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Analysis of possible use of pyrolytic products as binders in asphalt mixes

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Preliminary report

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Analysis of possible use of pyrolytic products as binders in asphalt mixes

New binders are produced in order to reduce dependence on crude oil. This study aims to evaluate pyrolytic products and their blends with bitumen, as a possible substitution for bitumen. Bitumen of the penetration grade 70/100 was used as a matrix for the blends. Standard test were conducted in order to determine mechanical properties of the reference bitumen, pyrolytic products, and their blends. The results obtained during this study indicate that pyrolytic products can be used as a substitution for bitumen originating from crude oil or, in proper proportions, as an additive to that bitumen.

Key words:

bitumen, pyrolysis, scrap tyres, plastic waste

Prethodno priopćenje

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Analiza mogućnosti primjene produkta pirolize kao zamjene za bitumen u asfaltnim mješavinama

Novе vrste veziva se proizvode kako bi se smanjila upotreba sirove nafte. Ispitivanje je provedeno s ciljem analize proizvoda pirolitičkog spaljivanja i njegove mješavine s bitumenom, kao moguće zamjene za bitumen. Kao kontrolni bitumen korišten je cestograđevni bitumen 70/100. Uobičajenim ispitivanjima utvrđena su mehanička svojstva kontrolnog bitumena, proizvoda pirolize i njihovih mješavina. Rezultati ispitivanja pokazuju da se proizvod pirolize može primjenjivati kao zamjena za bitumen ili, u prikladnom omjeru, kao dodatak bitumenu.

Ključne riječi:

bitumen, piroliza, otpadne gume, plastični otpad

Vorherige Mitteilung

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Analyse der Anwendungsmöglichkeiten von Pyrolyseprodukten als Binder in Asphaltmischungen

Neue Arten von Bindemitteln werden hergestellt, um die Anwendung von Rohöl zu verringern. Die Untersuchungen wurden durchgeführt, um Produkte pyrolytischer Zersetzung und ihre Mischungen mit Bitumen als möglichen Ersatz für Bitumen zu analysieren. Als Kontrollbitumen wurde Straßenbaubitumen 70/100 angewandt. Durch herkömmliche Prüfungen wurden die mechanischen Eigenschaften des Kontrollbitumens sowie der Pyrolyseprodukte und ihrer Mischungen ermittelt. Die Resultate zeigen, dass Pyrolyseprodukte als Ersatz für Bitumen, oder in entsprechendem Anteil als Zusatz zu Bitumen, angewandt werden können.

Schlüsselwörter:

Bitumen, Pyrolyse, Altreifen, Plastikabfall

1. Introduction

Although binder is the smallest part of the asphalt mixture, its role is very important. Binders influence behaviour of mixtures (both at low and high temperatures [1]), susceptibility to damage caused by moisture [2], and their durability.

Bitumen is the most frequent type of binder. Bitumen consumption has been decreasing in recent years in Europe. Nowadays, bitumen used in paving industry is mainly produced from crude oil. Bitumen distilled from crude oil represents on an average 3 - 4 % of all refinery production. Out of 1300 (or more) types of crude oil, only approximately 10 % can be used to produce bitumen that meets stringent engineering requirements of the present-day end users [3]. Because of an increasing demand for different types of fuel, only a small percentage of crude oil is intended for the production of bitumen. These are the main reasons for recent rises in the price of bitumen [4].

In the past years, many researchers have tried to find a suitable alternative for bitumen in asphalt mixtures. As a direct result of their studies, several alternative binders have been produced from different raw materials, e.g.: biomass (agricultural crops, forestry by-products) [5], wood [6, 7], swine manure [8], or cooking oil [9, 10]. Alternative binders, produced from biological products, are sometimes called bio-binders [5]. Alternative binders are used for full or partial substitution of bitumen, or as an additive (e.g. rejuvenator added to an aged, reused bitumen [11]). The bio-binders are characterised by lower viscosity, [12] and asphalt mixtures exhibit better performance at both low and high temperatures [6, 13, 14]. In our research, we investigated pyrolytic products as a possible bitumen substitution, or additive to bitumen. The pyrolytic product tested in the scope of this research was produced through pyrolysis of scrap tyres and plastic waste.

Increasing amounts of waste are produced each year in Europe. Thus a total of 2.52 billion tonnes of waste was generated in the European Union (28 countries) in 2012, out of which plastic waste accounted for 18.6 million tonnes [15]. In 2010, 3.3 million tonnes of scrap tyres were collected in Europe (EU 27 + NO + CH). Most of the scrap tyres are being reused either as a material or for energy recovery. Nevertheless, 157 thousand tonnes are stored in landfills [16].

Waste legislation applicable in the European Union dictates the following waste-management hierarchy: reuse, recycling, recovery and, as the last option, disposal [17]. This is the reason why waste was used as raw material for the new product in our study. The pyrolytic product was produced from plastic waste and scrap tyres. The analysis was made to determine whether the pyrolytic product can be used as an adequate substitution for bitumen binder. Alternatively, a partial replacement of bitumen with our product was analysed. The third option was to use the pyrolytic product as an additive only. In our case, the focus was on determining whether the adhesion between the aggregate and the binder was better after the pyrolytic product was used instead of the pure bitumen only.

Physical properties of pyrolytic products were studied and compared with properties of the reference bitumen made of crude oil.

2. Materials and methods

2.1. Materials

Both pyrolytic products tested in the scope of this study (designated as PP 1 and PP 2) were obtained by pyrolysis of scrap car tyres, using also a negligible part of plastic waste. The products were generated by condensation of vapours from a batch reactor, where the pyrolysis was conducted at 450 °C. The vapours came from the reactor at 370 °C, and were then cooled and condensed at 260 °C. The final product was extracted from the condenser and cooled. By repeating the procedure with slight variations, two samples were obtained, namely PP 1 and PP 2. The only difference between them was that the pyrolysis took longer in case of PP 2 sample. Laboratory equipment used in pyrolysis is shown in Figure 1.



Figure 1. Laboratory equipment for pyrolysis

Bitumen with the penetration grade of 70/100 (B 70/100), produced by MOL, Hungary, was used as the matrix for the blends.

Bitumen was mixed with PP 1 and PP 2 samples that were obtained by pyrolysis. All blends were produced in laboratory by adding a controlled quantity of PP 1 and PP 2 to the bitumen. Bitumen and PP sample blends were produced by mixing the two components with a ratio of 1:1 or 9:1. Each blended sample weighed 100 g, i.e. 50 g of PP sample and 50 g of bitumen, and 90 g of bitumen and 10 g of PP sample, respectively. The

blending process consisted of heating both components to the mixing temperature for 60 minutes, and pouring the required mass into a small container. The two components were then manually stirred together for approximately 60 s in order to produce a uniformly distributed binder blend. The blends were then poured into sample containers, and stored at 5 °C prior to testing. The mixture 1 is a blend of bitumen and PP 1, while the mixture 2 is a blend of bitumen and PP 2.

The aggregate fraction 8/11 from Bleiburg, Austria, was used in order to test the binder and aggregate affinity. It was assumed that the aggregate was silicate rock by chemical composition. The test samples and their proportions are presented in Table 1.

Table 1. Samples examined in research

Sample	Proportion of bitumen B 70/100 [%]	Proportion of pyrolytic product [%]
Bitumen B 70/100	100	0
PP 1	0	100
Mixture 1 (9:1)	90	10
Mixture 1 (1:1)	50	50
PP 2	0	100
Mixture 2 (9:1)	90	10
Mixture 2 (1:1)	50	50

2. 2. Methods

Standard testing methods normally applied for bituminous mixtures, as defined in European standards, were used to characterize the samples. The penetration, softening point, ductility and the Fraass breaking point were determined in order to make comparison between the bitumen, pyrolytic products, and their blends.

2.2.1. Density

The density of the samples was determined with the capillary – stoppered pycnometer as described in EN ISO 3838:2004 [22].

2.2.2. Viscosity and mixing temperatures

The viscosity of the binders was assessed, and the mixing temperatures were determined using the rotational viscometer Haake RS50.

2.2.3. Penetration test

The needle penetration test according to EN 1426:2007 was used to determine consistency of samples and their blends [18]. Samples were tested with a standard needle weighing 100 g, which was applied vertically onto the sample for 5 s at 25 °C. Test results are presented as penetration in tenths of a millimetre.

2.2.4. Softening point temperature

The Ring and Ball method as determined in EN 1427:2007 was used to assess softening properties of samples [19]. The softening point was determined by heating the samples cast in brass ring to the point where the steel balls, lying on top of the binder, became enveloped and eventually fell through the binder for a prescribed distance of 25 ± 0.4 mm.

2.2.5. Fraass test

The brittleness at low binder temperatures was determined by the Fraass breaking point test according to EN 12593:2007 [20]. A binder sample was evenly applied onto a metal plate. This plate was then cooled at a constant temperature and flexed repeatedly until failure of the binder layer. The temperature at which the first crack occurs is referred to as the Fraass breaking point.

2.2.6. Testing tensile properties

Tensile properties of the binders were determined by the force ductility method in accordance with EN 13589:2008 [21]. After casting binder, moulds were transferred to traction plates and then stretched at the prescribed speed at 25 °C up to the elongation of 1170 mm. Brittle break occurred when the specimen failed before reaching the prescribed elongation.

2.2.7. Affinity between aggregate and binder

The last test was carried out in order to determine affinity between aggregate and binder in accordance with EN 12697-11:2012 [23]. This affinity was assessed by visual examination of aggregate coated with binder, following mechanical stirring in the presence of water. The degree of binder coverage was determined as an average proportion of the surface area of aggregate particles covered with binder (expressed in %).

3. Results and discussion

3.1. Density

The density of bitumen and pyrolytic product was determined in the first stage. The results are given in Table 2. These results show that the pyrolytic products, i.e. both PP1 and PP2, have a greater density compared to density of bitumen.

Table 2. Density of reference bitumen and pyrolytic products

Sample	Density [g/ml]
Bitumen B 70/100	1.0170
PP 1	1.0905
PP 2	1.1300

3.2. Viscosity and mixing temperatures

The viscosity was measured using the rotational rheometer Haake RS50 in order to determine the mixing temperature for each of the pyrolytic products. The mixing and compaction temperatures of asphalt mixtures were determined at elevated temperatures from the plain asphalt viscosity – temperature charts at 0.170 ± 0.02 Pas and 0.280 ± 0.03 Pas, respectively [24]. The rotational viscometer test method is presently considered to be the most practical for determining the viscosity of bitumen, since the instrument allows testing various binders over a wide range of temperatures [25]. The viscosity is the ability of a liquid to resist flow. Therefore, the binders with a high viscosity have difficulty with flowing, while the binders with low viscosity tend toward the state of a Newtonian liquid [11]. The mixing temperature for bitumen B 70/100 is $T_{B\ 70/100_mix} = 124.1$ °C. For PP 1 the temperature is $T_{PP1_mix} = 92.6$ °C, and for PP 2 the temperature is $T_{PP2_mix} = 133.1$ °C. Figure 2 shows dependence of viscosity on temperature for bitumen B 70/100, for samples PP 1 and PP 2, respectively. The slopes of both pyrolytic products are steeper than those representing bitumen. This is contrary to our expectations because Airey et al. discovered in their research that synthetic binder has a flatter slope, compared to the curve for bitumen derived from crude oil [26]. A steeper slope means smaller temperature range of applicability, but at the same time the

binder is more susceptible to the change of viscosity with temperature [27].

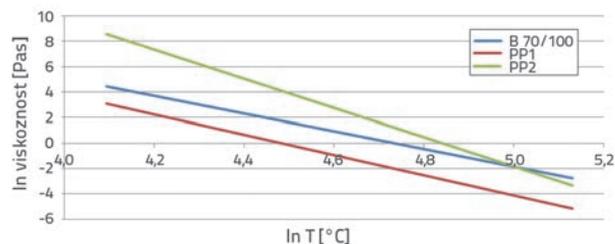


Figure 2. Viscosity at various temperatures

Viscosity values are not constant simply because they are affected by many conditions. The most important ones are temperature and shear rate. Figure 2 shows the Heukelom diagram, which represents the dependence of viscosity and temperature. As already mentioned, viscosity is also shear-rate dependent.

The dependence of shear rate on viscosity is shown in Figure 3. The left side of Figure 3 shows that bitumen and PP 1 are ideally viscous fluids at $T = 80$ °C and at shear rate from 0 to 10 1/s. This means that the viscosity of ideally viscous fluids or Newtonian fluids is shear-independent. PP2's viscosity is shear-rate dependent, and decreases with an increase in shear rate. This behaviour is called shear-thinning or pseudo-plastic flow behaviour.

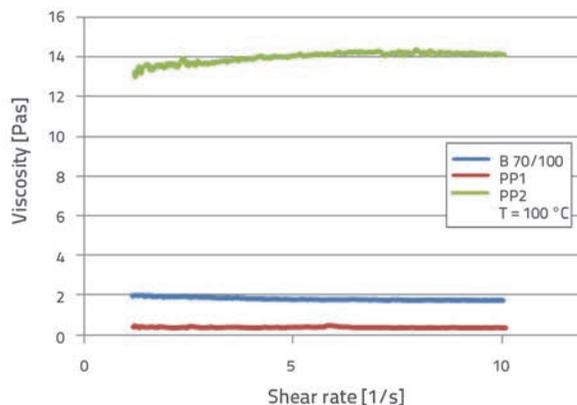
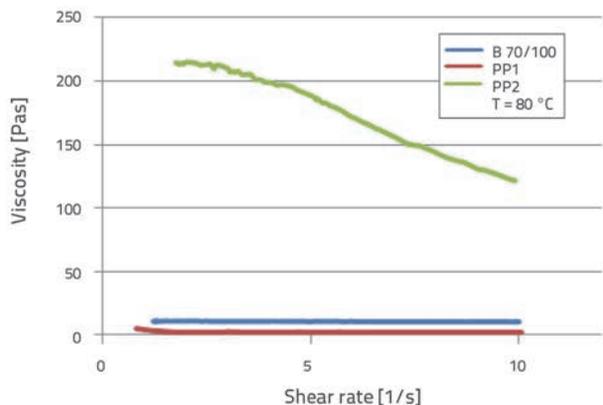


Figure 3. Viscosity curve of tested samples at $T = 80$ °C (left) and $T = 100$ °C (right)

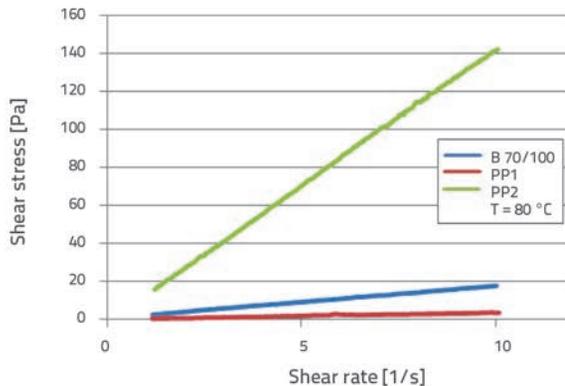
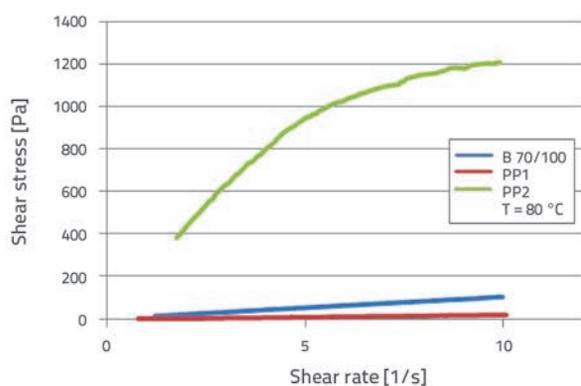


Figure 4. Flow curve of tested samples at $T = 80$ °C (left) and $T = 100$ °C (right)

The right side of Figure 3 shows the dependence of shear rate on viscosity at a higher temperature, $T = 100\text{ }^{\circ}\text{C}$. At this temperature B 70/100 and PP 1 are ideally viscous liquids. But at the temperature $T = 100\text{ }^{\circ}\text{C}$, PP 2's viscosity is shear-rate independent and it is also an ideally viscous liquid.

Figure 4 shows flow curves of tested samples at two temperatures. At the lower temperature, $T = 80\text{ }^{\circ}\text{C}$ (left), the flow curve of B 70/100 and PP 1 exhibits an ideally viscous behaviour, and PP 2 shows a pseudo-plastic behaviour, as observed at viscosity curves. Flow curves at $T = 100\text{ }^{\circ}\text{C}$ (right) also confirm our findings, i.e. the curves show an ideally viscous behaviour of tested samples.

3.3. Penetration test

Penetration test results are presented in Table 3. The penetration of the reference bitumen is 80 (1/10 mm). PP 1 is softer as its penetration value is 125.3 (1/10 mm), while PP 2 is harder as its penetration value is 13.3 (1/10 mm). The penetration of the mixture increased after various proportions of PP1 were added, which is an indication that the product softened the bitumen. The mixture 1, 1:1 had the highest penetration value amounting to 140.7 (1/10 mm). The penetration value of the mixture was higher than that of the two components from which the blend was made. Typically, the mixing of two bitumens with different penetration values produces a bitumen characterised with an intermediate penetration [28].

But in this case, the objective was not to mix two bitumens, but bitumen and a pyrolytic product. One of possible explanations for the results obtained would be that our PP 1 sample has a lower glass transition temperature than the bitumen (which is in agreement with the softening point results). The chosen proportion of PP1 in bitumen of the mixture 1 1:1 decreases glass transition of the blend, which explains the softening [28]. Blends of the bitumen and the PP 2 have a similar penetration value (75.9 and 73.1 tenths millimetres), which means that the proportion of the additive was not relevant for the results.

We expected that PP 1 will make the bitumen softer, because of its high penetration value, and that PP 2 will make the bitumen harder, because of its low penetration. The results confirmed our predictions, but are contradictory to the findings presented by Chaala et al. [29].

After 14 days, the depth of penetration decreased for all samples with the exception of PP 2, for which the penetration value actually increased.

The values written in brackets are the values calculated according to the mixing law specified in EN 13108-1:2006/AC:2008 [30]. The penetration values were calculated using the following Eq. (1):

$$\log(\text{pen}_m) = \frac{B_0}{100} \log(\text{pen}_0) + \frac{B_{PP}}{100} \log(\text{pen}_{PP}) \quad (1)$$

where are:

B_0, B_{PP} - percentages of the bitumen and pyrolytic product in the blend

$\text{pen}_0, \text{pen}_{PP}, \text{pen}_m$ - penetration values for bitumen, the pyrolytic product and the blend.

The comparison of measured and calculated results show that the results do not match well for PP 1, but that the match is adequate for PP 2, even though the differences in values for the reference bitumen and PP 2 are much higher than for the reference bitumen and PP 1.

3.4. Softening point temperature

The softening point test is used to determine the binder temperature when it reaches the flow state. The softening point is also an indication of the point in time when the road pavement will likely deform due to high temperatures. The temperature sensitivity of asphalt is relatively low when the softening point is high [26]. The softening point results are presented in Table 4. The softening point of PP 1 is lower and the softening point of PP 2 is higher than the reference bitumen's softening point. The blends with the pyrolytic product have a softening point similar to that of the bitumen. The same applies even for the mixtures with the ratio of 1:1. Chaala et al [28] also obtained similar results about the softening point of the blends.

The softening point was also calculated using the mixing law [30]. The results are given in brackets in Table 3. The softening point was determined by the Eq (2):

$$T_{R\&B,m} = \frac{B_0}{100} T_{R\&B,m} + \frac{B_{PP}}{100} T_{R\&B,PP} \quad (2)$$

Table 3. Penetration test results

Sample	Penetration depth [1/10 mm]	Penetration depth [1/10 mm] after 14 days
Bitumen B 70/100	80.0	-
PP 1	125.3	109.2
Mixture 1 (9:1)	89.8 (80.9)	73.2
Mixture 1 (1:1)	140.7 (106.1)	134.4
PP 2	13.3	16.0
Mixture 2 (9:1)	75.9 (79.6)	66.2
Mixture 2 (1:1)	73.1 (76.5)	68.0

Table 4. Softening point and Fraass breaking point

Sample	Softening point [°C]	Fraass breaking point [°C]
Bitumen B 70/100	47.6	-17
PP 1	39.6	-8
Mixture 1 (9:1)	45.7 (47.4)	-16
Mixture 1 (1:1)	46.6 (47.1)	-14
PP 2	75.1	+7
Mixture 2 (9:1)	46.9 (47.5)	-17
Mixture 2 (1:1)	49.2 (48.4)	-11

where are:

B_{σ} , B_{pp} - percentages of bitumen and pyrolytic product in the blend
 $T_{R\&B,\sigma}$, $T_{R\&B,PP}$, $T_{R\&B,m}$ - softening-point temperatures for bitumen, pyrolytic product and blend.

Just like in case of penetration, calculated results match better with measured ones for PP 2 compared to PP 1.

3.5. Fraass test

The Fraass breaking point results shown in Table 4 prove that pyrolytic products exhibit worse low temperature properties, but that their blends with the bitumen are comparable to bitumen properties. Similar deviations from expected values were noted for all tests on the mixtures of bitumen and pyrolytic product. It can be seen from the properties of mixtures that they do not act as a mixture of two physical fluids without interactions. Furthermore, it can be observed that even when added to bitumen in small quantities, the pyrolytic product, PP 1, and

even more PP 2, act like plasticizers [31], reducing viscosity and making the mixture more fluid.

3.6. Tensile properties testing

All tested samples exhibit little or no tensile properties. The results are presented in Table 5. Only the sample of the reference bitumen was elongated to maximum length. PP 1 and mixtures with PP 1 could stretch more than PP 2 and its blends. The largest force was measured on PP2, and it yielded at the shortest elongation of 4.1 cm, which means that PP2 is the most brittle product. This also confirms our Fraass test results. It was expected that pyrolytic products will have better tensile properties. It can only be assumed that the composition of scrap tyres was completely changed by pyrolysis. The results of the mixture 1 (1:1) are contrary to our expectations. After the results of B 70/100 and PP 1, we assumed that their blend would have the elongation that is at least equal to that of PP 1. This can be explained by assuming that the blend is affected by interactions between bitumen and the pyrolytic product. The

Table 5. Tensile properties of binders

Sample	Force [N]	Elongation [cm]	Energy [J/cm ²]
Bitumen B 70/100	1.1	117.0	0.1
PP 1	0.3	97.02	0.0
Mixture 1 (9:1)	0.7	88.21	0.0
Mixture 1 (1:1)	0.5	39.99	0.0
PP 2	72.7	4.1	2.4
Mixture 2 (9:1)	1.2	45.4	0.1
Mixture 2 (1:1)	1.2	30.1	0.0

Table 6. Affinity between aggregate and binder

Sample	0 [h]	6 [h]	24 [h]	48 [h]	72 [h]
Bitumen B 70/100	100	90	70	40	30
Mixture 1 (1:1)	100	90	80	55	45
PP 1	100	95	90	70	50
Mixture 2 (1:1)	100	95	85	70	50
PP 2	100	95	60	40	25



Figure 5. Affinity between bitumen (left), mixture 1:1 (centre), PP 1 (right) and aggregate after the 72 h test according to EN 12697-11

blend's cohesion has decreased compared to the cohesion of its components.

3.7. Affinity between aggregate and binder

The composition of aggregates used in road pavements varies considerably, and ranges from principally siliceous to entirely calcareous materials [30]. Prior to our testing, the aggregate was washed in order to remove all dust particles from the aggregate's surface. The aggregates are highly heterogeneous. Although aggregates can be classified as the limestone, granite, or gravel, each of these materials can in fact be composed of a variety of minerals. This variety in the composition and, hence, in chemistry is reflected in the aggregate's adhesion behaviour. It should be noted that the aggregate composition and type have a much stronger influence on the adhesion and bonding of the asphalt to the surface, than does the binder [31]. The results of the affinity between the binder and aggregate are presented in Table 6. The affinity of bitumen was comparable to the affinity of PP 2. PP 1 and the blends exhibit similar results, which are better than the results of PP 2 and bitumen. This is also shown in Figure 5.

The difference in stripping began to show after 24 hours. At the end of the test, the blends and PP 1 covered 45 % to 50 % of the aggregate. That is almost double of what was registered for bitumen and PP 2, namely 25 % to 30 %. Figure 5 shows the results obtained by testing after 72 hours. As already explained in previous section, the bitumen and pyrolytic product blending weakens cohesion of the blend. Therefore, the ductility test

results are worse. According to affinity test results, it can be assumed that the blends have a better adhesion compared to bitumen and PP2.

4. Conclusions

A pyrolytic product was tested as a possible substitution for binders produced from crude oil. Two different pyrolytic products, the first one softer than the second, were tested. The utilisation of PP 1 as a substitution for bitumen would be appropriate, because it exhibits similar or even better mechanical test results compared to bitumen. The main weakness of PP 1 is its smaller temperature range, since it has a lower softening point and a higher Fraass breaking point when compared to bitumen.

PP 2 is too hard to be used as a full substitution for bitumen. PP 2 has the best results for the softening point but, at the same time, it has the highest Fraass breaking point. We can also use the pyrolytic product as a partial substitution for bitumen (blends containing 50 % of PP). In this case, the mixtures exhibit properties similar to bitumen, and they have a better affinity with the aggregate compared to bitumen. The effects of reduced viscosity and improved affinity between the binder and aggregate also point to the possibility of using pyrolytic products as rejuvenators. The effect of reduced cohesion of the blends could prove problematic for any future testing. Tests of asphalt mixtures containing pyrolytic product are currently planned to obtain further information on this issue. Besides, by decreasing dependency on crude oils, the new product could also have a positive effect on the environment since it is produced from waste material.

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