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Research Article

Laboratory Evaluation of Aging Behaviour of SBS Modified Asphalt

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To study the effect of aging SBS modified asphalt on the performance of asphalt pavement, aging at various times and temperatures was conducted with thin film oven, and then tests were made about the penetration, softening point, ductility, viscosity, toughness, and fluorescence microscopy of modified asphalt with different aging levels. The results show that, with the increasing of aging time, the penetration and ductility of modified asphalt decrease while its softening point and viscosity increase, and the variation trend of the toughness and tenacity is related to the aging temperature; the aging dynamic model with viscosity as parameter can well characterize the aging process of modified asphalt; at microlevel, with the decreasing of SBS particle size, the uniformity of particle size is better. Analysis of macroscopic properties, microscopic characteristics, and significance shows that the SBS particle area ratio has a significant correlation with tenacity as the aging temperature changes. When the aging temperature is 163°C, the SBS particle area ratio still has a significant correlation with tenacity as the aging time changes.

1. Introduction

SBS modified asphalt is widely used in asphalt pavement of high-grade highway in China, due to its excellent performance. But it will get aging inevitably after a period of time [1–3]. For SBS modified asphalt, its aging includes the aging of polymer SBS as well as the aging of the matrix asphalt itself [4, 5]. As waste pavement construction materials, aging SBS modified asphalt still has high recycling value. While designing the recycling of discard SBS modified asphalt mixture, the aging effect of SBS modified asphalt must be understood. Though many studies have been made on the aging of SBS modified asphalt [6, 7], the existing findings have not well explained the unique aging phenomenon of SBS modified asphalt. Also, the researches of evaluation and prediction of aging properties of SBS modified asphalt with different aging levels are relatively less [8]. As we all know, the toughness test is used to evaluate the modified properties of modified asphalt, mainly used for SBR modified asphalt. But there are few researches on SBS modified asphalt, especially for the aging properties.

SBS modified asphalt is a multiphase mixture system composed by polymer SBS and matrix asphalt. Its performance is not only affected by the matrix asphalt but also closely related to the state of SBS in asphalt [9–11]. Most researches on modified asphalt aging did not consider this factor, the multiphase characteristics, or the effect of changing polymer SBS particles on the modified asphalt performance in the aging process [12, 13].

With this consideration, this paper conducts an in-depth research on the aging effect of SBS modified asphalt. The aging process of SBS modified asphalt was simulated with thin film oven test (TFOT). The macroscopic properties and fluorescence microstructure characteristics of modified asphalt with different aging levels were conducted. By analysing variation rule of macroscopic properties and microscopic characteristics, this paper attempts to provide a further understanding about the aging mechanism of SBS modified asphalt. In conclusion, this research aims to provide a theoretical foundation for improving the service life of SBS modified asphalt pavement and recycling discard SBS modified asphalt mixture.

Table 1: Main technical indexes of matrix asphalt.

Indexes	Test results	Specification
Penetration (25°C)/0.1 mm	61.3	60~80
Softening point/°C	50.7	≥46
Ductility (15°C)/cm	>100	≥100
TFOT		
Quality change/%	−0.214	≤±0.8
Residual penetration ratio (25°C)/%	67.7	≥61
Residual ductility (10°C)/cm	11.1	≥6

Table 2: Main technical indexes of modified asphalt.

Indexes	Test results	Specification
Penetration (25°C)/0.1 mm	56	40~60
Softening point/°C	60.5	≥60
Ductility (5°C)/cm	29.5	≥20
Viscosity (135°C)/Pa·s	1.550	≤3
TFOT		
Quality change/%	−0.252	≤±1.0
Residual penetration ratio (25°C)/%	78.6	≥65
Residual ductility (5°C)/cm	17.5	≥15

2. Materials and Methods

2.1. Materials. SBS modified asphalt in test is made of TIPCO 70# asphalt. Its main technical indexes are shown in Table 1. The modifier is Yueyang Petrochemical linear SBS (YH-791), with content of 3.5% while the content of rubber oil and stabilizer is 3% and 1‰, respectively.

2.2. Sample Preparation. At first, matrix asphalt was heated to 165°C, and the rubber oil was added to the matrix asphalt according to preset content. Then the polymer SBS was added. The mixture was stirred uniformly at speed of 1100 r/min for 7 h at about 180°C. Then the stabilizer was added in batches. Stir continued for 2 h to obtain SBS modified asphalt sample. The main technical indexes are shown in Table 2.

Table 2 shows that the main technical indexes meet the technical requirements for SBS modified asphalt I-D by the current specification "Technical Specification for Construction of Highway Asphalt Pavement" in China.

2.3. Experimental Design. As modified asphalt production and construction temperature is generally controlled at 160°C~180°C, TFOT aging test temperature was set to 163°C, 173°C, and 183°C, and aging time to 3 h, 5 h, 7 h, and 10 h.

Macroscopic properties mainly focus on 25°C penetration, softening point, 5°C ductility, Brookfield viscosity, toughness, and tenacity, and the microscopic properties focus on fluorescence microscopic characteristic. The fluorescence test is mainly used for appearance analysis and quantitative analysis of modified asphalt.

3. Test Results and Analysis of Macroscopic Properties

3.1. Impact of Aging on the Three Major Indexes of Modified Asphalt. Figure 1 shows the variation of SBS modified asphalt penetration, softening point, and ductility with aging time under different temperature conditions.

Figure 1 shows that, with the increasing of aging time, the penetration and ductility of modified asphalt decrease gradually. When the temperature arrives at 173°C and 183°C, ductility tends to be zero after 10 h. The specimen is close to brittle fracture. On the contrary, the softening point continues to increase. In addition, the difference of penetration and softening point gradually increases under different aging temperature. This indicates that temperature has increasing impact upon the two indexes.

3.2. Impact of Aging on Toughness and Tenacity of Modified Asphalt. Asphalt toughness test, a rapid tensile test under specific conditions (25°C, 500 mm/min), is applied to measure the impact resistance and adhesion property of asphalt. It is effective in evaluating modified effect of the asphalt. The maximum tensile deformation ability of modified asphalt is evaluated with toughness, while the delay damage property and the resistance to fracture after tensile yielding are evaluated with tenacity.

Figure 2 is the typical tension-deformation curves of SBS modified asphalt and matrix asphalt, respectively, in the toughness test.

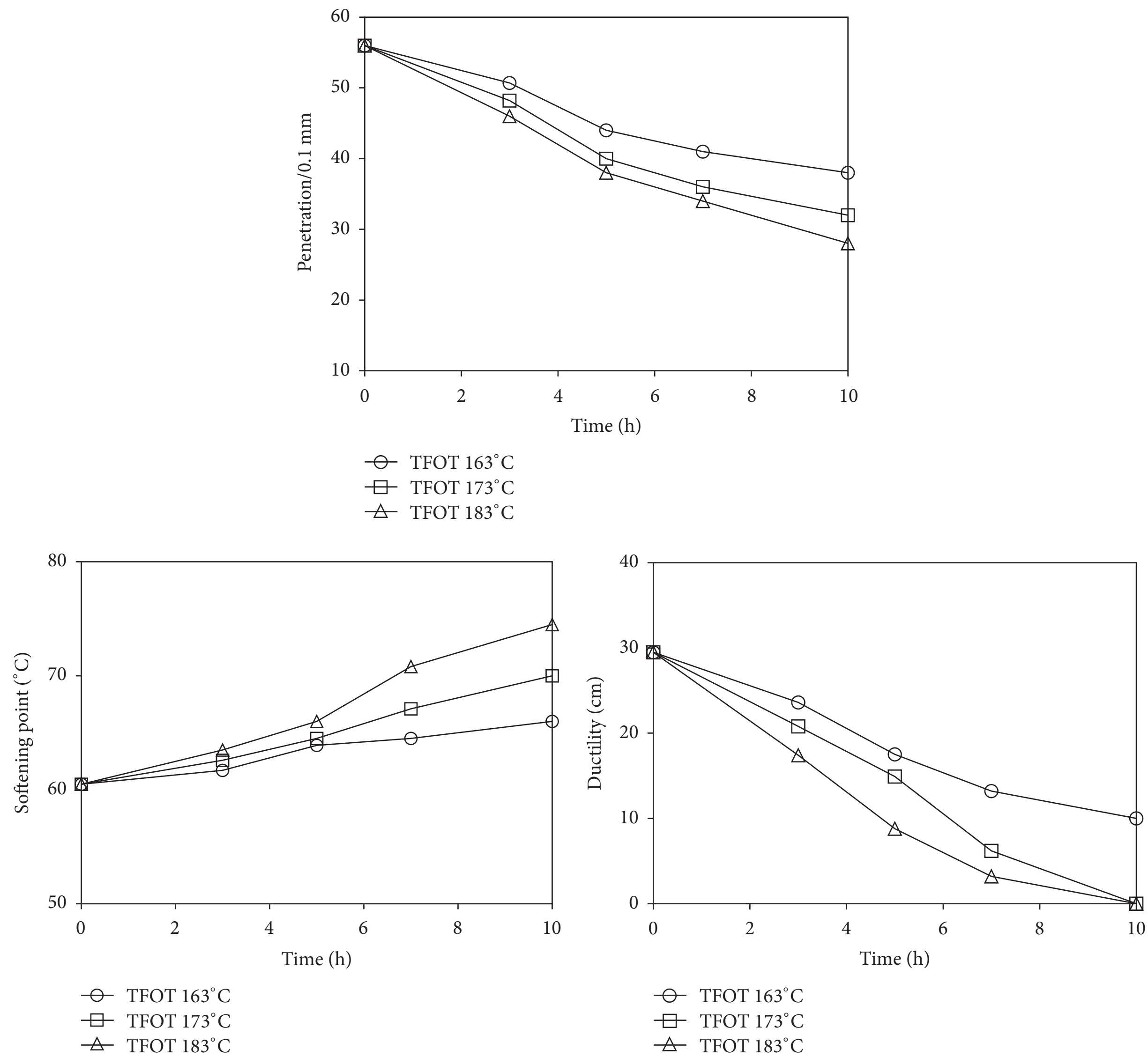


Figure 1: Correlation of three major indexes with aging time.

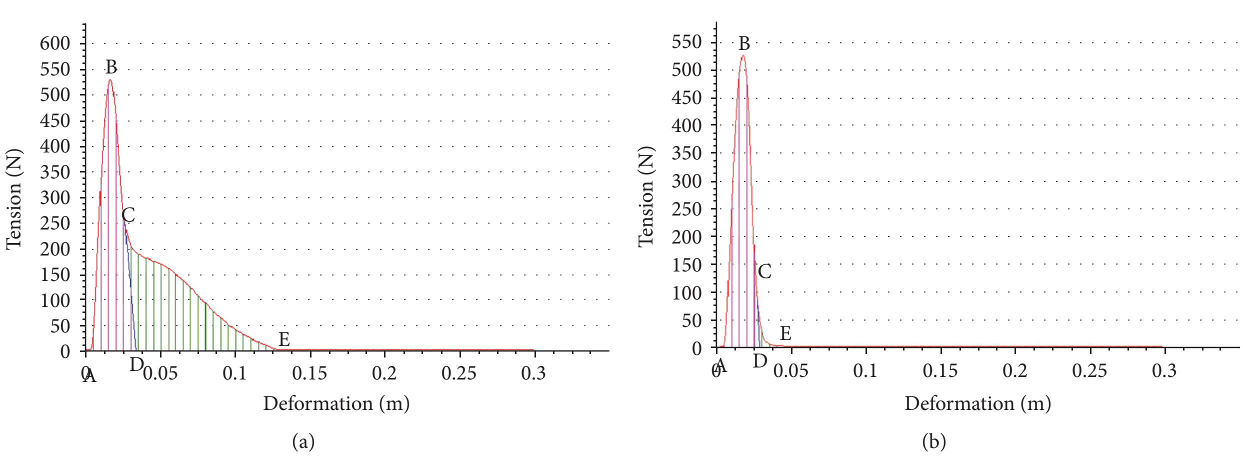


Figure 2: Tension-deformation curves of SBS modified asphalt and matrix asphalt.

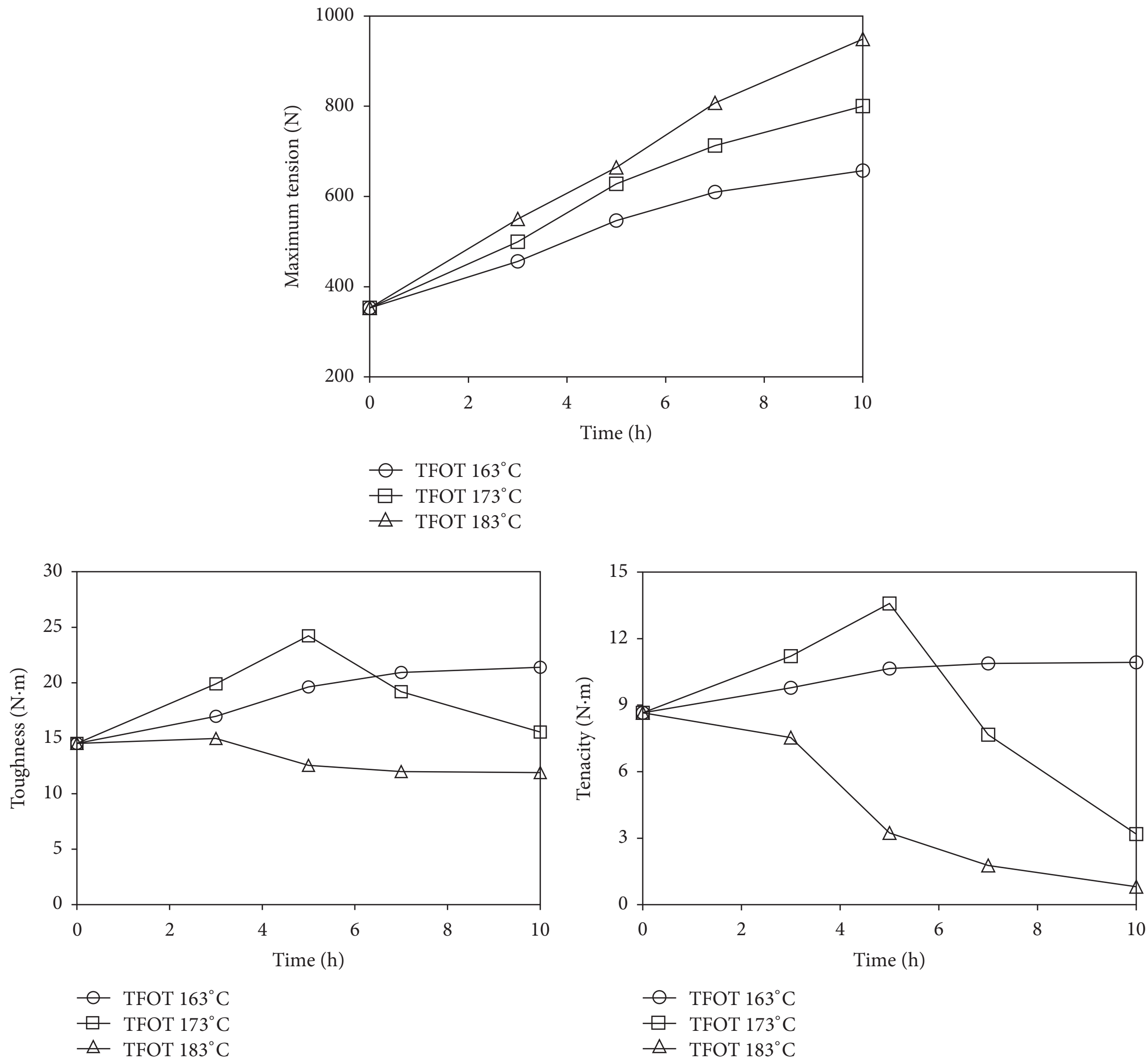


Figure 3: Variation curve of maximum tension, toughness, and tenacity of modified asphalt with the change of aging time.

Figure 2 shows that the tension increases rapidly with the increasing of deformation in the asphalt tensile process and it reaches the maximum point B when deformation reaches about 2 cm. Different from matrix asphalt, an obvious delayed fracture stage (CE) exists in the modified asphalt. The tension decrease rate slows down in this stage, while great deformation appears. Thus, a preliminary judgement can be concluded: toughness is mainly affected by the SBS.

The variation curve of the maximum tension, toughness, and tenacity of modified asphalt with the change of aging time is shown in Figure 3.

Figure 3 shows that, as the aging progresses, the maximum tension of modified asphalt increases gradually. The trend of toughness and tenacity is distinct at different aging temperatures. With aging temperature at 163°C, toughness and tenacity increase gradually. The main reason probably is that the SBS continues to have swelling reaction and thus enhances antideformation capacity of modified asphalt. On the contrary, when the aging temperature is 183°C, toughness and tenacity continuously decrease, mainly because the excessive temperature results in series of reaction such as degradation and chain scission of SBS, overaging of matrix asphalt. With aging temperature at 173°C, toughness and tenacity show a two-phase variation trend: increasing at first and then decreasing. The reason may be that, during the early stage of aging, SBS continues to have swelling reaction, and consequently antideformation ability improves. As the time exceeds 5 h, the SBS chain segment breaks and loses constraints on the asphalt. The antideformation ability reduces, leading to the decreasing of toughness and tenacity.

3.3. Impact of Aging on the Viscosity of Modified Asphalt. Asphalt aging usually involves a series of reactions such as fracture, reorganization, and polymerization of chemical bonds, thus inevitably leading to changes of viscosity. Therefore, researches on the changes of viscosity during the aging

Table 3: Dynamic parameters of aging reaction of modified asphalt.

Aging temperature/°C	$v/10^{-3}\cdot h^{-1}$	Correlation coefficient	A	Ea/KJ·mol^{-1}
163	11.1	0.93		
173	36.9	0.91	540906	23.91
183	75.8	0.95		

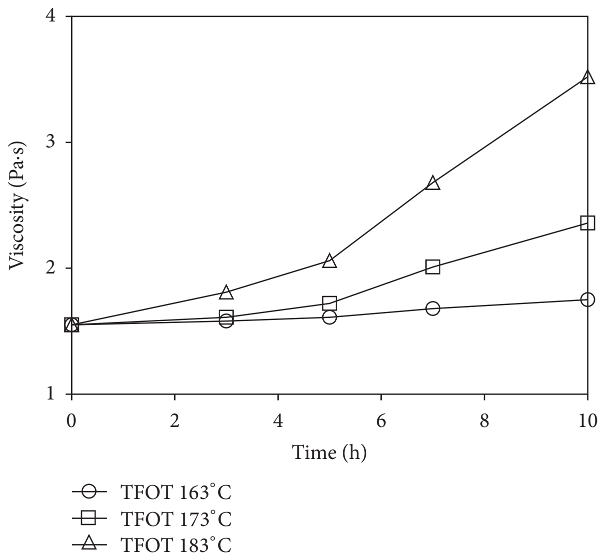


Figure 4: Variation curve of viscosity of modified asphalt with the change of aging time.

process of modified asphalt can facilitate the understanding of aging mechanism. Figure 4 shows the variation curve of viscosity of modified asphalt with the change of aging time.

Figure 4 shows that the viscosity of modified asphalt increases gradually during the aging process and increases with an increasingly large extent. In addition, viscosity difference of modified asphalt increases significantly at different temperatures. This indicates that high temperature accelerates aging.

3.4. Establishment of Dynamic Model of Modified Asphalt Aging. Asphalt aging is an irreversible first-order dynamic reaction [14]. With the assumption that aging of SBS modified asphalt is also a first-order reaction, the dynamic equation is

$$-\frac{dy}{dt} = vy, \quad (1)$$

where y is asphaltene reactant concentration in modified asphalt, mol; t is the aging time, h; v is the reaction rate constant, h^{-1}.

Experiments show that the reactant concentration is inversely proportional to the apparent viscosity (where $y = a/\eta$, η is apparent viscosity). Substituting the apparent viscosity into equation (1) with integral available, (2) is obtained:

$$\ln \frac{\eta}{\eta_0} = vt, \quad (2)$$

where η_0 is the initial apparent viscosity; η is the apparent viscosity at time t, Pa·s.

Brookfield viscosity in test belongs to apparent viscosity. So the reaction rate constant of modified asphalt under different aging time is calculated by Brookfield viscosity. Linear fitting of $\ln(\eta/\eta_0)$ with aging time and the slope of fitting regression line is the reaction rate constant of modified asphalt under the corresponding temperature condition. According to the Arrhenius equation,

$$\ln v = -\frac{Ea}{(RT)} + \ln A, \quad (3)$$

where Ea is activation energy of modified asphalt aging reaction, KJ·mol^{-1}; R is molar gas constant, value 8.314 J/(mol·K); T is aging temperature, °C; A is preexponential factor.

The activation energy of modified asphalt aging reaction can be obtained by fitting the reaction rate constant and aging temperature with (3). The fitting results are shown in Table 3.

From Table 3 the correlation coefficients of the three aging temperature regression lines are relatively large, indicating that the aging of modified asphalt is a first-order reaction. When the aging temperature rises, v becomes larger, indicating that the aging rate increases. Substituting the Arrhenius equation into (2),

$$\ln \eta = \ln \eta_0 + Ate^{-Ea/RT}. \quad (4)$$

Then substituting the modified asphalt dynamic parameters into (4),

$$\ln \eta = 0.43825 + 540906te^{-2875.9/T}. \quad (5)$$

The viscosity of modified asphalt at different aging temperatures and time can be calculated with (5). Since (5) was obtained based on the measured viscosity at 163°C, 173°C, and 183°C, the calculated viscosity values at the same temperatures from this equation are of less significance to be compared. So the other temperatures (158°C, 168°C, and 178°C) and aging time (8 h) were supplemented. Results are shown in Table 4.

Table 4 show that the calculated values are in good agreement with the measured values with the maximum relative error only 12.6%. This indicates that the aging dynamic equation based on apparent viscosity better reflects the aging process of modified asphalt and helps to evaluate and predict its antiaging property.

4. Microstructure Characteristics Analysis

Polymer SBS in asphalt absorbs light oil to form polymer phase. Under the fluorescence excitation, polymer phase

Table 4: Calculated and measured values of apparent viscosity of modified asphalt (pa·s).

Aging time/h	Calculated values	Measured values	Relative error/%	Calculated values	Measured values	Relative error/%	Calculated values	Measured values	Relative error/%
		163°C			173°C			183°C	
0	1.55	1.55	0	1.55	1.55	0	1.55	1.55	0
3	1.61	1.58	1.9	1.71	1.61	6.2	1.98	1.81	9.4
5	1.64	1.61	1.9	1.82	1.72	5.8	2.32	2.06	12.6
7	1.68	1.68	0	1.95	2.01	3.0	2.73	2.68	1.9
10	1.74	1.75	0.6	2.15	2.36	8.9	3.48	3.52	1.1
Supplements		158°C			168°C			178°C	
8 h	1.64	1.56	5.1	1.82	1.66	9.6	2.35	2.39	1.7

reflects light with relatively long wavelength, appearing white. On the contrary, the asphalt phase does not reflect any light, appearing black. So by fluorescence microscopy the asphalt phase and polymer phase can be clearly distinguished. As the reflected light field is applied, there is no damage to the distribution of polymer phase in the asphalt. Hence, the fluorescence microscopy gets a lot of good reproducibility asphalt microimages. By analysing the structure of polymer SBS morphological and dispersion state of asphalt, it is accessible to effectively evaluate the mechanical properties of modified asphalt and establish the relationship between macroscopic mechanical properties and microstructure characteristics.

To ensure the reliability of fluorescence images and avoid the particularity of individual samples, three slides were made in parallel for each sample. Four representative fluorescence images were taken for each slide.

4.1. Phase Structure Analysis of Modified Asphalt. Fluorescence microscopic images of unaged and aged modified asphalt are shown in Table 5.

Table 5 shows that polymer SBS in the unaged modified asphalt are distributed in the asphalt in the form of spherical particles, and this modified asphalt belongs to single-phase continuous structure with polymer SBS as dispersed phase and asphalt phase as continuous phase. Aging will not change its phase structure. The SBS particle size is relatively large before modified asphalt undergoes aging process. After aging, the particle size significantly decreases; the uniformity of particles gets better. At the same aging time, when the aging temperature reaches 183°C, the particle size is the smallest. At the same temperature, with prolonged aging time, the particle size significantly decreases and the distribution is more intensive.

4.2. Quantitative Analysis of the Microstructure of Modified Asphalt. The performance of modified asphalt is closely related to its phase property which is connected with the distribution of SBS particles in asphalt. Therefore, professional image processing software (Image-Pro Plus) is used to analyse and process the fluorescence images of modified asphalt, which segment, count, and measure the image. The average area and area ratio of polymer SBS particles are chosen as the quantitative evaluation indexes to describe the aging mechanism of modified asphalt. Here the area ratio refers to the percentage of polymer SBS particle area to total area, indirectly reflecting the SBS swelling degree in asphalt.

4.2.1. Correlation of Microstructure Quantitative Indexes, Toughness, and Tenacity with Aging Temperature. According to the results of macroscopic properties and microscopic characteristics, compare the relationship between microstructure quantification indexes and macroscopic properties which is based on the connection points of aging temperature and aging time. The results show that the variation of microstructure quantitative indexes is consistent with toughness and tenacity.

Figures 5–8 show that the variation of microstructure quantitative indexes, toughness, and tenacity at different aging temperature and different aging time.

Figures 5–8 show that when the aging time is 3 h and 5 h, the area and area ratio of polymer SBS particles increase as the aging temperature increases. When the temperature exceeds 173°C, these two indexes decrease. When the aging time is 7 h and 10 h, the two indexes decrease with a progressively slow rate as the aging temperature increases.

The results show that when the aging time is less than 5 h and aging temperature is 163°C~173°C, the polymer SBS continues to conduct swelling reaction; the SBS molecular chain segment in asphalt increases, which leads to the increase of particle area and area ratio and the restriction effect of polymer on the asphalt. Finally the macroscopic properties show greater toughness and tenacity. When the aging temperature exceeds 173°C, polymer SBS has a series of reactions such as degradation and chain scission and the particle area and area ratio decrease, leading to the decreasing restriction effect on asphalt. Therefore, the toughness and tenacity of modified asphalt decrease.

Only from Figures 5–8, conclusion can be drawn: with the increasing of aging temperature, the change law of the area and area ratio of SBS particles are in conformity with that of toughness and tenacity, but it may be just an apparent phenomenon. Therefore, this paper uses the professional data analysis software to carry on a bivariate significance test with the significance level of 0.05. The results are shown in Table 6.

Table 6 shows that the correlation coefficients of micro- and macroindexes are very high and all indexes are positively correlated; only the significant parameter between area ratio and tenacity is less than 0.05. Therefore, when the aging time

Table 5: Fluorescence micrographs of modified asphalt.

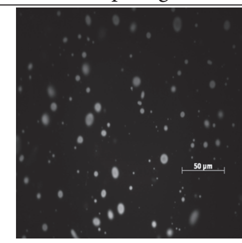
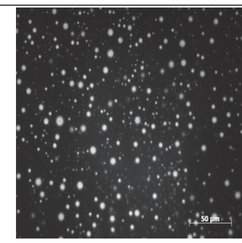
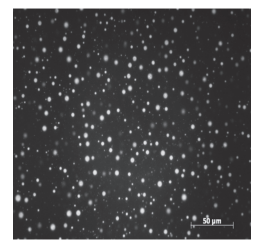
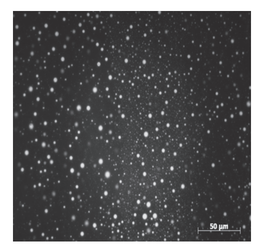
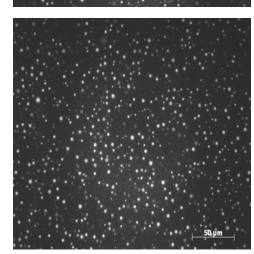
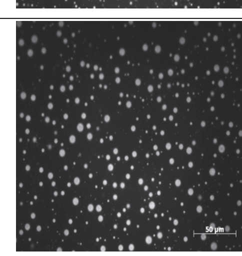
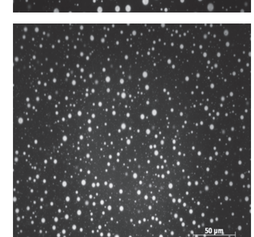
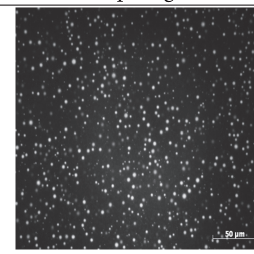
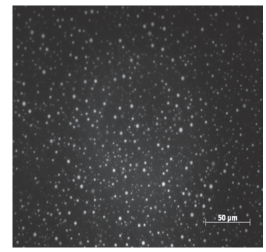
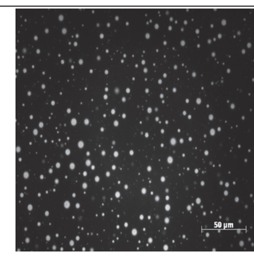
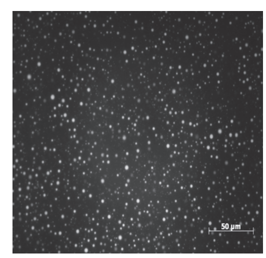
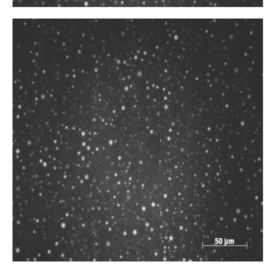
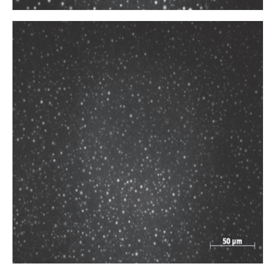
State	Sample figure
Unaged	
TFOT (163°C, 3 h)	
TFOT (163°C, 5 h)	
TFOT (163°C, 7 h)	
TFOT (163°C, 10 h)	
TFOT (173°C, 3 h)	
TFOT (173°C, 5 h)	

Table 5: Continued.

State	Sample figure
TFOT (173°C, 7 h)	
TFOT (173°C, 10 h)	
TFOT (183°C, 3 h)	
TFOT (183°C, 5 h)	
TFOT (183°C, 7 h)	
TFOT (183°C, 10 h)	

is 3 h, 5 h, 7 h, or 10 h, the area ratio of SBS particles has a significant correlation with tenacity as the aging temperature increases. Theoretical analysis shows that the particle area ratio is an integral index of modified asphalt. The larger the particle area ratio, the greater the specific surface area. Polymer particles will absorb more light oil in asphalt to reduce the surface energy. This results in more developed colloidal structure and increasing antideformation ability. In

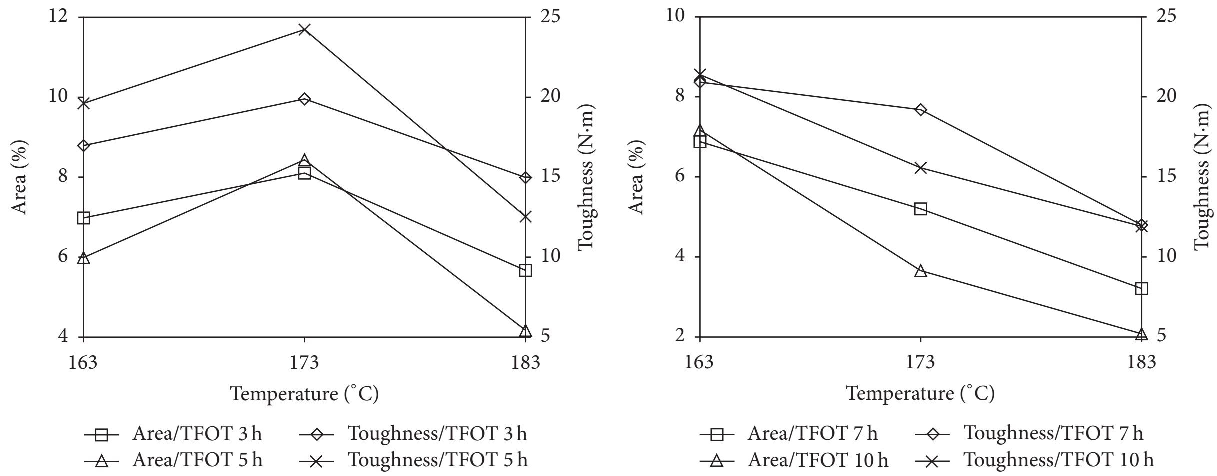


Figure 5: Correlation of SBS particle area and toughness with aging temperature.

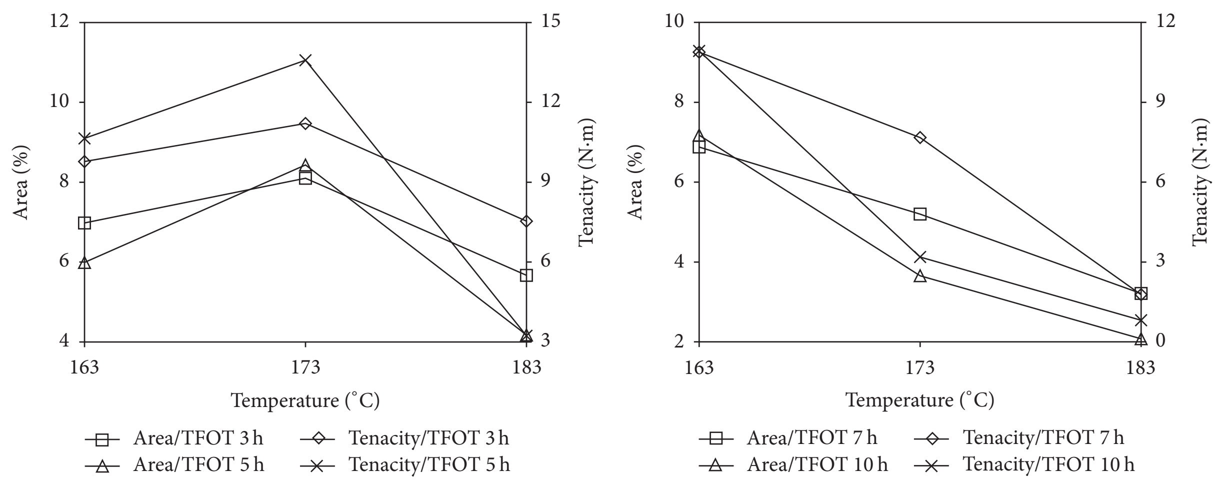


Figure 6: Correlation of SBS particle area and tenacity with aging temperature.

the macroscopic perspective, the tenacity of modified asphalt increases.

4.2.2. Correlation of Microstructure Quantitative Indexes and Tenacity with Aging Time. According to the above analysis, the correlation between the toughness and microscopic characteristics of modified asphalt is poor. So this section only analyses the correlation of polymer particle area ratio and tenacity with different aging time. The results are shown in Figure 9.

Figure 9 shows that when the aging temperature is 163°C or 173°C, the change law of particle area ratio and tenacity is basically identical; when the aging temperature is 183°C, due to high temperature, polymer SBS conducts a series of reactions such as degradation and chain scission, resulting in the continued decreasing of the particle area ratio and a different change law from that of tenacity.

Similarly, the significance test was carried out and the calculation method was similar to the previous section. The results are shown in Table 7.

Table 7 shows that the correlation between micro- and macroindexes is relatively small. Only when the aging temperature is 163°C, the significant parameter is less than 0.05. With increasing aging temperature, the comprehensive aging effect of polymer SBS and asphalt complicates antideformation capacity of modified asphalt, leading to the irregular changes of particle area ratio and tenacity.

5. Conclusions

In this paper, the main technical indexes of homemade modified asphalt satisfy the specification requirements, belonging to single-phase continuous structure, with polymer SBS as dispersed phase and asphalt phase as continuous phase.

(1) Aging has a significant influence on modified asphalt properties. It results in decreasing penetration and ductility and increasing softening point and viscosity. With the prolonging of aging time, the difference of penetration, softening point, and viscosity increases gradually at different aging temperatures.

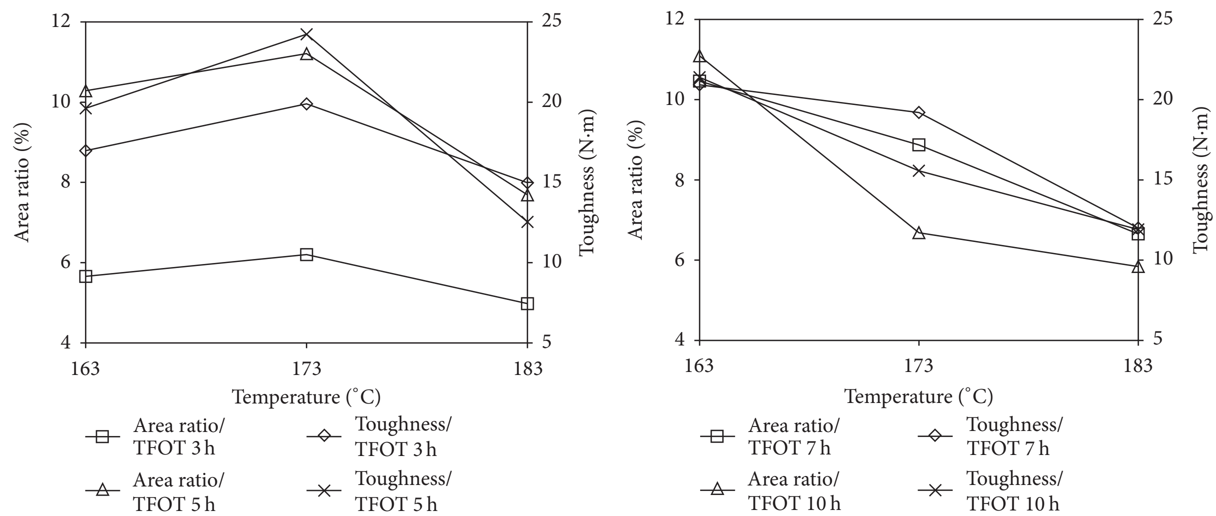


Figure 7: Correlation of SBS particle area ratio and toughness with aging temperature.

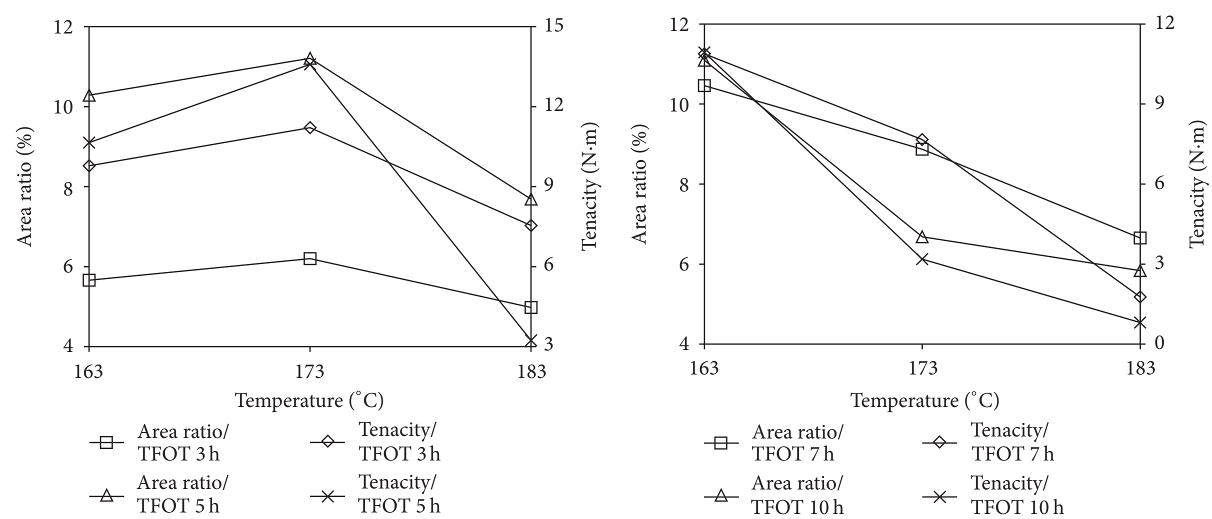


Figure 8: Correlation of SBS particle area ratio and tenacity with aging temperature.

Table 6: Correlation of microstructure indexes, toughness, and tenacity with aging temperature.

Aging time	Microindexes	Macroindexes	Correlation coefficient	Significance
TFOT 3 h	Area	Toughness	0.988	0.097
		Tenacity	0.995	0.052
	Area ratio	Toughness	0.985	0.111
		Tenacity	0.994	0.039
TFOT 5 h	Area	Toughness	0.979	0.129
		Tenacity	0.947	0.210
	Area ratio	Toughness	0.989	0.095
		Tenacity	1	0.015
TFOT 7 h	Area	Toughness	0.958	0.186
		Tenacity	0.992	0.076
	Area ratio	Toughness	0.970	0.156
		Tenacity	0.996	0.047
TFOT 10 h	Area	Toughness	0.996	0.054
		Tenacity	0.997	0.051
	Area ratio	Toughness	0.971	0.154
		Tenacity	0.997	0.049

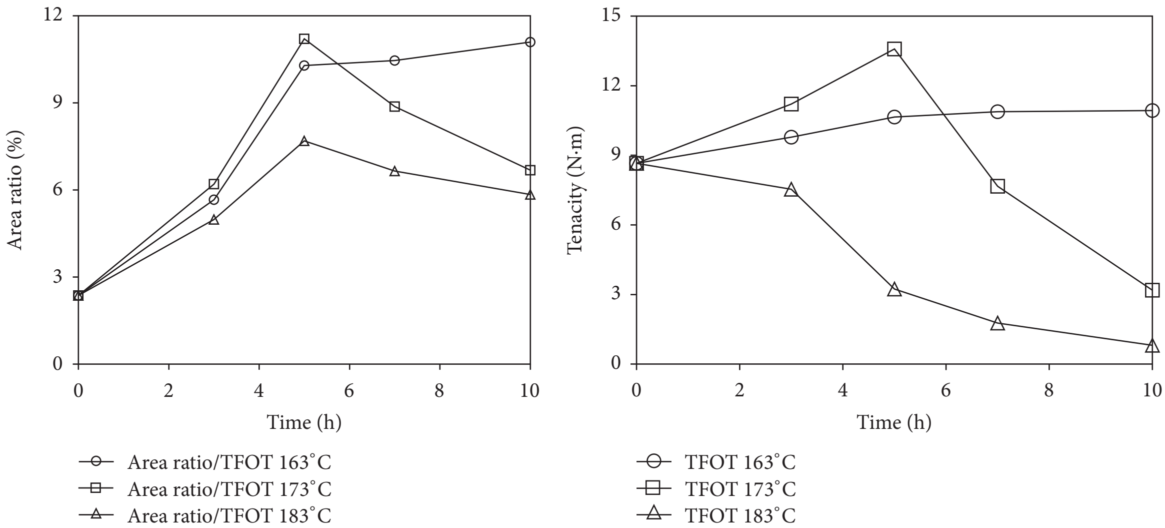


Figure 9: Correlation of SBS particle area ratio and tenacity with aging time.

Table 7: Correlation of microstructure quantitative indexes and tenacity with aging time.

Aging temperature	Microindex	Macroindex	Correlation coefficient	Significance
TFOT 163°C	Area ratio	Tenacity	0.991	0.012
TFOT 173°C	Area ratio	Tenacity	0.359	0.552
TFOT 183°C	Area ratio	Tenacity	−0.768	0.129

(2) The toughness test shows that, different from matrix asphalt, an obvious delayed fracture stage exists for modified asphalt, and thus the aging characteristics of modified asphalt can be effectively evaluated. At different aging temperatures, the toughness and tenacity of modified asphalt show different trends.

(3) The aging dynamic model of modified asphalt based on Brookfield viscosity is simple and reliable. It reflects well the aging process and is conducive to evaluating and predicting the antiaging property.

(4) There is no marked relationship between phase structure and aging of modified asphalt. As aging temperature or aging time increases, SBS particle size decreases and the particle uniformity is better.

(5) When the aging time is constant, the changing pattern of area and area ratio is in conformity with that of toughness and tenacity with the increasing of aging temperature. However, only the SBS particle area ratio has a significant correlation with tenacity. When the aging temperature is 163°C, the SBS particle area ratio also has a significant correlation with tenacity as the aging time increases. The results prove the possibility of establishing a link between microscopic characteristics and macroscopic mechanical properties of modified asphalt and provide a new method for evaluating mechanical properties of modified asphalt from microscopic aspect.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

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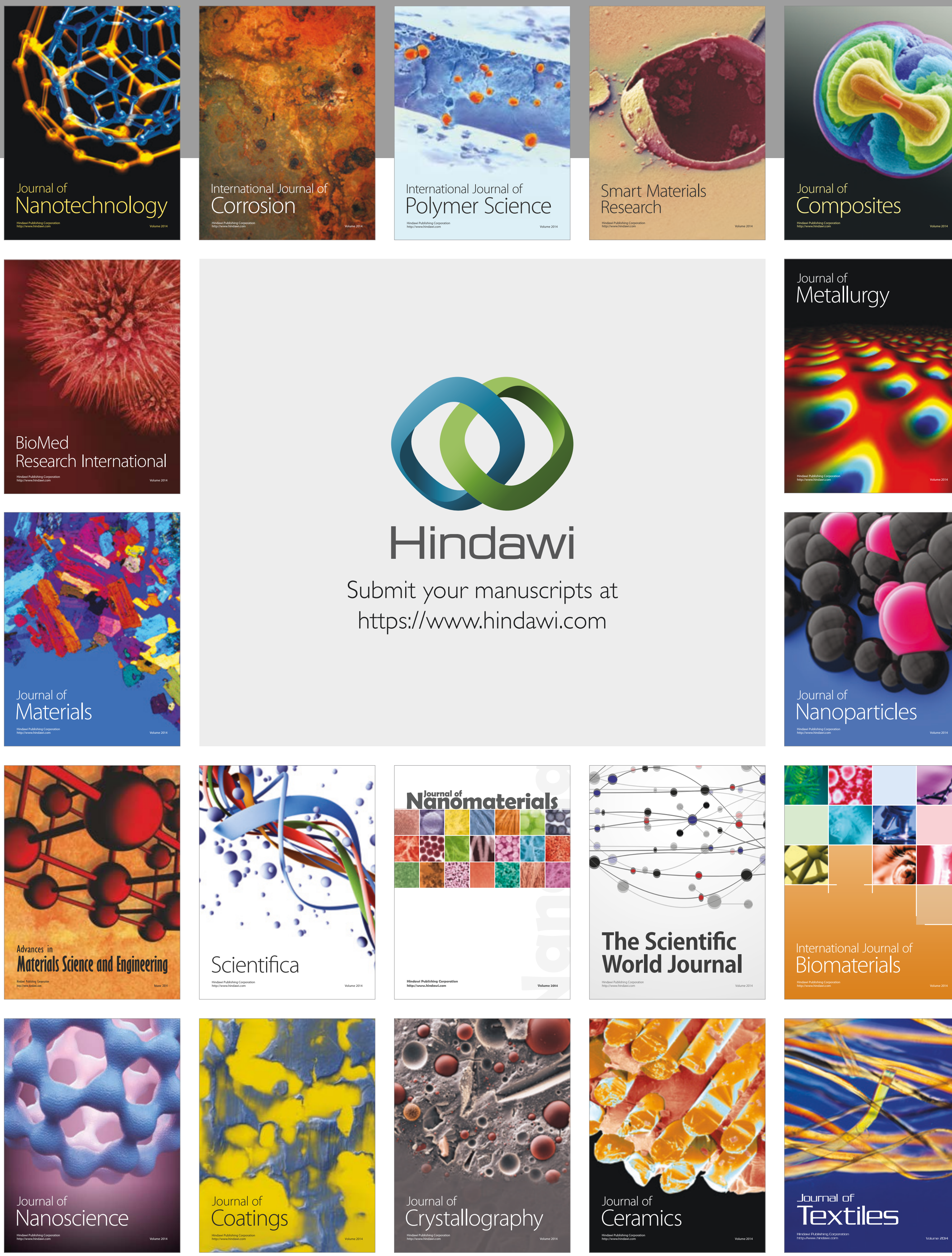

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