Asphalt production, paving and compaction techniques

Morpho-chemical characterization of fine and finest rock particles in asphalt

Martin Radenberg, Sören Holzwarth
Ruhr-Universität Bochum, Lehrstuhl für Verkehrswegebau

Abstract

The granulometry of the rock particles is of particular interest for use in road construction. The granu-lometric properties of a particle collective can be differentiated depending on parameters such as grain size and grain shape. For the fine and coarse aggregate there are already standardized measur-ing methods for the evaluation of these characteristic values. The filler has especially in dense asphalt concepts high mass fractions in the mixture. An in-depth knowledge of the properties of various fillers would be of great practical importance in terms of the optimum binder requirement, the required compaction regime and the performance characteristics of the asphalt. Sieve analyzes, sedimentation analyzes and innovative test methods such as laser diffraction analysis or photo-optical particle measurement can be used to determine the particle size distribution of fillers. Of particular importance in road construction seems to be the specific surface area of the fine particles. The finer the rock, the higher the specific surface area. For the analysis of the grain shape of fillers, there is currently no test method. This should be made testable by photo-optical analysis. In the course of this work, a test procedure will be developed, which will then be used to compare different fillers and fine aggregates. Characteristics such as particle size distribution, specific surface area, roundness and length / width ratio are to be systematically analyzed. In addition to the morphological parameters, the chemical composition of the aggregate can be de-termined by various test methods. One method in this context is the assessment by means of Raman spectrometry. Using a high-power laser, selected rock particles are irradiated and the scattered pho-tons are compared with a database and chemically identified. On the basis of Raman spectrometry, the proportion of different rock particles can be measured by isolated observation of rock particles.

This paper was first published in 2020 in version 1.0 of the proceedings of the 7th E&E Congress. This second edition (version 1.1.) of the proceedings is published in June 2021. The content of this paper has not changed, but the author(s) may have provided additional information in the form of an annex.

1 INTRODUCTION

The performance of an asphalt layer depends decisively on the components aggregates, binder and additives as well as their composition. The evaluation of binders using rheological test methods is becoming increasingly important, so that an extensive state of knowledge on binder properties is already known.

In order to ensure good asphalt performance, the interaction of all components in the asphalt must be considered. The binder content must be specifically adapted to the aggregate composition.

[1] shows the interaction of the asphalt components in a multi-scale model (Figure 1). By adding filler to bitumen (nano level) the mastic (micro level) can be created. By the addition of fine aggregates the mastic becomes mortar (Meso 1 level). The most important element for performance is the asphalt (Meso 2 level), which results from the properties of the sublevels.

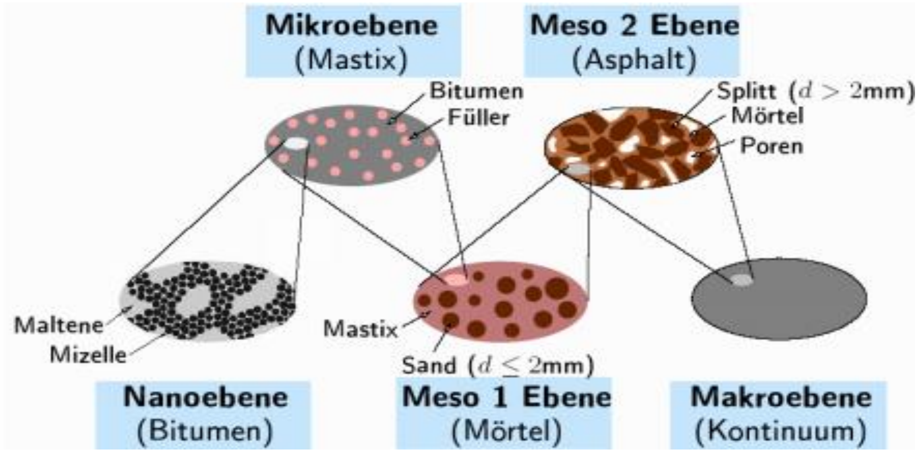


Figure 1: Multi-scale model for the visualization of the use behavior of asphalt [8]

Requirements for coarse and fine aggregates are already made with regard to granulometry, such as the particle size distribution, the proportion of broken grain surfaces, the flow coefficient, the content and quality of the filler and the particle shape according to DIN EN 933-3 [2] and 933-4 [3].
The filler is analyzed according to DIN EN 933-10 [4] by air jet sieving with regard to its grain size distribution. However, the test specification only differentiates between grain sizes up to 0.063 mm, 0.063-0.125 mm and 0.125-2 mm. For the proportions < 0.063 mm, a sieve passage of 70-100 M.-% is required without a deeper analysis of these high mass proportions. It is known from research projects that finest particles in particular have a very high specific surface and therefore a higher binder requirement than coarser particles. So far the granulometry of the finest particles cannot be examined more closely and is assessed purely qualitatively by procedures which make use of auxiliary variables of granulometry.
One such method deals with the stiffening properties of the filler by determining the void content according to rigden [5]. Another is the determination of the softening point increase "Delta Ring and Ball" of filler for asphalt according to DIN EN 13179-1 [6]. These stiffening properties have a significant influence on asphalt performance and are attributable to the granulometry of the fillers.

In particular, dense asphalt mix compositions have a high proportion of fillers and fine aggregates. For example, mastic asphalt is designed with up to 32 M.-% filler and up to 41 M.-% fine aggregate. Such dense construction methods are particularly dependent on the properties of asphalt mastic and mortar. In order to design this optimally, the components of the filler, the fine aggregate and the binder must be adapted to each other. The viscosity of the mastic and mortar is determined by the surface of the aggregate and the amount of binder available for coating. The larger the surface area of the aggregates, the more binder can be bound. This increases the yield point of this asphalt phase as a function of the aggregate/bitumen ratio. In order to guarantee a sufficient viscosity, a sufficient binder film must be present on the aggregates. Cubic aggregates have a better sliding ability than particles with a large angularity, which have a higher binder requirement to cover and slide the edges sufficiently (Figure 2).

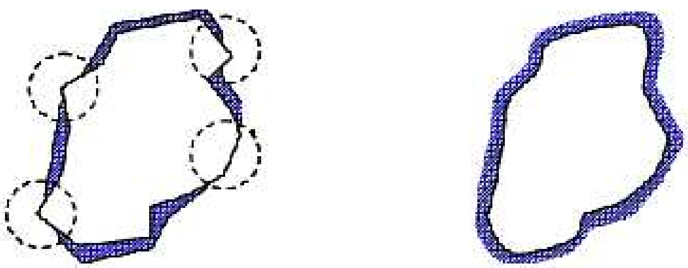


Figure 2: binder requirement of a rock grain for rock particles of different angularity [7]

DIN ISO 9277 [8] defines a test method for determining the specific surface area of solids by gas adsorption according to the BET method. The specific surface of limestone powder can also be determined using the Blaine method, which, however, is optimized for the measurement of cement with a predominantly homogeneous grain shape.

The photo-optical analysis of particles has been carried out for several decades in many areas of materials analysis. Due to technical progress, this method can characterize particles down to 0.5 µm precisely regarding particle size, shape and other granulometric properties.

One approach of this work is to make use of the potentials of photo-optical measurement methods for the evaluation of fillers in road constructions.

2 STATE OF RESEARCH

The properties of asphalt are primarily determined by the properties of the binder. As [9] has already stated, the "binder" should not only be considered as the bitumen but rather as the filler/bitumen mixture (mastic). The filler can only play a minor role in terms of the mechanical properties of the mastic due to its bitumen coating, but it has been shown that it has a significant influence on the mastic in terms of viscosity, stiffness and low-temperature performance, and thus also on the mechanical properties of the asphalt mix. As early as 1987, the "Recommendations" of the Belgian "Centre de Recherches Routières" confirmed that fillers can be used as a fund of stiffening and reducing temperature-induced stress.

Already [10] investigated the influence of the filler's granulometry on the properties of asphalt mastic. However, the methods available in 1937 were unable to prove any clear dependence.

In recent decades, researchers have focused primarily on the performance of asphalt mastic without analyzing the granulometric properties of the filler in depth. The primary input parameter for the filler was the void content according to rigden [5], which was used to analyze the dependencies between mastic and filler properties.

[11] investigated the influence of a limestone filler with different filler/bitumen ratios (f/b) on the viscosity of the mastic in the rotational viscometer. It was found that the variation of the f/b ratio leads to different viscosity-temperature behavior, especially at higher temperatures. As the proportion of limestone powder increases, both the dynamic modulus of elasticity and the dynamic viscosity increase. [12] also dealt with the influence of the f/b ratio. In the course of this research a clear dependence of the deformation resistance of asphalt on the f/b ratio could be derived.

Further analyses have been carried out in the recent past. Thus [13] dealt with the fatigue resistance of mastic in the dynamic shear rheometer. The research project was able to show that a high content of fine particles has a positive effect on the durability of mastic. The correlation analysis of the determined DSR results with the characteristic values of the aggregate showed that the grain fraction <20 µm has a significant influence on the fatigue resistance. The evaluation also shows that a more detailed analysis of the granulometry of the ultrafine particles could supplement the correlation by parameters such as grain shape, specific surface, etc. and thus possibly further specify them.

In addition to the physical properties of the fillers, the chemical component must also be considered. Thus in [14] the influence of filler chemistry on the rheological properties of mastic was determined by shear tests. The adhesion and cohesion properties could be traced back to the proportion of calcium oxide (CaO). The higher the CaO content, the higher the stiffness and the better the adhesion. [15] dealt with the influence of moisture on mastic behaviour and also came to the conclusion that cohesion and resistance to moisture were primarily due to the composition of asphalt mastic (chemical composition and surface area).

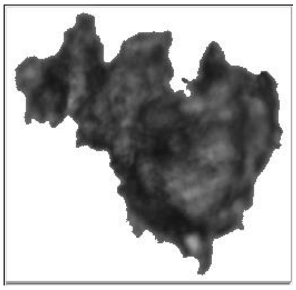
These findings make clear the need for a more in-depth analysis of the filler in terms of granulometry and chemistry. Technological progress is to be used, which makes it possible to view particles down to 0.5 µm very precisely photo-optically regarding particle size, shape and other granulometric influencing factors. An additional device component (Raman spectroscopy) can be used to check the chemical composition of the aggregate particles, which provides important extra information for assessing the performance of the mastic.

3 EXAMINATION

This research work deals with the physical-chemical evaluation of fillers. The findings will be used for a systematic analysis of dependencies between filler and mastic properties.

For the analysis, the Morphology G3ID test device from Malvern was used. This enables the photo-optical observation of ultrafine particles from 0.5 µm grain size. The measuring method is static and for the measurement a sample quantity between 1 and 3 mm³ is applied to a lens carrier by means of compressed air. The acquisition can then be carried out by different lenses, whereby each captured particle is assigned a running ID and, as shown in Figure 3, is analyzed for different aggregate characteristics. In addition, some of the relevant parameters are defined below.

Field	Value
Area (Pixels)	204229
Area (µm²)	15850,19
Aspect Ratio	0,735
CE Diameter (µm)	**142,06**
Centre X Position (µm)	66214,250
Centre Y Position (µm)	58476,773
Circularity	0,569
Convexity	0,683
Edge stitched particle	0
Elongation	0,265
Fiber Elongation	-1,000
Fiber Straightness	-1,000
Fiber Total Length (µm)	-1,00
Fiber Width (µm)	-1,00
HS Circularity	0,323
Id	85040
Intensity Mean	66,614
Intensity SD	21,869
Length (µm)	185,66
Magnification	10,00
Major Axis (°)	35,04
Max. Distance (µm)	191,52
Perimeter (µm)	782,34
SE Volume (µm³)	1501120,50
Solidity	0,797
Width (µm)	136,42



Convexity – surface roughness of a particle

Circularity – how close the shape is to a perfect circle

Circle Equivalent – the diameter of a circle with the same area as the 2D image

Aspect-Ratio – the ratio of the Width to the Length of the particle

SE Volume – The volume of a sphere with the same CE Diameter

Intensity Mean – average of the pixel greyscale levels

Figure 3: Acquisition of a particle together with its characteristic values in the test device Morphology G3ID

That device offers the possibility to determine the chemical properties of a particle by Raman spectroscopy. The Raman effect is an inelastic scattering of light by matter, which, unlike the (elastic) Rayleigh scattering, is partly absorbed by the scattered medium. With molecules it happens in the form of rotation and vibration energy and with solids this is added by energy quanta to the lattice vibration, the phonons. The subsequent energy balance results in a frequency shift of the scattered light. The determined wavelengths of the scattered light can be spectroscopied by means of atomic energy levels.

After measuring the dispersed rock sample, individual particles can be selectively targeted by selecting the particle ID on the sample plate and analyzed by Raman spectroscopy. Figure 4 shows a particle of a diabase filler with the detected Raman spectrum.

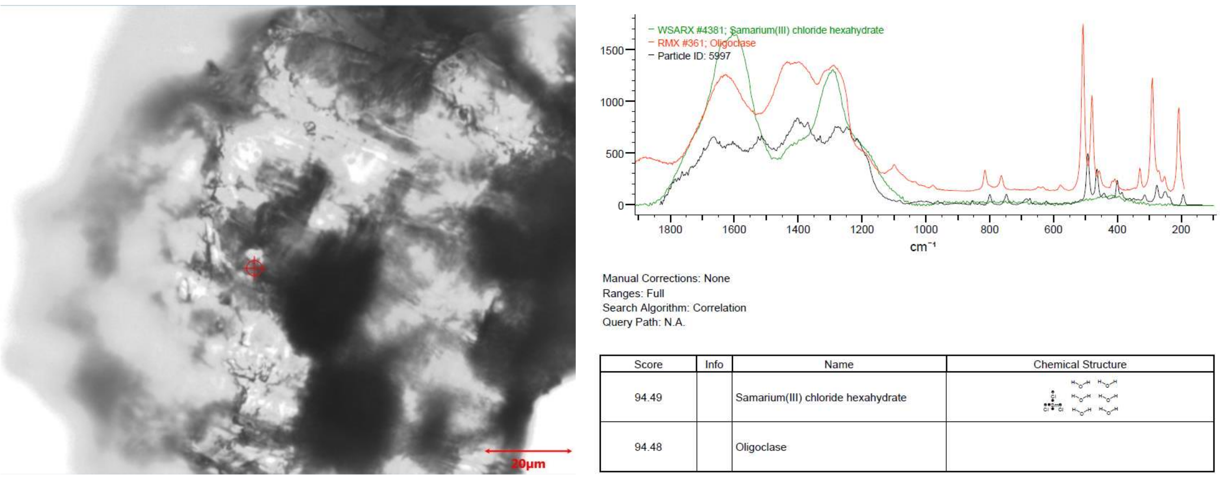


Figure 4: Manual focus of the rock surface to perform a Raman spectroscopy with the associated spectral distribution

The aim of this research activity was to develop a test procedure with which the properties of fillers could be reproducibly recorded. Subsequently, seven different fillers (basalt, limestone, andesite, rhyolite, porphyrite) were measured and their properties compared.

In the first phase, the preliminary investigations, the measuring parameters of the testing device were adapted to fillers for road construction. By adjusting the size of the measuring area, the image overlap, the number of planes in which measurements are made (depth of field) and the selection of objectives, uniform test boundary conditions can be defined.

For the preliminary investigations, three fillers (a diabase filler and two limestone fillers with different stiffening properties) were specifically varied with regard to the above-mentioned parameters, as shown in Figure 5.

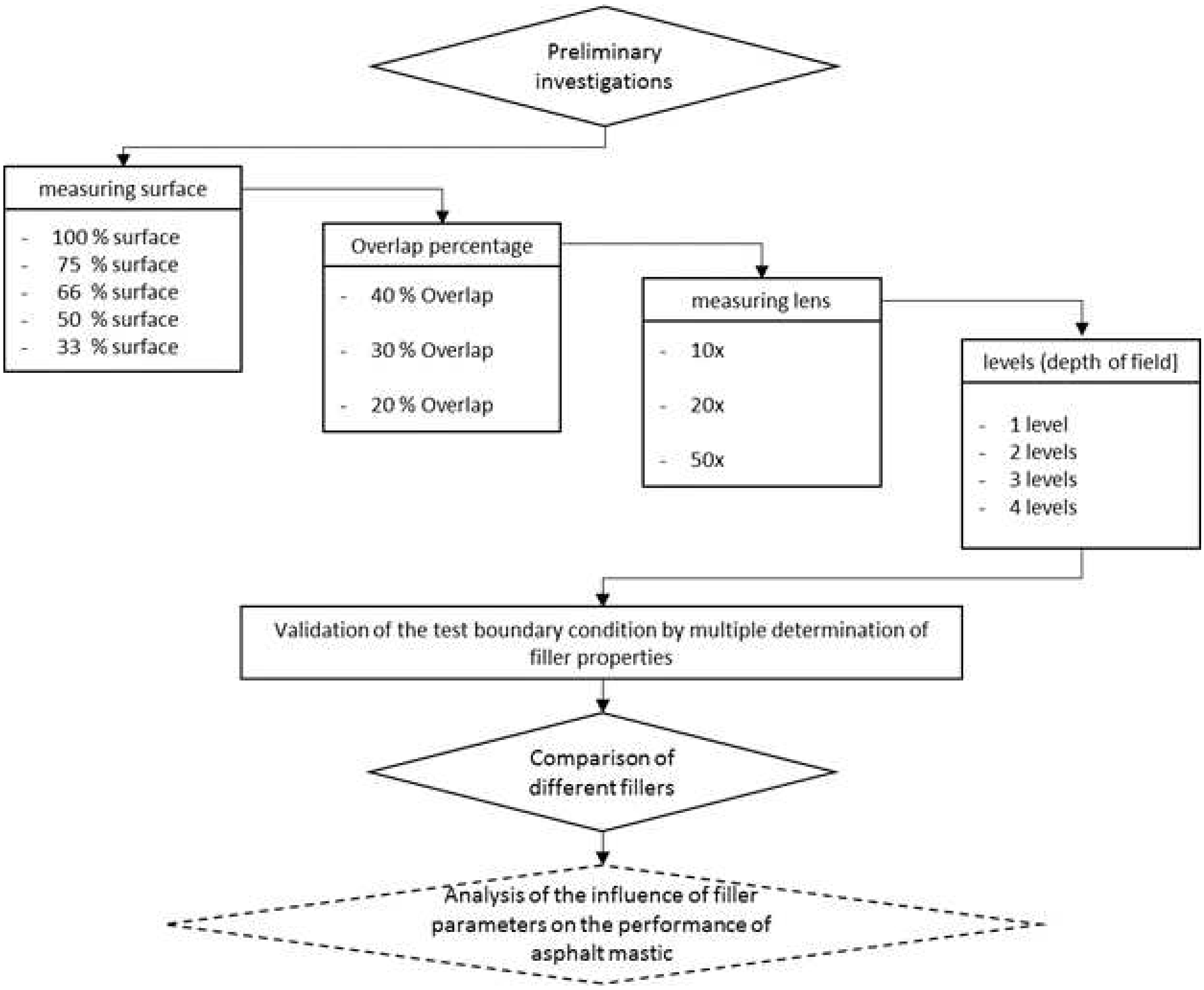


Figure 5: Flow chart of the research activity carried out

The results of the preliminary investigations aim to define a standard operating procedure with which the seven fillers of the main investigation should then be analyzed.

4 RESULTS

4.1 Preliminary investigations

The results of the preliminary investigations were systematically examined with regard to the varied test conditions. The used testing device can evaluate the results by comparing the parameter area, CE diameter, length, circularity, aspect ratio, etc.) as shown in Figure 6.

The measurement of the entire sample area with all available objectives would take several days per sample, which makes it necessary to adjust the test boundary condition so that a sample can be examined with the necessary precision and as quickly as possible.

In the following, the evaluations of the four variation steps are shown according to Figure 5. For this purpose, the determined percentage grading curve with the corresponding "parameter variability" is displayed in figures 6 to 10.

In the first step, the necessary measuring surface was determined which is required for sufficient precision.

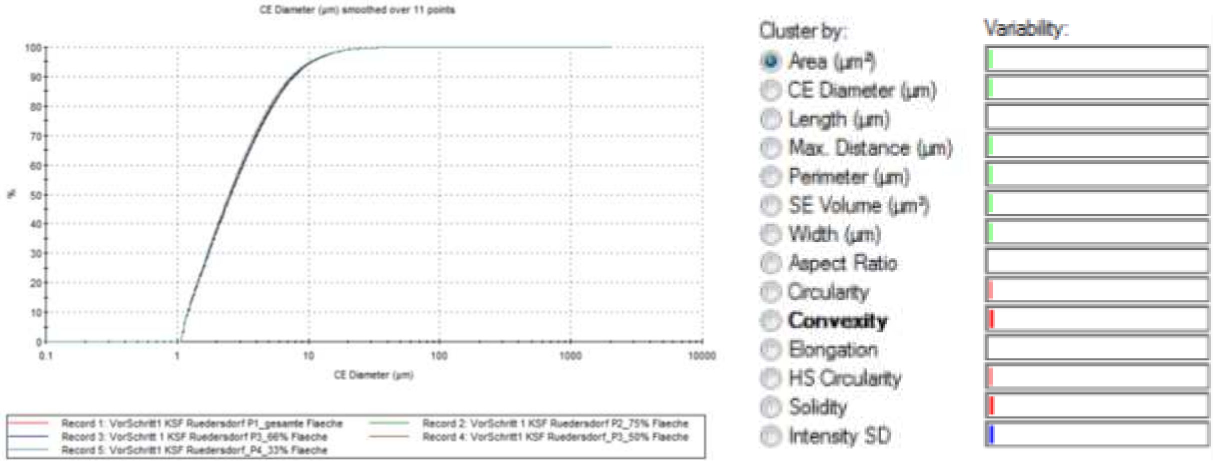


Figure 6: Preliminary investigation with variation of the size of the measuring surface using the example of a limestone filler

On the results of the variation of the measuring surface it could be determined that a variation of the surface between 33 % and 100 % results in only minimal differences. The sample quantity was dispersed on the sample plate and was measured with the different surface proportions. The same dispersed sample was always considered and not redispersed in order to create ideal conditions for comparison. The area proportions (≤ 75 %) were distributed over the total area in such a way that both edge areas and central areas were measured. Thus it can be excluded that an inhomogeneity during dispersion of the sample falsifies the measurements.

Based on the results of all three filler investigations with variation of the measuring surface, the measuring time per lens could be reduced by approx. 65 % without loss of precision.

In the second step, the influence of the overlap percentage on the image analysis was investigated (Figure 7). The results of all three fillers confirmed that an overlap of 20 % was sufficient for typical fillers in asphalt road construction. The variation also showed only minimal deviations. As a result, the test duration per lens could be reduced by approx. 45 %.

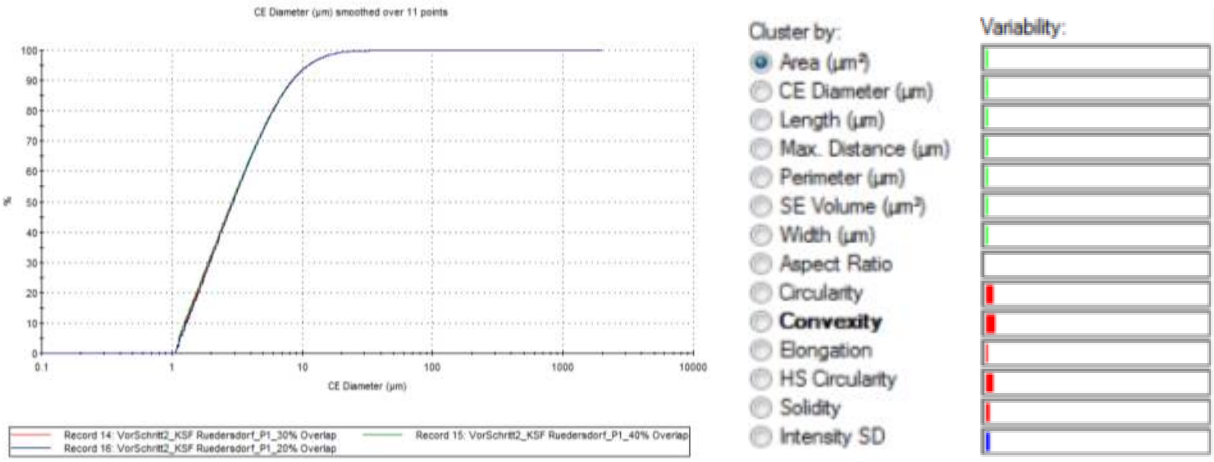


Figure 7: Preliminary investigation with variation of the image overlap using the example of a limestone filler

In the third step of the preliminary investigations, the variation of the measuring lens was carried out. As specified by the manufacturer, the various lenses can achieve optimum results in the areas indicated in Figure 8. According to DIN EN 933-10 [4], fillers must have a proportion of 70-100 M.-% < 0.063 mm. The appropriate lenses selected were those with 50x, 20x and 10x magnification. For the examination of the three fillers only one sample was dispersed and measured with the different oculars. The results are shown in Figure 9 together with the variability of the measurement results.

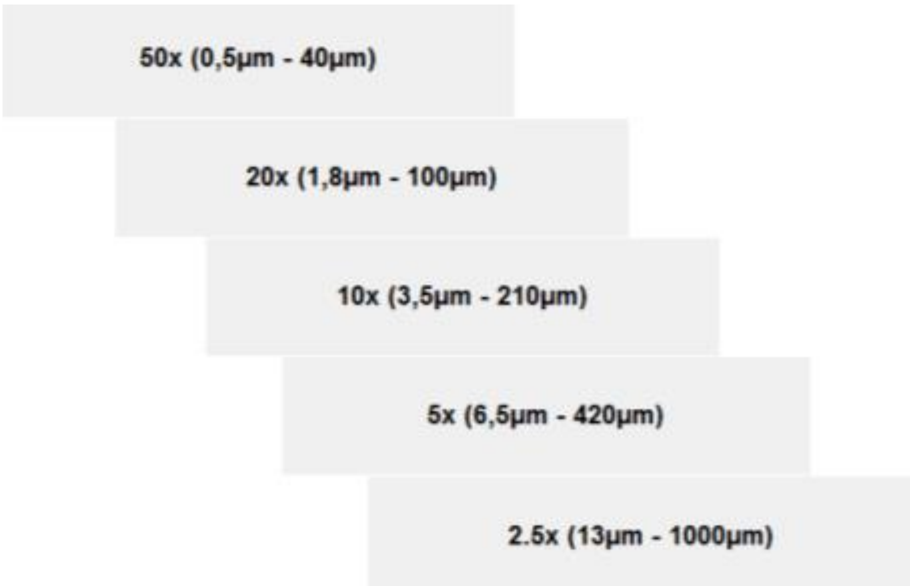


Figure 8: Lenses installed on the testing device with specified measuring range

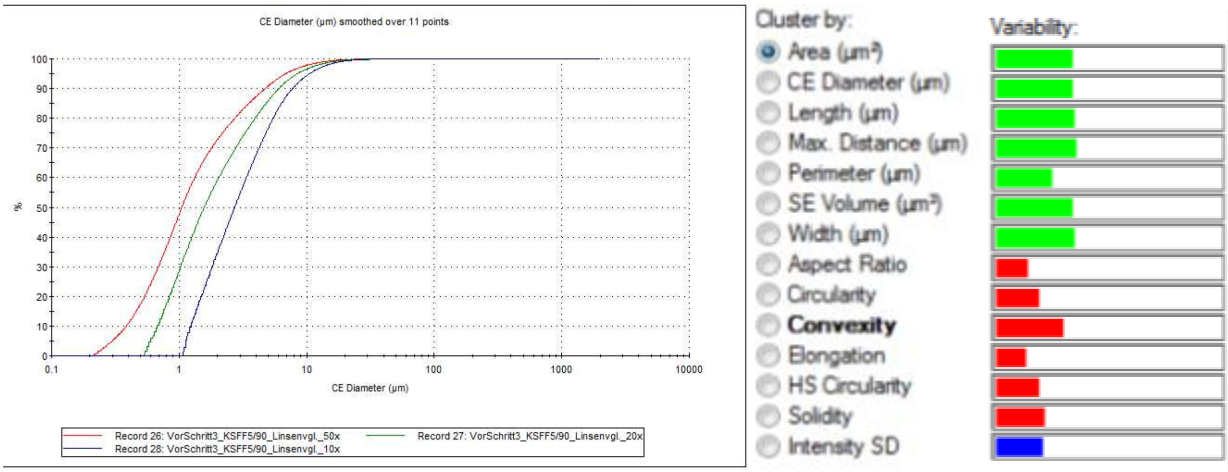


Figure 9: Preliminary investigation with variation of the lens used using the example of a limestone filler

The evaluation results in a clear discrepancy of the results depending on the measuring objective. This can be attributed to the fact that the different lenses in the peripheral areas of the measuring precision can only detect and evaluate the particles very blurred. It was found that each sample must be measured with all three lenses and systematically assembled. For the ranges between 0 and 20 µm the 50x objective has to be used, for 20 to 80 µm the 20x objective and for particles > 80 µm the 10x objective. A computer-aided evaluation tool is therefore used for the final evaluation and a procedure is programmed there which takes into account the linking of the three magnification lenses.

In the last step, the depth of field was varied with different numbers of measuring levels. At this point, a sample with 1-4 levels was also evaluated under comparison conditions. Figure 10 shows exemplarily the results for the limestone filler with its test variability.

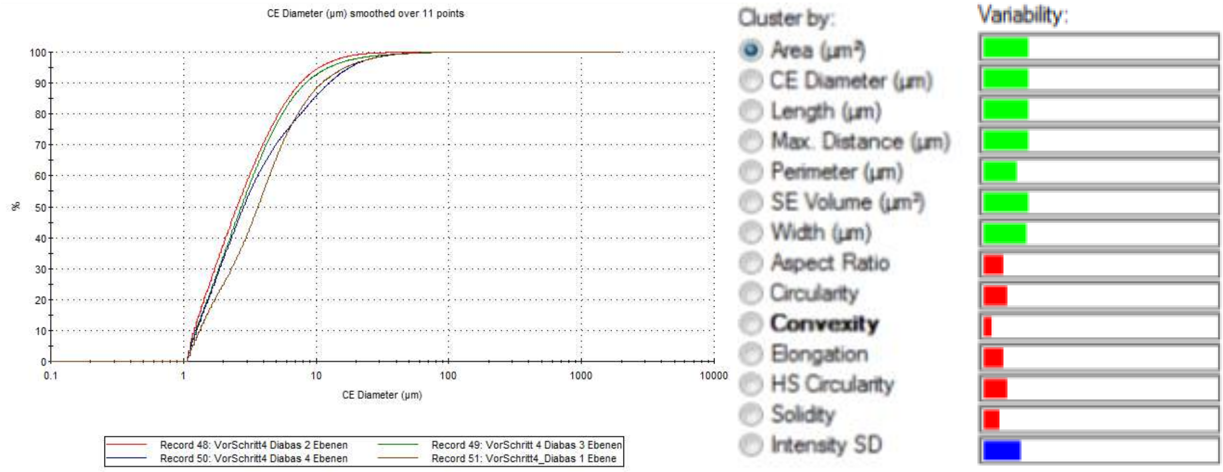


Figure 10: Preliminary investigation with variation of the number of depth of field planes using the example of a diabase filler

Based on the measurement results, it could be determined that the test results with only one measuring level clearly differ from those with 2-4 measuring levels. No further improvement was achieved with three or more measuring levels.

A standard operation procedure could be defined taking into account all results of the procedural steps. For the measurement of fillers, measurements with the objectives 50x, 20x and 10x on 33 % of the sample area at 20 % image overlap each and in three measuring levels (depth of field) are to be carried out.

For evaluation, the measurement results shall be systematically combined. For the range 0.5 to 20 µm the 50x is used, for 20 to 80 µm the 20x and from 80 µm the 10x magnification is used. The combined results can be systematically evaluated in a software tool regarding the filler characteristics.

4.2 General inspections

Seven different fillers were examined in the course of the main inspections. The results of the investigations are shown in Figure 11 and were evaluated with regard to a grading curve in [M.-%] (top left), convexity (top right), circularity (undersize curve center left; frequency curve center right), length/width ratio (bottom left) and color identification (bottom right). The grading curve determined in asphalt road constructions refers to a mass, while the testing device outputs cumulative frequencies. A conversion could be performed on the sphere-equal volume output with the aid of the bulk density of the respective filler.

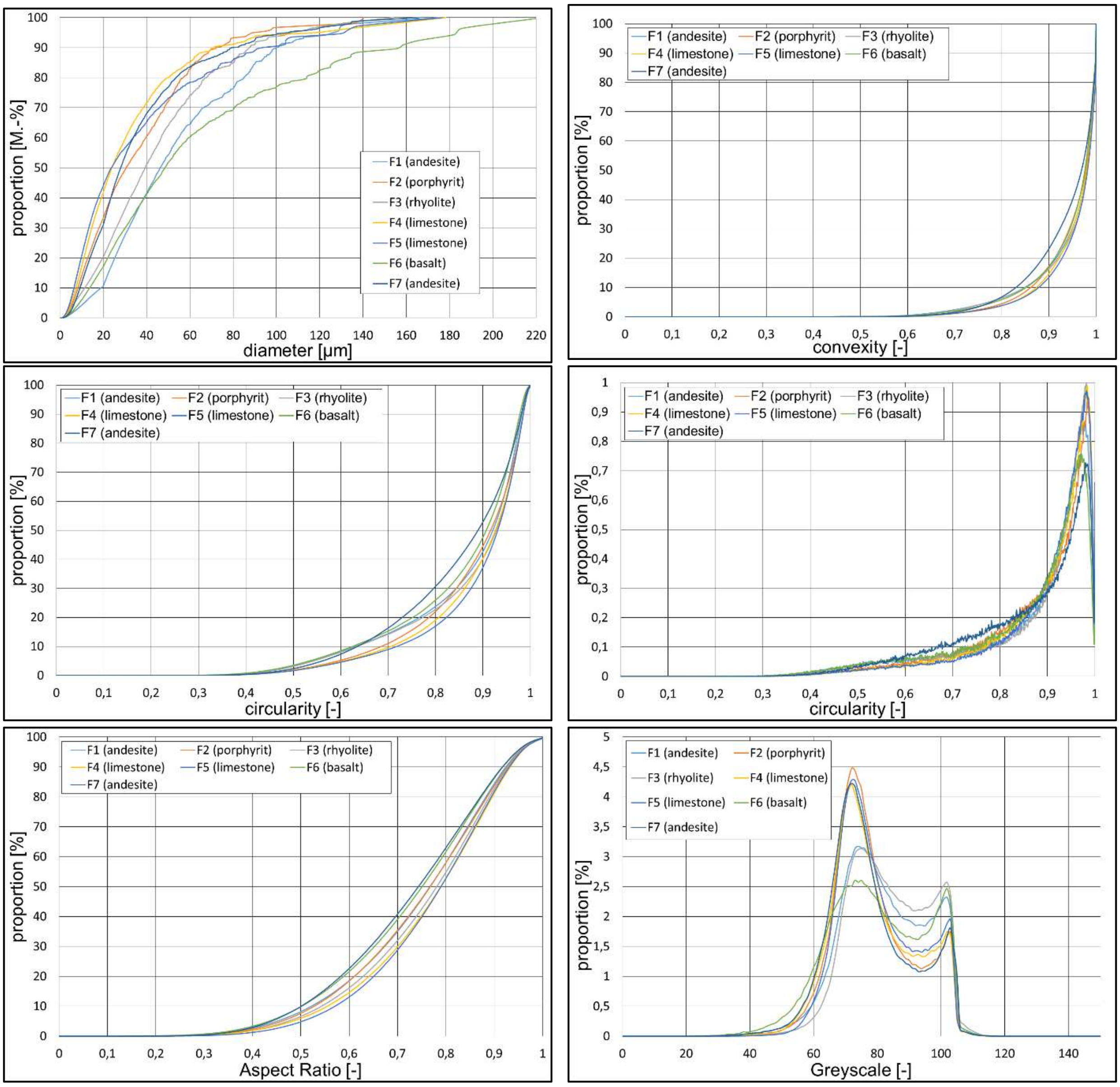


Figure 11: Results of the investigations on seven fillers used in road construction

Clear differences could be determined, especially with regard to the particle size distribution. Table 1 gives an overview of the characteristic values of the fillers.

Table 1: Characteristic values for the assessment of the seven fillers of the main inspections

filler	rock type	No. of particles	Σ surface	proportion			Circularity <0,8	Aspect Ratio at 0,6	Convexity at 0,9
				<20 μm	<40 μm	<63 μm			
[n]	[-]	[n]	[mm²]	[M.-%]	[M.-%]	[M.-%]	[%]	[%]	[%]
1	andesite	539,314	9,87	10,6	41,6	67,4	23,8	18,6	16,9
2	porphyrit	420.872	6,93	33,4	60,4	85,4	22,1	18,5	16,8
3	rhyolite	935.641	8,18	20,3	51,1	76,2	22,8	16,2	16,3
4	limestone	595.402	8,28	41,8	71,6	87,2	19,1	14,6	14,7
5	limestone	1.034.912	10,72	44,0	65,1	79,1	17,0	13,1	13,5
6	basalt	420.095	8,57	17,1	41,2	61,8	25,9	21,6	17,3
7	andesite	645,524	14,51	31,0	68,0	84,7	30,7	22,7	23,2

5 SUMMERY

Based on the results of these investigations, it can be summarized that technical progress allows a detailed examination of the filler component by means of photo-optical analysis. This static test method can comprehensively analyze the granulometry with regard to grain size and shape. In addition, the Raman spectroscopy integrated in the test device can be used to perform a chemical characterization of the aggregates.

By means of a specific preliminary investigation program, it was possible to develop a uniform standard operating procedure with which the fillers can be reproducibly examined. For this purpose, measurements are carried out with three lenses (50x, 20x and 10x magnification) in order to be able to accurately describe aggregate particles in a range from 0.5 µm to 210 µm and with a good object sharpness. The measurement in different depth of field planes also allows additional precision of the results.

Current research activities are increasingly focusing on asphalt mastic regarding its performance properties. In this context, [13] were able to establish a clear reference to the granulometry of the filler particles, in particular the <20 µm content. These findings can now be analyzed in greater depth using further granulometric properties.

For this purpose, a current research project is systematically analyzing the influence of filler granulometry on the properties of asphalt mastic. On the one hand, the stiffening properties are to be determined by the softening point increase "delta ring and ball" of filler for asphalt according to [6], on the other hand, innovative tests of the mastic properties are to be carried out in the dynamic shear rheometer and bending beam rheometer.

6 REFERENCES

[1] Lackner, R., Blab, R., Jäger, A., Spiegl, M., Kappl, K., Wistuba, M., Gagliano, B. & Eberhardsteiner, J.: Multiscale modeling as the basis for reliable predictions of the behavior of multi-composed materials, Progress in Engineering Computitional Technology, Saxe-Coburg Publications, ISBN 1-874672-22-9, 2004

[2] DIN Deutsches Institut für Normung e. V., DIN EN 933-3: Tests for geometrical properties of aggregates – Part 3: Determination of particle shape - Flakiness index, 2012

[3] DIN Deutsches Institut für Normung e. V., DIN EN 933-4: Tests for geometrical properties of aggregates – Part 4: Determination of particle shape – Shape index, 2008

[4] DIN Deutsches Institut für Normung e. V., DIN EN 933-10: Tests for geometrical properties of aggregates – Part 10: Assessment of fines – Grading of filler aggregates (air jet sieving); 2009

[5] DIN Deutsches Institut für Normung e. V., DIN EN 1097-4: Tests for mechanical and physical properties of aggregates – Part 4: Determination of the voids of dry compacted filler; 2008

[6] DIN Deutsches Institut für Normung e. V., DIN EN 13179-1: Tests for filler aggregate used in bituminous mixtures – Part 1: Delta ring and ball test; 2013

[7] Geisenhanslüke, C.: Influence of the granulometry of fine particles on the rheology of pastes; Structual Materials and Engineering Series; No. 13; University Kassel, 2008

[8] DIN Deutsches Institut für Normung e. V., DIN ISO 9277: Determination of the specific surface area of solids by gas adsorption – BET method, 2010

[9] Cupo-Pagano, M., D'Andrea, D., Cantisani, G.: The role of filler in bituminous mastics and concretes, Mechanical Tests for Bituminous Materials, Di Benedetto & Francken, RILEM, ISBN 90 5410 876 2, 1997

[10] Traxler, R.N.: The Evaluation of Mineral Powders as Fillers for Asphalt, Journal, Association of Asphalt Paving Technologists, Vol. 8, 1937

[11] Pritz, T., Zakar, P.: Dynamische viskoelastische Kennwerte von Bitumen-Kalksteinmehl-Gemischen, Bitumen 2, 1984

[12] Moreno, E., Contreras, V., Carrera, V., Palleiro Costa, L.: Influence of the mastics on the permanent deformation of asphalt pavements, Paper on 6th Euroasphalt & Eurobitume Congress in Prague, 2016

[13] Hospodka, M., Hofko, B., Blab, R.: Zur Ermüdungsbeständigkeit von Asphaltmastix – Prüfmethodik und Einflussparameter, Technische Universität Wien, Institut für Verkehrswissenschaften, Paper on 5. Dresdner Asphalttage, 2017

[14] Zhang, J., Fan, Z., Hu, D., Hu, Z., Pei, J., Kong, W.: Evaluation of asphalt-aggregate interaction based on the rheological properties, International Journal of Pavement Engineering, 19:7, 586-592, DOI: 10.1080/10298436.2016.1199868, 2016

[15] Chaturabong, P., Bahia, H.: Effect of moisture on the cohesion of asphalt mastics and binding with surface of aggregates, Road Materials and Pavement Design, 19:3, 741-753, DOI: 10.1080/14680629.2016.1267659, 2016