

SOME RESEARCH RESULTS OF CONVENTIONAL MINERAL FILLERS USED IN ROAD CONSTRUCTION

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Abstract: This paper presents and summarizes some research results which were carried out by the Institute of Ceramics and Polymer Engineering, University of Miskolc in the last few years.

In order to achieve the best results extended literature review was made. Mineral fillers were tested from different Hungarian locations in order to reveal their role in asphalt pavements. Particle size distribution, morphology, specific surface area and air void of fillers were determined, as well as different adsorption tests were also made.

The performed tests give useful information about the behaviour of mineral fillers in asphalt pavements, especially about the cohesion between the particles and bitumen.

Keywords: asphalt, hydrophilic, microstructure, mineral filler, oil adsorption, Rigden void, surface

INTRODUCTION

Asphalt: the most common pavement material

Asphalt materials consist of a mixture of mineral aggregates and bitumen. The mineral skeleton of this composite system builds up from different fractions of rocks. The maximum carrying capacity of asphalt pavements can be reached by a precisely controlled technology which consists not only of the mixing mineral aggregates with bitumen but appropriate compacting [1].

The finest parts of mineral materials in asphalt pavements are called fillers of which particle sizes are under 0.063 mm [2]. Their particle size distribution (PSD) is extremely important in asphalt technology. Particle size and polydispersity, morphology, volume fraction and packing highly affect both the viscosity of bitumen and the performance of pavement [3, 4, 5, 6]. The presence of fines – owing to their surface properties – also affects the adsorption ability of bitumen [7].

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The properties of fillers fundamentally affect the contact with bitumen, therefore the properties of asphalt mastics (= bitumen + filler) and pavements [8, 9]. Cohesion, contact and absorption ability depend mainly on particle size (polydispersity), fineness modulus, specific surface, porosity and mineral composition (possibly the CaO content [10]) of fines. According to Clopotel and Bahia [11], the adsorption of bitumen components is proportional to the specific surface area (SSA) of fillers and at the same time it is independent from the mineral composition. Morphology, i.e. the geometry and shape of fillers affect the internal friction of mixture [12].

The role of mineral fillers in asphalt mixtures has been expansively investigated by several researchers. In their work, Grabowski and Wilanowicz [4] looked for quantitative relationship between some features of limestone fillers (such as particle size distribution, Rigden Void between filler particles, sphericity, surface texture and specific surface) and their stiffening properties. Their results showed significant difference between the structure of fillers and the above mentioned features. A number of tests were made by Ishai and Craus [6] on different mineral materials. They used the packing volume concept and described specific rugosity as an important parameter of packing which characterizes well the geometrical irregularity of mineral materials and eventually affects the properties of asphalt mixtures. Shashidar and his colleagues [13, 14] tested the effect of particulate mineral materials of asphalt mixtures on the rheological properties in their researches. According to them, the interaction between the particles, their agglomeration, particle shape and orientation in a given volume decrease packing, which affect the stiffness of asphalt pavements. Liao and his associates [15] tested the role of mineral fillers in asphalt mastics in point of stiffening effect and failure. According to their results, the amount of fillers significantly affects the stiffening of mastics. The higher the amount of filler is the higher is the stiffness of mastics.

According to Sakharov [16], the effect of fillers is based on the fact that they play an interactive role with their fineness and surface characteristics. They have a relatively high specific surface area in the mineral skeleton of asphalt mixtures, therefore they are able to adsorb a great part of bitumen.

Lisihina [17] executed tests on different (basic and carbonate) rocks concerning the adsorption of bitumen. According to the Author, these materials are able to absorb much more binders on their unit surfaces than acidic rocks. Lisihina stated that fillers with high porosity can absorb the components of bitumen in a selective way (selective diffusion).

In addition, a number of researchers [4, 6, 18, 19, 20] have also made experiments on asphalt mixtures with the use of mineral fillers. According to Hu et al. [21] the strength and structure of asphalts are highly affected by the contact between fillers and bitumen. The greater the binding force between mineral particles and bitumen is, the higher the strength of asphalt pavement is.

According to the above mentioned, the contact between fillers and bitumen depends on the adsorption processes taking place on their interface. Physical adsorption is affected by the porosity and the specific surface area of the fillers. According to numerous research articles, the bond between bitumen and siliceous

fillers is mechanical (generated by the Van der Waals forces) while the bond between bitumen and another filler (for example limestone) is chemical.

The amount of pores, cracks and small crystals stuck on the surface of filler particle increase the specific surface area, therefore affect the adsorbable amount of bitumen. Compared to other particles, specific surface area of fillers in mineral skeleton of asphalt mixture is considerably higher, therefore they are able to adsorb great part of bitumen in compound. It is practical to use material from ground basic, carbonate rocks as fillers. These rocks are hydrophobic which is favourable in view of adsorption of binder material [8]. When choosing fillers, mineral composition has a great importance, that is, material has to be pure. Presence of clay minerals is unfavourable for instance, as it decreases mechanical strength, resistance to mechanical weathering, and finally leads to failure of pavement sooner than planned.

Geometry, size, and surface activity of fillers all affect the relation with bitumen. Examination of physical-chemical mechanisms on the surface was made by Craus et al. [22]. They stated that greater surface activity of mineral materials promotes the strength of bond on filler-bitumen boundary and increase quantity of adsorbed bitumen.

According to Maidanova and Rozental [23], the following factors affect the adsorption of bitumen: specific surface area and activity of mineral filler, as well as the amount of SARA (saturates, aromatics, resins and asphaltenes) fractions of bitumen. In their opinion, there is an adsorption boundary within the bitumen-filler structure which influences the specific surface area of filler. They also stated that aromatic oils play a significant role in the adsorption phenomena. Aromatic oils and saturates form the greater part of bitumen. These components are responsible for plasticity, fluidity and elasticity of bitumen [24].

In his summary work, Lesueur [25] characterized limestone as a kind of “active filler”. In his opinion, asphaltenes building up bitumen presumably surround limestone particles through an adsorption layer. He also assumed that these adsorption layers are formed not only in case of limestone but every time independently from filler type.

Gou et al. [26] examined the effect of adsorption ability of mineral fillers on chemical and rheological properties of bitumen. Their results showed, which were obtained on asphalt mastics samples, that polar fractions of binder material can absorb on the surface of fillers. They also stated that the amount of adsorption is significantly influenced by the mineral composition of surface and also the specific surface area of fillers. In another research work of Gou et al. [27], the bitumen-filler interaction was observed by atomic force microscopy. They found that the effective thickness of bitumen on the filler surface was around 1 μm .

Taking into account the above literature results and methods, the test were performed. The experimental results may promote more accurate experience in the cohesion between bitumen and mineral fillers and their applicability in road construction.

1. MATERIALS AND METHODS

1.1. Sample preparation

During the research the Authors have examined conventional mineral materials used in asphalt technology (limestone, andesite, dolomite). The samples are originated from Hungarian locations. Limestone fillers were previously ground by the manufacturers. Andesite and basalt rocks were available in 2/4 mm fractions, which were ground in planetary ball mill after washing and drying. After grinding, fine fillers were fractioned by a standard sieve of 0.063 mm nominal particle size. Samples were then dried to weight constancy and closed air-proof for further examinations.

1.2. Testing methods

Particle size distribution (PSD) of the ground material was measured by a HORIBA LA-950V2 laser diffraction particle size analyzer in wet mode using distilled water as dispersing media and sodium-pyrophosphate as dispersing agent applying the Fraunhofer-theory as evaluation method. Before the measurements 1-minute ultrasonic treatment was applied on the samples for better dispersion.

For morphological tests Carl Zeiss EVO MA10 scanning electron microscope (SEM) was used. High resolution electron micrographs were taken in different magnifications for further observation. The aim of this technique was to observe morphology and geometrical features of fillers.

Specific surface area (SSA) of the fillers (by BET-method) was determined by TriStar 3000 instrument (manufactured by Micromeritics). The method of this instrument is based on physical absorption and capillary condensation, and it works in volumetric mode. The fine filler samples were prepared with the use of SMARTPREP instrument. Adsorption-desorption isotherms, BET-specific surface area of samples were determined.

Hydrophilic coefficient of fillers was determined by sedimentation method. 5 grams of fine materials were weighed and put in a 25 ml measuring cylinder. In parallel, 10 ml of water and 10 ml of kerosene were poured into the cylinder. Fillers and liquids were thoroughly mixed together then the cylinders were filled with more liquid and closed with plugs. Volume of fillers was determined after 72 hours of sedimentation.

The air voids (or Rigden Void, RV) of dry compacted fillers were measured using the Rigden-apparatus, according to MSZ EN 1097-4:2008 standard [28]. To calculate RV, specific gravity of fillers is required, which were measured by pycnometric method. The knowledge of RV is useful in designing asphalt mixes and it is a good indicator for estimating stiffness.

German Filler Test [12] was done on the samples in order to determine the ability of oil adsorption. 15 grams of oil was mixed together with 45 grams of filler. A ball was tried to be formed from the mixture. If it was successful and the particles were held together by the oil, more filler – in 5 grams increments – was added. If the

mixture did not stick together anymore, the cohesion between the particles and oil was loosen. The reason of this is that oil filled the voids between the particles.

2. RESULTS AND DISCUSSION

Table 1 contains all of the relevant test results, which were obtained during the researches.

Table 1
Test results of mineral fillers

	Mineral filler			
	Limestone #1	Limestone #2	Andesite	Basalt
Average particle diameter, d_{50} [μm]	16.82	13.77	5.58	10.45
BET specific surface area, SSA [m^2/g]	0.93	1.55	4.62	2.99
Specific gravity, ρ [g/cm^3]	2.717	2.820	2.791	2.960
Rigden Void [%]	41.5	47.9	40.6	40.2
Oil adsorption (according to German filler test) [g]	95	50	75	75
Hydrophilic coefficient, η [–]	0.73	0.59	0.83	0.69

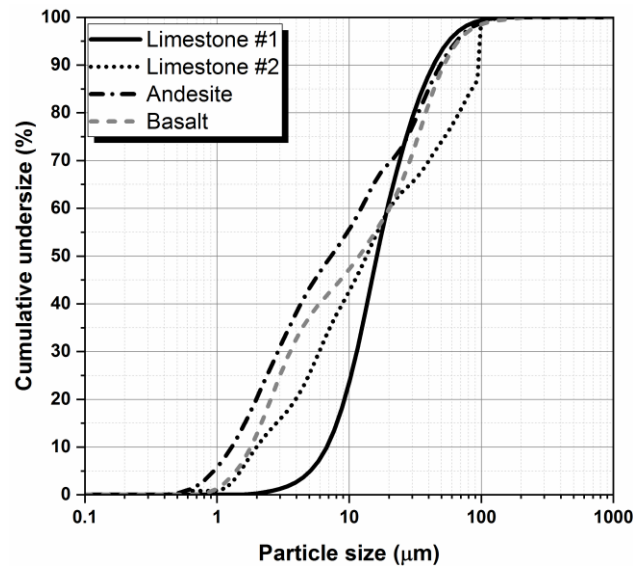


Figure 1
Particle size distribution of fillers

Figure 1 demonstrates the particle size distributions of fillers. According to the particle size distributions 80% of particles are smaller than 0.063 mm, in all cases. Based on the given data, it can be stated that andesite has the lowest average value (average particle size, d_{50}), with 5.58 μm . Limestone #1 has the highest median value with 16.82 μm . The reason for the differences between the average particle sizes of fillers is the way the rocks were ground. Limestones were ground by the manufacturers using industrial apparatus, while andesite and basalt were ground by the Authors in a lab scale.

In SEM micrograph (Figure 2. a) and b) of the limestones, relatively bigger, solid calcite particles can be seen. Small particles adhered on the surface of bigger particles. Bigger planar faces and planar fractures formed on other particles during grinding, especially in case of Limestone #1. Based on this, we can conclude a relatively small surface area which is favourable in view of bitumen quantity to be used.

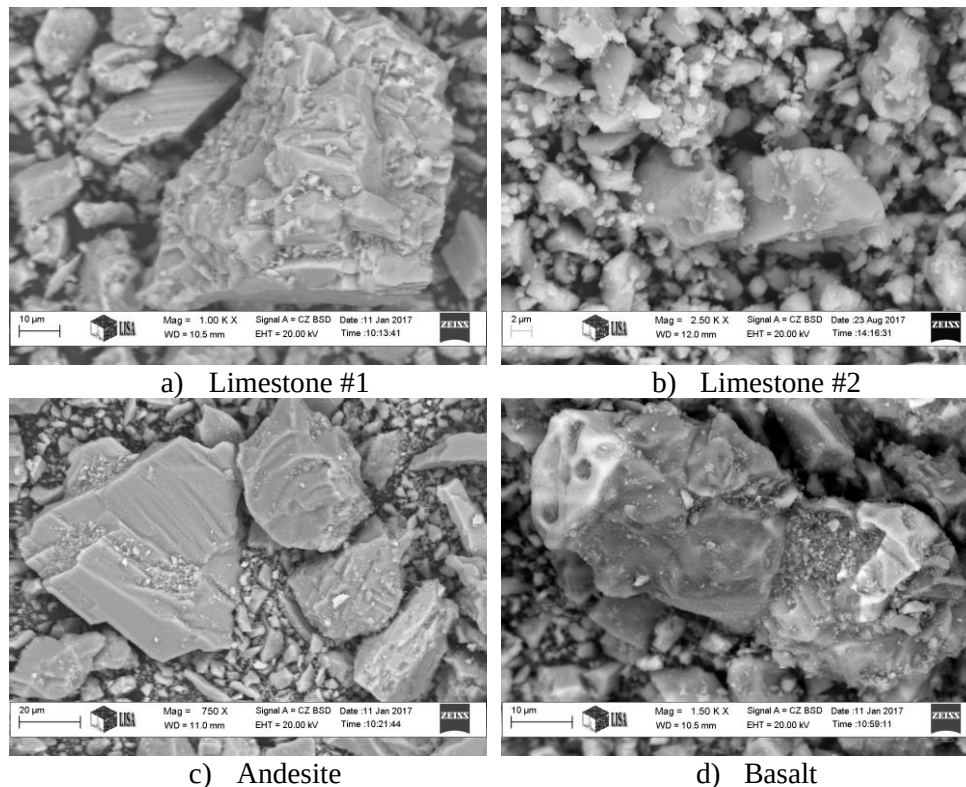


Figure 2
SEM micrographs of fillers

A considerable polydispersity of the sample is well visible on the micrograph of andesite (Figure 2. c). Besides the bigger, planar faced particles a great quantity of fine particles also occurs, but great part of them is of submicron size. We can observe

that fine particles adhered to the surface of the bigger particles. Particles formed during grinding have characteristically sharp edges and uneven fracture surfaces. Particles are considerably solid, there is no obvious trace of large pores, nevertheless the great number of submicronic particles refer to extensive specific surface area.

Polydispersity of the material fraction can also be observed on the SEM micrograph of basalt (*Figure 2. d*). Just like in case of andesite, basalt also has those submicronic particles which may refer to extensive specific surface.

According to the N₂ adsorption tests the lowest specific surface area belongs to the Limestone #1 (0.93 m²/g), while andesite has the highest SSA value (4.62 m²/g). SSA values of the limestones are favourable referred to the quantity of bitumen to be used in asphalt mixture production. In case of using limestone as filler we do not need to calculate with excess of bitumen. Nevertheless, application of andesite may result in increased bitumen requirement, which affects, mainly worsens other features of the asphalt mixture (bleeding, rutting).

According to the void volume of the dry compacted fillers, Basalt has the lowest (40.2%), Limestone #2 has the highest (47.9%) RV values, which is related to particle size distribution. As Limestone #2 contains a relatively higher amount of coarser particles than the other samples, therefore the space between the particles (interparticle void) is higher.

Based on the results of Kandhal et al. [12], the German filler test is on one hand a standard for the rate of the Rigden Void and on the other hand provides information about the adsorption ability of the fillers. During the use of the method, hydraulic oil fills the air void among the particles of the filler used in the maximum extent. Besides, the oil layer formed on the surface of the particles presumably promotes better adherence of the particles to each other.

Research results showed that Limestone #1 was able to bind unit value of oil in the greatest volume (95 g), while Limestone #2 was able to bind the smallest volume (50 g). This presumes that voids content among Limestone #1 particles is small, therefore oil is able to hold a greater number of particles. However, voids content of Limestone #2 is high as it is able to hold only a minimal number of particles.

Determination of hydrophilic coefficient is based on the comparison of water and oil adsorption ability of the filler. Oil, like bitumen is an apolar material. However, during the examinations the Authors have used kerosene instead of oil, as this liquid has the same features as oil in view of the experiment, but due to its lower density sedimentation of the particles is faster. This is rather important, as value of the hydrophilic coefficient can be defined from the volume ratios (i.e. $V_{\text{water}}/V_{\text{kerosene}}$) of sedimented particles in these liquids. If the ratio value is more than 1, filler is hydrophilic; if it is less than 1, it is hydrophobic. Based on Ref. [8], hydrophilic coefficient of the good filler used in asphalt pavements is 0.7–0.85. Based on the results all the examined fillers are hydrophobic. Limestone #2 has the lowest ($\eta = 0.59$) hydrophilic coefficient, while andesite has the highest ($\eta = 0.83$). Hydrophilic coefficient of Limestone #1 and Andesite are in the optimal range, therefore these materials are able to form a good relationship with bitumen.

Based on a previous research result of the Authors [29] hydrophilic coefficient value of fillers ground from carbonate rocks (limestone, dolomite) is affected also by particle size. They found that the highest the ratio of fine particles in the given fraction is, the lowest the hydrophilic coefficient value will be. Liquid surrounds particles as a kind of layer. The thicker the layer surrounding the particles in the kerosene is, the better relation between this apolar liquid and the mineral material can be formed. Therefore, the higher the sedimentation volume of the particles in the kerosene is, the lower their hydrophilic coefficient will be.

CONCLUSIONS

The aim of this article was to present some research results, which were carried out on different mineral materials used in asphalt pavements. It can be stated, that particle size distribution have a great influence not only on the specific surface area and air void, but the cohesion between the fillers and bitumen. The results of the performed tests, contribute to a better understanding of the interaction between filler and bitumen.

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