Rheological characteristics of bitumen mastic depending on composition and filler dispersity

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Rheological characteristics of bitumen mastic depending on composition and filler dispersity

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Abstract. As fillers in asphalt concrete are used for bitumen viscosity increase, structure modification it is necessary to estimate the filler structuring effect in bitumen binder. For this purpose the influence of phase and size parameters of fine-dispersed powdered fillers on rheological characteristics of bitumen mastics (bitumen–filler composition) is studied. Maximal effect for bitumen properties is observed for such fillers as kaolin and chalk-stone. These two fillers have the highest specific surface area (SSA) and the highest void content. So, they are able to absorb a higher content of bitumen vs. fillers with lower SSA and void content values. Variations in Delta Ring and Ball values for kaolin- and chalk stone-based mastics at 83°C and 24°C, respectively are significant vs. other compositions (up to 8–10°C). It can be associated with structuring effect under high temperature. Significant change in bitumen physical and mechanical properties can be occurred when kaolin application is connected with thin flake form of its particles. Study of rheological characteristics of bitumen mastics by the dynamic shift remoter under high temperatures demonstrates lower values of complex modulus for bitumen mastics containing lime stone and sand fillers with medium and high dispersity vs. chalk-stone- and kaolin based analogues.

1. Introduction

Fillers added to asphalt concrete are fine-dispersed powders, produced by grinding of rocks or powdered industrial by-products. They are used as modifier for bitumen to increase the binder viscosity (i.e. structuring effect) as well as to expand of operating temperature range. The resulting structured disperse system acts as binding component in bitumen concrete. It leads to reducing of creep deformation (plastic yield) and increasing of bitumen admixture. It is generally believed, that structuring capacity of filler is associated with highly-developed surface.

However, it should be noted that not all disperse materials similarly effect on bitumen due to the following reasons: mineral and chemical compositions, dispersity, particle shape and surface reactivity of them. These parameters are interacted, so it difficult to determine particular effect of each parameter on general structuring effect of asphalt-based materials.

Degree of structuring of organic binder containing filler is associated with particle distribution in bulk i.e. packing density. Also this parameter could be determined by particle-size distribution i.e. filler dispersity as well as particle shape [1, 2]. In case of multimodal particle distribution it is the number of interpartial contacts, packing density of them and strength of system [3]. Due to complex

interdependencies between such parameters as particle-size distribution, particle shape and packing density, it is easier to measure packing density than to calculate it. Packing density is characterized by interpartial pores presence, that generally determined by Rigden voids method [4].

According to Rigden theory filler can keep some quantity of bitumen in pore space [5] that initiates increasing of bitumen viscosity. Extra bitumen content is called «free bitumen». This quantity depends on type of filler. So, fillers those are differed in composition and dispersity have different content of air pores in «dry» packing density and different amount of «fixed» bitumen in admixture. That leads to difference in structuring capacity that should be determined according to rheological characteristics.

2. Experimental Part

2.1. Materials

Fillers, applied in this work, were obtained by grinding of different types of rocks such as carbonates (limestone and chalk-stone), silicates (hydrothermal quartz, kaoline). According to XRD-analysis, carbonates consists of calcite $CaCO_3$, hydrothermal quartz – quartz SiO_2, and kaoline – kaolinite ($Al_4[Si_4O_{10}](OH)_8$) as well as nacrite, dickite, halloysite. The above materials are «pure» or monomineral. Construction bitumen BND 60/90 (Russia) was used as binding component.

2.2. Methods.

Real density was measured with helium pycnometer Pycnomatic ATC.

Dispersity of fillers is determined by following parameters:

– Particle-size distribution and specific surface area measured with laser particle analyzer FRITSCH Analysette 22 NanoTec plus;

– Specific surface area measured by Blaine method as well as by BET method based on nitrogen adsorption using SORBI-M equipment.

Mastics for test procedure were prepared by joint mixing of bitumen and fillers at 150–160 °C, stirring rate is 100 r/min with following component ratio (%): filler/bitumen – 37.5 /62.5 (Table 1).

Table 1. Composition of bitumen mastics with different fillers

Filler		Limestone medium-dispersed LM	Limestone fine-dispersed LF	Chalk-stone C	Hydrothermal quartz medi-um-dispersed QM	Hydrother-mal quartz fine-dispersed QF	Kaoline K
bitu-men/filler ratio	vol. %,	62.5/37.5					
	wt. %,	38.3/61.7	38.3/61.7	38.2/61.8	38.8/61.2	38.6/61.4	39.2/60.8

Rheological characteristics of mastics, obtained by mixing of bitumen and filler were measured with standard methods (penetration test, melting point-test). Variation in penetration and melting point was fixed as difference between bitumen and mastics values. Flow behavior of bitumen and based mastics were studied in wide temperature range using rotational viscometer Rheotest RN4.1.

Standard test system of rheometer consists of two parallel metal plates (upper moving plate with diameter of 12 mm), temperature monitoring sensor. Choice of measuring system is based on gypothesis of higher viscosity of mastics vs. free-of-filler bitumen. Thickness of test fused beads for bitumen and mastics was 1 mm. Rheological characteristics were studied in temperature range of 45–80^{o}C with step size of 5^{o}C. So, controlled parameters were followings: temperature, applied stress (i.e. torque moment was 100 μmN·m) and loading period with frequency of 1.59 Hz. Test was realized in CSS regime with controlled shearing stress. Output parameter was deformation (taking into account that applied stress was fixed) that was used for calculation of complex modulus G* as «amplitude stress – deformation» ratio [6].

Microstructure of bitumen mastics was studied with laboratory microscope AXIO SCOPE A1 (Carl Zeiss) in transmission regime. Test samples were thin layers of mastics at glass slides.

3. Results and Discussions

3.1. Characteristics of fillers

According to grain size analysis, all fillers are characterized by polydispersed particle size distribution (Fig.1). They can be graded by size in following sequence: QM (size range is 0.01–80 μm) → K (size range is 0.1–50 μm) → LM (size range is 0.01–35 μm) → QF (size range is 0.01– 30 μm) → LF (size range is 0.01–30 μm) → C (size range is 0.01–20 μm). It should be noted that high dispersity of C with average particle size of 1.52 μm and content of particle smaller than 100 nm is 2.73 % takes place.

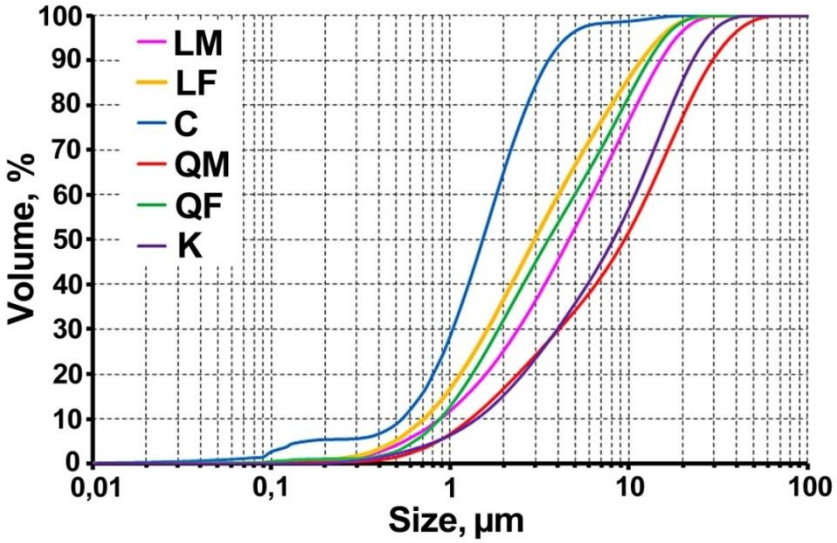

Figure 1. Integral curves of particle size distribution for the fillers

Specific surface area (SSA) is also the basic parameter especially for characterization of fillers in bitumen (Table 2). This parameter, measured by Blain method is in agreement with data of particle size distribution analysis (Figure 1, Table 2) besides for K.

Table 2. Characteristics of fillers

Filler	Real density, g/cm^3	Specific surface area, m^2/kg			Void content, vol. %
		by Blaine method	by BET method	Particle Size Distribution	
LM	2.713	703.8	5500±1200	1041.9	58.3
LF	2.710	978.8	4100±600	1412.9	55.6
C	2.718	1520.7	9700±300	2760.4	70.5
QM	2.658	329.7	1600±200	711.1	44.8
QF	2.675	885.4	3000±400	1244.6	61.3
K	2.613	1357.1	10200±1100	714.5	74.8

SSA for kaolin by Blain method is 1357.1 m^2/kg and by BET method is also high (10200 m^2/kg). It is associated with morphology of the filler: separate slides of kaolinite with high surface area those are collected in «packs». That explains an appearance of large particles (Fig. 1). Reduction of BET-measured SSA when increasing of grinding period for Limestone (Table 2) can be associated with particle aggregation due to extra energy. Actually, both two methods of SSA measurement characterize air pore content in powdered material that explains data of SSA and void content. For

exp., void content, calculated for fillers is similar to Rigden porosity, applied in other countries [4, 5]. So, void content will be strongly effect on rheology of mastics, produced by bitumen and filler mixing.

3.2. Standard rheological tests for bitumen mastic

Due to small variations in real density of fillers, the «bitumen-filler» (B/F) ratio in mastics is varied from 38.2/61.8 to 39.2/60.8 (Table 1). I.e. bulk and weight concentrations of filler in bitumen are comparable in all mastics compositions.

According to data obtained, K and C maximally influent on bitumen properties (Table 3). Two both fillers are characterized by the highest SSA and void content values. So they can absorb more of bitumen vs. others fillers. This parameter significantly effects on melting point gradient (for exp., 24°C for C-based mastics and 83°C for K-based mastics and 8–10°C for other mastics compositions). Relative viscosity (penetration) gradient is less visible, but also takes place at 25 °C and at 0 °C (Table 3).

Table 3. Physical and mechanical characteristics of mastics with different fillers

Parameters	Filler					
	LM	LF	C	QM	QF	K
Penetration gradient						
At 25 °C	42	43	54	45	48	56
At 0 °C	9	7	8	8	7	12
Delta Ring and Ball	10	8	24	10	10	83
Penetration index	-0,71	-1,13	0,57	-0,98	-1,06	6,27

High correlation between fillers porosity, melting points gradients (values of Delta Ring and Ball) (Table 2) and penetration index (Table 3) at 25°C for two types of fillers is observed (Figure 2).

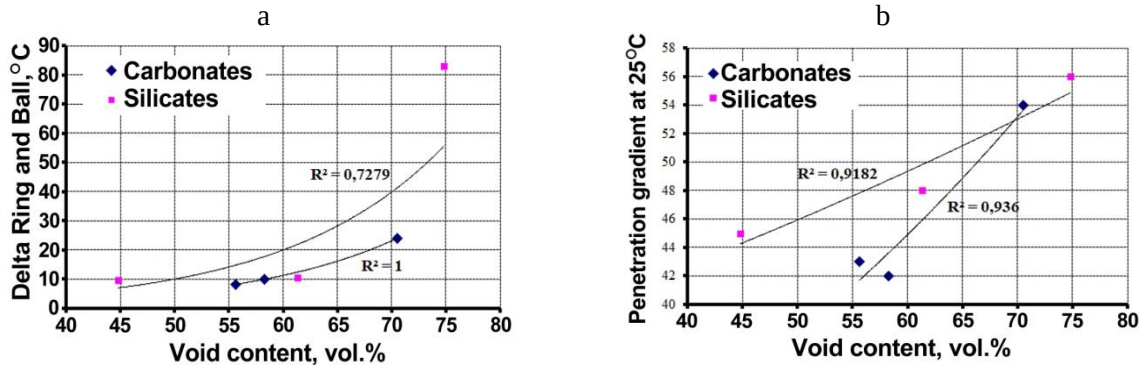

Figure 2. Correlation between fillers porosity, melting points gradients obtained (a) and penetration gradient at 25°C (b) for filler-containing bitumen mastics

In case of melting point as a relative viscosity factor at high temperature vs. penetration test we can see, that growing of temperature leads to increasing of gradient in mastics viscosity. In can be associated with intensification of structuring effect at higher temperatures due to higher void content. This relationship is clearly represented by penetration index values (Figure 3): introduction of carbonate and silicate fillers effect on this parameter vs. reference bitumen in range of ±0.24 (penetration index =-0.95). At the same time C and K initiate penetration index up to 0.57 and 6.27, respectively. Penetration index demonstrates thermal sensibility of bitumen and based mastics. Increasing of penetration index leads to reducing of sensibility of bitumen viscosity of temperature and increasing of heat-resistance of material. These results are confirmed by others studies [7, 8].

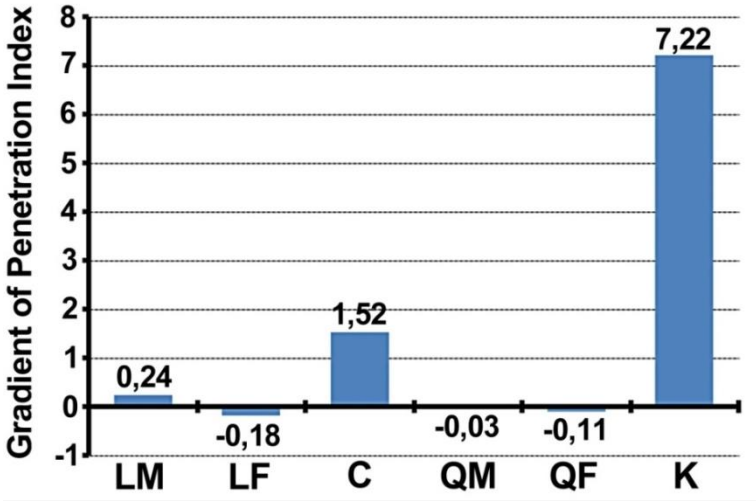

Figure 3. Dependence of penetration index for bitumen on fillers with different phase and size heterogeneity. Carbonates: LM, LF, C. Silicates: QM, QF, K

Significant variation in physical and mechanical properties of bitumen when kaolin (K) introduction is associated with plate-like particle shape. The results obtained are in agreement with earlier studies [1], devoted to effect of particle shape of fillers on rheological properties of bitumen mastics.

3.3. Rheological tests of bitumen mastics by vibrational method

According to results of the above studies, physical and mechanical characteristics of bitumen mastics should be investigated more deeply at temperature conditions.

Data from Figure 4 demonstrates, that complex shear modulus for mastics based on limestone and quartz mineral fillers is significantly lower vs. C- and K based ones but higher than for bitumen. It is confirmed by data of penetration gradient at 25°C, melting points and penetration index.

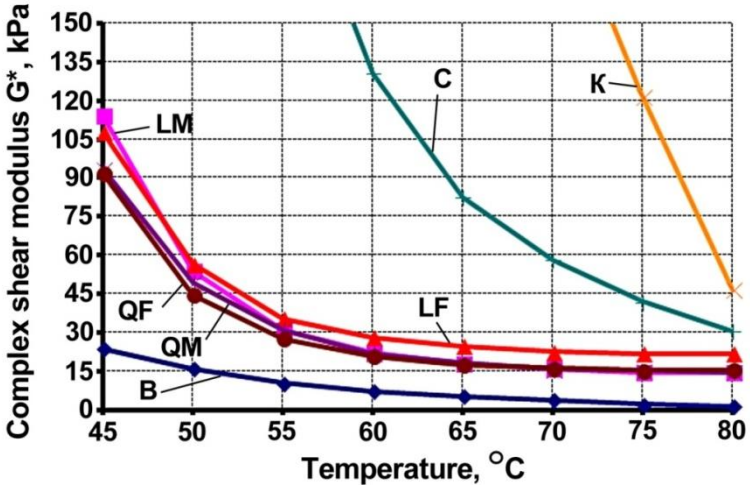

Figure 4. Effect of temperature of complex shear modulus G* for bitumen (B) and mastics with different fillers

C and K based mastics demonstrate the max reduction of temperature sensitivity vs. bitumen. It can be initiated by small-size particles (for C) and high SSA (for C and K as well) and leads to dramatic

structuring effect to bitumen. The most clearly this effect is realized at high temperature (Figure 4), especially, for limestone and less, for hydrothermal quartz.

3.4. Microstructure of bitumen mastics

Different structuring effect of fillers on bitumen is confirmed by images in Figure 5. Microstructure for C based mastics is presented at higher resolution due to small-size particles (up to 5 μm).

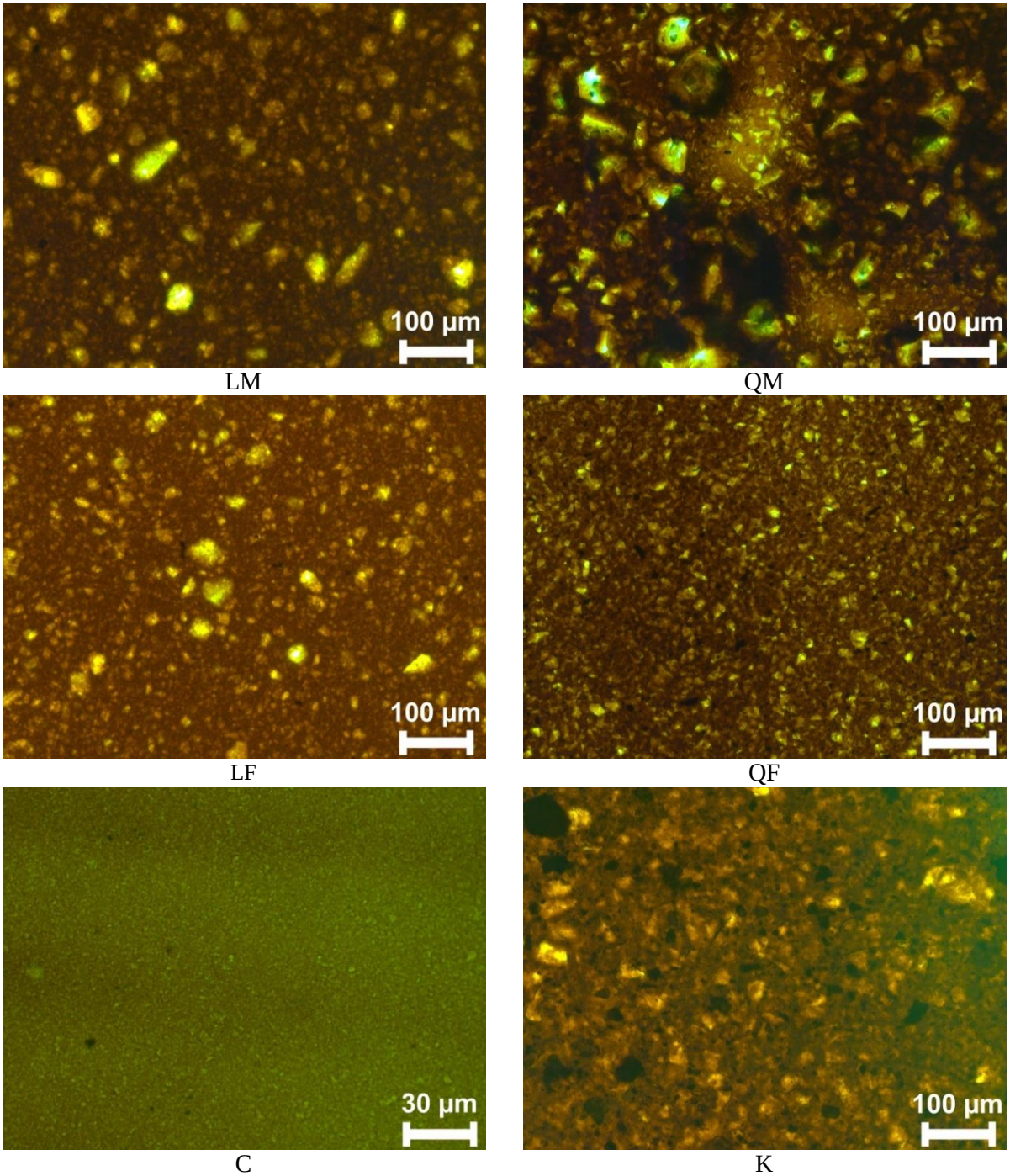

Figure 5. Microstructure bitumen-mineral mastics

According to images in Figure 5, structure of all experimental mastics is characterized by homogeneous distribution of filler in bitumen. Microstructure of limestone- and hydrothermal quartz-based mastics demonstrate a presence of large particles having a little effect on rheological characteristics as great part of bitumen is unbonded. For C and K based mastics thickness of intergrain bitumen layer is significantly lower leading to formation of more compact structure. For K based mastics filtration of bitumen into interlayer space of kaolin particles as well as distribution of separate layers in bitumen matrix takes place. But due to small-size particles of fillers, it is difficult to study structure formation with optical microscopy.

4. Conclusions

Results of study of effect of composition and dispersity of fillers on rheological characteristics of bitumen mastics demonstrate strong correlation. For example, variation in dispersity leads to transformation of composite structure, in generally. The rheological results demonstrate that the max reducing of temperature sensitivity is characterized for mastics contained fine-dispersed fillers with high SSA due to small-size particle (for C) and shape of particle (for K). These fillers are characterized by the highest concentration of air pores initiating a dramatical structuring effect on bitumen, especially, at high temperature.

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