Alternative modifications of bituminous binders for mastic asphalt mixtures

Jakub Šedina¹, Jan Valentin¹ and Lucie Benešová¹
¹Faculty of Civil Engineering, Czech Technical University in Prague, Thakurova 7/2077, 166 29 Prague 6 – Dejvice, Czech Republic

E-mail: sedinjak@fsv.cvut.cz

Abstract. This paper focuses on potential benefits of alternative bituminous binders for mastic asphalt mixtures, which were modified by new type of low viscosity additives or activated rubber powder. Paper presents results of laboratory investigation on mixtures with standard bituminous paving grade bitumen 20/30 and mixtures with modified bituminous binders. The reference bitumen 20/30 was modified by micromilled activated rubber powder, by a new generation of synthetic waxes (WE-CM20, WE-BM), or by the combination of synthetic wax and micromilled rubber powder. Comparison of different mastic asphalt mixtures was based on laboratory testing (indentation test, compressive strength test, bending (tensile) strength test, stiffness modulus test and cyclic compression test). Possible uses of these applications is for example in pavement structures for bridge decks, or for effective sealing of expansion joints on bridges. Mastic asphalt characteristics are compared with selected characteristics of used bituminous binders (complex shear modulus, rotational viscosity, etc.).

1. Introduction

The increasing traffic loading acting on pavement structures not only in the Czech Republic puts generally more and more emphasis on the quality of the asphalt mixture for road construction which should demonstrate improved durability and, primarily, overall asphalt pavement lifetime. At the same time, there are a number of pavement structure components where the above applies even more urgently, for instance also due to difficult installation or the impossibility of compacting a traditional mixture to a sufficient quality level. In many cases, traditional bituminous binders fail to meet more strict performance requirements and their properties have to be modified by appropriate additives or polymers. This has been known in the road construction industry for over 30 years; nevertheless, with the development of a number of new additives or traditional modifier variations, the options have grown wider. Modifications of common paving grade bituminous binders allow influencing asphalt mixture parameters like the processability, load-bearing capacity and durability. Similarly, specific characteristics in need of significant improvement (e.g. resistance to permanent deformation etc.) can be handled as a priority. Bituminous binder modification is common in all types of applications; the general effort strives to improve the parameters of the traditional bituminous binders, thus consequently improving the asphalt mixture quality. The experimental study presented herein focuses on the use of modified binders in mastic asphalt technology, particularly on the modifiers with a potential to influence both the working temperatures and the overall energy requirements of the
technology and, on the other hand, on the monitoring of some alternatives to the chemical modification of bituminous binders as such. Due to that, the impact of bituminous binders modified by micromilled activated rubber powder (ARP) has been examined in tests where bituminous binders were partly substituted with secondary material – crumb rubber from processing of old tires in a non-standard format.

Subsequently, bituminous binders are applied to mastic asphalt mixture MA8 where the objective is utilising such mastic asphalt for special applications like narrow pavement lanes close to bridge expansion joints. Although this application is the primary target, the use of such mastic asphalt for large-area structural layers as commonly applied is viable as well.

2. Asphalt Mixture Design
The appropriate combination of crushed aggregate and sand was chosen for mixture MA8 to meet the requirements of technical standards with respect to grading characteristics while being economically viable from the perspective of the quantity of bituminous binder additions; the selected version is marked FSV MA8. The optimal quantity of binder was determined according to CSN 73 6160 [1]. In the case of modification by micromilled active rubber power (ARP), a version with a higher binder content was tested as well since the rubber-modified binders demonstrated poorer workability. The preparation of MA mixtures used aggregate according to the definition stipulated by CSN EN 13043 [2] and CSN EN 13108-6 [3].

| Mixture | Bitumen content | Filler | Crushed aggregate 4/8 Zbraslav | Sand 0/8 Uhy | Sand 0/4 Uhy |
|---------|-----------------|--------|-------------------------------|-------------|-------------|
| MA8 FSV | 7.40%           | 22.24% | 42.12%                        | 3.70%       | 24.54%      |

3. The modifiers or additives used

3.1. Synthetic waxes WE-CM20 and WE-BM
Bituminous binder modification by synthetic waxes is based on the need to modify the current bituminous binders to reduce their working temperatures, or to improve workability of the resulting asphalt mixture (lower temperature, easier application etc.). The mastic asphalt technology per se has high-energy requirements (approx. 2.5 times more energy needed than for compacted asphalt mixtures). The mastic asphalt working temperatures reach up to 250 °C, so the search for a more favourable solution, for instance in the form of wax-modified bituminous binders, is logical – even a slight reduction of working temperatures by 10-15 °C has a rather distinctive economic impact. The use of various wax types for mastic asphalts has been researched since late 1980’s, with the primary aim of improving asphalt mixture processability. In the last ten years, it has been applied as a tool to lower working temperatures and reduce emissions, which might be released at higher temperatures. The synthetic waxes used are innovative products not commonly used in road construction (to improve bituminous binder properties) yet. The product was sourced from a collaborating partner in Germany.

3.2. Micromilled active rubber powder (ARP)
From both the environmental and economical perspective, modification by rubber powder has a considerable potential that has already been demonstrated in the effort to include the material in bituminous binders for standard, compacted asphalt mixtures. With respect to the increasing demand for management of non-renewable sources and with an emphasis on waste recycling, the processing of (not only) used tyres for bituminous binder modification purposes constitutes an interesting option. It must be nevertheless emphasised that from the industrial as well as broader social perspective, the activated material should be tested for reuse in the primary application area – i.e. for rubber products.
The added values of ARP modification of bituminous binders is presently seen in hot mix asphalts (HMA) where such modification positively affects the rheological and mechanical-physical properties of bituminous binders. Asphalt mixtures with modified bituminous binders demonstrate improved resistance to plastic deformation (improved stiffness of the mixture) and increased resistance to frost cracking (behaviour of the binder and, consequently, the mixture in the low temperature range). Some experts also declare a positive effect in the form of a partial improvement of the acoustic parameters, although no major noise reductions can be expected.

The variants depicted below work with a bituminous binder modified by active rubber powder of 0-0.8 mm particle size (adjusted by high-speed milling) with 5 % of a butadiene-based activator. The bituminous binder was modified using the wet process where activated rubber powder is added to the heated bituminous binder (at least 170 °C are required), in our case to the quantity of 15 M% of the bituminous binder. Mixing was carried out in the laboratory with approx. 450 revolutions per minute for at least 30 minutes.

4. Descriptions of the bituminous binders and modifiers applied

The comparison of individual variants involved the standard bituminous binder commonly applied to mastic asphalts – hard paving grade bitumen 20/30 according to CSN EN 12591. The alternative solution and further improvements to common binders consisted of modifying or adjusting the bituminous binders by additives based on synthetic waxes, amide waxes and pulverized (micro-milled) active rubber powder from old tyre processing. The active crumb powder (ARP) used herein showed particle sizes up to 0.8 mm and has been obtained by a specific high-speed milling method as defined by the Czech utility model No. 29199. ARP was supplied by Lavaris s.r.o., which was, in the course of 2016, also involved in the development of a material alternative to the bridge expansion joint damper profile. To improve ARP stability in the bituminous binder, a special butadiene-based activator was added to the entire composition.

Table 2. Characteristics of experimentally assessed bituminous binders.

| Bitumen          | Penetration [0.1 mm] | Softening point [°C] | Penetration index [-] | Rotational viscosity @ 6.87 s⁻¹ |
|------------------|----------------------|----------------------|----------------------|--------------------------------|
| 20/30            | 20.5                 | 62.8                 | -0.4                 | 1.5                             | 0.8                             |
| 20/30 + 3% WE-CM | 17.8                 | 79.9                 | 2.0                  | 1.1                             | 0.6                             |
| 20/30 + 3% WE-BM | 21.9                 | 63.6                 | -0.1                 | 1.0                             | 0.7                             |
| 20/30 + 15% CR (APR5AK) | 15.0            | 81.3                 | 1.8                  | 13.8                            | 12.4                            |
| 20/30 + 15% CR + 2% WE-CM | 14.2            | 87.9                 | 2.5                  | 13.8                            | 11.6                            |

Table 2 indicates the basic characteristics of the modified or doped bituminous binders designed and tested. Besides the conventional tests, the bituminous binders were also described by the penetration index that, aside from auxiliary material characterisation, also provides a simple way to define thermal susceptibility of the bituminous binder. As the workability parameters were important for mixing and further processing of this type of mixture, rotational (dynamic) viscosity was also determined since it could hint at the future behaviour of the mixture with respect to mixing, or any potential reductions in the working temperature and temperature for paving of the mastic asphalt.

What is significant from the point of view of penetration is that the additives applied do not fundamentally modify this characteristic. The synthetic waxes in the test slightly increased the value of penetration while ARP has a stiffening effect, which is completely common for this type of modification. In the case of the softening point, the effect of synthetic wax WE-CM is interesting, raising the value only slightly. The ARP modifier has a more pronounced effect – the softening point is increased considerably. No less important is dynamic viscosity measured by a rotating spindle. For the selected temperature, the effect of the waxes is minimal; it only becomes important from the perspective of mastix in mastic asphalts. On the other hand, the application of micromilled rubber
results in an enormous viscosity increase which means deteriorated workability under the temperature concerned, and for the sake of mastic asphalt, it indicates that higher energy might be required under standard working temperatures $>200 \, ^\circ\text{C}$ when compared to the reference bituminous binder. The application of wax to an ARP-modified binder obviously reduces dynamic viscosity slightly.

5. **Mastic asphalt test specimen preparation**

The preparation of MA mixtures governed by CSN EN 12697-35+A1 [7] defines the temperatures of individual components in the mixture as well as the method and time of mixing. In the case presented by this paper, the important issue is manual mixing where we faced a disproportionately longer mixing time along with a rather major problem of keeping the working temperature within the required range. The preparation of the cubic test specimens followed CSN EN 12697-20 [4] in the case of the specimens for indentation test and compressive strength tests. The same method was applied to cylindrical test specimens for the stiffness modulus test according to CSN EN 12697-26 and the cyclic compression strength according to CSN EN 12697-25 to define the deformation behaviour under repetitive stress. The preparation of the beams for the bending tensile strength test utilised re-heated mixture according to ČSN 73 6160 [1] was used. The mixture obtained from the specimens already tested for the indentation number and compressive strength was used to prepare the beams.

6. **Workability**

The mastic asphalt workability test can be conducted according to ČSN 73 6160 [1], and allows declaring a potential reduction of the working temperatures. Unfortunately, the road construction laboratory of the Civil Engineering Faculty, Czech Technical University, is not sufficiently equipped to perform this test at this moment. Therefore, the individual options are currently only assessed subjectively depending on the demands of manual mixing effort under the selected working temperature, and only serves as an informative indicator. The aggregate and bituminous binder mixture FSV MA8 as designed demonstrated relatively inferior workability where the mixture needed a long time to mix, roughly 10 minutes and more while the standard mixing time should amount to 5 minutes or so. It was also important to pay increase attention to the compaction of the material in the moulds; some specimens in this respect had to be ruled out due to excess air bubbles. From the point of subjective assessment of workability, mixtures with ARP-modified bituminous binders had the poorest behaviour as expected; this was the reason to apply a synthetic wax in combination with ARP to improve workability while mixture properties remain unchanged.

7. **Tested MA mixture parameters**

The evaluation of suitability of modified binders for MA mixtures depended on the indentation, compressive strength and bending tensile strength tests. The set of basic tests was further accompanied by the findings of the stiffness modulus and cyclic compression tests.

| Bitumen         | Flexural strength, 100 mm/min $22^\circ\text{C}$ (MPa) | Modulus of elasticity $22^\circ\text{C}$ (MPa) | Compressive strength $a=40$ mm $22^\circ\text{C}$ (MPa) | Indentation number, 30 min (mm) | Indentation number increment, 60 min (mm) |
|-----------------|------------------------------------------------------|---------------------------------------------|--------------------------------------------------------|--------------------------------|------------------------------------------|
| 20/30           | 7.4%                                                 | 6.83                                       | 245                                                    | 7.69                           | 6.85                                     | 1.33                                    | 0.14                                    |
| 20/30+CR        | 7.4%                                                 | 6.78                                       | 340                                                    | 9.61                           | 12.27                                    | 0.39                                    | 0.04                                    |
| 20/30+CR        | 7.9%                                                 | 9.35                                       | 519                                                    | 10.01                          | 9.05                                     | 0.35                                    | 0.25                                    |
| 20/30+CM        | 7.4%                                                 | 11.05                                      | 485                                                    | 11.64                          | 8.97                                     | 0.95                                    | 0.03                                    |
| 20/30+BM        | 7.4%                                                 | 10.74                                      | 433                                                    | 11.18                          | 9.26                                     | 0.95                                    | 0.14                                    |
| 20/30+CR+CM     | 7.4%                                                 | 6.33                                       | 630                                                    | 11.86                          | 12.73                                    | 0.25                                    | 0.02                                    |
7.1. Indentation test
The fundamental parameter for mastic asphalt assessment is the indentation test performed on cubes. The process of specimen preparation, storage and testing is described by ČSN EN 12697-20 [4]. For mixtures with aggregate of 0/8 mm grading, the standard stipulates a (recommended) indentation number within the range of 1.0 to 5.0 mm after 30 min (0.6-6.5 mm for verifications); the indentation number increment should not exceed 0.6 mm. The data presented does not comply with one part of the margin conditions. The values measured for mixture FSV MA8 variants with improved or modified bitumens demonstrate lower values after 30 minutes than the minimum indentation number requirement of 1.0 mm for a mixture design according to the ITT (the aforementioned minimum threshold value is required in order to reduce the risk of asphalt mixture brittleness and to prevent cracking). In case the criteria for verification tests are applied, the variants with micromilled ARP modification fail. Despite these results, certain clear dependencies on the choice of bituminous binder and the potential benefits of individual bituminous binder modifications or improvements can be seen. Mixtures modified by micromilled rubber (Figure 4, var. 20/30+CR) demonstrate a distinctive improvement in the indentation number with a reduction of approximately 60 % compared to the reference mixture. Modification by waxes also contributes to lower the indentation number value. Bituminous binder modified by the combination of ARP and synthetic wax showed the best results.

![Figure 1. Results of indentation test for mixture variants FSV MA8.](image)

7.2. Compressive strength
Another parameter for the purposes of evaluating suitability of modified binders for mastic asphalt layers is the compressive strength, tested on cubes. The compressive strength was measured both on standard dimension cubes (a=70 mm), and on cubes obtained from the remains of the specimens for the bending beam (flexural) strength test (a=40 mm). The absolute values of the two measurements differed to a point; nonetheless, the trend for comparisons of the individual bituminous binders is similar in both cases. Compared to the reference mixture, the variants with improved or modified binders demonstrate significantly higher compressive strength values. The most preferred variant is, again, the bituminous binder modified by a combination of micromilled rubber and synthetic wax. Even in this case, the modification had a positive effect on the parameters examined. There is a clash with the recommendation given by the existing standards where the resulting compressive strength should fall within the 3.5 – 8.0 MPa range. In this case, too, the threshold value is set to prevent excess stiffness of the material, which is usually linked to increased brittleness, and increased risk of cracking under lower temperatures.
7.3. Bending tensile strength

The test of bending tensile strength, or the calculation of the derived modulus of elasticity characteristic, also clashes with the limits set by the applicable national standard where the mixtures with wax-modified bituminous binders exceed the maximum recommended value of 7.0 MPa. If this value is disregarded, bituminous binder modification or additivation achieved an improvement of tensile strength and, when compared to the reference mixture, even the elasticity modulus increased. In this case, mixtures with bituminous binder modified by WE wax achieved the best results. Higher values were also noticed for a mastic asphalt mixture with increased quantities of ARP-modified bitumen. Both mixtures demonstrate an increase in bending tensile strength by approx. 40 %.

7.4. Stiffness modulus

The determination of stiffness is described in ČSN EN 12 697-26 [6]. The test method C was applied, i.e. the non-destructive test of repetitive tensile stress. Generally, the stiffness modulus is not a traditional characteristic to test in mastic asphalt mixtures. In the case of stiffness, we can see the positive impact of ARP-modified bituminous binders where the test specimens demonstrated significantly better results in stiffness testing under higher temperatures – this potentially indicates their suitability for applications where permanent deformation (rutting) should be reduced. A very interesting parameter is thermal susceptibility, which indicates quality of asphalt mixture. Thermal susceptibility is expressed by the ratio of the stiffness modules measured at 0 °C and the stiffness modulus at 27 °C and 40 °C. It is expected, that higher quality of asphalt mixtures is characterized by
lower temperature sensitivity. The modified mixtures demonstrated a significant reduction of thermal sensitivity in comparison to the reference mixture. In this case, bituminous binder modification by synthetic waxes had a rather negative effect with respect to the stiffness determined under higher temperatures, and to poorer thermal susceptibility. When the bituminous binder was modified by synthetic wax WE-BM, the higher thermal susceptibility resulted from the high stiffness modulus at 0°C, which acts as the basis for the thermal susceptibility test. From the perspective of stiffness values at higher temperatures, the results were comparable to the reference mixture.

Table 4. Results of stiffness modulus determination for mixture variants of FSV MA8.

| Bituminous binder | Stiffness modulus 0 °C (MPa) | Stiffness modulus 15 °C (MPa) | Stiffness modulus 27 °C (MPa) | Stiffness modulus 40 °C (MPa) | Thermal susceptibility S0/S27 | Thermal susceptibility S0/S40 |
|-------------------|-----------------------------|-------------------------------|-------------------------------|-------------------------------|-----------------------------|-----------------------------|
| 20/30             | 7.4%                        | 19157                         | 10247                         | 5155                          | 2124                        | 3.7                         | 9.0                         |
| 20/30+CR          | 7.4%                        | 19344                         | 13101                         | 8500                          | 5037                        | 2.3                         | 3.8                         |
| 20/30+CR          | 7.9%                        | 17597                         | 11738                         | 6612                          | 2998                        | 2.7                         | 5.9                         |
| 20/30+CM          | 7.4%                        | 19504                         | 10047                         | 4406                          | 1553                        | 4.4                         | 12.6                        |
| 20/30+BM          | 7.4%                        | 20113                         | 10776                         | 5507                          | 2139                        | 3.7                         | 9.4                         |
| 20/30+CR+CM       | 7.4%                        | 19504                         | 14857                         | 8363                          | 3826                        | 2.3                         | 5.1                         |

Figure 4. Stiffness modulus determined for mixture variants of FSV MA8.

7.5. **Cyclic compression test**

The test method defined in ČSN EN 12697-25 [5] determines mixture resistance to permanent deformation caused by cyclic stress in a cylindrical specimen. In the case of mastic asphalt mixtures, the dynamic indentation number can express sufficiently the permanent deformation according to the aforementioned test. The results obtained from the measurements correspond with the indentation number measurement where the dynamic hardness number is considerably superior for MA mixtures with ARP-modified bituminous binder. This test shows that increased quantities of CRMB binder in mastic asphalt mixtures do not induce any changes in the characteristics tested. Contrastingly, the combination of micromilled ARP and synthetic wax brings on a slight reduction of the characteristic in the case of creep modulus.
Figure 5. Creep modulus and dynamic indentation number from the cyclic compression test (@ T=60 °C) for mixture variants FSV MA8.

8. Conclusions
The variants of the new MA8 mixture designs with modified or additivised bituminous binders demonstrated an interesting potential where the addition of modifiers positively affects the resulting parameters of assessed mastic asphalt types.

The positive effect of binder modified by micromilled active rubber powder was clearly proven when the mixtures modified thereby demonstrated increased strength characteristics, low thermal susceptibility, high stiffness modulus even under high temperatures, high resistance to cyclic stress and to permanent deformation. Unfortunately, the improved parameters came at the expense of subjectively poorer workability where the mixtures showed higher demands on mixing energy during test specimen preparation.

Bituminous binder modification by a new generation of synthetic waxes, WE-CM and WE-BM, also delivered a benefit in the form of improved strength parameters, particularly bending tensile strength. From the perspective of the stiffness modulus, thermal susceptibility and resistance to permanent deformation, mastic asphalts seem to be more favourably enhanced by WE-BM wax modification where the mixtures with this binder achieved equal or slightly better parameters than the reference mixture. The effect of the waxes on processability, and thus potential working temperature reduction, has not been verified or sufficiently proven, and remains to be examined further.

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