Thermal aging of bitumen mixtures with crushed sand

Mukhammat Salikhov¹ and Evgeniy Veyukov¹
¹Volga region state University of technology, Yoshkar-Ola, Russian Federation

Corresponding author: VeukovEV@volgatech.net

Abstract. Public roads with advanced types of coatings are mainly constructed of asphalt concrete. Along with good performance, they have some disadvantages. One of them is temperature aging. The aging process is inevitable. However, to increase the stability of bituminous mixtures, it is important to study the structure formation and aging processes of bitumen films on the surface of all components separately, including the areas of contact with crushed sand. In order to study the aging processes, the methods of studying the aging of bituminous mixtures have been critically analyzed. Considering the shortcomings of the methods available, a new method has been developed. Based on the experiments performed using this method, we studied the aging processes of bituminous mixtures at the temperature of +150° C for various periods of time. The resulting mathematical models describe the aging process based on the considered factors. The experimental results were analyzed in accordance with the theoretical concepts of the interaction of bitumen films on the surfaces of crushed sand particles of amorphous rocks. The conducted research has practical conclusions.

1. Introduction

It is known that 60-93% of public roads are constructed using improved type of pavement with coatings made of asphalt concrete and other bituminous mixtures [1], [2]. Along with clear advantages comparing to other structural road construction materials, the latter have a quite short service life. There is a lot of research and practical experience studying the performance of asphalt concrete pavements in various road-climatic zones [3], [4], [5], [6], [7], [8]. But there is little study of the possible effect of the aging of bitumen in bituminous mixtures on its service life. There is high probability that it is the stability of the bitumen coating under the influence of road, climatic and other factors that causes changes in the most important physical, mechanical and operational properties of asphalt concrete over time.

It is known that bituminous mixtures are predominantly multicomponent systems with macro, micro and ultra microstructures formed inside them. They have organic binders (such as oil bitumens distributed in thin layers (form films) of various thicknesses structured on the surfaces of mineral components of various mineral composition, sizes and shapes. Due to this, we can assume that under the environmental influences bitumen films undergo changes depending on their thickness, structuredness, size, duration and intensity of the influencing factors, etc. Since bitumen films at different stages of the structure formation and temperature act as a lubricant for the surfaces of mineral materials and glue them together, they ensure stability of the entire system and joint performance of all components in the resilient-elastic stage. The longer these conditions are provided, the higher performance of this material in the structural layer, the higher stress resistance to internal and external factors. However, long exposure to climatic factors (water temperature, solar radiation, mechanical
stresses, etc.) and fatigue over time results in changes in the chemical and physicochemical state of bitumen films. They undergo irreversible changes and eventually lose their adhesive-cohesive properties. It is important to determine the general laws of the change in time of the effect on the properties of bitumen films by studying the changes in physical and mechanical properties of bituminous mixtures.

The purpose of this research is to study the temperature aging processes of viscous petroleum bitumens in mixtures with crushed sand made from screenings of crushing peddle and gravel raw materials. The object of research is a road; the subject is the road surface.

2. Methods

Screenings were made during the processing of pebble and gravel materials, consisting of metamorphic rocks, of the M 800 crushability mark, F100 frost resistance and I-1 abrasion resistance.

Some characteristics of the studied materials are presented in Table 1 and Table 2.

Table 1. Granulometric composition of crushed stone

| Size of sieves, mm | 10  | 5  | 2.5 | 1.25 | 0.63 | 0.315 | 0.16 | 0.071 | < 0.071 |
|-------------------|-----|----|-----|------|------|-------|------|-------|---------|
| Private balances, % | 0   | 1.55 | 34.83 | 19.68 | 19.31 | 10.78 | 6.17 | 6.80 | 0.89 |
| Total balances, %  | 0   | 1.55 | 36.38 | 56.06 | 75.37 | 86.15 | 92.31 | 99.11 | 100 |
| Full passing, %    | 100 | 98.45 | 63.62 | 43.94 | 24.63 | 13.85 | 7.69 | 0.89 | 0 |

Table 2. Some characteristics of bitumen 70/100

| Indicator | Standard requirement | Actual value of the indicator |
|-----------|----------------------|-----------------------------|
| 1. Needle penetration depth at temperature +25 °C, 0.1 mm | 71-100 | 78 |
| 2. Softening point, °C, not lower | 47 | 48 |
| 3. Tensile at °C, cm, not lower | 3.7 | 3.7 |
| 4. Fragility temperature, °C, not lower | -18 | -19 |
| 5. Flash point, °C, not lower | 230 | 308 |
| 6. Change in sample mass after aging, %, not more than | 0.6 | 0.3 |

To select methods of the experimental research of aging of the bitumen-sand mixture, we proceeded from the fact that the basis method is standard method. First, they do not allow studying individual influences of the type and size of the mineral lining on the bitumen film aging [9], [10]. Secondly, the influence of the bitumen film thickness on this process is not taken into account. Thirdly, special equipment is required [11], [12]. To solve these problems, a new method was developed [13]. This method allows accelerated study of bitumen aging within its combination with any of the studied components of the bituminous mixture or in their entire combination. There was also a comparative analysis of this process in time using a dimensionless parameter – the aging coefficient \( K_{\Delta t}^{t_i} \) to the indicator of physical and mechanical properties under study according to the formula:

\[
K_{\Delta t}^{t_i} = \frac{I_i^{t_i, t_{wu}=t_i}}{I_i^{t_i, t_{wu}=0}},
\]

where \( I_i^{t_i, t_{wu}=t_i} \) and \( I_i^{t_i, t_{wu}=0} \) are values of the indicators of the studied physical and mechanical properties of the bituminous mixture before and after heating at high temperature over a period of time from 0 to \( t_i \).
The aging dynamics is assessed according to the values of the aging intensity $I_{AG}^{ki}$ and comparative analysis of the aging coefficient values with an acceptable value of $[K_{AG}^{ki}]$ and $[t_{wu}]$. The latter must be proved separately. To prove the value of the duration of preheating mixtures at the given heating temperature, we can tentatively take the time when the sample reaches the maximum permissible value of the considered physical and mechanical indicator according to the current standards of the documents for the studied material.

$$I_{AG}^{ki} = \frac{\Delta K_{AG}^{ki}}{\Delta t_{wu}},$$  \hspace{1cm} (2)

where $\Delta$ is the change in the value of the aging coefficient by the $i$-th indicator for the considered period of time: $\Delta t_{wu} = t_{wu,i} - t_{wu,i-1}$.

Performing experiments and processing results

The testing samples were prepared by sampling viscous bitumen 70/100, crushed sand, drying them to constant weight and determining some of their physical and mechanical properties according to current standard methods. Samples of viscous bitumen of crushed sand were heated to operating temperatures, thoroughly mixed together in the intended proportions, and standard cylindrical samples were formed according to the standard method with a diameter and height of 50.5 mm. The taken bitumen to sand ratios were proved taking into account the fact that in the asphalt concrete mixes of classical types A and B according they range from 1:22...1:15, for types G and D – 1:9.3... 1:10. According in pebble and gravel raw materials the B:P ratios are 1:2.2...1:3.2. According to this conclusions, it was decided to study the ratio of bitumen to sand mixtures (B:P) equal to 1:9, 1:10, 1:11, 1:12.

3. Results and Discussion

Studies of the aging process of samples of bitumen sand mixtures were performed according to the proposed method [14]. At the same time, the values of the aging intensity coefficients are based on the key indicators adopted for the study or a number of indicators determined during standard tests according to any domestic or foreign standards. The use of the dimensionless indicator ($I_{AG}^{ki}$) allows analyzing the aging process, regardless of the scale factor.

In this study, the value of the temperature of technological aging is considered to be $+150 \, ^\circ C$, since it is recommended to use the operating temperature of making bituminous mixtures within $+140 \ldots +160 \, ^\circ C$.

The results of the experimental work and some calculations are presented in Table 3.

| Par. No | B:P ration, fraction of units by weight (% of bitumen content) | Heating duration, hour | Average density g/cm$^3$ | Compressive strength limit at $R_{CS}^{+50^\circ C}$, MPa | Index aging ratio to the indicator $R_{CS}^{+50^\circ C}$ | Aging intensity, hour$^{-1}$ |
|--------|-------------------------------------------------|------------------------|--------------------------|---------------------------------------------|-----------------------------------------------|-----------------------------|
| 1      | 1:9 (B=11.1)                                    | 0                      | 2.32                     | 0.78                                        | 1.00                                          | 0.00                         |
| 2      | 1:9                                             | 1                      | 2.42                     | 1.55                                        | 1.98                                          | +0.98                        |
| 3      | 1:10 (B=10.0)                                   | 3                      | 2.20                     | 1.03                                        | 1.32                                          | -0.11                        |
| 4      | 1:11                                            | 5                      | 2.37                     | 1.42                                        | 1.82                                          | +0.16                        |
| 5      | 1:12                                            | 7                      | -                        | -                                           | -                                             | -                            |
| 6      | 1:10                                            | 0                      | 2.48                     | 1.37                                        | 1.00                                          | 0.00                         |
| 7      | 1:11                                            | 1                      | 2.39                     | 1.40                                        | 1.02                                          | +0.02                        |
| 8      | 1:12                                            | 3                      | 2.35                     | 1.97                                        | 1.43                                          | +0.14                        |
| 9      | 1:13                                            | 5                      | 2.40                     | 2.75                                        | 2.00                                          | +0.20                        |
| 10     | 1:14                                            | 7                      | 2.36                     | 1.47                                        | 1.37                                          | -0.05                        |
Next, a number of mathematical models was obtained using a software environment CurveExpert. Here are some of them:

a) for B:P = 1:9 (s = 0; r = 1.0):

\[ y = a + b \cdot \cos(c \cdot x + d), \]  

(3)

where \( y \) – is the aging coefficient value in terms of compressive strength at +50 °C; \( x \) – is the duration of the preliminary heating of the mixture at +150 °C per hour; \( a, b, c, d \) – empirical coefficients:

\( a = -9.49; b = 11.21; c = 0.06; d = -0.003 \).

b) for B:P = 1:9 (s=0.1281; r=0.9875):

\[ y = a + b \cdot \cos(c \cdot x + d), \]  

(4)

where \( a = 1.44; b = 0.51; c = 0.78; d = -3.82 \).

c) for B:P = 1:11 (s=0.2722; r=0.4465):

\[ y = a \cdot e^{-(x-b)^2 / 2c^2}, \]  

(5)

where \( a = 1.281; b = 4.216; c = 6.507 \).

d) for B:P = 1:12 (s=0; r=1.0):

\[ y = a + b \cdot \cos(c \cdot x + d), \]  

(6)

where \( a = -35.327; b = 38.475; c = 0.1005; d = -0.3163 \).

Changes in the values of the aging coefficient regarding the indicator \( R_{CS}^{50^\circ C} \) after heating at +150 °C for 5 hours depending on changes in the ratio of bitumen and sand (B:P = 1:9...1:12) have the following regularity (s = 0; r = 1 , 0):

\[ y = a + b \cdot \cos(c \cdot x + d), \]  

(7)

where \( x \) – is the content of bitumen in the mixture, % of the weight of sand; \( a = 1.915; b = 65.506; c = 2.871, d = -5.021 \).

The analysis of Table 3 shows that at changing the amount of bitumen in unheated bitumen-sand mixtures, the highest value of the tensile strength and compressions at +50 °C is at a flow rate of 9.1...10% of the mass of sand. Moreover, for these ratios, respectively, higher than the value of the aging coefficient after 5...7 hours of heating. Therefore, this provides the highest resistance to the mixture aging of the mixture.

The resulting mathematical models show that the longer the preheating time and the content of bitumen in the mixture, the faster and more intensively the process of temperature aging of bitumen-sand material.

For better understanding and creating a stable system against aging processes during the preliminary heating of mixtures in Table 4 bitumen calculations of the sand for individual classes of particles.

According to Table 4, the calculated and actual values of the bitumen intensity of the sands (bitumen consumption) were not exactly the same. This can be explained by the fact that, firstly, when considering the calculated value [15], particles of diorite rock were taken, and in this case, crushed sand was made from a mixture of various metamorphic rocks; secondly, the source [15] does not specify – these data are for crushed or natural materials. It is known that crushed materials usually
require a greater consumption of binder. To further clarify the possibility of using the theoretical calculation method in the future, the additional research should be performed.

**Table 4. Calculation of bitumen capacity of the sand under study**

| Par. № | Content of particles in 1 kg of crushed sand per class kg·10⁻³ | Content of particles in 1 kg of crushed sand per class Shares | Specific bitumen capacity, ·10⁻² | Bitumen capacity, ·10⁻² | Actual bitumen intensity (according to selection) |
|--------|------------------------------------------------------------|------------------------------------------------------------|---------------------------------|------------------------|------------------------------------------------|
| 1      | less than 0.71                                           | 8.90                                                       | 0.89                           | 16.5                   | 0.17                                                          |
| 2      | 0.071-0.16                                               | 68.0                                                       | 6.80                           | 10.0                   | 0.68                                                          |
| 3      | 0.16-0.315                                               | 61.7                                                       | 6.17                           | 9.0                    | 0.56                                                          |
| 4      | 0.315-0.63                                               | 107.8                                                      | 10.78                          | 7.9                    | 0.85                                                          |
| 5      | 0.63-1.25                                                | 193.1                                                      | 19.31                          | 7.0                    | 1.35                                                          |
| 6      | 1.25-2.5                                                 | 196.8                                                      | 19.68                          | 5.9                    | 1.16                                                          |
| 7      | 2.5-5                                                    | 348.3                                                      | 34.83                          | 5.6                    | 1.95                                                          |
| 8      | 5-10                                                     | 15.5                                                       | 1.55                           | 4.5                    | 0.70                                                          |
| Total  | 100.00                                                   |                                                            |                                | 7.42                   | 10.00...9.10                                                  |

At the same time, we can assume that the system stability against destruction also depends on the structure of the bitumen film on the surfaces of sand particles.

It is known [16], [17], [18] that the structures of the bitumen film on the surfaces of mineral grains consist of tightly bound, loosely bound (weakly oriented) and free zones [19], [20]. When heated, the most surface-active constituents of bitumen, asphaltic acids and their anhydrides (C₉H₁₇COOH) are concentrated by a monomolecular layer in the intercontact zones. In the relatively weak «-OH» bonds, hydrogen atoms are cleaved and replaced by oxygen atoms. This, along with the processes of oxidation, polyoxication, evaporation of maltene groups and some others, results in an increase in the content of asphaltenes. These processes also decrease adhesive properties of the bonds of bitumen films with the surfaces of mineral particles and the adhesion-cohesive property of bitumen. Accordingly, during the heating process, there is a high possibility of a general decrease in the resistance of the samples under compression confirmed by the performed experiments. We should note that there may be an increase in the resistance of the samples to destruction due to an increase in the fragility of bitumen in the initial period of heating (from 1 to 3 hours).

4. **Conclusions**

Results of the operational study of the aging processes of viscous bitumen as a part of bitumen-sand mixtures:

1. The optimum ratios of bitumen in the composition of bitumen-sand mixtures based on crushed San with the highest resistance to aging processes when heated at high temperatures were determined;
2. Mathematical models describing the aging processes of samples of bituminous mixtures, depending on the duration of heating at high temperatures and the content of bitumen in them were obtained.
3. The mechanism of the aging process of bitumen films on the surfaces of crushed sand particles from metamorphic rocks is suggested.

**Acknowledgements**
The authors thank the staff of the Department of construction technologies and highways of the Volga state technological University (Yoshkar-Ola, Russian Federation).
References

[1] Essawy A I, Saleh A M M, Zaky M T, Farag R K and Ragab A A 2013 Environmentally friendly road construction Egypt. J. Pet.
[2] Zhurinov M Z, Teltayev B B and Kalybai A A 2019 Characteristics of road bitumen modified with carbon nanopowder News Natl. Acad. Sci. Repub. Kazakhstan, Ser. Geol. Tech. Sci.
[3] Korolev E V., Gladkikh V A and Khusid D L 2016 Resistance of sulfur-extended asphalt to rutting Vestn. MGSU
[4] Liu Tingguo ., Zankavich V N, Aliakseyeu Y N and Khroustalev B M 2019 Recycling of Materials for Pavement Dressing: Analytical Review Sci. Tech.
[5] Pshembaev M K and Kovaley Y N 2018 Optimization of preservative for protection of concrete pavement of highways Sci. Tech.
[6] Manzone M and Ruffinengo B 2019 Performance of a snow blower prototype mounted on different vehicles type J. Agric. Eng.
[7] Vuye C, Musovic F, Tyszka L, Van Den Bergh W, Kampen J, Bergiers A, Maeck J, Buylaert A and Vanhooreweder B 2016 First experiences with thin noise reducing asphalt layers in an urban environment in Belgium Proceedings of ISMA 2016 - International Conference on Noise and Vibration Engineering and USD2016 - International Conference on Uncertainty in Structural Dynamics
[8] Loise V, Vuono D, Policicchio A, Teltayev B, Gnisci A, Messina G and Oliviero Rossi C 2019 The effect of multiwalled carbon nanotubes on the rheological behaviour of bitumen Colloids Surfaces A Physicochem. Eng. Asp.
[9] Baek C, Underwood B and Kim Y 2012 Effects of oxidative aging on asphalt mixture properties Transp. Res. Rec.
[10] Chomicz-Kowalska A, Gardziejczyk W and Iwański M M 2016 Moisture resistance and compactibility of asphalt concrete produced in half-warm mix asphalt technology with foamed bitumen Constr. Build. Mater.
[11] Wei H, Bai X, Qian G, Wang F, Li Z, Jin J and Zhang Y 2019 Aging mechanism and properties of SBS modified bitumen under complex environmental conditions Materials (Basel).
[12] Saedi S and Oruc S 2020 The influence of SBS, viatop premium and FRP on the improvement of stone mastic asphalt performance Fibers
[13] Wasilewska M, Małaszkiwicz D and Ignatiuk N 2017 Evaluation of Different Mineral Filler Aggregates for Asphalt Mixture IOP Conference Series: Materials Science and Engineering
[14] Teltayev B B, Amirbayev E D and Radovskiy B S 2018 Viscoelastic characteristics of blown bitumen at low temperatures Constr. Build. Mater.
[15] Zakrevskaya L, Handelsman I and Provatorova G 2018 The effect of modification of binders on technological and operational properties of composite construction materials MATEC Web of Conferences
[16] Teltayev B and Radovskiy B 2018 Predicting thermal cracking of asphalt pavements from bitumen and mix properties Road Mater. Pavement Des.
[17] Szerb E I, Nicotera I, Teltayev B, Vaiana R and Rossi C O 2018 Highly stable surfactant-crumb rubber-modified bitumen: NMR and rheological investigation Road Mater. Pavement Des.
[18] Teltayev B B and Seilkhanov T M 2018 NMR-spectroscopy determination of fragmentary composition of bitumen and its components Eurasian Chem. J.
[19] Teltayev B B and Amirbayev Y D 2016 Evaluation of rheological characteristics for bitumen binders at different longivities of thermal condition News Natl. Acad. Sci. Repub. Kazakhstan, Ser. Geol. Tech. Sci.
[20] Porto M, Caputo P, Loise V, Teltayev B B, Angelico R, Calandra P and Rossi R C 2019 New experimental approaches to analyse the supramolecular structure of rejuvenated aged bitumens News Natl. Acad. Sci. Repub. Kazakhstan, Ser. Geol. Tech. Sci.