Sustainability & Environment

The use of lignin as bio-binder in asphalt applications

Jeroen Besamusca[1], Paul Landa[2], Rop Zoetemeyer[3], Richard Gosselink[4], Bram Lommers [5], Martin Junginger[6], Martijn Verschuren[7]
[1]Q8Research, [2]AKC, [3]Biobased Delta, [4]Universiteit Wageningen, [5]Mourik bv, [6] Universiteit Utrecht, [7]H4A

Abstract

Several investigations have shown interest in the use of durable material for road constructions, including bio-based oil, The introduction of lignin in asphalt applications opens the door for a new bio-binder in infrastructure. With a 50/50 blend of bitumen and lignin, the laboratory tests revealed equal performance with regular asphalt. Several trials started already all in top-layer applications and including a test at the harbour of Antwerp for heavy-duty performance. The total amount of bitumen used worldwide is estimated on 70 million ton annually. The current amount of lignin produced worldwide is probably 50 million ton and will increase. The advantage of using lignin is the positive impact on our environment. Lignin is a natural resource released during the production of pulp in the paper industry. It is an extensive waste stream, which is mainly used for energy production. Paper is formed on the carbon dioxide in our atmosphere and therefore binding carbon in a positive way. The waste of pulp production is not a production as such; it is a left over from the use of bio-material. By using the lignin in road constructions, the bound carbon will stay captured. The use of lignin contributes to lower emission because of the lower production temperature of asphalt production. Predictions show that bitumen-producing refineries will decrease resulting in a lack of supply for infrastructure. Recycling and use of durable material will be the answer for our children and their future.

This paper was first published in 2020 in version 1.0 of the proceedings of the 7th E&E Congress. This second edition (version 1.1.) of the proceedings is published in June 2021. The content of this paper has not changed, but the author(s) may have provided additional information in the form of an annex.

1. Introduction

Globally, policies aiming to achieve the 2015 Paris agreement to limit climate change to 2° C or lower are targeting the phasing out of fossil fuels for energy and materials [1].

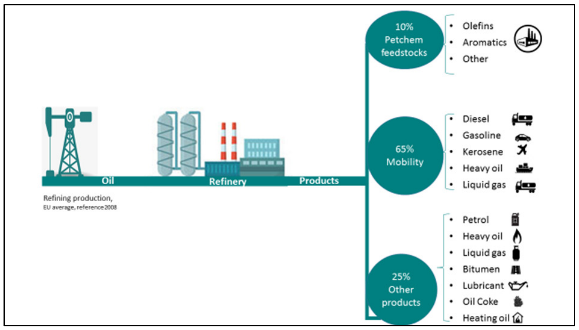

Figure 1: Refinery output [1]

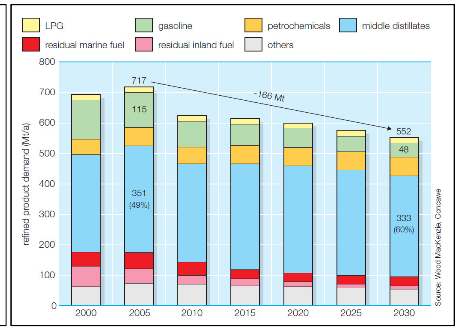

Figure 2: Prospect change for EU [1]

For the construction sector, and specifically the constructions of roads, this is a tremendous challenge, as asphalt contains significant amounts of bitumen. Even though the use of bitumen is not emitting major GHG emissions directly (as most fossil carbon is fixed), the construction sector is looking for renewable alternatives. In addition, refineries are increasingly using furnaces to crack most or all of the low value bottoms from crude oil to achieve higher yield of lighter products. This however means increasing less volumes of high quality bitumen on the market, which triggered price increases with 20% of bitumen in e.g. the Netherlands from 2017 to 2018. Thus, there is also an economic incentive to search for alternatives for bitumen.

One such alternative could be lignin. Lignin is one of the most abundant natural polymers (next to cellulose and hemicellulose) present in plant material and consists of aromatic building blocks. It occurs in woody and non-woody cellulosic biomass and acts as a binder to provide strength and rigidity to the plants, provide UV stability and resistance against microbial attack. In the bio-economy, lignin is generated as a side stream in the production process for pulp and paper and as a non-fermentable side stream in the production of cellulosic bioethanol from lingo-cellulosic biomass such as wood, straw and grasses [2]. Ligno-sulphonates, resulting from sulphite pulping, are produced as a commodity at over 1 million tonnes per year. Kraft lignins are produced by several companies via the Kraft process over 100,000 tonnes per year and isolation of Kraft lignin is realised by several technologies such as lignoboost [3]. Despite its properties as a valuable biopolymer, currently lignin is mainly burnt as low-value fuel. The energy value of dry lignin is around 24 MJ/kg.

Its availability in Europe is significant:

a. from Kraft pulping operations approx. 55 million tonnes/year of which a fraction is recovered from black liquor; Kraft lignin, has a characteristic strong odour. The sulphate and the odour results in a poor usefulness of this type of lignin.
b. from sulphite pulping operation approx. 1.2 million tonnes/year of lingo-sulphonates are recovered. They are applied as additives in cement and drilling fluids and agents in the chemical industry.
c. as a by-product of innovative biorefineries approx. 225 million tonnes/year estimated by 2030 with the current target to replace 30 % fossil-based fuels by bioethanol. The applied processes are mainly based on steam explosion and enzymatic hydrolysis, and the soda process, resulting in sulphur free lignins. This soda lignin is being applied in phenol-formaldehyde dyes, composites, antioxidant, antibiotics, animal feed, concrete and agents in the chemical industry.

Lignin has good adhesive properties and can resist high temperatures. This makes it interesting as a (partial) replacement as a binder in asphalt. However, so far, limited work has been published on lignin as (partial) bitumen replacement. In the Netherlands, a number of field trials have been carried out since 2015, and considerable experience has been gained with various types of lignin as binder in asphalt. The aim of this paper is to present an overview of the current state-of-the-art of the technical, economic and environmental performance of novel lignin-based asphalt.

In the following section, a literature overview on the performance of lignin is presented to show the current state of knowledge. Next in section 3, the state of lignin use in the Netherlands is presented, providing amongst others an overview of the existing next plots and the technical performance of various types of lignins as binder in asphalt. Section 4 explores the potential greenhouse gas reduction of lignin based asphalt in comparison to fossil based asphalt. Finally, the paper provides discussion, conclusions and recommendations for further research needs in section 5.

2. **Literature review**

In the search for alternatives for bitumen as asphalt binder several bio-based binders were tested in the past. Binders developed from micro-algae [4], vegetable oil streams [5,6], wood derived bio-oil [7,8] and oil from waste [9,10,11,12] led to some improvements in an asphalt binder, but only the vegetable oils were used as rejuvenating agents to compensate for aged bitumen quality decrease. A different alternative bio-based material to substitute bitumen is lignin, which is the primary bio-based material studied in this work.

Lignin from biomass has been studied and is known as a potential substitute for part of the bitumen in asphalt and as substitute in other applications such as resins, carbon fibres, polyurethanes, and composites.
The reason to apply lignin as partial substitute for bitumen is its functionality as binder, UV stability properties and it has structural similarities to fractions of bitumen, as lignin contains similar unsaturated aromatic rings joined by alkyl chains. Next to that is the potentially large availability of lignin of over 70 million tonnes an important factor [13]. Several references have been published on the use of lignin in bitumen application for hot asphalt application [14,15]. Recent publications are WO 2015/137813 [16] and WO 2019/092278 [17]. Both patents describe the use of lignin as partial replacement of the binder in asphalt up to 50% substitution.
Next to these, several papers have emerged describing the use of lignin in asphalt binders as an anti-oxidant (Akusar et al.) [18], anti-aging (Xu et al. [19], Arafat et al.[20]), and modifier (Xie et al.[21]). Also in 2019 Yue et al. [22] have published a short review. All these scientific papers describe the use of lignin in bitumen from small amounts (0.2% to max 20%). This paper describe substitution degrees of up to 50%, which is far more significant in the search for alternatives for the current bituminous asphalt binder. Ten demonstration roads in the Netherlands and one in Belgium with a focus on the SMA top layer of roads have been performed.

3. Lignin use in the Netherlands

Since 2015 there are about 15 types of lignins tested in hot mix asphalt (HMA). Porous Asphalt (PA 16) was used as a first screening test because of the fact that this material is very prone for water sensitivity due to the open structure. The tensile strength is immediately under “attack” when non compatible materials are used in the recipe. To fulfill the Dutch specification for the Indirect Tensile Strength Ratio (ITSR) a minimum of 80% should be retained. The ITSR test was also executed on a stone mastic asphalt (SMA 8) formula with lignin in a certain percentage in the recipe. Bitumen substitutions between 30 to 50% were tested. A few types of lignin were not feasible in HMA. Very often, this was a result of impurities and organic rest material in the lignin.

In the first demonstration road, a lignin-based asphalt binder with 50% lignin / 50% bitumen was used to construct a top-layer on a road of 70 m length in the Netherlands in June 2015. This innovative asphalt binder can be processed at lower temperatures (130-140°C) compared to traditional asphalt (170-180°C). The industrial road is used every day by heavy trucks and cars, and after 4 years, the top-layer stays in excellent condition. Later on, more demonstration roads have been paved with the use of different lignin types such as soda, kraft and hydrolysis lignins.

An overview of the demonstration roads (SMA toplayers) in the Netherlands/Belgium is given in table 1.

Table 1. Demonstration roads of lignin based asphalt realised in the Netherlands and Belgium

Location	Name	Road type	Length (m)	Lignin used	Bitumen substitution (%)	Year of installation
Sas van Gent	Wervenweg	Industrial	70	soda	50	2015
Terneuzen	Europaweg	Regional	400	Kraft	45	2016
Terneuzen	Finlandweg	Industrial	100	Kraft	45	2017
Wageningen	Bornsesteeg	Cycling path	1000	Soda, Kraft, hydrolysis [18]	45	2017
Beek en Donk - Boxmeer	N272	Regional	2500	Kraft	32	2017
Oostburg	Rondweg	Regional	1000	Kraft	45	2018
Vlissingen	Schotlandweg	Industrial	500	Kraft	45	2018
Vlissingen	IJslandweg	Industrial	400	Kraft	45	2018
Zevenaar	Witte Kruis	Cycling path	500	Soda	50	2018
Gent (B)	Industrieterrein	Industrial	200	Kraft	45	2018
Goes	Joachimkade	Industrial	300	Kraft	45	2019

Lignin might have different purities, due to their extraction process (figure 3). Impurities like carbohydrates, ash and proteins can strongly influence the binder properties, compatibility with bitumen and moisture resistance.

Measurements of noise showed a reduction of 4.2 dB(A) [23] together with a reduction of rolling resistance of 3.4% [24]. On top of that, the brake deceleration with Lignin-asphalt showed 15% better performance than the minimum specified values [25].

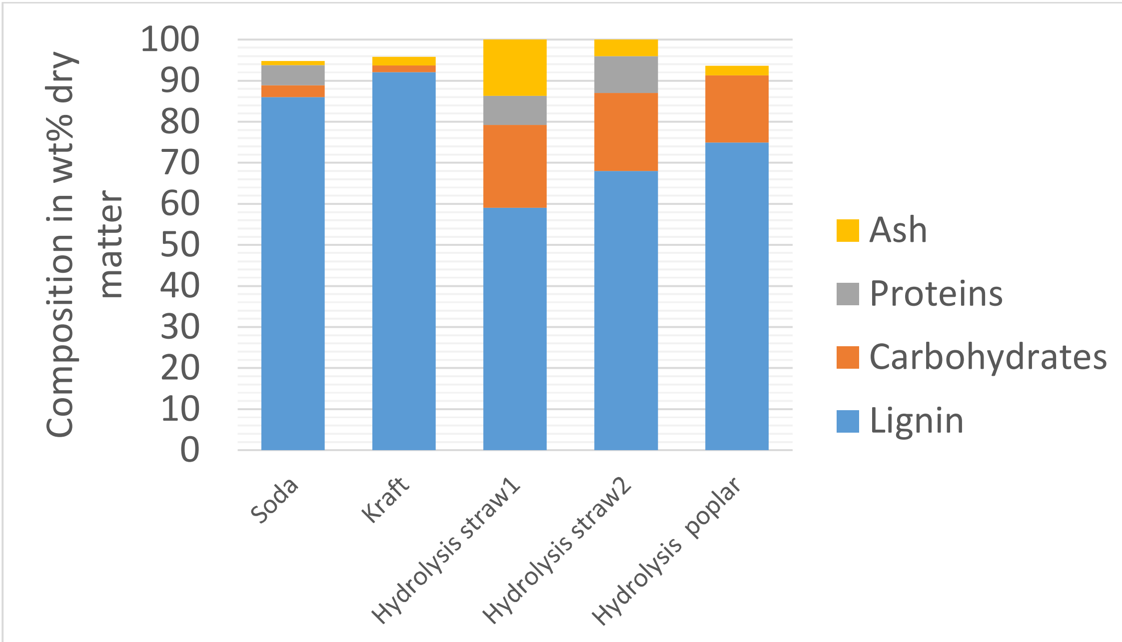

Figure 3: Composition of different lignin raw materials [17]

The results of the strength properties of different asphalt test specimens, in which different lignins are used in the binder, before and after submerging in water of 40°C during 21 days are shown in figures 4 and 5. These figures show that not all technical lignins tested meet the criteria of 80% retained strength (ITRS) for porous and stone mastic asphalt. A hydrolysis poplar lignin, most probably, loses its wet strength due to the presence of carbohydrates, mainly cellulose, and larger molecular structure.

The porous asphalt consisted of PA 16 according to the Dutch RAW standard with 5.3 % mass of binder. The binder consisted of 50% mass 70/100 bitumen and 50% mass lignin.
The SMA was formulated according to the Dutch RAW standard for SMA 8 with 6.8% mass of binder. The binder consist of 50% mass of 70/100 bitumen and 50% mass lignin.

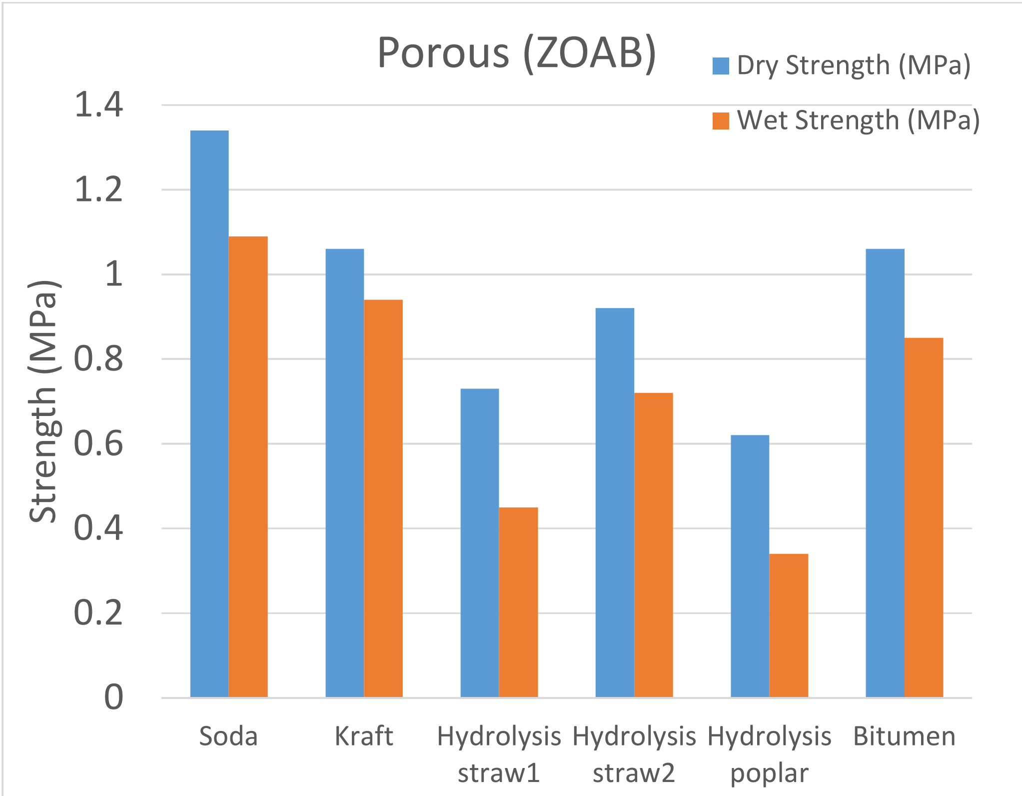

Figure 4a: Dry / wet strength of lignin-bituminous binder in porous asphalt (ZOAB).

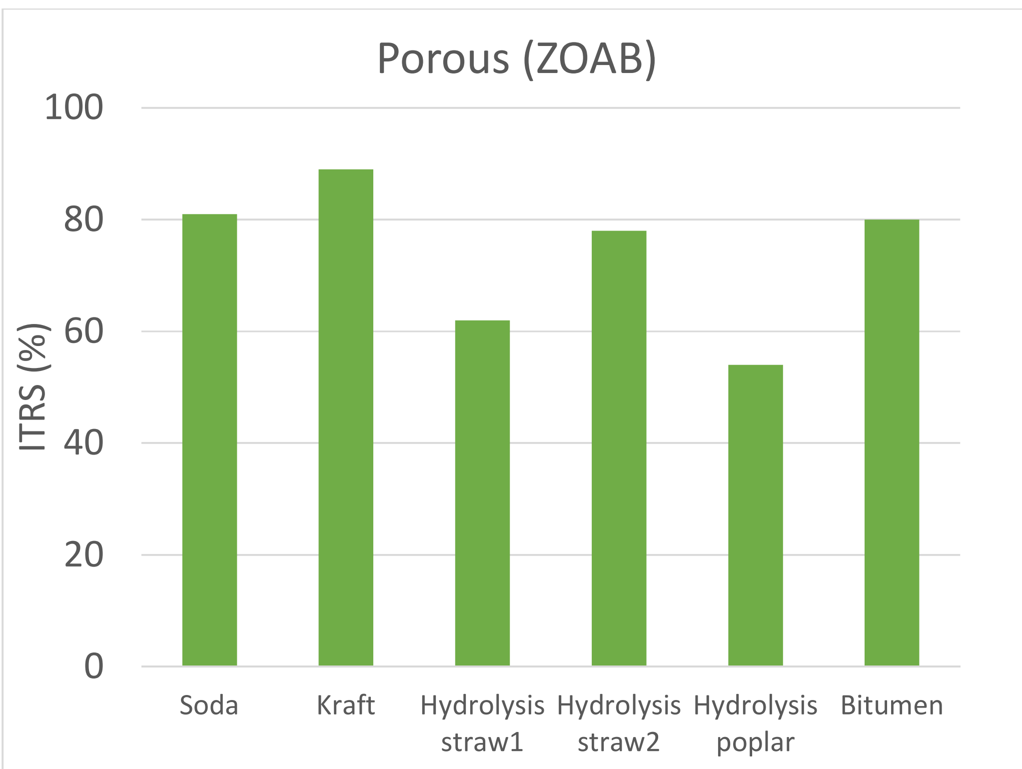

Figure 4b: ITSR of lignin-bituminous binder in porous asphalt (ZOAB).

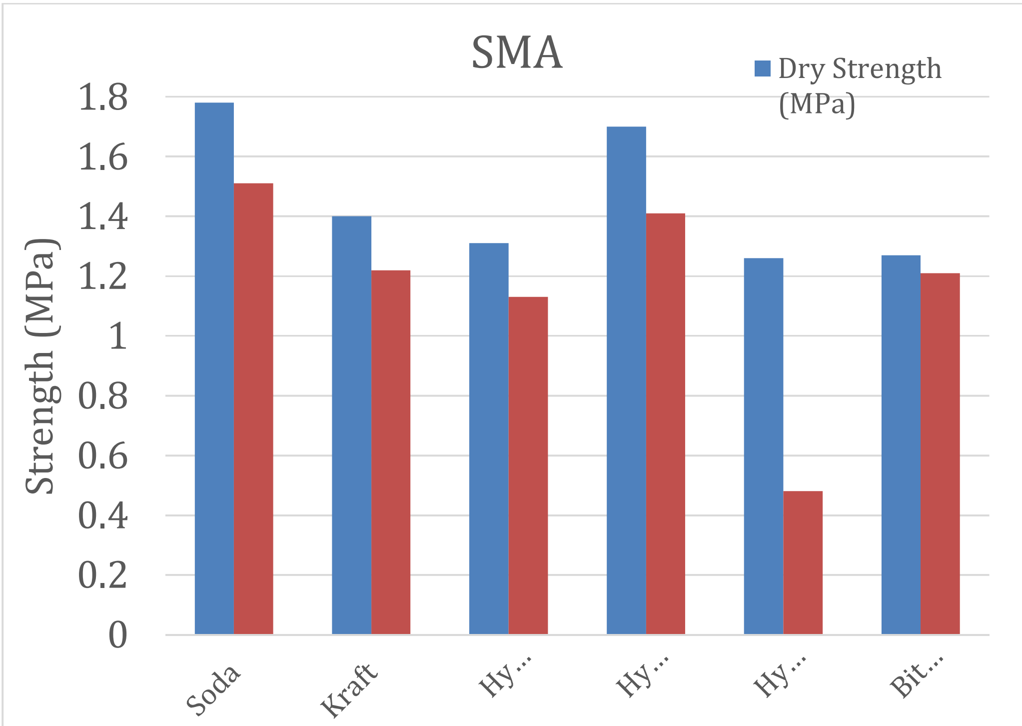

Figure 5a: Dry / wet strength of lignin-bituminous binder in dense asphalt (SMA).

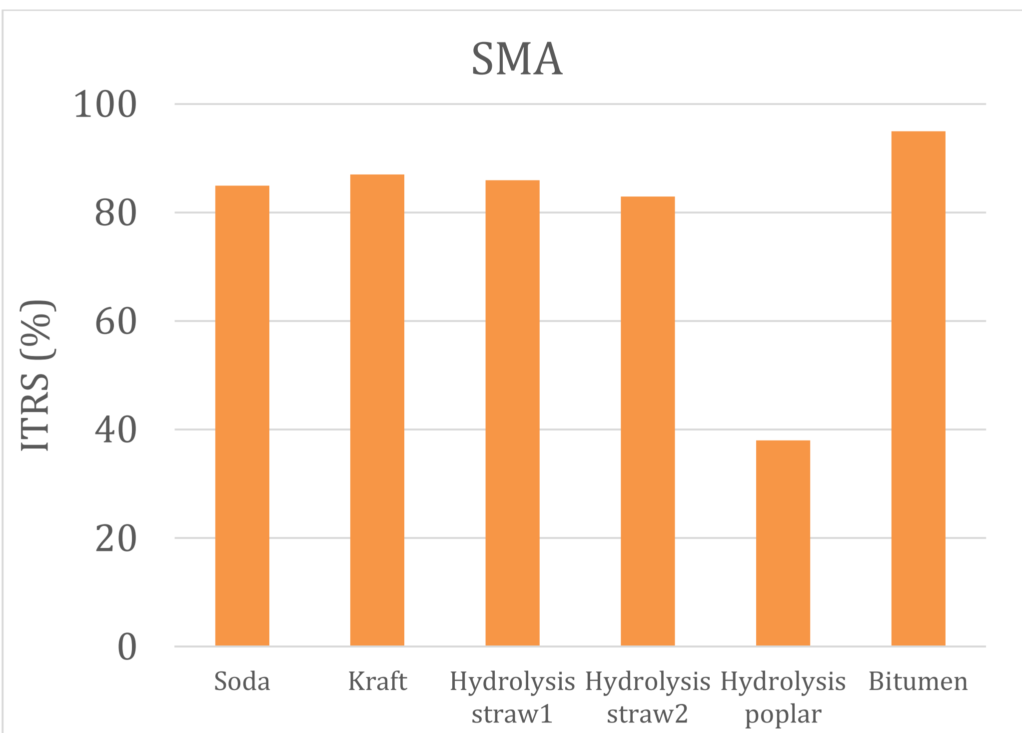

Figure 5b: ITSR of lignin-bituminous binder in dense asphalt (SMA).

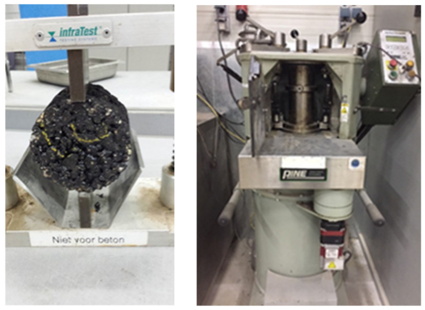

Figure 6: Test set-up of dry / wet strength and ITSR of lignin-bituminous binder

4. Environmental impact of asphalt production

One of the primary reasons to investigate the use of lignin as replacement for bitumen in asphalt is reduction of Greenhouse Gas (GHG) emissions, and ultimately switching form a fossil to a renewable feedstock for the production of asphalt. The transport sector (within the infrastructure sector) uses many fossil fuel based products, especially in the construction of asphalt roads that contain a high proportion of fossil-fuel derived products. Taking the Netherlands as a case study, traditional asphalt production is responsible for 550 kton CO_2 eq./ year; equivalent to 23% of overall GHG emissions of the infrastructure sector in the Netherlands [26,27]. Bitumen (fossil based derivative) in the asphalt mix accounts for 15% of total GHG emissions of the production of ZOAB (Zeer Open Asfalt Beton, i.e. porous asphalt) in the Netherlands [25]. Lignin, could be used as an alternative for bitumen [21,22,29] to reduce GHG emissions.

Khandelwal [31] assesses the GHG emission mitigation from lignin use in asphalt (ZOAB). They explore the use of two types of lignin:

1. Kraft lignin - Kraft is the dominant pulping process in the world, existing mills in Scandinavia are used for the input data [3,30,32]
2. Steam explosion bio refinery lignin – there are several initiatives in the EU to build lingo-cellulosic bio refineries. One of them, Biobased Delta, intends to develop a lingo-cellulosic bio refinery in the southwest of the Netherlands utilizing the steam explosion process, which is used for the input data for the calculation of GHG emission [33].

Khandelwal [31] determined the GHG emissions reduction (direct and indirect) if 50% of fossil-based bitumen is replaced with lignin (cradle-to- gate). To this end a cradle-to gate consequential life cycle analysis (CLCA) is performed following ISO14040 guidelines. The choice for a consequential LCA is based on the fact that this LCA explores the changes if lignin is introduced on a large scale to substitute bitumen in the Netherlands and the indirect substation effects it may trigger. Due to limited data and time availability, only global warming potential was investigated as impact category. The techniques used for lignin isolation investigated are the 'Ligno-boost' process (Kraft paper mill) and 'Steam explosion' technology (lingo-cellulosic bio refinery).

The research analysed two possible scenarios for assessing the GHG emissions using LCA (cradle-to-gate). In the first scenario (called baseline), a marginal amount of lignin is extracted from the Kraft pulp mill and SE bio refinery to replace fossil bitumen and the remaining lignin is incinerated to provide heat and electricity. In this baseline scenario, the existing use of lignin in pulp mills for bioenergy purposes is not changed.

In the second scenario, lignin is completely isolated in the pulp mill and the bio refinery and exported. This requires the use of additional energy for the supply of heat and electricity in the respective refinery and pulp mill, which can be met either with additional natural gas or additional woody biomass. In the calculation of GHG emission, lignin which is a biogenic product stores carbon and thus negative emissions, are allocated to it [34]. The only environmental impact considered in this research was global warming potential (GWP). The functional unit is kg CO_2 eq./tonne of ZOAB (ZOAB is the Dutch abbreviation for very porous asphalt).

Within the first scenario, the baseline case uses mass allocation, where a marginal amount of lignin is extracted for the ZOAB mix and rest is used for internal heat and power, causing no indirect effects in the bio refinery and kraft pulp mill. Energy and economic allocation were also used to evaluate the impact on the results. In the second scenario of complete lignin isolation, forest residues or natural gas were used for internal energy of the plants and the impact of different allocation mechanism (such as mass, energy and economic allocation) was explored. The study used a mix of public literature and (partly confidential) data from industry to provide foreground data. Most background data was taken from various public databases, and incorporated in SimaPro. For more details, see Khandelwal [31].

The cradle-to-gate assessment (cradle-to-gate) sums up GHG emission as 53.3 kg CO_2 eq./tonne of ZOAB (Kraft process) and 64.5 kg CO_2 eq./tonne of ZOAB (Bio refinery lignin) as compared to the 72.2 kg CO2 eq./tonne of ZOAB (ZOAB regular) for 100% bitumen based ZOAB regular. For the base cases, the reduction of GHGs for ZOAB bio refinery lignin is low (10.7%) compared to ZOAB Kraft lignin (26.2%), as lignin isolation in the Steam explosion process requires more energy than the Ligno-boost process. In addition, it was assumed that the woody biomass for the bio refinery is imported from the US to the Netherlands, whereas for Kraft lignin, import from a Scandinavian pulp mill was assumed (causing lower GHG emission in the logistic chain).

One key contributor to lower GHG emission is the storage of biogenic carbon in the asphalt, which is counted as a negative emission in the cradle to gate approach. The biogenic carbon stored in the bio refinery lignin is larger than Kraft lignin due to the carbon composition of the feedstock used in the process.

Another key aspect explored in this study is diversion of lignin from existing uses and the subsequent fuel substitution at the pulp mill / bio refinery. As seen in Figure 6, largest GHG emission reductions are achieved if the pulp mill / bio refinery uses additional biomass (forest residues). In this case, using mass allocation, a GHG emission of 52.8 kg CO_2 eq./tonne of ZOAB is the optimal solution, achieving a reduction of 26.9%.

Using other allocation methods and natural gas as alternate fuel yields lower GHG emissions reductions (see figure 5).

Figure 5 compares ZOAB regular (left) with ZOAB containing 50% lignin derived from Kraft mills and lingo-cellulosic bio refineries. The base cases assume no changes at the pulp mill/ bio refinery, the five cases on the right show the effect of fuel substitution at the pulp mill / bio refinery using different fuels and allocation methods.

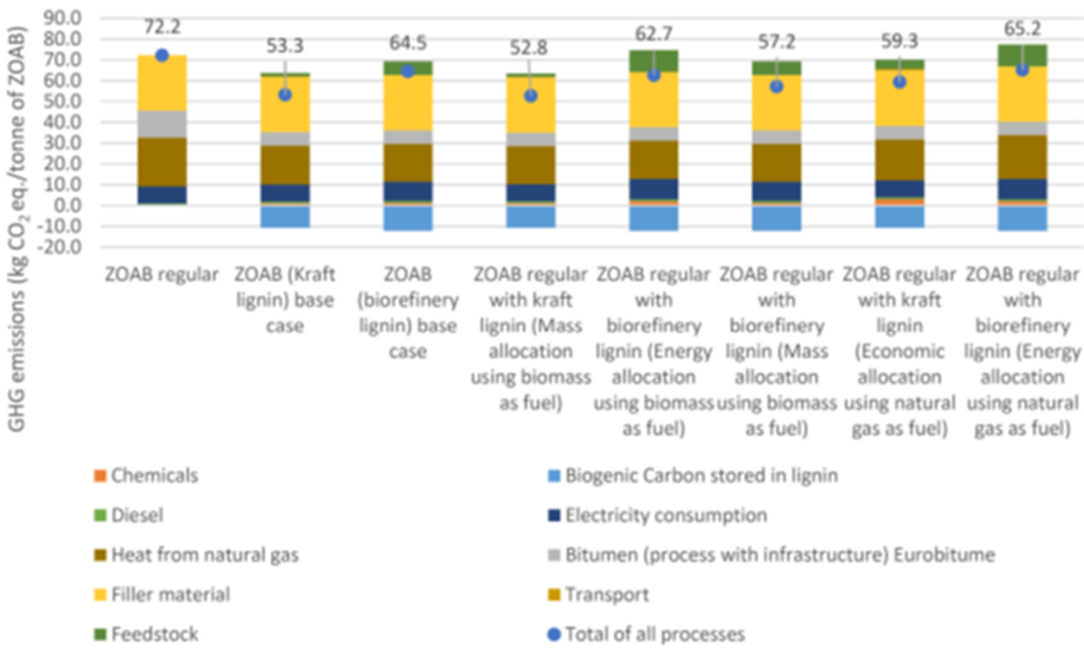

Figure 5. Cradle-to-gate comparison of ZOAB regular (left) with ZOAB containing 50% lignin. The ZOAB regular reference is based on Vos-Effting et al [35]

The results indicate that using lignin to substitute bitumen may result in promising GHG reductions. If deployed on a larger scale in the Netherlands, it could (as a rough indication) reduce overall GHG emission of the infrastructure sector by 1.3-4%. In addition, the long term storage of biogenic carbon in asphalt represents negative emissions which - if deployed at global scale – could help sequester carbon for long periods of time. However, many questions remain unanswered, such as the overall environmental performance of lignin-based asphalt (such as impacts on land use, water, biodiversity), the durability and lifetime of biobased asphalt, possibilities for recycling and other end-of-life options etc., which will ultimately significantly impact on the full cradle-to-grave results of a biobased asphalt.

5. Discussion

While the promise of constructing roads with bio-based raw materials sounds appealing, still many questions remain with regard to the benefits and potential drawbacks. Lignin can reduce the application of fossil bitumen, thus reducing GHG emissions of road construction and road use. But the source and alternative uses of lignin can have a major impact on the overall performance at the GHG-balances. As lignin is mostly used as a fuel in industrial processes, the question arises what substitution effects may occur if lignin is increasing levered for asphalt production. While there is room for further efficiency improvements in many pulp mills, use of natural gas (or coal) should be avoided.

Next to potential GHG benefits, claims exist on the reduction of noise and rolling resistance and an increase of brake deceleration. However, this is based on an individual observation, without further scientific evidence. Thus, further monitoring and analyses are required to verify these claims.

6. Conclusion

Within the last four years in the Netherlands, ten road sections with lignin as a binder in asphalt have been constructed to demonstrate that lignin can partially substitute bitumen. The total length of the sections is about 7 km. The most applied type of lignin is Kraft lignin, as it the best available type of industrial lignin. This lignin has a high purity and shows good performance in the laboratory tests and in the demonstration roads up till now. However, other types of lignin also show promise for use in asphalt applications to reduce dependence of crude oil. On the other hand, lignin application in asphalt may result in the reduction of noise and rolling resistance and an increase of brake deceleration. Why, how and when this occurs, has not yet been investigated in detail.

Based on a cradle-to-gate LCA, the maximum GHG impact for road construction [31] has been calculated as a reduction of 27% compared to fossil asphalt. The main benefit is due to the storage of biogenic carbon in the asphalt, extracting it from the atmosphere. However, GHG balances about the use and end-of-life phase (including requirements for maintenance, lifetime and possibilities for recycling of the asphalt) still lack, as do the potential impacts on health and on the environment. New road sections with lignin for further investigation are planned.

Research needs

In order to quantify the total benefits and impacts of lignin-based asphalt, a structured research program will be required. Items to investigate in this program should include:

1. A full life cycle assessment, including GHG-balances, for the logistics and processing of raw materials, the phase of using roads, maintenance and recycling. Very important is the impact on prolonging of the period for using a road, as this saves money, energy and GHG-emissions. In many production processes, like the paper and pulp industry, lignin is being used for energy generation for the process. Application of lignin for road construction will cause a shift in the energy sources for its production that need to be analysed. Also other environmental impacts should be assessed and compared with equivalent fossil asphalt, such as resource depletion, acidification, eutrophication, human- and eco-toxicity). In addition, also more fossil bitumen reference systems for a wider geographic scope and various allocation methods could be evaluated.
2. The impact of lignin based asphalt on health, safety and environment. Unknown are the emissions and leakage of substances during processing and usage. Also unknown are the impacts on braking path, slippery, icing, noise reduction and rolling resistance.
3. Lignin quality. Every plant or tree has a different type of lignin. The processes for extracting lignin (Kraft, steam explosion, soda, enzymatic hydrolysis, organosolv, etc.) also result in different types of lignin qualities and chemical composition. Kraft lignin is the most common lignin, but the potential impact of e.g. sulphur needs to be explored further.
4. Lignin volumes. What would be the impact of large-scale utilisation on the availability and prices of lignin?
5. Contractors. Lignin is a powder, while bitumen is a (very viscous) liquid. Most blenders and contractors do not have experience with lignin and need to find out how installations, processes and execution should be adjusted.
6. Fossil bitumen. The reference for road construction is fossil bitumen. LCAs, GHG-balances, the impacts on braking path, slippery, etc. with lignin-asphalt should be compared with asphalt from fossil bitumen, taking into account the types of asphalt (DAB, ZOAB), SMA, the types of roads (municipalities, highways, cycling tracks, etc.).
7. International. All over the world asphalt is being used for road construction. Every country has different processes, due to factors such as climate, raw materials, traffic and traditions. Thus, experiences gained in the Netherlands may not be transferable one-on-one to other countries.

In the Netherlands the Biobased Delta Foundation started the multi-year program CHAPLIN (Collaboration in asHalt APplications with LIgniN) to get these questions answered. Many governmental organisations, contractors, universities and research institutes join this program. As asphalt is an international building material, the Biobased Delta Foundation is interested in spreading this program over Europe and looks for interested partners to join.

ACKNOWLEDGEMENTS

This work has been funded via several initiatives and the following parties were acknowledged for their partial funding: Zeeland province, Zeeland Seaports, Ministry of Economic Affairs, Gelderland province, municipality of Wageningen, WUR, Biobased Delta.

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Annex

1. ANNEX ID0386 THE USE OF LIGNIN AS BIO-BINDER IN ASPHALT APPLICATIONS

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1.1. Extension of test tracks

The period between submitting the paper and now have seen an extension of test tracks in the Netherlands. A lot of experience has been gained in the Netherlands with lignin asphalt [1]. The first test strip on the road dates from 2015 and a lot of laboratory research has been carried out. Since that first test strip, the number of lignin asphalt strips has increased considerably. More than 20 have now been constructed: cycle paths, regional and industrial roads with different types of lignin asphalt. The overview of the test strips is listed in table 1.

Table 1. List of test tracks in the Netherlands

Location	Name	Road type	Length (m)	Lignin used	Substitution (%)	Year of installation
Sas van Gent	Wervenweg	Industrial	70	soda	50	2015
Terneuzen	Europaweg	Provencial	400	kraft	45	2016
Terneuzen	Finlandweg	Industrial	100	kraft	45	2017
Wageningen	Bornsesteeg	Cycling path	1000	soda, kraft, hydrolysis	45	2017
Beek en Donk	N 272	Regional	2500	kraft	32	2017
Oostburg	Rondweg	Provincial	1000	kraft	45	2018
Vlissingen	Schotlandweg	Industrial	500	kraft	45	2018
Vlissingen	IJslandweg	Industrial	400	kraft	45	2018
Zevenaar	Witte Kruis	Cycling path	500	soda	50	2018
Gent (B)	Industrieterrein	Industrial	200	kraft	45	2018
Goes	Joachimkade	Industrial	300	kraft	45	2019
Vlissingen	Frankrijkweg	Cycling path	300	kraft	45	2019
Vlissingen	Oostenrijkweg	Industrial	250	kraft	50	2019
Sluiskil	Piet Heinstraat	Village	150	kraft	50	2019
Ijzendijke	Turkeijeweg	Provincail	800	kraft	50	2019
Vlissingen	Maltaweg	Industrial, 3 layers!	486	kraft	35/35/50	2020
Overijssel	N765 (Kampen)	Provincial	300	kraft	30	2020
Vlissingen	Frankrijkweg	Cycling path	163	kraft	40	2020
Vlissingen	Oostenrijkweg	Industrial	562	kraft	50	2020
Vlissingen	Europaweg	Cycling path	1169	kraft	40	2020
Vlissingen	Engelandweg	Industrial	137	kraft	50	2020
Sluiskil	Industrieweg	Provincial	588	kraft	50	2020

ACKNOWLEDGEMENTS

Acknowledgements to all participants in the Chaplin program.
Governmental: Rijkswaterstaat (department of Waterways and Public works; Provence of Gelderland, -North Brabant, -Overrijsel, -South Holland; Municipalty Wageningen and –Bergen op Zoom
Companies: H4A; Dura Vermeer; NTP; Latexfalt; Roelofs Groep; Vertoro; Aventum; Pray; Boskalis; Biondoil.
Knowledge Centre: Utrecht University; Wageningen Food & Biobased Research; TNO; Q8 Research; AKC.

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