Chapter

Rejuvenator Obtained by Pyrolysis of Waste Tires for Use in Asphalt Mixtures with Added Reclaimed Asphalt

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Abstract

Although in recent years, big progress has been made in the field of recovering waste tires, they still represent an unwanted waste and their production is constantly increasing. We can use waste tires as a raw material for a new product. In our study, multiple liquid products were produced by pyrolysis of waste tires. After extensive testing of their properties, we selected the most suitable pyrolytic product for the purpose of rejuvenation. Rejuvenators are designed to soften the old, brittle and stiff aged bitumen in reclaimed asphalt. Bitumen with its viscoelastic characteristics is the most important component of asphalt and dictates its behaviour. Commonly bitumen, after adding rejuvenator, becomes less viscous, more ductile and its coating properties are restored. By using a pyrolytic rejuvenator, the proportion of reclaimed asphalt added to the asphalt mixture was increased. The reuse of reclaimed asphalt and waste tires means a reduction in waste material and is therefore important for the preservation of the environment and sustainable development.

Keywords: pyrolyse, rejuvenator, bitumen, reclaimed asphalt, waste tires

1. Introduction

Bitumen combined with stone aggregate creates an asphalt mixture, which is the most common surface layer of the roads. Bitumen in asphalt mixtures ages already during the production, transport and installation of asphalt mixture and the process of ageing continues during the use of asphalt. Ageing processes are influenced by several factors, the most important of them are: temperature, UV radiation and oxygen exposure. Subsequently, bitumen becomes harder and more brittle, its viscosity increases, adhesion and cohesion deteriorate, which leads to the ravelling and the formation of cracks in asphalt mixture [1–3]. With the use of special additives, the so-called rejuvenator, bitumen restores its basic properties [4, 5]. Furthermore, rejuvenators allow mixing and installation at lower temperatures, which reduces production costs and energy consumption, therefore the production of asphalt is more environmentally friendly. The rejuvenator enables to re-install the aged bitumen from reclaimed asphalt (RA) into the fresh asphalt [6]. The first rejuvenator was used as early as 1960 [7]. Over the years, several different products have been used as rejuvenators: emulsions, oil components with a high content of maltene, soft bitumen with high penetration, tertiary amines, waste vegetable oils, waste motor oils and various combinations of these materials [8, 9]. A rejuvenator made from waste tires by a pyrolysis process, a pyrolytic rejuvenator, was developed in our study [10].

Pyrolysis in technical terms is a process in which the chemical decomposition of a substance takes place at elevated temperatures and without the presence of oxygen. Because of the increasing number of vehicles, demand for car tires is also increasing, and as a consequence their production is increasing (324 million tires were sold in 2019 in Europe [11]), resulting in an increasing number of waste tires at the end of their service life. European Directive on the landfill of waste 1999/31/EC [12] has prohibited the disposal of waste tires at landfill sites since 2006, as their improper storage may endanger human health and pose a threat to the environment, e.g. fire, the spread of rodents and insects and dangerous emissions that can be released into the air or the earth. In addition to this directive, waste management is also significantly influenced by the waste directive 2008/98/ES [13] which sets out a 5-step waste hierarchy, namely: prevention, preparing for re-use, recycling, other recovery (e.g. energy recovery) and disposal. Pyrolysis is a suitable process for the treatment of waste tires, as it has a small impact on the environment, and at the same time obtained products that can be used further. In the past, pyrolytic products from waste tires have been mainly used as additives to reduce the temperature sensitivity of bitumen [14–17]. The pyrolytic product from waste tires pyrolysis may contain high concentrations of polycyclic aromatic hydrocarbons (PAH). Therefore, the slow pyrolysis process of waste tires was performed at a relatively low temperature where the highest treatment temperature (HTT) did not exceed 500°C. Consequently, a product with low PAH content, i.e. lower than those reported in existing studies [18, 19] was obtained.

In the first phase of the presented study, several different pyrolytic products were developed [20]. Among them, the most appropriate for the purpose of rejuvenator was selected. In the second part, the influence of the pyrolytic rejuvenator on the properties of the non-aged and aged bitumen was examined. Results of the tests proved that the pyrolytic rejuvenator revived aged bitumen [21]. In the last phase, the pyrolytic rejuvenator was used in asphalt mixtures, to which the percentage of reclaimed asphalt was gradually increased [22].

2. Pyrolytic product for the purpose of rejuvenation

Pyrolytic products in the research were obtained by the process of slow pyrolysis. In the first phase, the conditions of the pyrolysis process e.g. the duration (from 10 min to 150min) and the pyrolysis temperature (from 280–500°C) were changed in order to obtain different pyrolytic products. In the second phase, the pyrolytic products were modified with various oils and crushed rubber. All composed pyrolytic products were similar to bitumen. Their properties and properties of their blends with reference bitumen were evaluated by standard European mechanical tests, which are usually used to determine the properties of bitumen. As reference bitumen and as a matrix of the blends a 50/70 penetration grade bitumen (B50/70) was used. All the blends were laboratory produced by adding a controlled quantity of the pyrolytic product to the bitumen. The blends of bitumen and pyrolytic product were commonly produced by mixing two of the components in ratios of 1:1 (labelling them B+No. of pyrolytic product). Only the blends of pyrolytic product No. 11 were also prepared in smaller concentrations (11_x%, where x represents a share of the pyrolytic product).

The softening properties were determined by using the Ring and Ball method (RB) according to the EN 1427 [23]. Fraass breaking point test according to the EN 12593 [24] was used to determine the brittleness of the products at low

	RB	Fraass breaking point	Force ductility				Mixing temperature (@ η = 0.17 Pas)	Compaction temperature (@ η = 0.26 Pas)
			T	Elongation	Force (max)	Energy $E'_{0,4}$-$E'_{0,2}$		
	[°C]	[°C]	[°C]	[mm]	[N]	[J/cm^2]	[°C]	[°C]
B 50/70	50.5	−11	25	1500*	0.97	0.07	141.2	121.9
1	44.6	−14	25	236	0.1	0.00	153.6	127.1
2	59.1	2	25	187	1.01	0.06	192.9	159.9
3	54.6	−18	25	137	0.34	0.02	169.2	141.2
4	61.3	−12	25	257	1.41	0.10	185.6	156.8
5	60.6	−8	25	354	1.41	0.12	187.8	158.6
6	47.3	−20	25	232	0.27	0.02	169.3	138.8
7	45.9	−14	25	827	0.41	0.02	139.4	116.4
8	65.6	+1	25	1309	12.59	0.91	177.6	153.1
9	42.0	−20	15	145	0.14	0.01	190.6	150.3
10	30.4	−23	15	202	0.04	0.00	155.7	121.5
11	—	−1	15	—	—	—	202.3	154.4
12	43.9	−21	15	325	0.07	0.01	212.4	173.6
13	54.9	−21	15	322	0.29	0.05	255.5	207.5
14	37.5	−22	15	230	0.0	0.00	150.3	122.4
B + 1	43.3	−13	25	902	0.31	0.02	152.1	125.8
B + 2	46.4	−8	25	534	0.52	0.03	146.0	123.3
B + 3	47.1	−18	25	335	0.58	0.04	143.6	121.9
B + 4	46.9	−11	25	1500	0.96	0.06	146.6	123.3
B + 5	46.6	−10	25	1492	0.82	0.05	152.1	128.0
B + 6	44.8	−18	25	760	0.43	0.03	142.2	119.2
B + 7	45.7	−14	25	1500*	0.72	0.04	140.1	118.2
B + 8	52.3	−6	25	1500*	3.15	0.22	150.7	129.1
B + 9	38.1	−24	15	1500*	0.67	0.10	158.6	129.3
B + 10	37.2	−22	15	1500*	0.61	0.06	135.3	113.8
B + 11	—	−24	—	—	—	—	140.3	105.2
11_5%	42.8	−17	15	1343	2.6	0.23	183.3	117.2
11_10%	36.6	−23	15	746	0.45	0.03	134.8	111.8
11_20%	22.4	−23	—	—	—	—	129.6	102.5
B+12	40.2	−21	15	1326	0.10		176.8	143.5
B+13	45.4	−14	15	1500*	0.28	0.03	169.9	140.8
B+14	42.6	−19	15	1122	0.20	0.00	139.6	117.2

** Maximum elongation.*

Table 1.
Results of standard mechanical tests of pyrolytic products and their blends with reference bitumen [20].

temperatures. The tensile properties of the bitumen and pyrolytic products were determined by the force ductility method in accordance with EN 13589 [25].

For the production of asphalt, it is important to determine the optimum mixing temperature for specific bitumen, which is the temperature to be maintained at the asphalt plant during mixing. For laying of asphalt the compaction temperature, at which the mixture has to be compacted on sites has to be determined. The mixing and compaction temperatures for pure pyrolytic rejuvenators and bitumen blends were determined with the rotational viscometer Haake RS50. Both temperatures determine recommended viscosity of bitumen: 0.170 ± 0.02 Pas form mixing and 0.260 ± 0.03 Pas for compaction, respectively [26].

Results in **Table 1** show most pyrolytic products lowered the softening point of the blends in comparison with reference bitumen. That means the pyrolytic product could also lower the high softening point of the extracted bitumen in reclaimed asphalt. For our product, this is a good feature, as we want to use it as a rejuvenator. In addition, almost all pyrolytic products lowered the Fraass breaking point, meaning the temperature range is extended.

Results of final elongation at force ductility tests show that all pyrolytic products broke before they reached the maximum possible length (1500 mm, designated 1500* in **Table 1**) to the contrary of reference bitumen. The pyrolytic products 4, 5, 7, 8, 9, 10, and 11 (in adequate concentration) retained the elongation ability of the reference bitumen in the blends. All other pyrolytic products shortened the elongation of the blends. Except for the pyrolytic product no. 8, all other values of

	Viscosity		
	@ T = 60°C	@ T = 100°C	@ T = 150°C
Label	**[Pas]**	**[Pas]**	**[Pas]**
B 50/70	209.37	2.03	0.12
B + 1	71.50	1.56	0.23
B + 2	104.19	1.67	0.17
B + 3	110.84	1.67	0.15
B + 4	92.59	1.59	0.17
B + 5	99.02	3.36	0.18
B + 6	61.04	1.31	0.14
B + 7	71.39	1.21	0.13
B + 8	234.85	2.60	0.21
B + 9	56.31	1.74	0.29
B + 10	30.91	0.90	0.12
B + 11	6.93	0.43	0.15
11_5%	73.03	1.35	0.12
11_10%	31.14	0.90	0.09
11_20%	9.01	0.53	0.09
B + 12	94.83	2.78	0.58
B + 13	144.06	3.33	0.45
B + 14	59.34	1.25	0.12

Table 2.
Results of viscosity measurements of the blends (pyrolytic products with reference bitumen) [21].

maximum force measurements were in the range around 1 N or even smaller. The ductility test was performed at 25°C for the pyrolytic products (and their blends) from 1 to 8. For others, the test temperature was lowered, to 15°C. Pyrolytic product 11 and its blends, B+11 and 11_20%, could not be tested even at the temperature of 15°C, so we did no performed test for those three samples.

The results of the mixing and the compaction temperatures of the pyrolytic products are higher than the reference bitumen's. Although this indicates that the pyrolytic products have a higher viscosity, also the homogeneity of the sample influenced the viscosity. Inhomogeneous samples (2, 9, 10, and 11) have higher viscosity. Blends of pyrolytic products 7, 10, 11 and 14 have lower mixing and compaction temperatures.

The viscosity of the blends was determined at three different temperatures and was measured at the constant shear rate. Results (**Table 2**) show that the viscosity of the bitumen and the blends is decreasing with increasing temperature. In general, the viscosity of all blends is lower than the viscosity of the references bitumen at 60 °C and 100°C. At 150°C viscosity of almost all blends was at least as high as the reference's bitumen; nevertheless, the absolute values of the viscosity were very small.

Based on the results of these tests we decided that out of the fourteen manufactured and modified pyrolytic products, the most suitable pyrolytic product for the role of a pyrolytic rejuvenator was number 14.

3. Effect of pyrolytic rejuvenator on non-aged and laboratory aged bitumen

After selecting the appropriate pyrolytic rejuvenator, the focus of the study was to evaluate its effect on paving grade bitumen. When reclaimed asphalt (with aged bitumen) is added to the new asphalt mixture, stone aggregate and a calculated amount of fresh (non-aged) bitumen are also added at the same time. Consequently, researches on non-aged bitumen and on laboratory aged bitumen were conducted. The short term ageing, to which bitumen is subjected during mixing, transport and installation of asphalt, is in laboratory simulated by rolling thin film oven test (RTFOT) method according to EN 12607 [27]. According to this method, bitumen is aged under the influence of high temperatures and constant air flow. The ageing conditions are not exactly the same as in asphalt production, but the ageing results are comparable [28]. Ongoing ageing of bitumen during road use was simulated by the pressure ageing vessel (PAV) method according to EN 14769 [29]. PAV ageing at elevated pressure and temperature was performed on RTFOT aged bitumen. PAV simulates ageing according to climatic conditions (temperature, UV, etc.), but cannot take into account variables in the asphalt, such as the proportion of air voids, the type of aggregate and the absorbency of the aggregate.

Blends of non-aged bitumen and pyrolytic rejuvenator were laboratory prepared in different concentrations: 3%, 5%, 10% and 20% rejuvenator based on the mass of the reference bitumen (**Table 3**). All prepared samples and concentrations of the blends are presented in **Table 3**.

In addition to before mentioned standard test on samples, determination of the sample's consistency by needle penetration test according to the EN 1426 [30] at 25°C was also performed. The elastic recovery of the samples was determined according to the EN 13398 [31]. According to this standard, a specimen was first elongated to 20 cm and then cut in the middle to obtain two halves of the thread. After the predetermined time (30 minutes) for recovery has elapsed, the shortening of the half threads was measured and expressed as the percentage of the elongation length.

To check the possible phase separation in the blend of bitumen and pyrolytic products, a storage stability test according to EN 13399 [32] was performed only for the blend of pyrolytic product No. 14 and reference bitumen. In the test, the sample of the blend is maintained in the vertical vessel at 180°C for three days. After the sample is cooled down, it is cut into three equal parts. The two ends (top and bottom) are further analysed to evaluate possible differences in characteristics. The affinity between pyrolytic rejuvenator and stone aggregate (limestone) was checked by standard rolling bottle method according to EN 12697–11 [33] and compared with the affinity of reference bitumen (non-aged) and the blend.

The properties of samples in the low-temperature range were characterized with the bending beam rheometer (BBR) according to EN 14771 [34]. Bending tests are suitable for testing brittle materials when measurements at tensile load do not provide insight into the properties of the material or are not feasible. The stress relaxation in bitumen is significantly slower at low temperatures, which can lead to the formation of cracks in the asphalt and loss of binder functionality. During the BBR test a bitumen beam is bent under a constant load and deformation of bitumen is measured. The flexural creep stiffness $S_m(t)$ is calculated at time t = 60 s. The characteristic parameter is also value m_{60}, the slope of the curve S (t) at t = 60 s, which indicates the relaxation capacity of the bitumen stress. We presumed that the adequate quality of bitumen at low temperatures is ensured by the maximum value of S_{60} = 300 MPa and the minimum value of m_{60} = 0.300.

The results of softening point, Fraass breaking point and penetration of reference bitumen (non-aged, 'B 50/70' and laboratory aged, 'B_PAV'), pyrolytic rejuvenator 'PR' and their blends are shown in **Table 3**. Comparison of penetration and Fraass breaking point between 'B 50/70' and pyrolytic rejuvenator 'PR' indicates on their different chemical composition. Pyrolytic rejuvenator shows a significantly lower value of softening point and Fraass breaking point and at the same time a much higher value of penetration.

	Label	Proportion of the reference bitumen	Proportion of the pyrolytic rejuvenator	RB	Penetration	Fraass breaking point
		[%](m/m)	[%](m/m)	[°C]	[1/10 mm]	[°C]
Non- aged bitumen	B 50/70	100	0	50.5	53	−11
	B50/70_3%	97	3	50.2	55	−11
	B50/70_5%	95	5	50.3	59	−20
	B50/70_10%	90	10	49.0	60	−16
	B50/70_20%	80	20	47.3	75	−16
	B50/70_50%	50	50	42.6	115	−19
Pyro-lytic rejuve-nator	PR	0	100	37.5	233	−22
Aged bitumen	B_PAV	100	0	70.4	21	−10
	B_PAV_3%	97	3	69.0	22	−6
	B_PAV_5%	95	5	67.7	22	−6
	B_PAV_10%	90	10	65.6	26	−7
	B_PAV_20%	80	20	61.6	34	−7

Table 3.
Tested blends of pyrolytic rejuvenator (labels, RB and penetration) [21].

Results on non-aged samples show the effect of the rejuvenator on the standard mechanical properties (penetration, softening point, Fraass breaking point) of bitumen. Penetration values increased, at the same time softening values decreased with increasing rejuvenator proportion. All added amounts of rejuvenator to non-aged bitumen decreased the values of the Fraass breaking point.

Original B 50/070 was RTFOT+PAV aged, simulating naturally aged bitumen in reclaimed asphalt. Then the same proportions of pyrolytic rejuvenator No. 14 (as in the case of non-aged bitumen) were added and the blends were tested.

A comparison of non-aged and aged bitumen blends shows the impact of laboratory ageing. Aged B 50/70 (B_PAV) and all bitumen blends became stiffer since the penetration of aged bitumen decreased and the softening point increased. Unexpectedly, all proportions of rejuvenator which were added to RTFOT+PAV aged B 50/70 decreased the values of the Fraass breaking point. It should be noticed that the repeatability of the Fraass breaking point test is 3°C.

Table 4 presents results of tensile properties of bitumen, pyrolytic rejuvenator and their blends. Non-aged bitumen and its blends with rejuvenator elongated to the maximum length (1500 mm). The elongation of aged bitumen was prolonged with the addition of a rejuvenator. When mixing materials such as bitumen, two consequences can be observed: the mixing effect (mostly linear change) and the structural-interaction effect (mostly nonlinear change) [21]. Maximum force decreased proportionally with the added rejuvenator indicating a linear change occurred. The effect of nonlinearity is not observed, as the elongation at maximum force, Fmax, is the same for all samples with non-aged bitumen, as well as for all samples with aged bitumen, regardless of the amount of rejuvenator added. The elongation of the non-aged bitumen was about 1.5 times greater than the elongation of the aged bitumen with rejuvenator, indicating that aged bitumen was not completely restored. The results show that due to the added rejuvenator, the mechanical properties of aged bitumen approached the values of non-aged bitumen, but a complete restoration was not achieved.

	Force Ductility at 25°C				Elastic recovery
	Elongation	Force Fmax	Elongation at Fmax	Energy	
Label	[mm]	[N]	[mm]	[J/cm^2]	[%]
B 50/70	1500*	0.97	17.18	0.07	/
B50/70_3%	1500*	1.58	7.22	0.11	13
B50/70_5%	1500*	1.27	7.21	0.10	13
B50/70_10%	1500*	0.99	7.21	0.07	12
B50/70_20%	1500*	0.71	7.11	0.05	12
PR	230	0.04	17.12	0.00	−38
B_PAV	184	21.66	12.01	1.34	30
B_PAV_3%	304	20.03	12.10	1.37	27
B_PAV_5%	469	17.08	12.40	1.28	21
B_PAV_10%	307	13.38	12.71	0.95	21
B_PAV_20%	370	8.27	12.89	0.62	22

** Maximum elongation.*

Table 4.
Tested blends of pyrolytic rejuvenator (ductility, elastic recovery) [21].

Temperature	Mixing temperature (η = 0.17 Pas)	Compaction temperature (η = 0.26 Pas)	BBR T at S_{60} = 300 MPa	BBR T at m_{60} = 0.300	Higher T (BBR)
Label	[°C]	[°C]	[°C]	[°C]	[°C]
B 50/70	141	122	−19.3	−20.8	−19.3
B50/70_3%	140	120	−19.6	−21.5	−19.6
B50/70_5%	140	120	−19.8	−22.0	−19.8
B50/70_10%	138	118	−20.7	−22.7	−20.7
B50/70_20%	139	118	−22.6	−24.5	−22.6
PR	150	122	−31.3	/	/
B_PAV	155	139	−16.1	−11.7	−11.7
B_PAV_3%	154	137	−16.9	−12.9	−12.9
B_PAV_5%	153	137	−17.5	−12.8	−12.8
B_PAV_10%	151	134	−18.3	−13.3	−13.3
B_PAV_20%	150	132	−19.6	−18.8	−18.8

Table 5.
Results of rheological tests [21].

The elastic recovery did not change significantly for non-aged bitumen regardless of the amount of rejuvenator. In the case of aged bitumen, the elastic recovery decreases with the amount of rejuvenator and thus approached the value of the reference aged bitumen. The elastic recovery of the rejuvenator had a negative value, so the sample was stretching after the test, meaning that the rejuvenator had no elastic properties and all energy was lost. We expected that due to rubber content in car tires some elasticity will remain in our product, but it is evident that all rubber from tires decomposed during the pyrolytic process.

Mixing and compaction temperatures (**Table 5**) of aged bitumen were higher than temperatures of non-aged bitumen, and in both cases, the temperatures decreased with the increasing amount of added rejuvenator.

S_{60} and m_{60} are criteria that determine the lower limit of the bitumen application temperature. When the conditions: $S_{60} \leq 300$ MPa and $m_{60} \geq 0.300$ are met at the same time, the bitumen shows sufficient low temperature resistance. **Table 5** shows the results, i.e. the minimum temperatures for all samples. For non-aged bitumen, the critical temperature was determined by S_{60}, while for aged bitumen the temperature at parameter m_{60} was decisive. With the addition of the rejuvenator, the critical temperature was lowered. Aged bitumen with the highest amount of rejuvenator additive achieved similarly low temperatures as the reference non-aged bitumen.

Storage stability was checked only on a blend with the highest amount of pyrolytic rejuvenator, B50/70_50%. The test results showed that although an immense quantity of the rejuvenator was added to the reference bitumen, the blend remained homogeneous. This was evident from the very small changes in the penetration and softening values of the binder in the upper and lower parts of the tube (**Table 6**). Results also show that due to storage at high temperature the characteristics (pen, RB) did not change much.

The result of the affinity test (**Table 7**) shows that after the first 6 hours there was no difference between the tested samples. The binder detached only slightly from the aggregate (limestone) in all samples. After one day of testing in the rolling

Label	Conditions	RB [°C]	Penetration [1/10 mm]
B50/70_50%	Ambient	42.2	115
B50/70_50% upper part	72 h at 180°C	43.0	118
B50/70_50% lower part	72 h at 180°C	42.8	117

Table 6.
Results of storage stability test [21].

Affinity [%]	Time of test [h]				
Sample	**0**	**6**	**24**	**48**	**72**
B50/70	100	95	80	55	40
B50/70_50%	100	95	80	50	30
PR	100	95	90	55	45

Table 7.
Results of affinity test.

bottle, differences between the samples appeared more obvious. The pyrolytic rejuvenator had the best affinity with aggregate. After two days, the reference bitumen and rejuvenator covered the aggregate equally well, and after three days the aggregate was best covered with pyrolytic rejuvenator and worst with a mixture of reference bitumen and pyrolytic rejuvenator.

To verify the relationship between rheological and mechanical measured properties, presented in **Tables 3–5**, linear relationships between individual properties were examined. Linearity between properties was evaluated with statistical parameter R^2 (**Table 8**). The results show that there is no linear relationship between BBR measurements and empirical mechanical tests. Correlation between the parameters S_{60} and m_{60} and the Fraass breaking point was expected since all measurements were performed in the low temperature range. Similarly, we expected a relationship between the properties measured in the medium and high temperature range. The softening point temperature was compared with the mixing and compaction temperatures. It turned out that there is no linear correlation for the samples of non-aged bitumen, and there is a good linear dependence for the samples of aged bitumen. There is also a good relationship between the results of the two most basic mechanical tests, penetration and softening point, for both non-aged and aged paving grade bitumen samples.

	RB – penetration (Table 3)	RB – mixing temperature (Tables 3 and 5)	RB – compaction temperature (Tables 3 and 5)	BBR S60 - Fraass breaking point (Tables 3 and 5)	BBR m60 - Fraass breaking point (Tables 3 and 5)
Non-aged bitumen	0.91	0.00	0.02	0.10	0.20
Aged bitumen	0.94	0.91	0.96	0.06	0.00

Table 8.
The R^2 values for various properties of non-aged and aged bitumen with rejuvenator.

4. Asphalt mixtures with reclaimed asphalt and pyrolytic rejuvenator

Asphalt mixtures have to withstand dynamic loads as well as high and low temperatures without cracking or rutting. Several standardized laboratory tests enable the evaluation of these asphalt characteristics. Before implementation, characteristics of reclaimed asphalt (RA) had to be established. Several samples of reclaimed asphalt from the same stockpile were sieved into sub fractions and extracted bitumen was investigated. The results showed that the bitumen content in RA was 4.5%. Based on this, the required amount of fresh bitumen and rejuvenator for each asphalt mixture type AC 8 surf were calculated.

The bitumen content of all prepared asphalt mixtures was determined at 5% of the mass regarding the total asphalt mixture mass. Regarding to the established effects of pyrolytic rejuvenator on (non-aged and laboratory aged) bitumen, the addition of 20% of pyrolytic rejuvenator to the bitumen from reclaimed asphalt was established. Control asphalt mixture of fresh materials (0% RA) and asphalt mixtures with 20%, 40% and 60% of RA according to the weight of the stone aggregate were prepared in laboratory (set of asphalt mixture samples is described in **Table 9**). Each asphalt mixture with RA was produced without and with a pyrolytic rejuvenator. All together seven asphalt mixtures were prepared with the same B50/70 bitumen (from the same producer) as used in previous research.

All asphalt mixtures samples were mixed in a laboratory and compacted in accordance with EN 12697–30 [35]. Basic information about asphalt mixtures was gained by determining bulk density, EN 12697–6 [36], maximal density, EN 12697–5 [37], void content, EN 12697–8 [38], and indirect tensile strength (ITS), EN 12697–23 [39]. ITS is maximum tensile stress applied to a cylindrical specimen loaded diametrically until the break. Cylindrical specimens (nominal diameter 100 mm) were for ITS compacted by 50 impacts on each side at temperature 150°C. Water sensitivity tests were completed according to EN 12697–12 [40], in order to evaluate the effect of moister. Water sensitivity is expressed by ITSR – indirect tensile strength ratio. Two sets of four cylindrical specimens were prepared, compacted by 35 impacts on each side at temperature 150°C. One set of specimens was conditioned in water for three days; the other set was kept dry. The ratio between their ITS values expresses water sensitivity.

For determination of characteristics at low temperature thermal stress restrained specimen test (TSRST, EN 12697–46 [41]) was carried out. Asphalt

Sample	Proportion of the RA	Addition of the pyrolytic rejuvenator	Measured bitumen content	The bulk density	The maximal density	Void content
	[%] (m/m)	/	[%] (m/m)	[kg/m^3]	[kg/m^3]	[%] (V/V)
0% RA	0	No	5.0	2398	2478	3.2
20% RA	20	No	5.0	2403	2490	3.5
20% RA + rej.	20	Yes	5.0	2407	2492	3.4
40% RA	40	No	5.0	2407	2494	3.5
40% RA + rej.	40	Yes	5.0	2394	2492	3.9
60% RA	60	No	5.0	2425	2502	3.1
60% RA + rej.	60	Yes	5.0	2417	2501	3.4

Table 9.
Standard properties of asphalt mixtures [22].

mixtures were compacted in form of slabs, EN 12697–33 [42], from which prismatic specimens were cut. In TSRST, the specimen, whose length is held constant during the test, is subjected to a temperature decrease with a constant temperature rate. Due to the confined thermal shrinkage, cryogenic stress builds up in the specimen. The results of the tests are the progression of the cryogenic stress over the temperature range until break, σcry(T), and the failure stress, σcry,failure, at the failure temperature, Tfailure.

To check the behaviour of asphalt mixtures at elevated temperatures the formation of wheel tracking was checked according to EN 12697–22 [43].

Extracted bitumen from all asphalt mixtures was investigated according to before mentioned standard testing methods.

The siewing curve had to be adjusted for each asphalt mixture (**Figure 1**) because different shares of RA were added. In all figures addition of rejuvenator is designated with RA+rej. The curves overlap with each other well and bitumen from RA does not affect the siewing curve. The inhomogeneity of RA was successfully solved by presiewing RA into sub fractions (0/2 mm, 2/4 mm and 4/8 mm).

The densities were determined according to standard procedures and the results are given in **Table 9**. The difference between the densities and void content of individual asphalt mixtures was very small. Results confirm that mixtures were comparable.

The presence of water in the asphalt is expected. Moisture is one of the most important factors influencing the durability of asphalt. The combination of excess moisture and traffic load shortens the life of the asphalt. Moisture in asphalt causes two main destructive mechanisms: deterioration of adhesion and cohesion. Strong deterioration of adhesion can be observed as peeling of bitumen from the stone aggregate, and deterioration of cohesion is observed as softening of the binder, which leads to lower strength of asphalt. The water sensitivity of asphalt mixtures is expressed by the ITSR quotient, which represents the ratio of indirect tensile strength of wet and dry specimens, expressed as a percentage. **Figure 2** shows the results of the water sensitivity test. For all tested asphalt mixtures indirect tensile strength increased with the addition of RA and decreased only slightly when rejuvenator was added. Asphalt mixtures were less sensitive to water after the addition of RA, as the ITSR ratio increased. No significant effect on ITSR was observed with the addition of a rejuvenator.

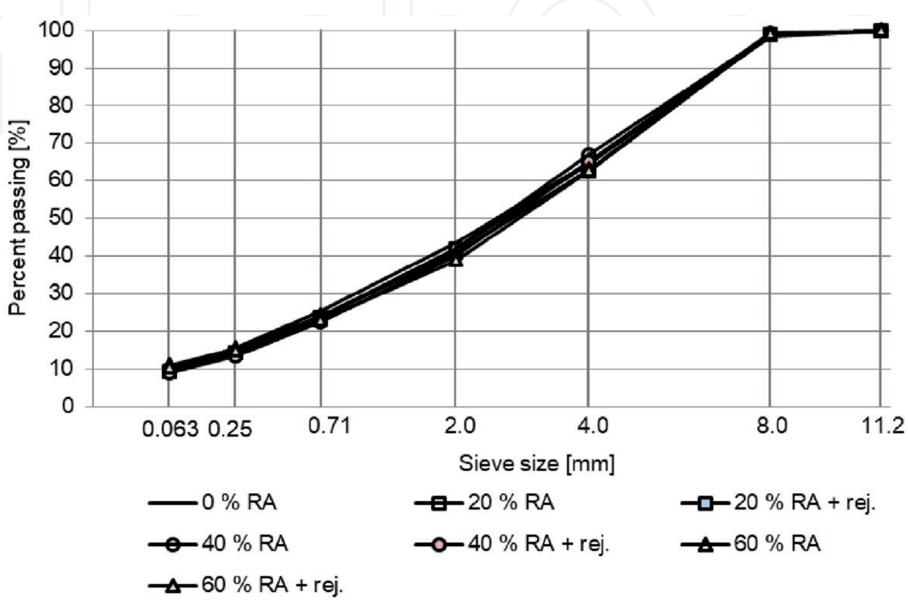

Figure 1.
The sieving curves of laboratory produced asphalt mixtures AC 8 surf [22].

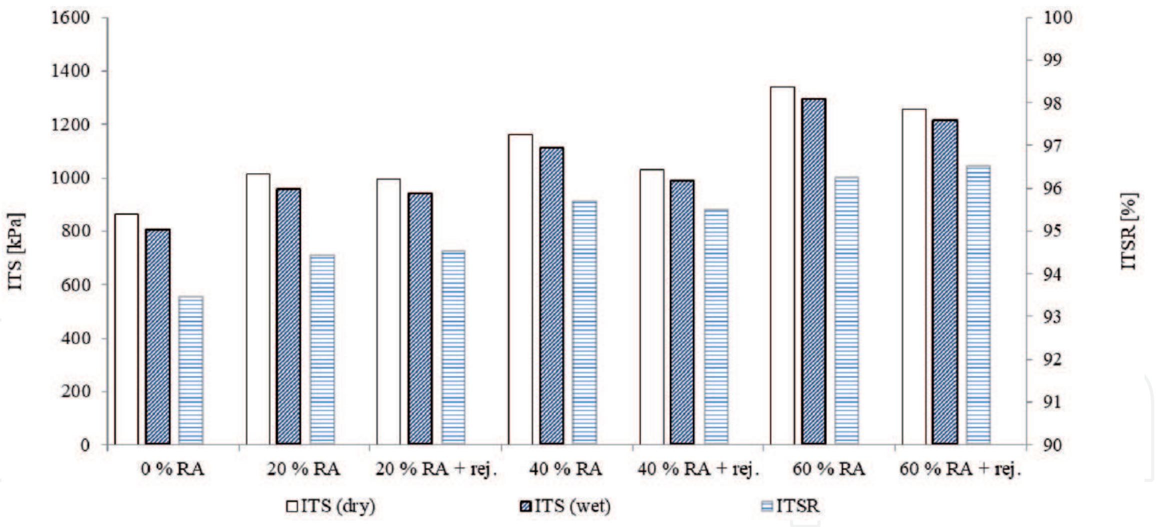

Figure 2.
The water sensitivity of the asphalt mixtures [1].

Testing of wheel track formation was performed at an elevated temperature of T = 50°C. The differences between the sample results (**Figure 3**) were not significant, indicating that the selected test temperature was too low.

Because the aged bitumen from RA increases the stiffness of the binder in the asphalt mixture, resistance to fatigue cracking is weakened in asphalt mixtures with a high amount of RA. In our study, the resistance to low temperature cracking was checked only on mixtures with the highest proportion of RA, as 60% of RA represents the worst conditions for asphalt resistance at low temperatures. The average results (three specimens were tested for each mixture) for three mixes are shown in **Figure 4**. Compared to the mixture with 0%RA, the failure temperature increased due to the added RA, which means deterioration of the mechanical properties of the asphalt mixture. The results for 60% RA mixture with the addition of pyrolytic rejuvenator show that the failure temperature decreased, and the resistance of the asphalt mixture with RA and the rejuvenator was therefore slightly better than the resistance of the basic mixture. The breaking stresses were similar for the three

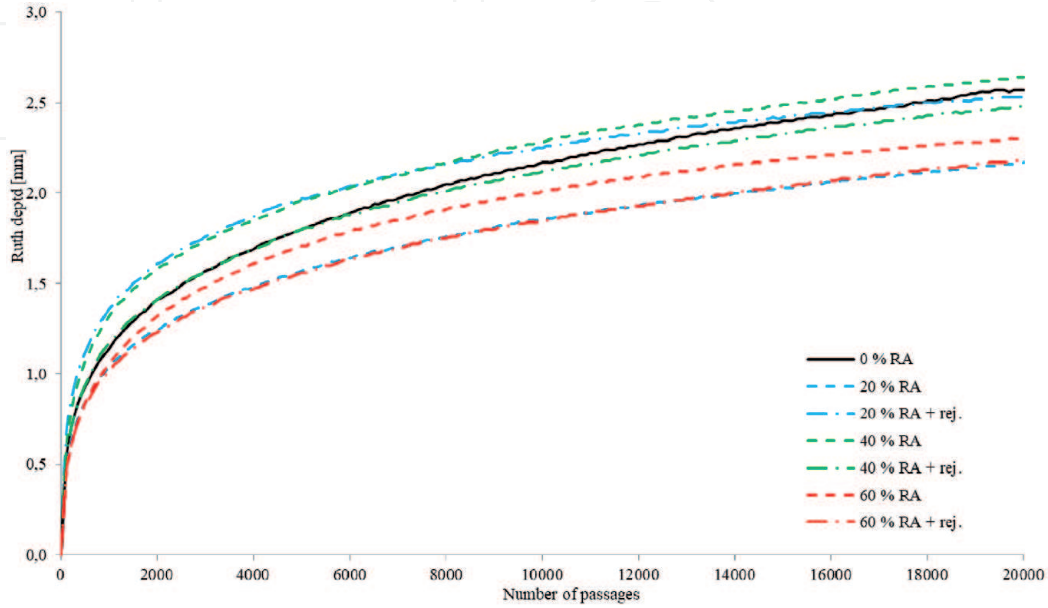

Figure 3.
Results of the wheel tracking test at T = 50°C.

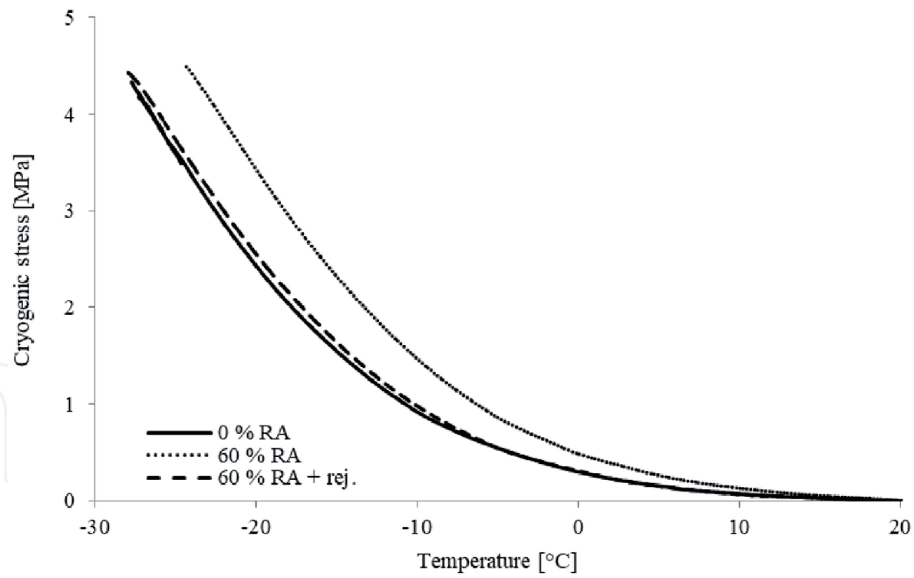

Figure 4.
Temperature dependency of cryogenic stress [1].

	RB	Fraass breaking point	Penetration
Label	**[°C]**	**[°C]**	**[1/10 mm]**
0% RA	58.6	−11	45
20% RA	59.4	−10	37
20% RA + rej.	58.2	−11	39
40% RA	60.8	−7	32
40% RA + rej.	58.3	−8	36
60% RA	63.3	−6	26
60% RA + rej.	60.7	−8	32
100% RA	65.3	−2	21

Table 10.
Results of standard mechanical tests of extracted bitumen [22].

tested mixtures, but the course of cryogenic stresses shows that the stresses were higher in the mixture with 60% RA than in the other two mixtures.

The extracted bitumen of asphalt mixtures was also investigated; the results are presented in **Table 10**. The highest value of the softening and Fraass breaking point was determined in bitumen from reclaimed asphalt (100% RA), meaning the bitumen was the most brittle and hard, as expected. The results showed that the addition of RA to asphalt mixtures without pyrolytic rejuvenator increased the softening point and Fraass breaking point and decreased penetration. However, the rejuvenator had a beneficial effect, as mixtures with the rejuvenator exhibited a lower softening point, Fraass breaking point and higher penetration than mixtures without pyrolytic rejuvenator. As expected, the bitumen became harder as the RA content increased, but the bitumen softened with the addition of a rejuvenator.

5. Conclusions

In the presented research, several pyrolytic products were tested. Based on the results, the pyrolytic product 14 was selected as the most suitable for the purpose of rejuvenating. It had a homogeneous structure, was solid at room temperature and it flowed at elevated temperatures. The indicator of suitability for use as a rejuvenator was the pyrolytic product's low softening and Fraass breaking point and high penetration value.

In mixtures with reference bitumen, the low Fraass breakpoint value was not maintained. The penetration of bitumen was increased by adding a pyrolytic rejuvenator and the softening point was decreased. The results of standard mechanical tests confirmed that the pyrolytic product softened the bitumen, which is the basic purpose of the rejuvenator.

The next part of the research was dedicated to the determination of the optimal proportion of rejuvenator that would regain the original properties of bitumen from reclaimed asphalt. This part of the research was performed on non-aged and laboratory aged bitumen. The addition of rejuvenator was limited at the upper limit, as the prepared pyrolytic product contained polycyclic aromatic hydrocarbons (PAHs). The results of decreased softening point and increased penetration show that the bitumen softened due to the rejuvenator. The Fraass breaking point increased in aged bitumen after the addition of a rejuvenator, indicating deterioration in the properties of the bitumen.

Despite the added rejuvenator the elongation in the ductility test was maintained for non-aged bitumen and increased for aged bitumen. However, in proportion to the increase in the rejuvenator, the maximum force in the sample decreased. The elastic recovery of non-aged bitumen did not change due to the rejuvenator, while in aged bitumen it decreased after the addition of the rejuvenator. The results of BBR confirmed our expectations, as resistance at low temperatures was increased by the addition of a rejuvenator. The addition of 20% of the pyrolytic product changed most properties of the aged bitumen in order to approach the characteristics of the non-aged bitumen. Although a complete recovery of the aged bitumen was not reached, the pyrolytic product can be successfully used as a rejuvenator. After the addition of pyrolytic product, the properties of aged bitumen were restored in the direction of the properties of the non-aged bitumen.

The asphalt mixtures were less water sensitive when RA and rejuvenator were added. Results proved RA improved the adhesion and cohesion of bitumen. Rejuvenator did not deteriorate the cohesion significantly, even though it made bitumen softer.

The results of the TSRST test revealed that RA deteriorates the properties of asphalt mixtures. However, low temperature cracking resistance improved with the addition of a rejuvenator. The improvement was made to such an extent that the asphalt mixture with the highest proportion of RA and rejuvenator had slightly better resistance than the control asphalt mixture.

On the basis of all the results, it can be concluded that pyrolytic rejuvenator enables the increase of RA share in asphalt mixture.

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