STRUCTURING OF POLYMINERAL HIGHLY CONCENTRATED POLYDISPERSION SYSTEMS BASED ON ORGANIC BINDERS

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Abstract

At present, the quality of bitumen-mineral mixtures both, in Russia and abroad, is estimated by a set of empirical methods based on many years of experience in using asphalt concrete in road building. Numerous attempts have been made over the years to theoretically substantiate the basic behavior patterns of the material under the influence of various factors. Thus, the creation of optimal conditions for the formation of microstructural contact bonds in a highly concentrated polymineral polydispersion system significantly contributes to the formation of asphalt mixes with necessary processing properties and asphalt concrete with required transport and performance characteristics. The article describes the solution of this problem according to the basic provisions of physical chemical mechanics of highly concentrated dispersion systems, taking into account peculiarities of contact interactions. The studies have allowed finding the quantitative correlations of indicators of physical mechanical and structural rheological properties being formed at compaction of bitumen mineral mixes as well as quantitative connections between formative and destructive factors with structural mechanical and construction technical properties of asphalt in road coverings. It has been established that the structural changes in asphalt concrete in road coverings under the influence of operational factors are physical chemical in nature and reliably described, taking into account the basic provisions of physical chemical mechanics and the theory of contact interactions.

Keywords: Bitumen mineral mixes, structuring, contact interactions, physical chemical and structural rheological properties

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I. Introduction

Asphalt concrete has complex rheological properties determining both, standard operating procedures and operational reliability of created structures. The procedural framework formed in different countries by now allows estimating quality data about bitumen mineral mixes.

In the United States, for example, the quality of asphalt concrete and asphalt concrete mixes is successfully estimated by Marshall’s method now embodied in the Super Wave technology. French road builders make use of Durieux’s method, whereas in Russia the uniaxial compressive strength is used for the purpose. Depending on operational conditions, each country poses its own requirements, i.e., the set of target physical mechanical parameters.

All the above enumerated methods are empirical and based on many years of experience in applying asphalt concrete in road building.

However, multiple attempts have been made over the years to substantiate the basic behavior patterns of the material under the influence of various factors. This has made it possible to find out that the change in proportion among the components and their quality condition in contact interphase and boundary layers during the structuring and formation of properties of bitumen mineral materials changes the state of structural bonds in the composite and, therefore, determines all its physical mechanical characteristics. It has been proven that additive properties of asphalt concrete and asphalt concrete mixes are determined by their macrostructure, mesostructure, and, especially, microstructure. Thus, professor I. A. Rybiev [I] proposed the theory of building conglomerates and professor V. P. Selyatin [II] developed the polystructural theory of building composites. Professor M. I. Volkov and his alumni studied the guided control over the properties of asphalt concrete by modifying the properties of the binder in the area of contact among mineral particles.

Asphalt concrete and asphalt concrete mixes used in road coverings are highly concentrated dispersion systems with a strongly developed interphase surface $S_{\text{un}}$ and a highly concentrated dispersion phase in liquid and gas dispersion media.

The creation of optimal conditions for forming microstructural contact bonds in a highly concentrated polymineral polydispersion system largely encourages the formation of asphalt concrete mixes with necessary processing properties and asphalt concrete with required transport and operation characteristics. The quantitative criteria of estimating the structural condition of road asphalt concrete and asphalt concrete mixes are yet to be introduced in the respective regulatory and engineering framework.

II. Materials and Methods

This task can be solved following the main provisions of the physical chemical mechanics of highly concentrated dispersion systems, taking into account peculiarities of contact interactions.
It has been established that at a high fraction of polymineral and polydispersion filler particles with structural shells with high-structure bitumen work in with each other in micro-contact areas with diameter $d_i[1]$. This encourages the formation of a new coagulation structure with active mineral filler grains as structuring centers. As the temperature rises, interphase contact interactions become weaker. The works by P. A. Rebinder, Ye. D. Shchukin, and L. Ya. Margolis have been used to propose using the porous body strength relation, taking into account the aggregate strength of elementary contacts as

$$P_m = k_1 \cdot F_{avg} \cdot n^{2/3} = k_2 \cdot \varphi \cdot S_{un}^{-\frac{2}{3}} = k_3 \cdot F_{avg} \cdot \varphi \cdot d_{equiv}^{\frac{2}{3}}$$

(1)

where $P_m$ is the critical shear stress, $F_{avg}$ is the average cohesive attraction of interparticle contact, $n$ is the average number of interparticle contacts per unit of volume ($m^3$), $\varphi$ is the relative structural density, $S_{un}$ is the unit surface of dispersion phase particles in the highly concentrated dispersion system, $d_i$ is the characteristic size of these particles. Modern equipment allows quantitatively estimating the critical shear stress of the highly concentrated dispersion system by various experimental methods.

The structural destruction of asphalt concrete and the ability of the material being or already formed to resist external operation actions depend on the conditions of its formation. In terms of quantity the condition of structure is characterized by $P_m$ (strength of the highly concentrated dispersion system), average cohesive attraction of interparticle contact $F_{avg}$, and average unit contact density $P_i$. The quantitative criteria characterizing the structure’s geometric parameters are equally important structural characteristics.

![Fig. 1: Changes in the number of contacts among ball-like particles with regard to their size](image-url)
They include the average number of these inter particle contacts per unit of volume \( n \), relative density \( \varphi \), unit surface of the mineral part of asphalt concrete mix \( S_{\text{un}} \), characteristic size of dispersion phase particles (mineral part of mix) \( d_{\text{equiv}} \).

As shown above in (1), the dispersion system strength depends on the average number of inter particle contacts. According to [XVII, XXXII], the number of contacts in highly concentrated dispersion systems is related to the average number of contacts per unit of volume \( n \).

First of all, the average number of contacts per unit of volume \( (\text{cm}^3) \) depends on the size or dispersion of mineral particles. Their size may range from 0.001 to 20 (or 40) mm, which depends on the kind and classifying feature of asphalt concrete mix.

\[ \text{Fig. 2: Change in the number of contacts among monodisperse ball-shaped particles, depending on their size} \]

This largely affects the number of single contacts. In Fig. 2 the change in the number of contacts among monodispersion ball-like particles is shown for a relative packing density of 0.8 and 0.9. As the particle size decreases within the indicated limits, the number of inter particle contacts may change by ten orders of magnitude.

The nomographic chart suggested by A.D. Zimon and Ye. D. Andrianov [XXXII, XXXV]
on the basis of the globular model of the porous structure of the dispersion system allows quantitatively estimating the average number of contacts among particles of various sizes, depending on the relative structural density (Fig.3).

To find the average number of contacts among dispersion particles per unit of volume, one can use the empirical expression derived with assistance from N. B. Ur'yev, V.N. Finanshin, and V. Ye. Chernomaz [XXIII]:

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Fig. 3: Zimon-Andrianov nomographic chart where $n$ is the average number of interparticle contacts per unit of volume, $\varphi$ is the relative density of compacted (compactable) asphalt concrete mix, $e$ is the Napierian base, $d_{\text{equiv}}$ is the characteristic particle size of the mineral part of asphalt concrete mix.

The interparticle distance is a no less important factor characterizing the conditions of interaction among the particles in the dispersed phase. The true strength of a single contact depends on the conditions of its formation; they determine the fixed equilibrium thickness of the liquid (bitumen) binder phase streak between mineral grains, first of all, in the formation of micro contact structure by fine-dispersed particles. This was confirmed in the various aspects of the porous body strength theory used in works by E. D. Yakhnin, A. F. Polak, N.B. Ur’yev, A.A. et al. [XXIII, XXXIV, XXV]. The difficulty with establishing the strict theoretical and functional relations among the critical shear stress, the average cohesive attraction of interparticle contact, and the average number of contacts per unit of volume ($P_m$ to $F_{\text{avg}}$ and $n$).

\begin{equation}
  n = 6.7373 \cdot 10^{10} e^{(5.44\varphi_1)} \cdot d_{\text{equiv}}^2
\end{equation}
stems from the fact that the derived regularities were theoretically substantiated and experimentally confirmed as the rule for monodisperse monomineral spherical particles [XXV]. However, in actual asphalt concrete mixes it was necessary to take into account the polydispersion and polyminerality of random odd-shaped particles. The average cohesive attraction of interparticle contact \( F_{\text{avg}} \) can be found by calculation using the experimental values of the critical shear stress and the known geometric parameters of the analyzed dispersion system according to E. D. Yakhnin’s refined formula [XXIII]. If to consider that \( \varphi_i = \rho_i / \rho \) and \( \varphi_p = \rho_p / \rho \), then

\[
F_{\text{avg}} = P_m \cdot d_{\text{equiv}}^2 \cdot \rho_i \cdot \frac{(\rho - \rho_p) \cdot (\rho_i - \rho_p)}{\rho^2 (\rho_i - \rho_p)}
\]  

(3)

Otherwise, expression (3) can be modified using relative densities and recorded as

\[
F_{\text{avg}} = P_m \cdot d_{\text{equiv}}^2 \cdot \rho_i \cdot \frac{(1 - \varphi_i) \cdot (1 - \varphi_p)}{\varphi_i \cdot (\varphi_i - \varphi_p)}
\]  

(4)

where \( \rho \) is the density of the mineral part of asphalt concrete, kg/m\(^3\); \( \varphi_i \) is the density of asphalt concrete; \( \rho_p \) is the density of asphalt concrete achieved by technology influences, kg/m\(^3\), \( \varphi_p \) is the average density of non-compacted asphalt concrete mix, kg/m\(^3\), that is found by experiment and depends on the average density and weight proportion of the original components of asphalt concrete mix; \( d_{\text{equiv}} \) is the equivalent size of particles in the mineral part of asphalt concrete mix; \( \varphi_i \) is the relative density achieved by exposing asphalt concrete mix to technology influences; \( \varphi_p \) is the relative density of absolutely crumbly asphalt concrete mix. Since the average single contact area is found proceeding from the strength additivity of elementary contacts of the dispersion system (1), the aggregate total area of elementary (single) contacts can be found as

\[
\sum S_i = \frac{1}{k_i} = \left( \frac{F_{\text{avg}}}{P_m} \right) \cdot n^{2/3}
\]  

(5)

Then, the average single contact area \( (S_i) \) at the overall number \( n \) of single contacts is

\[
S_i = \frac{\sum S_i}{n} = \left( \frac{F_{\text{avg}}}{P_m} \right) \cdot \frac{n^{2/3}}{n} = \left( \frac{F_{\text{avg}}}{P_m} \right) \cdot n^{-1/3}
\]  

(6)

The average single contact size can be found if the form in which this contact interacts with the mineral particle surface is known; in its respect, this depends on the formative conditions of the contact, mineral particle size and shape, bitumen viscosity, and a lot of other factors not yet yielding to direct quantification. For more sim-
plicity it is assumed that a coagulation inter particle contact forms along the round surface; in this case, the (average) size of a single elementary contact is

\[ d_i = \frac{4 \cdot S_i}{\pi} \]  \hspace{1cm} (7)

Average single contact density \( P_i \) is found as

\[ P_i = \frac{F_{avg}}{S_i} = P_m \cdot n^{2/3} \]  \hspace{1cm} (8)

The procedure of quantitatively estimating the structural formation of asphalt concrete mixes in the process phase and the destruction of the formed asphalt concrete structure affected by operational factors has been developed in the laboratory, production, and field environment of the laboratory of organic binders and asphalt concrete of the MADI together with the laboratory of highly concentrated dispersion systems of the Russian Academy of Sciences. For that purpose, the critical shear stress \( (P_m) \) was found by experimental methods. For example, fine-dispersed and micronized model compounds were exposed to rotational viscosimetry and conical plastometry [XXXI]. Framed compounds were processed by plate-bearing tests, standard methods, and the procedures suggested at different times by G. N. Kiryukhin [XIX], Yu. Ye. Nikolsky [XXII], V.M. Goglidze [V]. That way, the values of \( F_{avg}, n, P_i, d_i \) were found.

Various researchers have tried at different times to interpret the interaction among mineral particles via coagulation contacts from the standpoint of physical chemical mechanics.

According to S. Ya. Shalyt, N. V. Mikhaylov, and P. A. Rebinder [XXX], a particle of filler gets covered with a lime soap film, whereas bitumen becomes exponentially more viscous with the approximation to the particle from distance \( h_0 \) (Fig.4).

\[ \text{Fig. 4: Bitumen film structure in the contact area according to S. Ya. Shalyt, N. V. Mikhaylov, and P. A. Rebinder [XXX]} \]
The interaction in the contact area was explained in terms of energy (Fig. 5) by A. F. Polak [XXIII], B.V. Deryagin and L.D. Landau [III]. That said, $h_0$ was found for remote (around $10^{-7}$ m) and close coagulation (about $10^{-9}$ m).

A similar quality pattern of interaction between bitumen and mineral particles is exposed in several works by professor M. I. Volkov and I. V. Korolev [XVI, XX].

All this allows supposing that the average cohesive attraction of interparticle contact $F_{avg}$ is the quantitative estimate characterizing the quality of coagulation bonds. This force depends on the contact formation conditions under technology influences, and adhesion of bitumen and asphalt-coated aggregates. This parameter also allows estimating the results of the in-operation exposure of bitumen mineral material to external factors.

As found out by experimental investigations of asphalt concrete mix formation in the laboratories of the Department of Road Construction Supplies (MADI) and highly concentrated dispersion systems of the Institute of Physical Chemistry and Electrochemistry of the Russian Academy of Sciences (IPCHECH RAS), the most significant and structuring parameters are the compaction temperature and the compacting load intensity and action period.

The analysis of the findings has allowed suggesting the theoretical model of formation of contact interactions and micro-contact structure of bitumen mineral composites. It has been established that in the process phase the structural formation of asphalt concrete develops gradually (Fig. 6) and the optimal structures of bitumen mineral material form in the second phase.
The traits typical of dispersion particles of any size during the formation of structure and properties of highly concentrated dispersion systems, including bitumen mineral mixes, are manifested forces of interaction among the particles and between them and the environment. The kind, force, and energy of interaction depend on their nature, shape, surface and on the mineral particle weight (volume) and properties of bitumen. The surface phenomena at the phase boundary between the binder and the fine-dispersed filler are determinative to fine-dispersed mineral particles. The gravitational interaction and strength of particles proper have little significance. The determinative parameters of coarsely dispersed hard-grained systems are their strength and packing density. The role of the surface factors is subordinate [X, XX].

Various authors have suggested at different times various theoretical provisions of asphalt concrete structuring. According to P. V. Sakharov, I. A. Rybiev, M. I. Volkov, N. V. Gorelyshev, I. V. Korolev, Ya. N. Kovalev, etc., optimal structures form at a specific optimal content of organic binder.

In his doctoral thesis and subsequent publications Professor I. V. Korolev analyzed the changes in the thickness of bitumen films on the surface of mineral particles various in origin, structure, and size and suggested the concept of bitumen capacity, having related it to the unit surface of mineral particles, which allowed the author to suggest the calculation procedure for determining the optimal bitumen content in bitumen mineral materials [XVII, XXIV].

One may suppose that the optimal (maximum) values of average cohesive attraction of inter particle contact are achieved exactly at the bitumen capacity values experimentally validated by I. V. Korolev for fractions of various materials, which allows calculating extreme strength values of the whole dispersion system (e.g., the critical shear stress) in the accepted conditions of mineralogical and structural monodispersity.

For many years the procedure was used in road labs and multiple scientific investigations. However, the calculations made using this procedure would yield somewhat erroneously low values of optimal bitumen amounts as compared with experimental findings attained by trial mixtures for certain physical and chemical properties.
As found out in the multiple experiments conducted in the MADI and the IPCHECH, the main reason for this mismatch is that I. V. Korolev did not have reliable equipment at his disposal and determined the fraction bitumen capacity by tests on particles larger than 71 μm in size.

It is known that the fine-dispersed filler (mineral powder) forming the microstructure of asphalt concrete contains fine-dispersed particles of 1 μm in size and even smaller. These particles are the ones with the best developed surface ensuring the physical chemical interaction with organic binder.

According to the design comparison of unit surfaces using the procedure from [XXIII, XXI], other things equal, the relative share of mineral powder surface in the total unit surface of mineral particles rises from 80 – 85 to 95 – 98%.

It is known that various researchers have tried to analytically describe the grain sizing of powders and inorganic binders this index cannot be estimated by the sieve method. For example, V. Z. Pirotsky [XXVI] estimated the activity and other properties of class A cement and related the sizes of its particles (smaller than 3 μm included) with the energy spent on grinding. However, it is inexpedient to suggest these procedures for broad application in on-job labs because determining the grain composition of fine-dispersed materials is labor-intensive and production labs have no equipment necessary for the purpose.

III. Results

The optimal bitumen content determined by calculation using to the procedure tested in the laboratory of organic binders and asphalt concrete of the MADI is highly convergent with the test results.

For that purpose it was necessary to determine the bitumen capacity of particles of 1 to 71 μm in size for materials of various chemical mineralogical composition and genetic origin. The source information used was the bitumen capacity values (Fig. 7) found by test by I. V. Korolev [VII, XVI].

To achieve the goal, the authors decided to use statistical methods of mathematical forecasting.

The source information used to determine the bitumen capacity of fine-dispersed particles were the bitumen capacity values found by test by I. V. Korolev [XVII, XXI] and A. M. Gridchin [VIII, VII].

The most exact and adequate description of that dependence was found on the computer by the correlation and regression analysis of various built mathematical models.

The expressions with a correlation factor below 0.70 were not considered in the further analysis. Several linear, exponential, power law, polynomial, and more complex mathematical models were tried and analyzed for various asphalt-coated aggregates.

The regression models built for each material in semi-logarithmic coordinates on the computer bind mineral particle sizes with bitumen capacity indicators.
Table 1: Regression model multiple correlation coefficients found for various materials

| Regression model | 1   | 2   | 3   | 4   | 5   | 6   | 7   | 8   | 9   | 10  | 11  | 12  | 13  | 14  | 15  |
|------------------|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|
| Dense lime stone | 0.9 | 0.9 | 0.9 | 0.8 | 0.8 | 0.8 | 0.8 | 0.9 | 0.9 | 0.9 | 0.9 | 0.9 | 0.9 | 0.9 | 0.9 |
| Granite M-1200  | 0.69 | 0.53 | 0.85 | 0.64 | 0.865 | 0.84 | 0.77 | 0.779 | 0.87 | 0.55 | 0.87 | 0.88 | 0.78 | 0.66 | 0.51 |
| Diorite M-1200  | 0.95 | 0.95 | 0.95 | 0.76 | 0.76 | 0.76 | 0.76 | 0.76 | 0.76 | 0.76 | 0.76 | 0.76 | 0.76 | 0.76 | 0.76 |
| Sandstone M-600 | 0.79 | 0.79 | 0.79 | 0.79 | 0.79 | 0.79 | 0.79 | 0.79 | 0.79 | 0.79 | 0.79 | 0.79 | 0.79 | 0.79 | 0.79 |
| Lime shell stone | 0.63 | 0.63 | 0.63 | 0.63 | 0.63 | 0.63 | 0.63 | 0.63 | 0.63 | 0.63 | 0.63 | 0.63 | 0.63 | 0.63 | 0.63 |
| Silica sand      | 0.77 | 0.77 | 0.77 | 0.77 | 0.77 | 0.77 | 0.77 | 0.77 | 0.77 | 0.77 | 0.77 | 0.77 | 0.77 | 0.77 | 0.77 |
| Barn dolomite dust | 0.72 | 0.72 | 0.72 | 0.72 | 0.72 | 0.72 | 0.72 | 0.72 | 0.72 | 0.72 | 0.72 | 0.72 | 0.72 | 0.72 | 0.72 |
| Smelting slag M-1000 | 0.67 | 0.67 | 0.67 | 0.67 | 0.67 | 0.67 | 0.67 | 0.67 | 0.67 | 0.67 | 0.67 | 0.67 | 0.67 | 0.67 | 0.67 |
| Basic smelting slag M-600 | 0.5 | 0.5 | 0.5 | 0.5 | 0.5 | 0.5 | 0.5 | 0.5 | 0.5 | 0.5 | 0.5 | 0.5 | 0.5 | 0.5 | 0.5 |
| Acid smelting slag M-1000 | 0.4 | 0.4 | 0.4 | 0.4 | 0.4 | 0.4 | 0.4 | 0.4 | 0.4 | 0.4 | 0.4 | 0.4 | 0.4 | 0.4 | 0.4 |
| Diorite          | 0.39 | 0.39 | 0.39 | 0.39 | 0.39 | 0.39 | 0.39 | 0.39 | 0.39 | 0.39 | 0.39 | 0.39 | 0.39 | 0.39 | 0.39 |
| Low quartzite    | 0.27 | 0.27 | 0.27 | 0.27 | 0.27 | 0.27 | 0.27 | 0.27 | 0.27 | 0.27 | 0.27 | 0.27 | 0.27 | 0.27 | 0.27 |
| Crystalline schist | 0.22 | 0.22 | 0.22 | 0.22 | 0.22 | 0.22 | 0.22 | 0.22 | 0.22 | 0.22 | 0.22 | 0.22 | 0.22 | 0.22 | 0.22 |
| Gravel-like      | 0.21 | 0.21 | 0.21 | 0.21 | 0.21 | 0.21 | 0.21 | 0.21 | 0.21 | 0.21 | 0.21 | 0.21 | 0.21 | 0.21 | 0.21 |
| Lime stone       | 0.03 | 0.03 | 0.03 | 0.03 | 0.03 | 0.03 | 0.03 | 0.03 | 0.03 | 0.03 | 0.03 | 0.03 | 0.03 | 0.03 | 0.03 |

Multiple correlation coefficients of regression models of changes in bitumen capacity against dispersion, found for various materials

Fig. 7: Correlation coefficients of various regression dependences attained for mineral powders of various materials

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According to the calculated data analysis, the least accurate results are derived using the linear and the logarithmic dependence and the most accurate results are observed in the polynomials of the fourth and higher orders. At the same time, the correlation coefficient found by applying the power law dependence to the materials various in genesis, nature, mineralogy, and petrography in a broad range of depressiveness varies from 0.27 to 0.99.

That said, the probable metrological error in the test conducted by I. V. Korolev [VII, XVI] and A. M. Gridchin [VIII, VII] was taken into account as well.

As shown by the multiple correlation coefficients of regression models found for various materials and exposed in Table 1 and Fig. 7, the dependence describing the bitumen capacity changes in various materials most accurately and adequately is

\[ BT_i = b_0 \cdot d^{b_1} \]  \hspace{1cm} (9)

where \( BT_i \) is the fraction bitumen capacity; \( d_i \) is the material particle size, mm, \( b_0 \) and \( b_1 \) are the regression coefficients. The multiple correlation coefficient of the bitumen capacity index for various materials was 0.86 – 0.98. It has been found out that the materials produced from volcanic rock and man-made waste with a higher density and strength (diorite M-1200, dense lime stone M-1000, smelter slag M-600-1000), granite M1200 have the multiple correlation coefficient of at least 0.95.

At the same time, the correlation coefficient among the sedimentary porous structured materials ranged from 0.85-0.88. It follows from their practical application in asphalt concrete mixes that these porous materials combined with bitumen show good adhesive properties.

**Table 2: Predicted bitumen capacity of geological materials with particle depressiveness taken into account**

| Particle size, mm | Dense lime stone M-1000 No. 1 | Granite M-1200 No. 2 | Diorite M-1200 No. 3 | Sand stone M-600 No. 4 | Lime shell stone M-400 No. 5 |
|-------------------|-------------------------------|----------------------|----------------------|------------------------|-------------------------------|
| 25                | 2.2                           | 4.02                 | 2.96                 | 3.39                   | 2.95                          |
| 15                | 2.5                           | 4.23                 | 3.26                 | 3.64                   | 2.5                           | 3.21                         |
| 10                | 2.9                           | 2.7                  | 4.5                  | 4.41                   | 3.3                           | 3.51                         | 4.0 | 3.84 | 3.9 | 3.43 |
| 5                 | 3.0                           | 3.2                  | 4.7                  | 4.73                   | 4.0                           | 4.0                          | 4.5 | 4.22 | 4.6 | 3.85 |

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The resulting mathematical expressions were used to predict by calculation the bitumen capacity of various materials, added to as fine-dispersed fillers to asphalt concrete, for particles of 1 to 40 μm in size.

The results of calculating the bitumen capacity, including predicted values for fine-dispersed particles, are provided in Figs. 8 and 9 and Table 2.

According to the comparison of the calculated and the test values of bitumen capacity of various materials, the largest absolute and relative deviations are observed for particles smaller than 0.071 mm.

| 2.5 | 3.2 | 3.7 | 5.2 | 5.08 | 4.5 | 4.55 | 4.8 | 4.64 | 4.9 | 4.32 |
|-----|-----|-----|-----|------|-----|------|-----|------|-----|------|
| 1.25 | 4.6 | 4.3 | 5.6 | 5.45 | 5.6 | 5.18 | 5.0 | 5.09 | 4.9 | 4.85 |
| 0.63 | 5.3 | 5.0 | 5.7 | 5.84 | 5.9 | 5.88 | 5.2 | 5.59 | 5.0 | 5.43 |
| 0.315 | 6.0 | 5.8 | 5.9 | 6.27 | 7.0 | 6.69 | 5.4 | 6.14 | 5.4 | 6.10 |
| 0.14 | 7.0 | 6.9 | 6.4 | 6.81 | 7.9 | 7.78 | 5.8 | 6.85 | 6.2 | 6.98 |
| 0.071 | 7.3 | 8.7 | 7.4 | 7.30 | 8.7 | 8.83 | 8.5 | 7.51 | 7.7 | 7.81 |
| 0.036 | 9.4 | 9.2 | 8.4 | 7.8 | 9.5 | 10.0 | 9.3 | 8.2 | 10.1 | 8.7 |
| 0.018 | 10.7 | 8.40 | 11.4 | 9.1 | 9.8 |
| 0.009 | 12.5 | 9.0 | 13.0 | 9.9 | 11.0 |
| 0.005 | 14.2 | 9.6 | 14.5 | 10.8 | 12.2 |
| 0.002 | 17.3 | 10.5 | 17.2 | 12.2 | 14.2 |
| 0.001 | 20.1 | 11.3 | 19.5 | 13.4 | 15.9 |
| 0.001 | 23.3 | 12.1 | 22.2 | 14.7 | 17.8 |

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### Table 2: (continued)

| Particle size, mm | Material | Silica sand No. 6 | Burnt dolomite dust No. 7 | Smelter slag M-1000 No. 8 | Basic smelter slag with silicate decomposition products M-600 No. 9 | Acid steelmaking slag M-1000 No. 10 |
|-------------------|----------|-------------------|---------------------------|---------------------------|---------------------------------------------------------------|-----------------------------------|
| 25                |          |                   |                           |                           | 4.6                                                           | 4.6                               |
| 15                |          |                   |                           |                           | 5.1                                                           | 5.1                               |
| 10                | 2.9      | 11.9              | 4.0                       | 4.2                       | 5.6                                                           | 5.5                               | 5.6                               | 5.5                               |
| 5                 | 3.6      | 12.9              | 4.8                       | 4.7                       | 6.1                                                           | 6.2                               | 6.1                               | 6.2                               |
| 2.5               | 2.9      | 4.4               | 14.1                      | 5.4                       | 5.3                                                           | 7.2                               | 7.0                               | 7.2                               | 7.0                               |
| 1.25              | 4.3      | 5.5               | 13.5                      | 15.3                      | 6.2                                                           | 5.9                               | 7.8                               | 7.9                               | 7.9                               |
| 0.63              | 4.5      | 6.8               | 14.3                      | 16.6                      | 6.8                                                           | 6.6                               | 9.3                               | 9.0                               | 9.3                               | 9.0                               |
| 0.315             | 4.5      | 8.5               | 15.2                      | 18.0                      | 7.5                                                           | 7.4                               | 10.5                              | 10.2                              | 10.5                              | 10.2                              |
| 0.14              | 5.4      | 11.0              | 15.6                      | 19.8                      | 8.1                                                           | 8.4                               | 11.2                              | 11.8                              | 11.2                              | 11.8                              |
| 0.071             | 8.1      | 13.6              | 16.0                      | 21.5                      | 9.0                                                           | 9.3                               | 12.0                              | 13.3                              | 12.0                              | 13.3                              |
| 0.036             | 14.0     | 16.8              | 22.5                      | 23.3                      | 10.4                                                          | 10.4                              | 13.5                              | 15.1                              | 13.5                              | 15.1                              |
| 0.018             | 20.9     |                  | 25.4                      | 11.6                      | 20.5                                                          | 17.1                              | 20.5                              | 17.1                              |                                  |                                   |
| 0.009             | 26.0     |                  | 27.6                      | 12.9                      |                                                              | 19.3                              |                                  |                                   |                                  |                                   |
| 0.005             | 31.3     |                  | 29.6                      | 14.2                      |                                                              | 21.5                              |                                  |                                   |                                  |                                   |
| 0.002             | 41.8     |                  | 33.0                      | 16.4                      |                                                              | 25.3                              |                                  |                                   |                                  |                                   |
| 0.001             | 52.0     |                  | 35.8                      | 18.3                      |                                                              | 28.7                              |                                  |                                   |                                  |                                   |
| <0.001            | 64.7     |                  | 38.9                      | 20.4                      |                                                              | 32.5                              |                                  |                                   |                                  | 32.5                              |
This is due to the fact that I. V. Korolev did not differentiate the bitumen capacity values for smaller particles. The test values found by him are the integral characteristic of the whole highly dispersed range of particles smaller than 71 μm.

**Table 2: (continued)**

| Particle size (mm) | Material | Diabite No. 11 | Low-ore quartzite No. 12 | Crystalline schist No. 13 | Gravel-like No. 14 | Lime stone No. 15 |
|-------------------|----------|----------------|--------------------------|--------------------------|-------------------|-----------------|
| 25                |          | 2.5            | 2.2                      | 3.1                      | 2.20              | 2.25            |
| 15                |          | 2.9            | 2.5                      | 3.4                      | 2.51              | 2.58            |
| 10                |          | 2.8            | 3.2                      | 2.6                      | 2.8               | 2.7             |
| 5                 |          | 3.8            | 3.9                      | 3.8                      | 4.1               | 3.7             |
| 2.5               |          | 5.6            | 4.6                      | 4.3                      | 5.0               | 4.9             |
| 1.25              |          | 5.9            | 5.5                      | 4.5                      | 5.2               | 5.6             |
| 0.63              |          | 7.0            | 6.6                      | 4.9                      | 5.4               | 6.3             |
| 0.315             |          | 7.9            | 7.9                      | 5.6                      | 5.8               | 7.3             |
| 0.14              |          | 8.7            | 9.8                      | 7.5                      | 8.3               | 8.5             |
| 0.071             |          | 9.5            | 11.6                     | 8.7                      | 9.2               | 9.7             |
| 0.036             |          | 16.5           | 13.9                     | 14.0                     | 14.5              | 11.0            |
| 0.018             |          | 16.6           | 13.3                     | 12.6                     | 12.0              | 14.9            |
| 0.009             |          | 19.9           | 15.8                     | 14.4                     | 17.9              | 18.8            |
| 0.005             |          | 23.2           | 18.3                     | 16.1                     | 20.9              | 22.0            |
| 0.002             |          | 29.4           | 22.9                     | 19.2                     | 26.7              | 28.1            |
| 0.001             |          | 35.1           | 27.2                     | 21.9                     | 32.1              | 33.8            |
| <0.001            |          | 42.1           | 32.4                     | 25.0                     | 38.5              | 40.7            |

Notes to Table 2: the experimental values of partial residues are presented in column 1, whereas the calculated values are presented in column 2.
Fig. 8: Predicted bitumen capacities of dispersion particle components (according to the test data from Professor I. V. Korolev)

A lot of algorithms and programs developed in recent years allow designing the grain composition of asphalt concrete mixes on the computer quickly and reliably.

That said, the probable metrological accuracy of the test conducted by I. V. Korolev [XVII, XVI] was also taken into account.

For the results of determining by calculation the bitumen capacity of various materials added to asphalt concrete as fine-dispersed fillers with the design prediction of values for dispersion particles of 1 to 40 μm in size see Figs. 10–12.

The calculated and test values of bitumen capacity in various materials are compared in Table 4. The largest absolute and relative deviations are observed for particles smaller than 0.071 mm in size. This stems from the fact that I. V. Korolev did not differentiate the bitumen capacity values for smaller particles and the test values found by him are the integral characteristic of the whole highly disperse range of particles smaller than 71 μm.
A lot of algorithms and programs developed in recent years allow designing asphalt concrete mixes on PCs quickly and reliably.

The suggested procedure allows determining by calculation the optimal bitumen content in asphalt concrete mix, fits well within the asphalt concrete mix design procedure accepted in Russia and other CIS states, makes possible more reliable calculations, and multiply increases the labor output of employees at road building labs.

Thus, to produce strong asphalt materials resistant to loads, temperature effects, water, alternate freezing and thawing, chemical agents, and other factors, one should choose mineral components, taking into account the surface activity and porosity of mineral powder, sand, and breakstone. Other things equal, equal results can be attained at an increased surface activity of grains of sands and breakstone (e.g., breakstone from lime stone, dolomite, blast-furnace slag) by using coarser mineral powder or from less active materials. When granite breakstone, siftings, and silica sand (i.e., acid materials) are used, the mineral powder shall be basic by nature, conform to quality requirements, and be finer-grained.
The structural parameters and properties of the highly concentrated dispersion system are determined via the unit surface of dispersion phase [XIX].

The dispersion phase unit surface is one of the determinative factors making it possible to explain the formation and destruction pattern of highly concentrated dispersion systems to which bitumen mineral materials belong as well. There are methods of determining the unit surface of solid dispersion materials based on air and water impermeability, adsorption of oil and colorants, and also design methods [XV, VI].

A significant practical interest is awakened by the design methods of estimating the unit surface of the mineral part of asphalt concrete mixes. The relation suggested by several researchers [VIII, XVII, XVI] at different times allow determining the unit surface of the mineral part of asphalt concrete mixes by calculation under known assumptions.

For example, the expression suggested by I. V. Korolev [XVI] for determining the unit surface of fraction \( I \) is

\[
S_i = \frac{a}{b} \cdot \frac{1}{d_i} \cdot \frac{1}{\rho_i}
\]  

(10)

where \( S_i \) is the fraction unit surface; \( a \) is the grain surface coefficient; \( b \) is the volume coefficient of the grains of mineral material; \( \rho_i \) is the density of the grains of the fraction’s mineral material; \( d_i \) is the fraction particle size.

The \( \frac{a}{b} \) ratio varies from 6 to 14.696, which depends on the grain shape (see Table 1.4).

Fig. 10: Predicted bitumen capacity of dispersion particle components (according to the experimental data from Professor A. M. Gridchin)
It follows therefrom that, other things equal, the design unit surface of mixes with cubical and tetrahedral particles is larger than the design unit surface of mixes with ball-like particles by 40 and 140 %, respectively.

### Table 3: Mineral particle shape coefficient

| Mineral material grains | \( \frac{a}{b} \) ratio |
|-------------------------|-------------------------|
| Ball-like               | 6.0000                  |
| Cubical                 | 8.4853                  |
| Tetrahedral             | 14.6960                 |

**Fig. 11:** Mineral particle shape coefficient

Several Russian and foreign researchers (S. Pashkovsky, M. Duriez, Goled, S. Voydanovsky, etc.) have suggested at different times their own design procedures of determining the unit surface of asphalt concrete mixes; however, in terms of accuracy and reproducibility these methods are inferior to the relation used by I. V. Korolev.

In all of the procedures it was necessary to take into account the disperse phase mineral particle density.

In this case relation (10) is recorded as

\[
S_i = k_\phi \cdot k_\rho \cdot \frac{1}{d_i}
\]  

(11)

where \( k_\phi \) is the shape coefficient and \( k_\rho \) is the density coefficient.
Table 4: Mineral particle density coefficient

| Mineral particle density, kg/m³ | 2400 | 2500 | 2600 | 2700 |
|-------------------------------|------|------|------|------|
| Density coefficient, $k_\rho = \frac{\rho}{2.5}$ | 0.97 | 1.00 | 1.03 | 1.08 |

For the plot of changes in the density coefficient against the mineral particle density of the mineral components of asphalt concrete mix see Fig. 12.

**Fig. 12:** Changes in the mineral particle density coefficient in expression

It is quite obvious that these design relations allow finding only an approximate averaged result, which is due to the impossibility to clearly take into account the shape of actual particles various in size and quantitatively estimate the degree of its approximation to ball-like, cubic or tetrahedral shape.

It is difficult to take into account the polydispersion of particles and possible degree of aggregation of high-dispersion mineral powder particles. Nonetheless, however, the indicated relations can be used to a certain degree of reliability for analytically comparing the structural mechanical properties of asphalt concrete and asphalt concrete mixes.

The aggregate unit surface of the dispersion phase, in other words, mineral part of asphalt concrete mixes, is found as

$$S_m = \sum_{i=m}^{\infty} k_\phi \cdot k_\rho \cdot \frac{1}{d_i \cdot 100}$$

(12)

Taking into account the indicated assumptions, the above specified relations were used multiple times to comparatively calculate the design unit surface of various kinds and types of asphalt concrete mixes. The limit curves regulated by GOST 9128 were used for the purpose. The design procedure consisted in determining total and partial residues of grain compositions of mixes of all kinds and types regulated by the standard against both, the upper and the lower limit. The unit surface of mineral par-
particles was determined for each fraction. Then the unit surfaces of all the materials were summed for all fine fractions from 40 mm to 0.071 μm.

The calculations performed using this procedure together with the test check of the designed compositions, followed by the trial mixture, yielded erroneously low results. As a rule, those compositions had a somewhat higher porosity and water absorption than the compositions selected following the standard procedure.

The formation and destruction pattern of highly concentrated dispersion systems, bitumen mineral materials included is explained by analyzing the change in the dispersion phase unit surface. The unit surface of the mineral part of asphalt concrete mix is found by relation (12).

As noted above, the design methods do not allow finding absolutely accurate solutions, which is due to the uncertainty in estimating the density and grain shape of particles of each size, petrographic and mineralogical peculiarities of the source subsurface rock in use, and several other factors. The calculated data are heavily affected by the mineral powder depressiveness. It is known that powder forms the structure of asphalt binder, which heavily affects all the physical mechanical properties of asphalt concrete mixes. It has been found out over the years that mineral powder performs the following main functions in asphalt concrete:

1. fine-dispersed filler of micro voids that makes the mineral aggregate of asphalt concrete denser;
2. having a very developed surface (90 to 98 % of the total surface of mineral particles contained in asphalt concrete), mineral powder forms asphalt binder and encourages the formation of a structured dispersion system;
3. highly dispersed mineral particles of the filler interact across the interlayers of low structured bitumen, whereas the change in viscosity is described by Einstein’s equation [I], which converts bitumen from the volumetric to the film-like state.

The depressiveness of mineral powder is characterized by its grain composition and unit surface.

The powder grain-size composition is determined by the sieve method or sedimentation. The former is easy to implement and included in regulatory instruments in force. Its main weakness is the impossibility to estimate the weight ratio of particles smaller than 71 μm in size. The second method requires using more complex laboratory equipment and hiring high skilled specialists; it is more labor-intensive and less productive, which does not allow recommending it for mass-scale application in construction labs.

In the standard procedure the grain composition of the mineral powder for asphalt concrete mixes is estimated by the sieve method making it possible to estimate the weight ratio of particles of 2.5 to 0.071 mm in size. One of the main quality criteria for the powder is the number of particles smaller than 0.071 mm. To quantitatively estimate the unit surface of the mineral part of asphalt concrete mix, it is necessary to build the full curve characterizing the powder grain composition, including highly dispersive particles smaller than 1 μm.

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It is known that there have been attempts to analytically describe the grain sizing of powders and inorganic binders when the description by the sieve method is impossible.

It was supposed that the full grain composition curve of the fine-dispersed filler could be described by a mathematical expression. The experimental data obtained by I. I. Dodkhoev in the laboratory of D. Mendeleev University of Chemical Technology of Russia [IX] were used for checking. For the results see Table 5.

The regression mathematical expressions derived by mathematical processing describe the changes in the grain composition of lime stone mineral powder, class A cement, and blast-furnace slag cement. It has been found out that the change in the grain sizing of fine-dispersed materials is described most accurately and adequately by second-degree polynomial (13). That said, the multiple correlation coefficient is 0.97 to 0.98.

\[ A_i = b_0 + b_1 \cdot d_i + b_2 \cdot d_i^2 \]  

(13)

where \( b_0, b_1, \) and \( b_2 \) are the regression coefficients, \( A_i \) are the total residues, % wt., \( d_i \) is the particle size in mm or μm. This has allowed supposing that the changes in the grain composition of fine-dispersed mineral powder are also subject to the derived regularity in a broad particle dispersion range. The resulting total residue values allow finding the partial residues in wt. % and then determine the unit surface of the fine-dispersed component of the mineral part of asphalt concrete mix and the total unit disperse phase surface of the highly concentrated dispersion system.
Table 5: Grain compositions of mineral powder, class A cement, and blast-furnace slag cement according to the data from I. I. Dodkhoev

| $d_i$ | Mineral powder | Class A cement | Blast-furnace slag cement |
|-------|----------------|----------------|---------------------------|
|       | $a_i$ | $A_i$ | $A_i$ abs. | $a_i$ | $A_i$ | $A_i$ abs. | $a_i$ | $A_i$ | $A_i$ abs. | $a_i$ | $A_i$ | $A_i$ abs. |
| 0,63  | 0,00  | 0    | 0    | 0    | 0,00  | 0    | 0    | 0    | 0,00  | 0    | 0    | 0    |
| 0,315 | 0,00  | 0    | 3    | -3   | 0,00  | 0    | 0    | 0    | 5,00  | 5    | 14   | 11   |
| 0,14  | 10,00 | 10   | 14   | -1   | 0,00  | 0    | 20   | -10  | 0,00  | 0    | 14   | 11   |
| 0,071 | 13,62 | 24   | 25   | 3    | 0,00  | 13,79| 24   | 14   | 0,00  | 0    | 25   | 11   |
| 0,036 | 10,58 | 34   | 36   | 11   | 0,00  | 25,39| 49   | 13   | 10,51 | 21   | 36   | 11   |
| 0,018 | 6,32  | 41   | 47   | -5   | 0,00  | 9,67 | 59   | 12   | 4,78  | 25   | 47   | 11   |
| 0,009 | 19,35 | 60   | 59   | 12   | 8     | 19,61| 71   | 11   | 13,50 | 39   | 59   | 12   |
| 0,005 | 19,71 | 80   | 70   | 12   | 8     | 10,14| 89   | 10   | 27,21 | 66   | 70   | 12   |
| 0,002 | 8,07  | 88   | 82   | 12   | -4    | 5,79 | 94   | 9    | 18,37 | 84   | 82   | 12   |
| 0,001 | 8,17  | 96   | 95   | 12   | -4    | 3,35 | 98   | 8    | 9,92  | 94   | 95   | 12   |
| 0,0005| 4,18  | 100  | 100  | 5    | -1    | 2,26 | 100  | 100  | 1,26  | 5,64 | 100  | 100  |

Notational conventions in Table 2.3: $d_i$ is the particle size in mm, $a_i$ is the partial residue on sieve in wt. %, $A_i$ is the total sieve residue in wt. %.

Thereunder, a design and test procedure of determining the grain composition of mineral powder was suggested [IV]. In the first phase the powder grain composition is determined according to the standard procedure. The experimental results are approximated on the computer to sizes of up to 0.001 mm.

The mathematical tools used for this purpose include correlation and regression analysis, which allows having the mathematical description and estimating the statistical reliability of the derived mathematical expressions of the experimental curve and its prediction part as well as the statistical characteristics that allow estimating the prediction for accuracy and adequacy.

The mathematical expression thus found is used to calculate total and partial residues of mineral powder particles of 2.5 to 0.001 (fine-disperse) mm in size. The design values of partial and total residues of particles of 2.5 to 0.071 mm in size are compared with the experimental data obtained according to the procedure, and then the prediction error is estimated. The calculations can be performed using linear, loga-
rithmic, exponential, superposed models as well as polynomial models of the second and higher orders.

As confirmed by the lab tests and processing of the results according to the suggested procedure, the satisfactory accuracy and adequacy for actual mineral powders are found while describing the grain composition of mineral powders by the regression model as a second-order polynomial (Table 5).

IV. Discussion

It has been found out by experiment that the increase in the highly dispersive filler content within 5 to 20 % wt. monotonously increases the density by 10 %. That said, the average number of interparticle contacts per unit of volume increases by 80 % and the average mineral particle contact size decreases by 120 %. These structural changes monotonously increase the critical shear stress; depending on the test temperature and speed, this stress may increase by 50 to 170 %, which is explained, first of all, by an increased number of interparticle contacts. The average cohesive attraction of interparticle contact monotonously decreases by 50 to 120 %. In our opinion, this is thermodynamically related to changes in the power conditions of structuring. Other things equal, with the high-dispersion phase content increasing, the total compacting load is distributed across a larger number of elementary contacts resisting structural state changes, and the formation of each single contact requires a weaker compacting effort. The integral strength of the highly concentrated dispersion system increases mainly through the advanced increase in the number of contacts. The average strength of single contact at all the test temperatures and speeds monotonously rises by 40 to 200 %. This circumstance corresponds well with the inferences of P. A. Rebinder, N. V. Mikhaylov, and S. Ya. Shalyt [XXX] about changes in the binder viscosity in the near-boundary interface between the dispersion medium and the dispersion phase and with the ideas of I. V. Korolev [XVI] that the structure of asphalt concrete contains volumetric and film-like bitumen.
Fig. 13: Changes in the structural mechanical parameters of asphalt concrete against the fine-dispersed filler content at different bitumen viscosities

Fig. 14: Influence of the mineral powder content on the critical shear stress (a), average cohesive attraction of interparticle contact (b), and average strength of one contact (c)

The bitumen viscosity significantly affects the structuring and formation of properties of asphalt concrete mixes. The viscosity of organic binders depends on the shear speed and temperature.

In the formation of asphalt concrete the bitumen viscosity at process temperatures determines the mobility and place ability of asphalt concrete mixes and, therefore, the energy level of structuring and the configuration and formative conditions of elementary contacts.

The changes in the petroleum asphalt viscosity in a broad range of process and operating temperatures were studied by experiment (Fig. 14).

With bitumen becoming more viscous, the density, average number of contacts per unit of volume, and average single contact area in mixes do not change significantly.
It has been found out that in the course of structuring the structural mechanical characteristics heavily depend on the bitumen viscosity and may change by 1.5 to 2 orders (Fig. 15). It is known that, in its respect, the viscosity of bitumen depends on its grade and the gradient of shift and temperature. The assumptions set forth above were confirmed by the statistical modeling by correlation and regression analysis.

This circumstance corresponds well with the inferences of P. A. Rebinder, N. V. Mikhailov, and S. Ya. Shalyt [XXX] about changes in the binder viscosity in the near-boundary interface between the dispersion medium and the dispersion phase and with the ideas of I. V. Korolev [XVI] that the structure of asphalt concrete contains volumetric and film-like bitumen.

V. Conclusion

1. The highly concentrated dispersion system structuring theory and peculiarities of contact interactions should be taken into account in the guided regulation of the structuring of asphalt concrete and asphalt concrete mixes.

2. This study has allowed finding quantitative relations of the indicators of physical mechanical and structural rheological properties of highly porous bitumen sand mixes formed by compaction and dense kinds of asphalt concrete various in composition on various binders to the process parameters of compaction.

3. The theoretical and experimental work done in the study has allowed discovering quantitative links between the structuring and destructive factors and the structural mechanical and construction engineering properties of asphalt concrete in road coverings. It has been established that the attainment of standardized asphalt concrete density at the compaction of asphalt concrete mixes in the structural layers of road re-
vetment does not guarantee that the whole set of required physical mechanical properties and material structure meeting the operational conditions will be attained.

4. The structural changes in asphalt concrete in road coverings in use under the influence of operational factors are physical chemical in nature and reliably described in terms of quantity, taking into account the basic provisions of physical chemical mechanics and the contact interaction theory.

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