White paper:
A Nynas bitumen breaking new ground for lower carbon roads

EXECUTIVE SUMMARY

Designing high performance construction materials that last longer and which are 100% reusable or recyclable allows the road construction industry to take a major leap in reducing its overall carbon impact. Materials that fulfil these requirements while having a reduced carbon footprint, break entirely new ground on the journey to contribute to reduce carbon and climate impact. An upcoming new Nynas bitumen product range is one step which Nynas has taken on this journey.

This paper describes the development and current status of a new type of Nynas bitumen. A high-performance polymer modified bitumen for road applications that increases pavement life when compared to unmodified bitumen while having a smaller carbon footprint than other polymer modified bitumen products.

The new range combines Nynas polymer modified bitumen knowledge with a renewable hydrocarbon material containing mostly biogenic carbon. Materials containing biogenic carbon, sustainably sourced, can significantly reduce the total product carbon footprint. The result is a range of carefully designed, groundbreaking modified bitumen with all the performance advantages of a polymer modified bitumen – and more – with a reduced carbon footprint

Contents

INTRODUCTION

Bitumen is a key ingredient in asphalt mixtures used to build roads or in roofing- and construction materials to waterproof our homes. Designed to last for a very long time, many of these materials also allow for 100% re-use or recycling at the end of their useful service life. This makes bitumen one of the most, if not the most sustainable road construction products.

For more than 50 years, polymers have been combined with bitumen to create high-performance polymer modified bitumen (PMB) for use in heavy duty applications and/or to extend overall service life. The downside of using polymers is that the overall carbon footprint increases significantly when compared to standard bitumen [1] as polymers themselves can be very carbon intensive.

However, combining bitumen with renewable hydrocarbon materials containing biogenic carbon can lead to significant reduction of the carbon footprint. This principle has been applied into the development of Nynas upcoming polymer modified bitumen range, which will contain products that are engineered mixtures of bitumen, SBS polymers and a renewable material containing biogenic carbon. The information given in this paper is based on Nynas work with one such material, tall oil pitch (TOP), the bottom fraction of the distillation of crude tall oil, obtained from the wood pulping process. TOP comes with carbon credits.

The upcoming products are high-performance polymer modified bitumen for road applications that increase pavement life when compared to standard bitumen while having a similar carbon footprint. Studies lead by Nynas have further shown that the use of TOP improves the resistance to ageing of bitumen, thereby contributing to an overall improved durability [2]. It has been shown that it is fully re-usable/recyclable at the end of its service life. This combination results in a long service life, durable and fully re-usable or recyclable bitumen with a significantly reduced carbon footprint when comparing with other polymer modified binders.

This paper gives a review of the development of, and details the carbon footprint calculations for, one of the new range of Nynas products.

DEVELOPMENT OF BITUMEN WITH LOW CARBON FOOTPRINT

Performance advantages of polymer modification

Numerous investigations have already shown advantages of using PMBs when compared to unmodified bitumen. Improvements in asphalt performance are found with respect to ageing (durability), permanent deformation (rutting), fatigue damage, and low temperature cracking. The improvements are verified by various full-scale tests and field projects. For example, it was shown by the American LTPP (Long Term Pavement Performance) programme that road sections with PMB mixes had much less fatigue cracking, thermal cracking and rutting compared to conventional companion sections; thus, the use of PMBs significantly extends pavement lifetime [5]. As another example, in test sections constructed in Sweden during 2003-2006, significantly enhanced fatigue life was found for the asphalt mixes with SBS modified binders [6]. When the reference sections are optimized with a design life of approximately 20 years, the sections with the SBS modified binder in the base course mix are predicted to last at least 30 years longer.

Performance in the lab

Comprehensive laboratory investigations have been carried out on different bituminous binders containing TOP [3, 4], including polymer modified binders (PMBs). It has been demonstrated that TOP is fully miscible with bitumen, and by a careful selection of the base bitumen type and dosage, desired standard paving grade specifications according to EN 12591 can be achieved. PMB products formulated with TOP also fulfil the CEN specification EN 14023.

Moreover, bitumen containing TOP was studied with regards to long-term ageing by Pressure Ageing Vessel (PAV) at the standardized conditions according to EN 14769. No detrimental effect on binder long-term ageing was found. On the contrary, TOP has been shown to improve the ageing resistance of bitumen [3]. The long-term durability of bitumen containing TOP is also illustrated by ΔT_c, a bitumen durability parameter which was determined using the bending beam rheometer (BBR), see Figure 2 below.

Performance tests on asphalt mixtures including adhesion, stiffness, resistance to fatigue cracking, and resistance to rutting were performed [4]. Bitumen containing TOP was found to show improved adhesion with mineral aggregates as evaluated by the rolling bottle test[1] and indirect tensile strength ratio (ITSR)[2]. With regards to stiffness[3], fatigue[4] and permanent deformation[5], no significant differences were observed between bitumen containing TOP and the reference bitumen [4]. These laboratory test results indicate that the in-field performance of bitumen containing TOP is at least as good as that of the reference bitumen. Therefore, this new bitumen can be expected to have at least a comparable, if not a longer, useful life.

Laboratory tests also demonstrate that PMBs with and without TOP perform equally; examples of test results are shown in Table 1.

Table 1. Performance testing of asphalt mixtures with PMB 40/100-75 with and without TOP

Asphalt mixture ABS 16	Wheel tracking test at 60°C		Water sensitivity		
	Rut depth at 20000 cycles (mm)	Wheel tracking slope (mm/1000 cycles)	ITS wet (kPa)	ITS dry (kPa)	ITSR (%)
Reference 40/100-75	2.6	0.04	1556	1650	94
40/100-75 containing TOP	1.9	0.05	1534	1571	98

[1] EN 12697-11
[2] EN 12697-23
[3] EN 12697-26, annex C
[4] EN 12697-24, annex E
[5] EN 12697-22

Before proceeding to full scale trials, fumes from bitumen containing TOP were studied because, during laboratory scale blending and asphalt production, a different smell had been noticed. Fumes from TOP, bitumen with and without TOP were generated in the laboratory at elevated temperatures and the fume composition was extensively analysed. No compounds were detected at any level that triggered concerns of increased health risks for asphalt workers. During subsequent full scale field trials, both in asphalt production and paving operation, the different smell was confirmed but it was not considered annoying or irritating.

Field trials

Two full-scale field trials were carried out on bitumen containing TOP [3]. The first trial was made with a 160/220 grade bitumen in October 2016 in a residential area in Västerås, Sweden. The asphalt surface layer was made with a dense continuous graded asphalt mix, and no reclaimed asphalt pavement (RAP) was used. The pavement surface has been inspected annually since 2016; no distress has been observed confirming the asphalt performance.

In September 2018, a second field trial was conducted with a 70/100 grade bitumen on a road with more traffic in Arboga, Sweden, see Figure 1 below. Four sections were constructed with a stone mastic asphalt (SMA) surface layer, with and without RAP. The whole process of mixing and paving went smoothly, no operative differences were reported. As with the 2016 trial, the sections have been inspected annually with all the sections remaining in a good condition.

Figure 1. Field trial of bitumen containing TOP and performance follow up

In connection with the second field trial, virgin binders and those recovered from laboratory aged loose mixes were analysed by BBR, see Figure 2 below. It was found that bitumen containing TOP has, in terms of ΔTc, enhanced durability and consequently may well improve asphalt performance in terms of resistance to surface cracking.

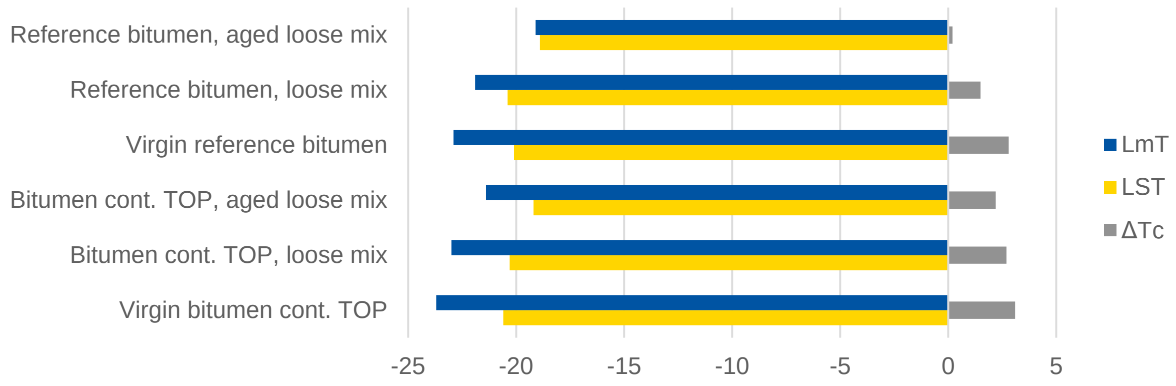

Figure 2. Analysis of 70/100 reference bitumen and 70/100 bitumen containing TOP by Bending Beam Rheometer

Follow up studies

Further extensive studies on bitumen containing TOP were carried out at Ancona University in Italy through a Ph.D project jointly supported by the university and Nynas [2,7,8,9,10]. The project mainly focused on the laboratory characterization of the properties of bitumen containing TOP, including chemistry, morphology, rheology, ageing susceptibility, adhesion with aggregates, moisture susceptibility and circularity aspects. The results of the chemical and rheological tests carried out indicated that the ageing susceptibility of the binder decreases as TOP content increases, see Figure 3. Bitumen containing TOP may have similar or even higher ageing resistance when compared to conventional bitumen of similar penetration grade. Moreover, from the performance point of view, the permanent deformation resistance and fatigue resistance of bitumen containing TOP were found to be comparable to those observed for the conventional bitumen of a similar penetration grade.

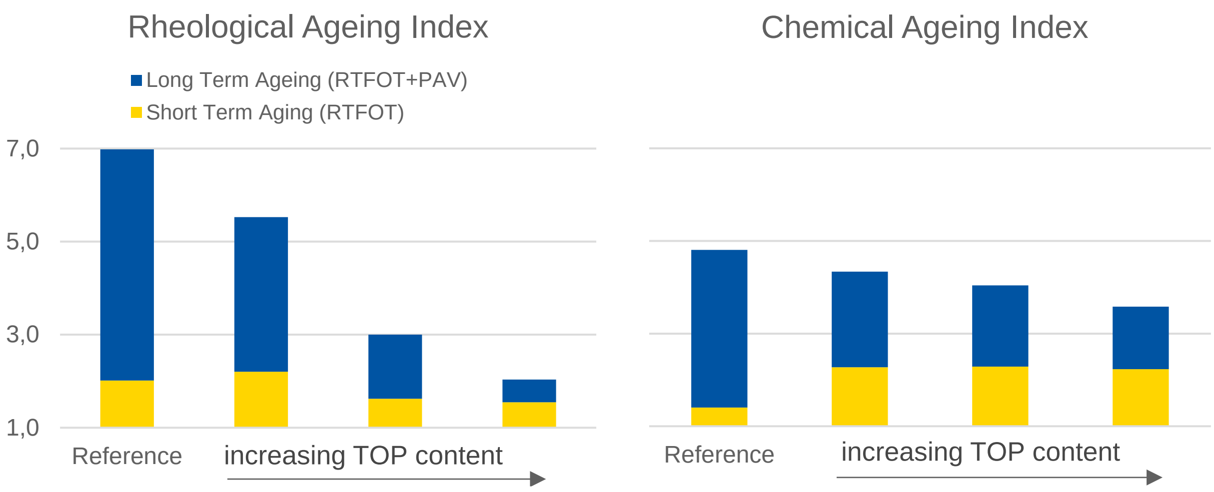

Figure 3. Rheological and Chemical Ageing Index as a function of TOP content

Furthermore, improved adhesion between bitumen containing TOP and aggregates and reduced moisture susceptibility of the systems were shown by binder bond strength (BBS) tests. In terms of asphalt viscoelastic properties and fatigue behavior, it was found that the long-term ageing behaviour of the asphalt mix with bitumen containing TOP was improved compared to the reference asphalt mix [2,7]. This points to potential fatigue performance benefits in the long term.

Overall, these findings confirm that TOP is suitable to be used combined with bitumen for road pavements. It is expected that bitumen containing TOP has at least comparable performance and perhaps even better durability and long-term performance compared to standard bitumen.

End of life and circularity

During its service life, bitumen is neither burned nor is it biodegradable [11]. Thus, bitumen is not turned into CO_2 during its useful life in asphalt roads. Furthermore, asphalt is 100% reusable [12]. Polymers like SBS blended with bitumen form an intrinsic part of the asphalt binder and are therefore re-used simultaneously and in the same way as bitumen.

Tall oil pitch, CAS number 8016-81-7, EINECS 232-414-4, on its own, is reported in ECHA's REACH dossier [11] not to be readily degradable nor readily biodegradable according to the Closed Bottle test OECD, 301D. It blends well with bitumen forming a homogenous stable mixture. Bitumen with up to 15% TOP is not expected to perform any differently from pure bitumen.

As part of the collaborative study with Ancona University, the recycling aspect of bitumen containing TOP has been investigated [8, 9]. The study focused on evaluating the effectiveness of bitumen containing TOP in the hot recycling of traditional RAP and their recyclability potential in a long-term perspective, in other words the circular propensity of this new material. A severely aged bitumen containing TOP was prepared in the laboratory using the Rolling Thin Film Oven Test[6] as short-term ageing test followed by extended Pressure Ageing Vessel[7] for long-term ageing, while a traditional RAP binder was recovered from reclaimed asphalts. Virgin binders, with and without TOP, were blended with the aged binders to simulate hot recycling. Mechanical and chemical properties, as well as ageing susceptibility of the blends, see Figure 4, were measured and compared with a reference bitumen. It was concluded that bitumen containing TOP is effective in the hot recycling of RAP and also completely recyclable.

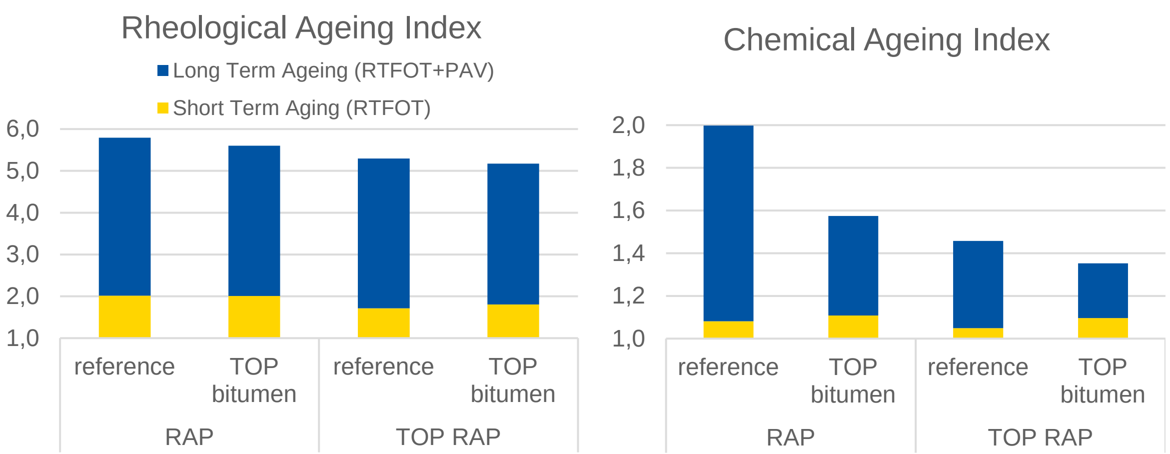

Figure 4. Rheological and Chemical Ageing Index of bitumen with and without TOP in RAP with and without TOP

Because of its extraordinary circular propensity, one can not really identify "end of life" for bituminous binders containing TOP in asphalt.

[6] RTFOT, EN 12607-1
[7] PAV, EN 14769

CARBON FOOTPRINT

Methodology

Public data from Life Cycle Inventory reports from raw material producers are used and combined to calculate the total carbon footprint of a typical product in the upcoming Nynas product range. The impact of secondary distribution is estimated based on public data and actual transportation distances. Combined these provide the total carbon footprint at the location of production. Relevant standards, assumptions and guidance can be found in references [13,14].

Raw material production

Typical cradle to factory gate $CO_{2\,eq}$ emission data reported for the raw materials bitumen, TOP and SBS are given in Table 2 in g/kg. This includes all raw material production, transport to and refining/manufacture of the components used at the place of their production.

Table 2. Cradle to gate $CO_{2\,eq}$ footprint of raw materials

	g/kg $CO_{2\,eq}$	
Bitumen	147 400 - 460 637 - 766	Eurobitume (2020) [15] Nynas own data (2021) Asphalt Institute (2019) [16]
TOP	740 600	Cashman et al (2015) [17] Supplier data [18]
TOP biogenic contribution	-3000 -2780	Elemental composition [3] Supplier data [18]
SBS	3716 2311 - 2500	IISR (1998) [19] Supplier data [18]

When forests grow, CO_2 is absorbed from the atmosphere and is bound. All pulping products harvested from the forest share that benefit. The elemental analysis [3] shows that TOP consists of about approximately 80% carbon. As a molecular weight ratio of CO_2 and C equals 3.7, 1 kg of TOP has bound approximately 3 kg CO_2 when the forest is growing. Therefore, the biogenic CO_2 content of TOP is -3000 g per kg TOP which may be subtracted from its production footprint. One supplier has reported a biogenic content of -2780 g $CO_{2\,eq}$/kg TOP manufactured at their production site and an embodied carbon content of 600 g $CO_{2\,eq}$/kg for production [18]. The net negative value suggests that CO_2 is stored by using the TOP in bitumen. Consequently, expressed in CO_2, the environmental benefit can be very significant.

For the new product, an SBS modified bitumen with TOP, the total $CO_{2\,eq}$ contribution of the raw materials is:

$$\%m\ bitumen\ * CO_{2\,eq}(bitumen) + \%m\ SBS\ * \ CO_{2\,eq}(SBS) + \%m\ TOP * \left(CO_{2\,eq}(TOP) - biogenic\ CO_2(TOP)\right)$$

Equation 1. Total $CO_{2\,eq}$ contribution from raw materials

For bitumen, the footprint data from the latest version of the Eurobitume Life-Cycle Inventory [15] has been used. However, there is new data available indicating that the $CO_{2\,eq}$ for bitumen is higher. Initial Nynas calculations indicate a range of 400 to 460 g $CO_{2\,eq}$/kg. For the purpose of this paper, the Eurobitume data is used. For TOP the supplier data [18] and for SBS the IISR data [19] is used.

Polymer modified bitumen production

Blending polymers into bitumen requires energy as a high shear mixing process is used. This process is estimated to have a carbon footprint of 10.5 kg $CO_{2\,eq}$ per ton of product [1]. This should be added to Equation 1 to represent the carbon footprint of the manufactured product.

Distribution

Equation 1 provides the $CO_{2\ eq}$ per ton of final product based on only raw materials. However, as raw materials are produced at different locations than the final product, the carbon emissions from the distribution/transportation of raw materials should be included. To calculate this contribution, considering different modes of transportation, data used is based on Cefic-ECTA reports [20, 21]. To build in a conservative margin, higher emission factors have been used in this report, see Table 3 below.

Table 3. $CO_{2\ eq}$ impact of raw material distribution

Transportation mode	$CO_{2\ eq}$ g/ton*km		Comment
	Used in calc.	Ref. Cefic-ECTA	
Road truck	100	70.3	Reference data based on empty return journey truck
Ship bulk	16	16	Overall emission factor for short-sea shipping
Ro-Ro ferry	50	21	Reference data is industry avg. intermodal road/short sea

Bitumen is normally transported by road truck or in bulk by ship. TOP is transported by road truck. Polymers are normally distributed by road truck, but some intermodal transportation occurs for which Ro-Ro ferry data is used.

Carbon footprint calculation of finished product: a real case

This case study example is based on actual data representing a real situation. Actual distribution distances and transportation modes of components of the new Nynas PMB in this example are given in Table 4.

Table 4. Distribution distances used per component and transportation mode

Raw material	Transportation Mode	
	Road Truck	Ship
SBS	3 000 km	
TOP	275 km	
Bitumen		635 km

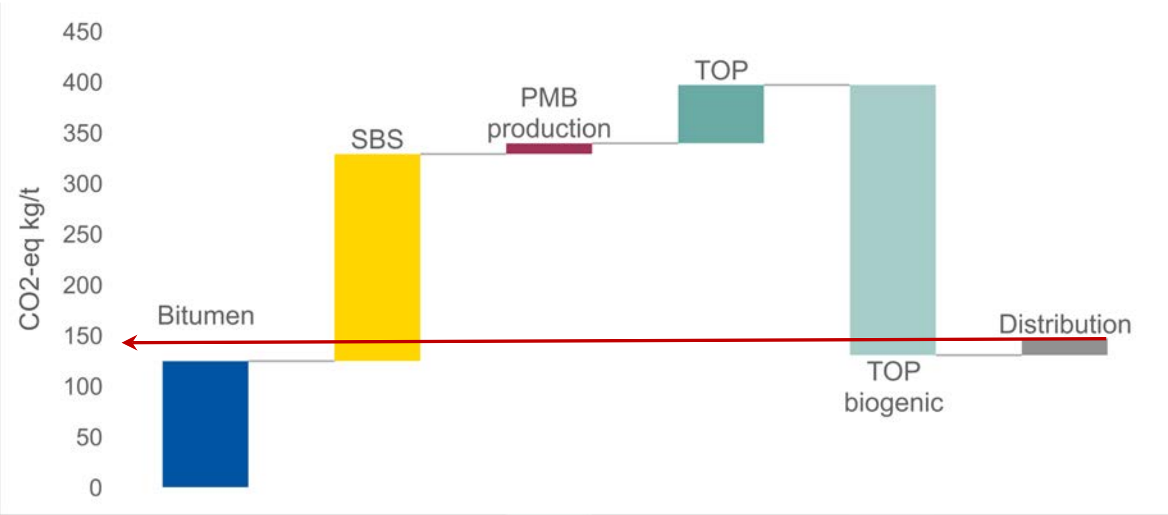

Figure 5. Total carbon footprint breakdown of finished product

The schematic representation of the total carbon footprint calculation for the finished product including contributions from the component materials, transportation of components and the PMB production is given in Figure 5. For this situation the total carbon footprint of the finished product is 147 kg $CO_{2\ eq}$ per ton product. The red line in Figure 5 indicates the carbon footprint of a standard paving grade bitumen, see also Table 2.

CONCLUSIONS

Bitumen combined with TOP can be designed to meet the requirements for standard paving grade bitumen and be used in asphalt mixtures for road applications. Laboratory investigations and field trials have confirmed at least equal performance and, for some properties, even better performance for bitumen containing TOP. Improvements were specifically identified in the areas of long-term durability and adhesion with mineral aggregates resulting in better water sensitivity of the asphalt mixtures studied. Bitumen containing TOP can be used in asphalt mix production using Recycled Asphalt Pavements. Furthermore, asphalt mixtures with bitumen containing TOP can be re-used or recycled themselves at the end of their useful service life.

The products in the upcoming Nynas range are PMBs containing TOP, engineered to have properties identical to those found in their corresponding PMBs without TOP. Modified bitumen, such as the Nynas Nypol range, is well known to significantly increase pavement life especially under heavy trafficked conditions creating more durable, long lasting asphalt pavements. Improving pavement life contributes greatly towards reducing the overall carbon footprint of the road. However, the use of carbon intensive polymers can increase the embodied carbon of asphalt. Using TOP in a polymer modified bitumen will compensate for this reducing the carbon footprint of an asphalt road even further. The net result for the new type of Nynas bitumen is a product with all the performance advantages of a polymer modified bitumen – and more – but with the carbon footprint of a standard paving grade bitumen.

The case study example presented in this white paper provides details of the cradle to gate carbon footprint of this modified bitumen.

Designing high-performance construction materials that last longer and that are 100% reusable/recyclable – at the end of a long service life – allows the road industry to take a major leap in reducing carbon impact. Engineering construction materials that achieve this which at the same time have a reduced carbon footprint, breaks entirely new ground on the journey to contribute to reduce carbon- and climate impact. The upcoming range of new PMBs is one step which Nynas is taking on this journey.

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