BITUMINOUS BINDERS EXTENDED WITH A RENEWABLE PLANT-BASED OIL: TOWARDS A CARBON NEUTRAL BITUMEN

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ABSTRACT

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Carbon footprint reduction is important for industry in general. Use of renewable/non-fossil materials is one option to achieve this for the asphalt industry. In this paper, a plant-based oil (PBO) from the forest and paper industry was systematically studied as a potential renewable bitumen extender. Comprehensive laboratory investigations were carried out on a selected PBO and on different bituminous binders prepared with this PBO. It was found that the PBO used was fully miscible with bitumen, and by a careful selection of the base bitumen type and the PBO dosage, desired standard paving grade specifications can be achieved. In terms of performance, improvements were found for the PBO-extended binders, including better long-term durability, and higher adhesion with mineral aggregates as evaluated by the rolling bottle test and higher indirect tensile strength ratio (ITSR). With regards to other asphalt performance properties (e.g. stiffness, fatigue and permanent deformation), no significant differences were observed between PBO-extended binders and reference bitumen. Good results were also obtained from full-scale field trials, both from pavement performance perspectives and Health, Safety & Environment (HSE) observations. A cradle-to-gate carbon footprint consideration of the PBO-extended binders (including polymer modified binders) shows that, depending on the proportion of PBO, the extended binders can even become carbon neutral.

INTRODUCTION

Since many years back there has been a lot of focus on reduction of carbon footprint and sustainable development. For road construction and asphalt industry, great efforts are already made to enhance pavement durability (Lu et al., 2011; Monismith, 2004; Nunn and Ferne, 2001; von Quintus et al., 2007), to reduce energy consumption (Sol-Sanchez et al., 2016; Wayman et al., 2014), to increase the use of recycled materials Celauro et al., 2010; Zaumanisa and Mallicka, 2015), to care for health, safety and environment (HSE), and to explore renewable and alternative materials (Fini et al., 2012; Ingrassia et al., 2019; Peralta et al., 2014; Xie et al., 2017). Undoubtedly, the long-term durability of asphalt pavements is a key aspect to sustainability, as longer lifetime means less maintenance, less use of materials, less energy, and certainly, less impacts on the environment. Also, an asphalt pavement is 100% recyclable, requiring that future recyclability must be ensured when new materials or wastes are added into bitumen and/or asphalt. With respect to energy consumption and HSE, different warm mix asphalt technologies have been developed to reduce temperature, for example by using chemical additives, organic additives, and foaming processes. To further reduce the carbon footprint of asphalt pavements, the use of renewable materials is another option for the asphalt industry.

In general, a biogenic material derived from plants which are sustainably managed and not subjected to depletion can be a potential renewable component for bitumen. There are many publications on studying different applications of plant-based oils (Ball et al., 1993; Cooper III et al., 2013; Fini et al., 2012; Kluttz, 2012; Oda et al., 2012; Seidel and Haddock, 2012; Yang et al., 2014), for example, as bitumen modifier, extender and emulsifier, or as a warm mix additive and rejuvenator.

Regarding a plant-based oil from the forest and paper industry, a comprehensive laboratory and field test program was already carried out in Finland in the late 1980s (Peltonen, 1989a, 1989b). It was shown that tall oil pitch extended bitumen displayed varied behaviors as compared with reference bitumen, including lower penetration index, higher stiffness at low temperatures, better adhesion, and similar aging sensitivity and deformation resistance. But increased fuming and smell were reported, and it was also claimed that quality of reclaimed asphalt pavement (RAP) was not good when test pavements with the extended bitumen were recycled. Another early experiment on tall oil pitch as bitumen extender was conducted in New Zealand in the 1990s (Ball, et al., 1993). At a proportion of up to about 32% (by weight), the extended binders did not separate with time, and were satisfactory in terms of short-term aging. However, long-term aging effect was not investigated in this study. More investigations on aging effect were also reported by Bearsley and Haverkamp (2007a). In their study, up to 25% tall oil pitch was blended with various grades of bitumen, and no positive effect was found. The same researchers also investigated the adhesive properties of tall oil pitch modified bitumen (Bearsley and Haverkamp, 2007b). It was shown that asphalt mixes made with the modified binders display higher resistance to moisture damage as assessed by tensile strength ratios. The improvement in adhesion was supported by an interfacial tension test on the binders, but not by Vialit test. Ahmedzade et al. (2007) observed that tall oil pitch, alone or together with SBS polymer, improved binder properties (e.g. temperature susceptibility and Fraass breaking point). Improvements were also shown for asphalt mixes in various tests such as Marshall stability and flow, compression strength, indirect tensile fatigue, and permanent deformation. In a recent technical report published by Austroads (Midgley and Urquhart, 2012), several alternatives and extenders to bitumen have been investigated and discussed, including Shale oil derived materials, alternative binder derived from raw materials of vegetable origin, and tall oil pitch. It was concluded that the binders extended with plant-based materials were very promising in terms of their future use as a direct replacement for conventional bitumen. However, according to the authors, these materials would be expected to have somewhat shorter service lives if used in sprayed seal applications.

In spite of such great research efforts, renewable materials generally have not been widely applied in bituminous binders and asphalts, especially when it comes to bitumen extender or as partial replacement of bitumen. Possible reasons could be that (a) the long-term performance of plant-based materials has not been demonstrated, (b) knowledge on their HSE aspects may be insufficient, (c) recyclability of such materials are unknown and needs to be investigated, and (d) there is a concern about availability of the materials, as well as their cost-effectiveness. In this paper, a plant-based oil (PBO) from the forest and paper industry has been systematically studied as a potential renewable bitumen extender. Laboratory analysis was conducted on PBO samples from different supply sources. Various bituminous binders were prepared with the PBO and studied extensively in terms of quality and performance. Full scale field trials were carried out and pavement performance has been followed up. A cradle-to-gate carbon footprint consideration of the PBO-extended binders (including polymer modified binders) was also done.

LABORATORY INVESTIGATIONS

Plant-based oils

Plant-based oil (PBO) samples from different manufactures were analysed by various physical property tests and chemical analyses. Those included viscosity, flash point, density, acid number, elemental composition, simulated distillation (SIMDIS), Fourier Transform Infrared Spectroscopy (FTIR), as well as generic fractions by thin-layer chromatography with flame ionisation detection or Iatroscan (IP 469). Typical properties and elemental composition of the PBO samples are shown in Table 1.

As indicated in Table 1, the PBO samples differ in viscosity. It is also well-known that the oil from the forest and paper industry is a complex mixture of numerous chemicals and consists of rosin acids, fatty acids and neutral compounds. This is reflected by a high acid number found for the material. As for the elemental composition, no significant differences are found between the PBOs. Compared to bitumen which typically consists of about 80 to 85% carbon, 10% hydrogen, 0.5% nitrogen, 0.5% oxygen, and 3 - 5% sulfur, the PBOs contain a similar level of carbon and slightly higher hydrogen, resulting in higher H/C ratios than that for bitumen (about 1.2). More importantly, these PBOs show much higher oxygen content, mostly in the form of acids and esters, as already suggested by the high acid numbers, as well as FTIR analysis on the functional groups.

Table 1. Properties and elemental composition of the PBO investigated

Parameter	PBO-1	PBO-2	PBO-3
Viscosity (EN 12595) at 60°C, mm^2/s	674	2235	712
Flash point (ASTM D 6450), °C,	247	247	> 220
Density (ASTM D 1475), g/cm^3	0.984 (at 50°C)	1.005 (at 15°C)	0.995 (at 50°C)
Acid number, mg KOH/g	37.7	89.1	--
Carbon, %	80.7	80.1	80.5
Hydrogen, %	11.2	11.3	11.3
Nitrogen, %	< 0.5	< 0.5	< 0.5
Oxygen, %	7.6	7.8	7.6
Sulfur, %	0.38	0.34	0.31
H/C ratio	1.66	1.69	1.67

The FTIR spectra of the PBOs used are quite similar, and a typical example is shown in Figure 1 for PBO-3. Identified functional groups are: methylene chains with four or more carbons at about 716 cm^{-1}, C-O at about 1160 cm^{-1}, C-H in methylene and methyl groups at 1450 and 1370 cm^{-1}, C=C (aromaticity) at 1600 cm^{-1}, C=O in carboxylic acids at 1700 cm^{-1}, C=O in esters at 1730 cm^{-1}, and CH_2 and $-CH_3$ at 2920 and 2850 cm^{-1}. As expected, esters and carboxylic acids are rich in the tested PBOs.

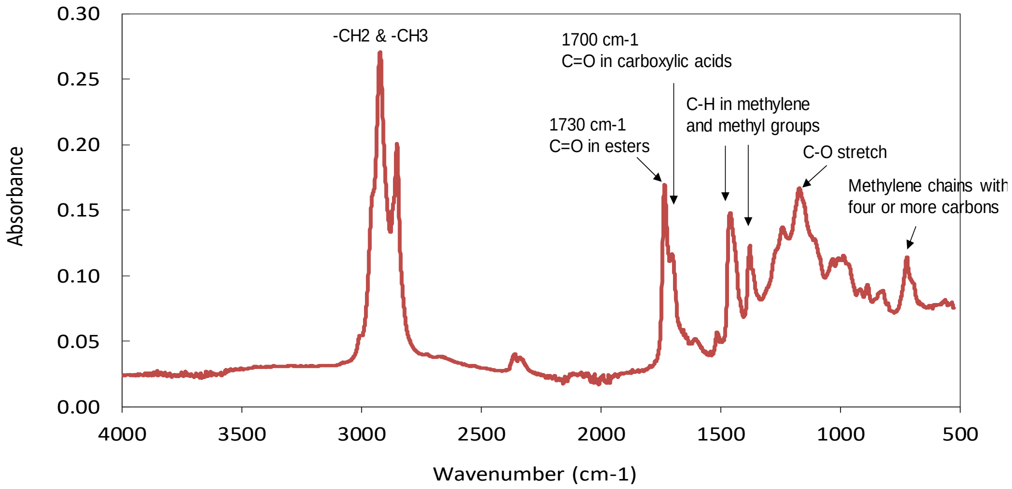


Figure 1. A typical FTIR spectrum of PBO

SIMDIS (ASTM D 6352) shows that, PBO-1, PBO-2 and PBO-3 have IBP (initial boiling point) of 384, 373 and 361°C, respectively, and at the boiling point of around 402°C (corresponds to normal paraffin C25), the recovered mass is 5 to 7 %. Considering their proportion (< 30%) in bitumen, this should suggest no negative impact from a fume emission point of view. Further analysis by Iatroscan (IP 469) shows that the PBOs mainly consist of aromatics and polars (I) or resins, while no saturates and very little polars (II) or asphaltenes are found, as illustrated for PBO-3 in Figure 2.

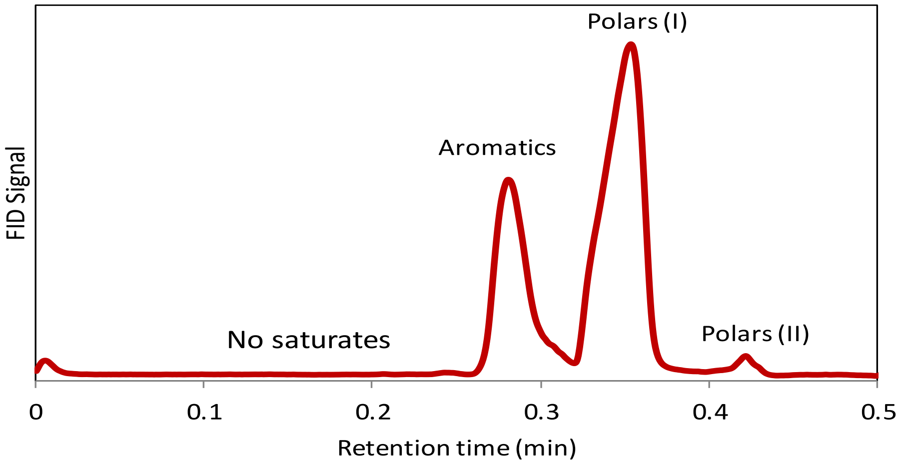


Figure 2. Iatroscan chromatogram of PBO

Bituminous binders extended with plant-based oils

Different base bitumen and different dosages (5 - 30%) of PBO were selected for sample preparation. To determine blending ratios, small amounts (< 100 g) of samples were prepared by manually homogenizing a hot bitumen (150°C) and PBO (100°C) for about 1 minute. For full binder analysis and asphalt tests, bitumen and PBO blends were prepared at 150°C using a laboratory low shear mixer with a speed of 500 rpm and a mixing time of about 10 min. The base bitumen used were normal paving grade bitumen complying to EN 12591. Blending proportions were made such that appropriate 70/100 and 160/220 grades according to EN 12591 (CEN, European Committee for Standardization) were obtained. In Table 2, test results are shown for several PBO and bitumen blends, one targeted to 70/100 (coded EB 80), and other four targeted to 160/220 (coded EB 200, EB 190, EB 180, and EB 170). All these blends fulfil the CEN specification.

Table 2. Binder analysis according to EN 12591

Parameter	Extended 70/100	Extended 160/220			
	EB 80	EB 200	EB 190	EB 180	EB 170
Penetration 25°C, 1/10 mm	75	198	190	182	167
Softening point, °C	46.4	35.8	36.4	37.8	38.4
Dyn.viscosity 60°C, Pas	213.2	61.1	62.5	74.7	51.3
Kin.Viscosity 135°C, mm^2/s	377.9	226	242	236	199
Fraass breaking point, °C	-13	-22	-23	-18	-23
Flash point COC, °C	292		294	266	310
Solubility in toluene, %	99.95	99.5	99.96	99.95	99.95
After RTFOT 163°C					
Change of mass, %	-0.02	-0.08	-0.24	-0.15	-0.12
Penetration 25°C, 1/10 mm	53	125	113	118	79
Softening point, ℃	50.4	40.6	41.8	42.4	45.8
Incr. in softening point, ℃	4	4.8	5.4	4.6	7.4
Retained penetration, %	71	63	60	65	47

Using penetration, softening point and viscosity at 135°C, two measures for temperature susceptibility were calculated: Penetration Index (PI) and Penetration Viscosity Number (PVN). A lower PI and lower PVN indicates higher temperature susceptibility. As illustrated in Figure 3, the PBO extended binders display lower PI but higher PVN when compared to the corresponding reference bitumen (Cf. Section 2.3).

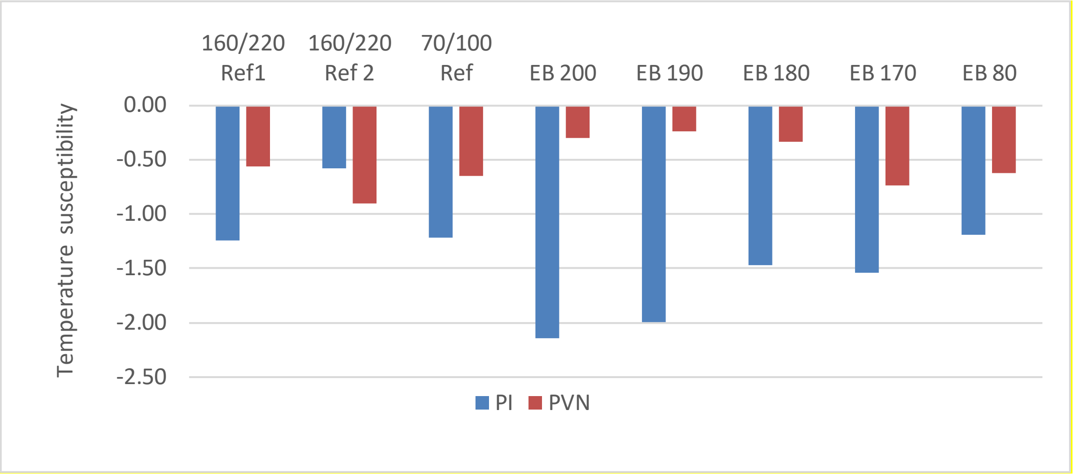


Figure 3. Temperature susceptibility as assessed by PI and PVN

The PBO used is also believed to be compatible with bitumen. To confirm this, two bitumen blends containing more than 10% PBO (by weight) were investigated by a storage stability test or phase separation test according to EN 13399. The tests were performed at 155°C using the same procedure as for polymer modified bitumen (PMB). No phase separation was observed during the hot storage of the blends.

The PBO extended binders are further investigated with regards to their rheological properties at different temperatures using a dynamic shear rheometer (DSR), as well as a beading beam rheometer (BBR). Examples of complex modulus versus temperature plots are shown in Figure 4. As can be seen, at high service temperatures (> 40°C), the unaged EB 200 is less temperature susceptible compared to the reference bitumen, which is opposite to the observations made by PI but in line with PVN observations. From the rheological measurements, effect of aging, especially long-term aging by PAV (pressure aging vessel), is evaluated. PAV test was conducted at the standardized conditions (i.e. 100°C and 20 h) and on samples after RTFOT. Complex moduli at 20°C and 10 rad/s were used to calculate long-term aging index, i.e. modulus ratio between PAV aged and RTFOT aged samples. Results are shown in Figure 5. In Figure 5, the PBO extended bitumen EB 200 (1) has a base bitumen of the same source as 160/220 Ref 1, while EB 200 (2) has a base bitumen of the same source as 160/220 Ref 2. Evidently, PBO has no detrimental effect on bitumen long-term aging when looking at stiffness moduli at 20°C. On contrary, it somehow improves the aging resistance of the bitumen.

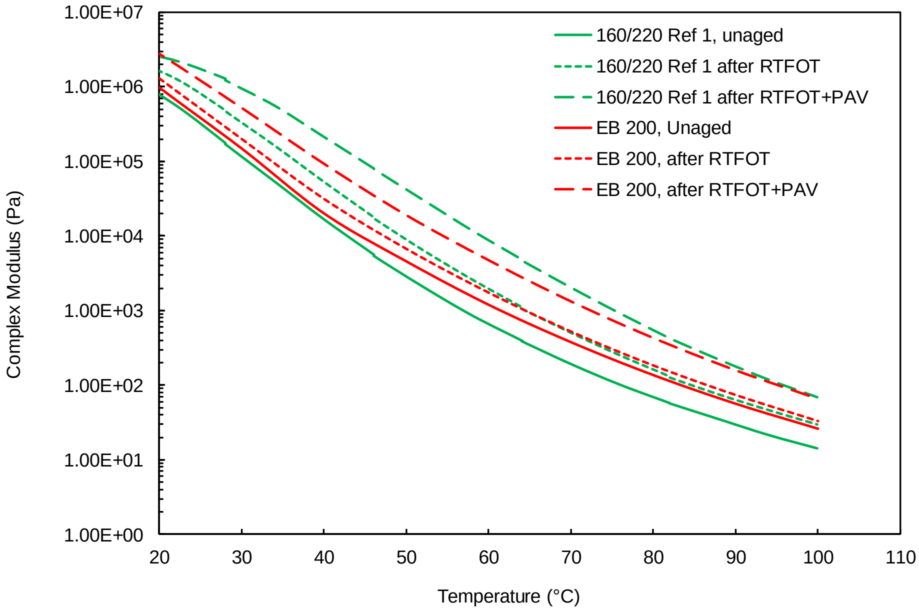


Figure 4. Complex modulus as function of temperature at 10 rad/s

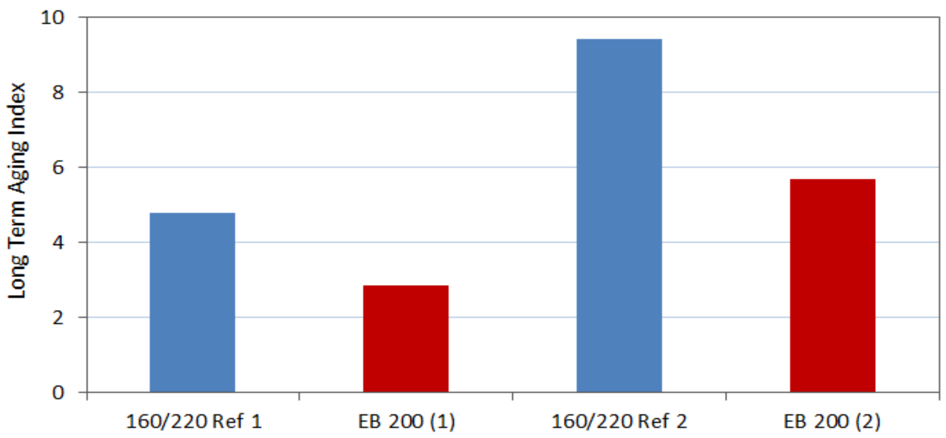


Figure 5. Effect of a plant-based oil on bitumen long-term aging

The long-term durability of the PBO extended bitumen is also shown by ΔTc, a binder durability parameter defined as LST (temperature at 300 MPa stiffness) minus LmT (temperature at 0.300 m-value) (Anderson et al., 2011), which in this study are determined from BBR. A higher (i.e. more positive) value of ΔTc means a higher durability. As an example, for EB 180 shown in Table 2, after RTFOT and PAV, ΔTc was found to be +1.9°C, which is significantly higher than many normal 160/220 penetration bitumen that generally have a negative ΔTc. This improvement will be further confirmed by analysing samples from a field trial in Section 3.

Evaluation of asphalt performance

In asphalt tests, five PBO extended binders and three reference bitumen were selected (see Table 3), covering 160/220 and 70/100 grades, also considering variations in bitumen origin, PBO source and dosage. The base bitumen used in EB 180, EB 190 and EB 200 have the same origin as the 160/220 Ref 1, whereas EB 170 has a base bitumen of a same origin as 160/220 Ref 2. Regarding 70/100 Ref and EB 80, they have the same origin as the 160/220 Ref 1.

Table 3. The binders used in asphalt tests

Binder	Penetration, 1/10 mm	Softening point, °C	Kin viscosity 135°C, mm^2/s	Fraass breaking point, °C
160/220 Ref 1	192	37.8	199	-21
160/220 Ref 2	167	40.6	181	-17
70/100 Ref	83	45.4	345	-17
EB 200	198	35.8	226	-22
EB 190	190	36.4	242	-23
EB 180	182	37.8	236	-18
EB 170	167	38.4	199	-23
EB 80	75	46.4	378	-13

Asphalts were prepared in accordance with a Swedish standard ABT11, which is a dense-graded asphalt with maximum aggregate of 11 mm, and of about 6.0% (by weight) binder content and 3.5% air voids. In a first phase evaluation, the aggregates used were crushed granite from Södertälje (Source 1) in Sweden. Performance evaluation includes adhesion, stiffness, resistance to fatigue cracking, and resistance to rutting. More asphalt evaluations were then performed using a reference aggregate (Source 2) from the Swedish Transport Research Institute (VTI). In both cases, no adhesion promoter was used in the asphalts.

Adhesion

The affinity between granite aggregate and bitumen was assessed using rolling bottle method according to EN 12697-11. In this study, the rolling of the bottles at room temperature was stopped after 6 and 24 hours, and two operators carried out independently a visual determination of the aggregate area covered by bitumen. Figure 6 shows results of the extended bitumen and corresponding reference bitumen. It is evident that the PBO studied significantly increases the coverage, indicating improved adhesion between the aggregates and the EB binders. The improvement was found to increase with concentration of the PBO.

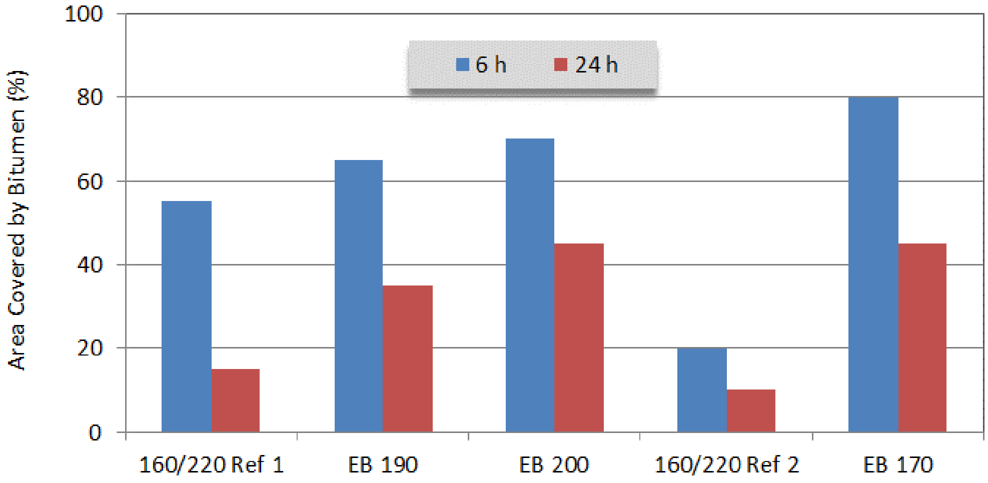


Figure 6. Results of rolling bottle test

Water sensitivity of asphalts was also evaluated by indirect tensile test (EN 12697-23) in accordance with Swedish standard TRVMB 704. For each asphalt mix, 10 specimens (diameter 100 mm, height 60 mm) were drilled from a laboratory compacted slab. A subset of five specimens was kept at room temperature (or dry conditions), and the other subset of five specimens was saturated (3 h) and then conditioned in a water bath at 40°C for seven days (wet conditions). The indirect tensile tests were performed at 10°C, and indirect tensile strength ratios (ITSR) were calculated. Figure 7a shows that all the asphalts display low values of ITSR, probably due to poor quality aggregates. However, the asphalts made with the extended bitumen show lower water sensitivity as compared to those with the corresponding reference bitumen. More ITSR tests were performed using a different aggregate from VTI (Source 2). As shown in Figure 7b, the EB binders behave quite similarly to the reference bitumen. At the same time, the overall levels of ITSR, as well as ITS values, are much better than those presented in Figure 6a, mainly reflecting aggregate effect.

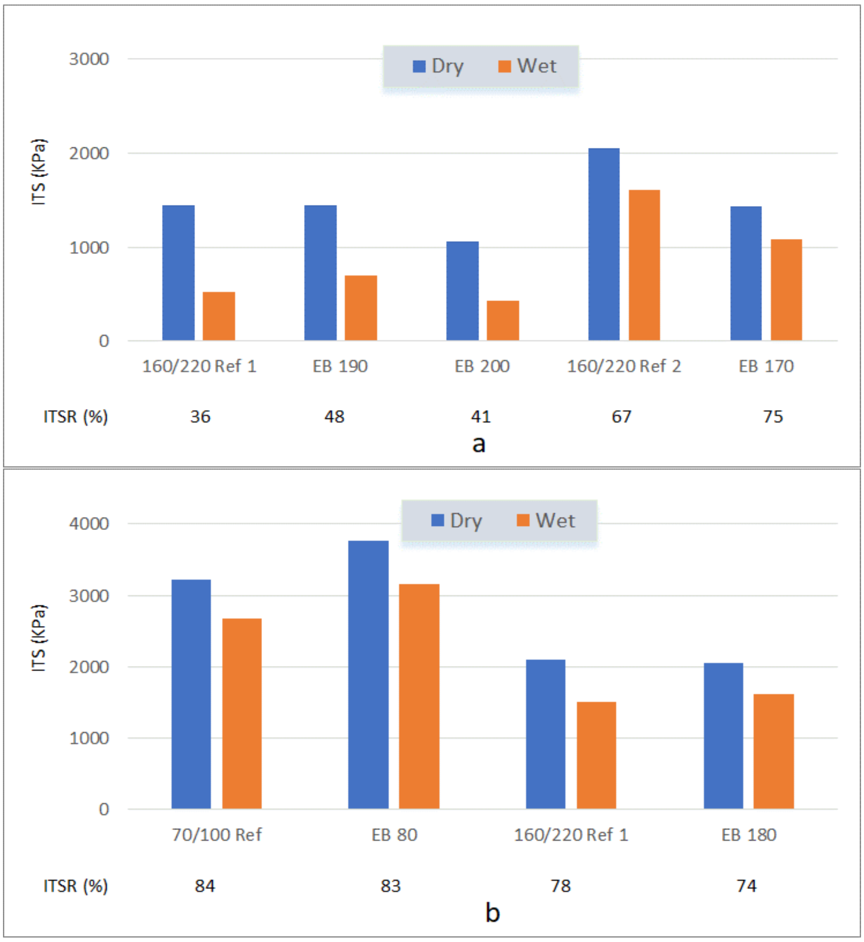
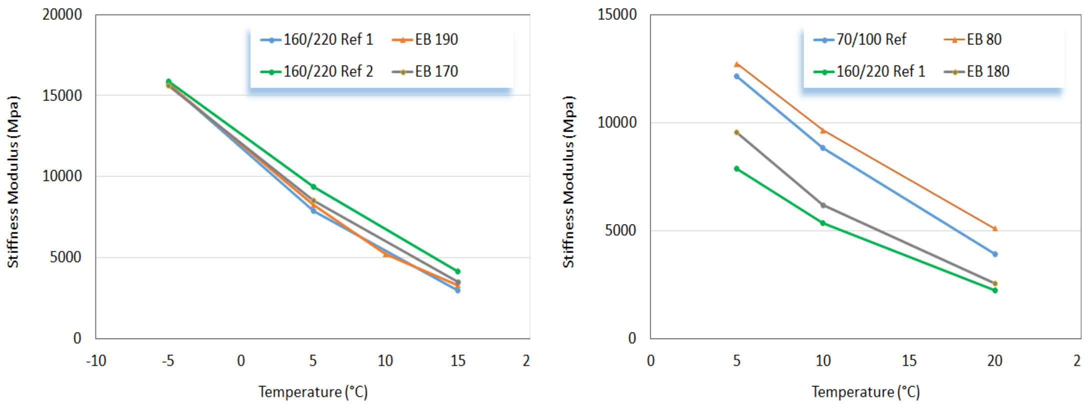


Figure 7. Asphalt water sensitivity assessed by ITSR
(a – aggregate from Source 1; b – aggregate from Source 2)

Stiffness

Stiffness measurements were performed at different temperatures using indirect tensile test (IDT) according to EN 12697-26 annex C. Specimens of 100 mm in diameter and 50 mm in height were prepared using a gyratory compactor. As illustrated in Figure 8, there are no significant differences in stiffness between the asphalts made with the extended bitumen and the corresponding reference bitumen. Moreover, these asphalts display similar temperature susceptibility, indicating that the tested PBO will most probably not affect asphalt low temperature cracking.



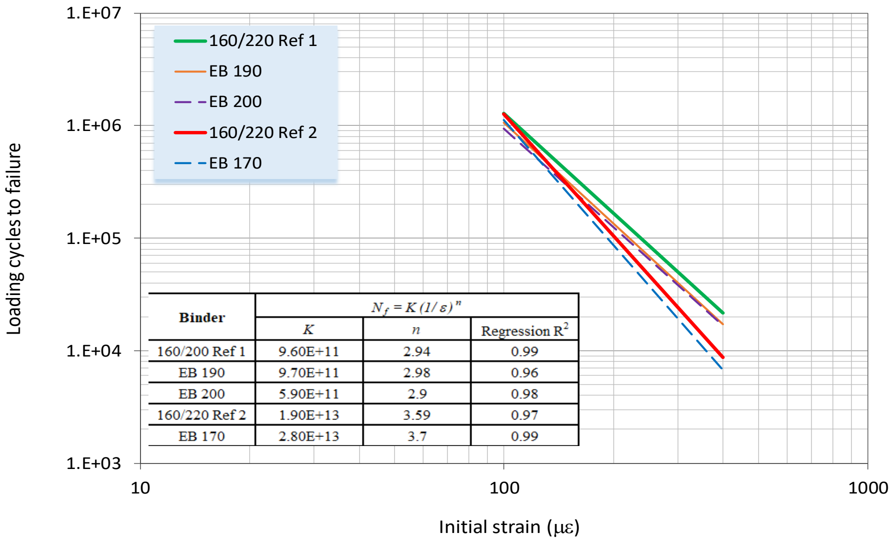
a

b

Figure 8. Asphalt stiffness as a function of temperature
(a – aggregate from Source 1; b – aggregate from Source 2)

Fatigue

Fatigue tests were carried out at 10°C on gyratory-compacted specimens (diameter 100 mm, height 50 mm) also using IDT as described in EN 12697-24 annex E. In the IDT fatigue test, a cylindrical specimen is exposed to a repeated haversine loading with a loading duration of 0.1 s followed by a 0.4 s rest period (a frequency of 2 Hz) through the vertical diametral plane. The resulting horizontal deformation of the specimen is measured and used to calculate tensile strain at the centre of the specimen. Fatigue life is defined as the total number of load repetitions when fracture of the specimen occurs. To establish the fatigue relationship at a test temperature, 12 specimens of each asphalt mix were tested under a range of loading levels. The obtained constants and fatigue lines are shown in Figure 9. As can be seen, the asphalts made with two reference bitumens behave differently; however, blending the PBO into the bitumen does not seem to have a significant effect on asphalt fatigue behaviour.



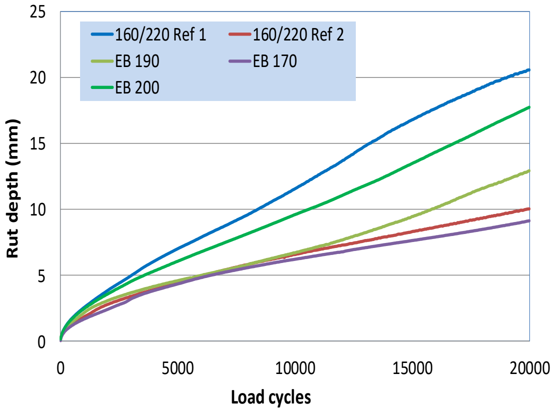
Binder	$N_f = K(1/\varepsilon)^n$		
	K	*n*	Regression R^2
160/200 Ref 1	9.60E+11	2.94	0.99
EB 190	9.70E+11	2.98	0.96
EB 200	5.90E+11	2.9	0.98
160/220 Ref 2	1.90E+13	3.59	0.97
EB 170	2.80E+13	3.7	0.99

Figure 9. Fatigue lines at 10°C for the asphalts made with the extended bitumen as compared to reference bitumen

Rutting

Resistance to rutting is evaluated using wheel tracking test typically at a temperature from 40 to 60°C. In this study, wheel tracking test was carried out at 45°C in air (dry) as well as in water with a small device according to procedure B described in EN 12697-22. Asphalt specimens were prepared by a roller sector compactor according to "TP Asphalt 33/2007", a compaction program pre-installed by the manufacturer of the compactor. Mixing temperature was 150 ± 5°C. The specimens have been "let to rest" at room temperature for seven days prior to testing. For each asphalt mix, two specimens were tested. The average data obtained at dry condition are shown in Figure 10. As indicated, the PBO extended bitumen show similar or slightly improved rutting resistance compared to the reference bitumen. It can also be seen that the two reference bitumen behave quite differently in the wheel tracking test; this could be attributed to their difference in softening point (160/220 Ref 2 shows about 3°C higher than 160/220 Ref 1, see Table 3). However, such interpretation cannot be made for the extended binders as their softening points are slightly lower, but still within specification, compared to the corresponding reference bitumen.

Regarding the tests in water condition, unfortunately, relevant results were not achieved, either because specimens failed earlier (< 10000 cycles) or repeatability between two specimens was too poor to make a reasonable average. It was felt that this type of test is probably not suitable for ABT 11 containing a soft bitumen like 160/220 at the same time without adding an adhesion promoter.



Samples	Rutting parameters		
	Proportional rut depth (%)	Ruth depth at 20000 cycles (mm)	Wheel track slope (mm/1000 cycles)
160/220 Ref 1	54.2	20.6	1.81
EB 190	33.1	12.9	1.25
EB 200	44.4	17.8	1.63
160/220 Ref 2	25.8	10.1	0.7
EB 170	23.4	9.1	0.58

Figure 10. Results of asphalt wheel tracking tests at 45°C

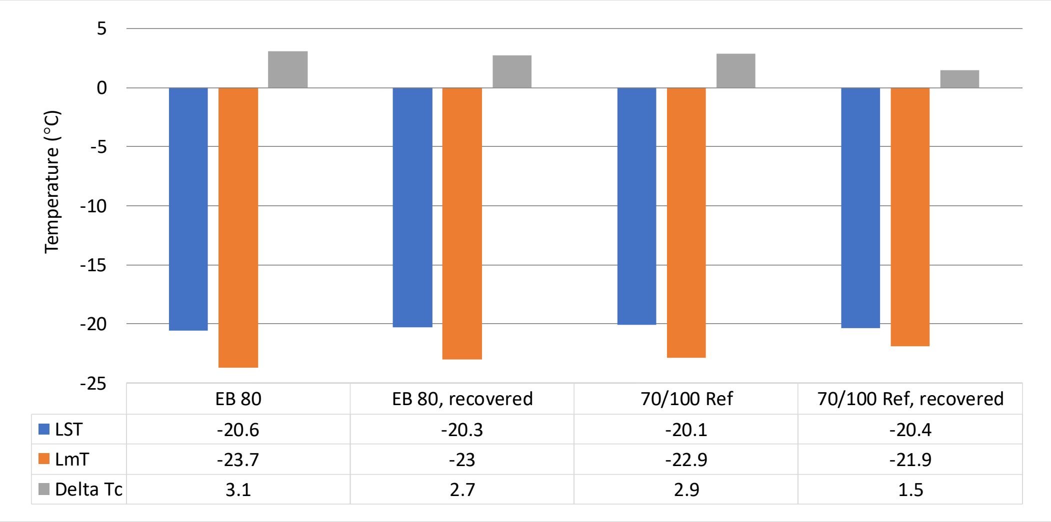
FULL SCALE FIELD TRIALS

Two field trials were carried out on the extended bitumen, EB 170 and EB 80, respectively. The trial on EB 170 was made in October 2016 in a residential area in Västerås. Asphalt layer was made with a dense mix (ABT11) with a nominal binder content of 6.0% and added with an adhesive agent. No RAP was used in the mix. During asphalt production and paving operation, a different smell was noticed, but it was not annoying or irritating. It should be pointed out that, prior to any field trial, fumes from PBO and PBO extended bitumen were generated in the laboratory at elevated temperatures, and the fume composition was analysed. No compounds were detected at any level that triggered concerns of increased health risk for asphalt workers.

Regarding performance, the pavement surface has been inspected yearly since 2016; no distress has been observed.

A second field trial was conducted in September 2018 on a road with more traffic in Arboga. Four sections were constructed with 40 mm asphalt layer: (1) SMA 16 with 70/100 Ref, without RAP; (2) SMA 16 with EB 80, without RAP; (3) SMA 16 with EB 80, with 20% RAP, and (4) SMA 16 with 70/100 Ref, with 20% RAP. The whole process of mixing and paving went smoothly, no operative differences were reported. Like the first field trial, the asphalt workers involved all commented that the smell was different but still acceptable. However, drawing a general conclusion on this aspect was not possible due to lack of quantitative data and only qualitative observations were available. The sections were also inspected in June 2019 after a first winter. As expected, all the sections remain in a good condition.

In connection to the second field trial, fresh binders and those recovered/extracted from loose mixes (without RAP) were analysed. Test results obtained by BBR are shown in Figure 11. As illustrated, the limiting temperatures at 300 MPa (LST) are quite similar for the extended bitumen and reference bitumen, and no significant differences are observed between fresh and recovered samples. However, relatively large differences are seen on LmT (temperatures at 0.300 m-value), resulting in a more positive ΔT_c for the extended bitumen than the reference bitumen, particularly for the samples recovered from loose mixes. This implies that the PBO may enhance bitumen durability, consequently, could improve asphalt performance in terms of resistance to surface cracking. To make further verification, follow-up on the field performance of the test sections will be carried out.



	EB 80	EB 80, recovered	70/100 Ref	70/100 Ref, recovered
■ LST	-20.6	-20.3	-20.1	-20.4
■ LmT	-23.7	-23	-22.9	-21.9
■ Delta Tc	3.1	2.7	2.9	1.5

Figure 11. BBR measurements of the binders used in the field trial in 2018

Carbon footprint

Based on CO_2 emission data published by Cashman et al. (2015), PBO has a cradle-to-gate carbon footprint of 740 g CO_2eq/kg, including raw material production, transport and refining. For bitumen, the following Eurobitume figure from 2012 without infrastructure has been used:

- CO_2: 174.244 g/kg GHG factor 1
- CH_4: 0.595 g/kg GHG factor 25
- CO_2eq for bitumen = 189 g CO_2eq/kg bitumen

When forest grows, CO_2 is absorbed from the atmosphere and is bound. All pulping products harvested from the forest share that benefit. The elemental analysis (Cf. Table 1) shows that the PBO consists of about 80% carbon. As a molecular weight ratio of CO_2 and C equals 3.7, 1 kg PBO has bound approximately 3 kg CO_2 when the forest is growing. Therefore, the biogenic CO_2eq content of PBO is -3000 g per kg PBO which may be subtracted from its production footprint. For an extended bitumen with 10% PBO, the total CO_2eq contribution is:

$$\%m\ \text{bitumen} * CO_2\text{eq bitumen} + \%m\ \text{PBO} * (CO_2\text{eq PBO} - \text{biogenic } CO_2) =$$
$$0.90*189 + 0.1*(740 - 3000) = -55.9\ \text{g}\ CO_2/\text{kg PBO-bitumen}$$

The negative value suggests that CO_2 is actually stored by using the PBO in bitumen. Consequently, the environmental benefit in terms of CO_2 reduction can be very significant. And a bitumen-PBO blend would be carbon neutral with about 7.5% PBO, as illustrated in Figure 12.

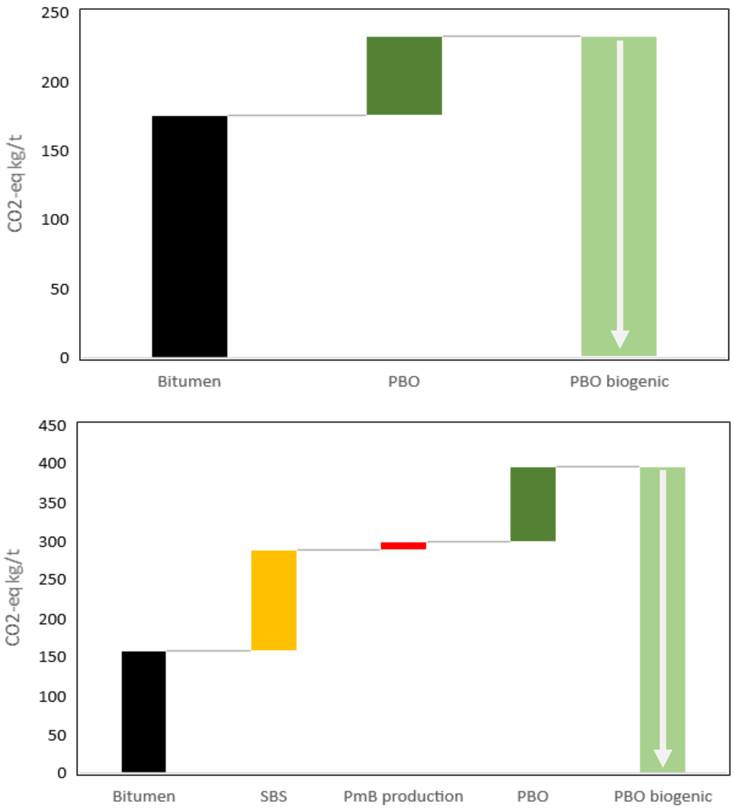


Figure 12. Illustrations of CO2 equivalent of bitumen (upper figure) and SBS polymer modified binder (lower figure) extended with PBO

A similar calculation was also made for SBS polymer modified binders based on the corresponding CO_2 data for the polymer and PmB production from Eurobitume LCI 2012. The CO_2 equivalents of different components are shown in Figure 12, suggesting that the polymer modified binder can become carbon neutral by selecting the right proportion of the PBO.

It should be noted that, for the above estimation, besides using the data from Eurobitume and from the literature (Cashman et al., 2015), assumptions are also made based on EN 15804:2012 and CEN TR 16970:2016, and that PBO is assumed to be manufactured from sustainably managed forests. The environmental and LCA standards and values may change in the future.

RECYCLABILITY

In a collaborative study with a university in Italy, the recycling aspect of PBO extended binders has been investigated (Ingrassia et al., 2020). The study focused on evaluating the effectiveness of the extended binders in the hot recycling of traditional RAP and their recyclability potential in a long-term perspective. A severely aged PBO extended binder was prepared in laboratory using RTFOT plus prolonged PAV, while a traditional RAP binder was recovered from reclaimed asphalts. Virgin binders (with and without PBO) were blended with those aged binders to simulate hot recycling. Mechanical and chemical properties, as well as aging susceptibility, of those blends were measured and compared with a control bitumen. It was concluded that the PBO extended binders are effective in the hot recycling of RAP and also completely recyclable.

CONCLUSIONS

This paper presents extensive investigations on a plant-based oil (PBO) as a potential renewable bitumen extender. The results obtained show that the PBO studied is a complex mix of numerous chemicals, including fatty acids and esters, and is fully compatible with bitumen and no phase separation occurs during handling or hot storage. The PBO extended bitumen fulfil specification EN 12591, with additional improvement on binder long-term durability.

In terms of asphalt performance, the PBO extended bitumen show improved adhesion especially when poor aggregates are used, as assessed by rolling bottle test and ITSR. Meanwhile, no significant differences in stiffness are found between the asphalts with the PBO extended bitumen and those with the corresponding reference bitumen. The asphalts also show similar temperature susceptibility, suggesting the PBO will most probably not affect asphalt low temperature cracking performance. In addition, blending the PBO into the bitumen does not seem to have a significant effect on asphalt fatigue behaviour. As for rutting resistance, the PBO extended bitumen are similar or slightly better when compared to reference bitumen.

Full scale field trials on the PBO extended bitumen are satisfactory from operation perspectives and HSE observations. All the test sections have been found to be in a good condition over time.

Importantly, a cradle-to-gate carbon footprint consideration shows that the PBO extended binders can become carbon neutral, depending on the proportion of PBO used even for SBS-polymer modified binders.

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REFERENCES

Ahmedzade P., Tigdemir M., Kalyoncuoglu S. F. (2007), “Laboratory investigation of the properties of asphalt concrete mixtures modified with TOP-SBS”, Construction and Building Materials, Vol. 21, pp. 626-633.

Anderson R. M., King G. N., Hanson D. I., Blankenship P. B. (2011), “Evaluation of the relationship between asphalt binder properties and non-load related cracking”, *AAPT*, Vol. 80, pp 615-663.

Ball G. F. A., Herrington P. A., Patrick J. E. (1993), “Tall oil pitch as bitumen extender”, *New Zealand Journal of Forestry Science*, Vol. 23, No. 2, pp 236-242.

Bearsley S. R., Haverkamp R. G. (2007a), “Age hardening potential of tall oil pitch modified bitumen”, *Road Materials and Pavement Design*, Vol. 8, No.3, pp.467-481.

Bearsley S. R., Haverkamp R. G. (2007b), “Adhesive properties of tall oil pitch modified bitumen”, *Road Materials and Pavement Design*, Vol. 8, No.3, pp.449-465.

Cashman S. A., Moran K. M., Gaglione A. G. (2015), “Greenhouse gas and energy life cycle assessment of pine chemicals derived from crude tall oil and their substitutes”, *Journal of Industrial Ecology*, Vol. 20, No. 5, pp 1108 – 1121.

Celauro C., Bernardo C., Gabriele B. (2010), “Production of innovative, recycled and high-performance asphalt for road pavements”, *Resources, Conservation and Recycling*, Vol. 54, pp 337–347.

Cooper III S. B., Mohammad L. N., Elseifi M. (2013), “Evaluation of asphalt mixtures containing renewable binder technologies”, *International Journal of Pavement Research and Technology*, Vol. 6, No. 5, pp 570-575.

EN 12591, “Bitumen and bituminous binders – Specifications for paving grade bitumens”, CEN, Brussels.

EN 12697-11, “Bituminous mixtures - Test methods for hot mix asphalt - Part 11: Determination of the affinity between aggregate and bitumen”, CEN, Brussels.

EN 12697-22, “Bituminous mixtures - Test methods for hot mix asphalt - Part 22: Wheel tracking”, CEN, Brussels

EN 12697-23, “Bituminous mixtures – Test methods for hot mix asphalt – Part 23: Determination of the indirect tensile strength of bituminous specimens”, CEN, Brussels.

EN 12697-24, “Bituminous mixtures – Test methods for hot mix asphalt – Part 24: Resistance to fatigue”, CEN, Brussels.

EN 12697-26, “Bituminous mixtures – Test methods for hot mix asphalt – Part 26: Stiffness”, CEN, Brussels.

EN 15804 (2012), “Sustainability of construction works — Environmental product declarations — Core rules for the product category of construction products”, CEN, Brussels.

Eurobitume (2012), “Life Cycle Inventory: Bitumen”, Published by the European Bitumen Association, Brussels, Belgium.

Fini E. H., Al-Qadi I. L., You Z., Zada B., Mills-Beale J. (2012), “Partial replacement of asphalt binder with bio-binder: characterisation and modification”, *International Journal of Pavement Engineering,* Vol. 13, No. 6, pp 515-522.

Ingrassia L. P., Lu X., Ferrotti G., Canestrari F. (2019), “Renewable materials in bituminous binders and mixtures: Speculative pretext or reliable opportunity?”, *Resources, Conservation and Recycling,* Vol. 144, pp 209–222.

Ingrassia1 L. P., Lu X., Ferrotti G., Conti C., Canestrari F. (2020), “Investigating the “circular propensity” of road bio-binders: effectiveness in hot recycling of reclaimed asphalt and recyclability potential”, *Journal of Cleaner Production,* Vol. 255, DOI:10.1016/j.jclepro.2020.120193.

Kluttz R. (2012), “Considerations for use of alternative binders in asphalt pavements – Material characteristics”, *Transportation Research Circular,* E-C165, pp 2-6.

Lu X., Redelius P., Soenen H., Thau M. (2011), “Material characteristics of long lasting asphalt pavements”, *Road Materials and Pavement Design,* Vol. 12, Issue 3, pp 567-585.

Midgley L., Urquhart R. (2012), “Investigation into the properties of alternative surfacing binders and bitumen extending binders”, Austroads Technical Report, AP-T206-12.

Monismith C. L. (2004), “Evolution of long-lasting asphalt pavement design methodology: A perspective”, *ISAP International Symposium on Design and Construction of Long Lasting Asphalt Pavements,* June 7-9, 2004, Auburn University, Alabama, USA

Nunn M. E., Ferne B. W. (2001), “Design and assessment of long-life flexible pavements”, *Transportation Research Circular,* Number 503, December 2001.

Oda S., Leomar Fernandes Jr. J., Ildefonso J. S. (2012), “Analysis of use of natural fibers and asphalt rubber binder in discontinuous asphalt mixtures”, *Construction and Building Materials*, Vol. 26, No. 1, pp 13-20.

Peltonen P. (1989a), “Research into the physico-chemistry of tall oil pitch modified asphalt composites”, *Wood and Pulping Chemistry, TAPPI Proceedings*, pp. 391-398.

Peltonen P. (1989b), “Finnish experience in tall oil pitch as asphalt substitute”, *Naval Stores Review*, September/October, 1989, pp.10-12.

Peralta J., Williams R. C., Silva H. M. R. D., Machado, A. V. A., (2014), “Recombination of asphalt with bio-asphalt: binder formulation and asphalt mixes application. *AAPT*, Vol. 83, pp 1-36.

Seidel J. C., Haddock J.E. (2012), “Soy fatty acids as a sustainable modifier for asphalt binders”, *Transportation Research Circular*, E-C165, pp 15-22.

Sol-Sanchez M, Moreno-Navarro F., Garcia-Trave G., Rubio-Gamez M. C. (2016), “Analysing industrial manufacturing in-plant and in-service performance of asphalt mixtures cleaner technologies”, *Journal of Cleaner Production*, Vol. 121, pp 56-63.

TR 16970 (2016), “Sustainability of construction works - Guidance for the implementation of EN 15804”, CEN, Brussels.

Von Quintus H. L., Mallela J. and Buncher M. (2007), “Quantification of effect of polymer-modified asphalt on flexible pavement performance”, *Transport Research Record*, No. 2001, pp 141-154.

Wayman M., Nicholls J. C., Carswell I. (2014), “Use of lower temperature asphalt in pavement construction: Demonstration site construction, in service performance and specification”, Transport Research Laboratory, UK.

Xie S., Li Q., Karki P., Zhou F., Yuan J. S. (2017), “Lignin as renewable and superior asphalt binder modifier”, *ACS Sustainable Chemistry and Engineering*, Vol. 5, No. 4, pp 2817-2823.

Yang X., You Z., Dai Q., Mills-Beale J. (2014), “Mechanical performance of asphalt mixtures modified by bio-oils derived from waste wood resources”, *Construction and Building Materials*, Vol. 51, pp 424-431.

Zaumanisa M., Mallicka R. B. (2015), “Review of very high-content reclaimed asphalt use in plant-produced pavements: state of the art”, *International Journal of Pavement Engineering*, Vol. 16, No. 1, pp 39–55.