown in Table 11.

Based on the test results of the modified asphalt prepared with the selected content and shear time, as shown in Table 11, the correlation coefficients and weightings of the evaluation indicators corresponding to the test values were calculated according to Table 6, and the comprehensive evaluation value was calculated as 78.92% using Formula (5). Compared with the predicted result, the error is 2.31%, indicating a close agreement between the predicted

and measured gray comprehensive evaluation values. Thus, the tires' pyrolysis carbon black can effectively improve the rheological properties of asphalt. From the test results, it can be seen that the TPCB/SCA composite-modified asphalt prepared using the improved RSM is the closest to the reference performance in Table 6, with superior high-temperature performance, qualified low-temperature ductility, good workability, and strong anti-segregation ability of the asphalt. Economic analysis needs to be carried out in the future for further promotion and application.

4 Conclusions

- 1) The present study explores the potential of waste TPCB as a reinforcing filler for asphalt modification. Additionally, SCA) has both inorganic and organic groups, which can improve the adhesion between asphalt and aggregates and achieve the characteristic of improving asphalt performance. A basic preparation process of composite-modified asphalt is proposed, and a novel TPCB/SCA composite-modified asphalt formulation is developed through systematic experimentation and analysis, ensuring the reliability of its preparation process.
- 2) To address the technical challenge of comprehensive evaluation involving multiple indexes, the gray correlation comprehensive evaluation method is incorporated into the RSM, leading to an improved RSM. By using this method, the comprehensive properties of asphalt, such as penetration, softening point, 10°C ductility, 135°C Brinell rotation viscosity, and storage stability were taken as response values to determine the optimal comprehensive performance indicators for composite-modified asphalt: the content of TPCB was 14.2%, the content of SCA was 2.1%, and the shear time was 56.3 min.
- 3) A highly accurate and reliable mathematical model is established for predicting and optimizing asphalt formulations. Performance verification tests reveal that the TPCB/SCA composite-modified asphalt exhibits significantly improved high-temperature shear deformation resistance, low-temperature cracking resistance, and storage stability, resulting in the highest gray comprehensive evaluation value among multiple indicators. This study sheds light on the potential of TPCB/SCA composite-modified asphalt for sustainable and high-performance road construction materials.

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