Collaboration in aspHalt APplications with LIgnin in the Netherlands - (CHAPLIN TKI)

Public report

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Contents

1 Work package 1 Lignin sources 6

1.1 Objectives & deliverables WP1 6
1.2 Methods 6
1.3 Results 6
1.3.1 Overview lignin sources in NL 6
1.3.2 Overview of lignin sources globally 7
1.3.3 Characterization of selected lignins 8
1.4 Discussion, conclusions and recommendations 11
1.5 References 11

2 Work package 2 Modification of lignin 12

2.1 Aim & deliverables 12
2.2 Methods 12
2.3 Results 12
2.4 Discussion, conclusions and recommendations 15
2.5 References 18

3 Work package 3 Asphalt testing on labscale 19

3.1 Inleiding 19
3.2 Doelstellingen en deliverables van WP 3 19
3.3 Werkwijze 21
3.4 Resultaten en discussie 21
3.5 Discussie, conclusies en aanbevelingen 28

4 Work package 4 Demonstration roads 29

4.1 Objectives & deliverables of WP 4 29
4.2 Methods 29
4.3 Results 30
4.4 Discussion, conclusions and recommendations 33

5 Work package 5 Monitoring of asphalt 34

5.1 Aim & deliverables WP 5 34
5.2 Methods 34
5.3 Results 35
5.4 Discussion, conclusions, recommendations 38

6 Work package 6 Techno-economic and Life cycle assessment of lignin-based asphalt 39

6.1 Objectives & deliverables WP 6 39
6.2 Methods 39
6.3 Results 40
6.4 Discussion, conclusions and recommendations 42
6.5 References 43

7 Work package 7 Project management, dissemination and roadmap 44

7.1 Objectives & deliverables WP7 44
7.2 Description of work 44
7.3 Results 45
7.3.1 Exploitation of CHAPLIN results & CHAPLIN roadmap for further testing of transects 47

8 Conclusions and recommendations 48

9 Annexes 49

Summary The asphalt industry is searching for durable roads for the future. With the expected scarcity of bitumen, the asphalt binder, and the quality changes, an alternative is needed to secure both the availability and quality of an asphalt binder. Lignin, as the binder in lignocellulosic biomass, could be this alternative thanks to its properties as natural binder and large availability from the pulp&paper and 2G cellulosic ethanol industries. In CHAPLIN TKI, a consortium covering the full value chain from biobased raw materials, asphalt expertise, R&D institutes and asphalt producers, worked further on the development of a lignin-based asphalt binder to bring this technology to TRL 6. In work package 1 several lignin resources have been selected to meet the criteria for an asphalt binder. These lignins were not only produced from the pulp&paper industry, but also from local biorefineries, in which lignocellulosic biomass is processed to produce carbohydrates and lignin (by Avantium and Vertoro). The selected lignins were successfully tested as asphalt binder in combination with bitumen, to substitute up to 50wt% with the AKC/WFBR technology. Blending results of the Vertoro lignin (25%) with bitumen showed however incompatible issues with the blending technology. Lignin introduces more stiffness to the asphalt binder, but could also give a stabilization effect to prevent bitumen oxidation during its life span. To further tailor the properties and compatibility the selected lignins have been modified by etherification in work package 2. In particular for one modification route the properties of the modified lignin brings the properties in a bitumen blend very close to bitumen 70-100. In work package 3 asphalt test blocks made with lignin-based binders showed good strength properties fulfilling the requirements in the Dutch situation. Also recycling of lignin-based asphalt seems possible with 25% old lignin-based asphalt in a bind (middle) layer. Additional work showed that a maximum of 50% biobased components shared between old and new binders is the limit. Release of formaldehyde was measured during production and pavement and the concentrations stay below the threshold values. This component can both be formed from bitumen as from lignin. In work package 4, selected lignins were tested in several demonstration roads of 300 meter length in Altena and Bergen op Zoom. In the road at Altena some parts were not performing best and this should be analyzed in more detail. A demonstration road in Gelderland could not be paved due to issues in the mixing of components in the asphalt mill. Unfortunately, the delivery of a hydrolysis straw lignin was delayed and could not be applied in a test section. In work package 5 the life time of lignin-based

asphalt was tested on binder and asphalt core level using traditional extraction methods such as dichloromethane. It is known that not all of the lignin dissolves in dichloromethane. Therefore the current method is not able to evaluate all of the bitumen/lignin binder. This fact should be taken into account when evaluating the life span results. For this, real core samples from existing demonstration roads were evaluated. Based on a limited set of data samples, the expected life time of the lignin-based asphalt is somewhat shorter (ca. 1.5 year) than for asphalt with a conventional bitumen binder. These results are based on the use of Lignoboost Kraft lignin. The results of work package 6 have demonstrated that significant GHG savings can be achieved if bitumen binders in asphalt are replaced with lignin. The GHG footprint of lignin-based asphalt in top-layers is between 30 to 75% lower than conventional asphalts. The current production and pavement cost of lignin-based asphalt are calculated to be 7 €/t to 17 €/t higher compared to conventional asphalt. The net difference in MKI score between lignin-based and conventional asphalt top-layers is small, and does, even in the best case, not make up for today's difference in production and pavement cost. The future scenarios are exclusively developed for surface layers in accordance with the scope of Chaplin-TKI. The overall indicative GHG savings potential is significant: 84 kt CO_2eq/yr in Best Estimate to 170 kt CO_2eq/yr in Ambitious scenario by 2050 compared to the Baseline scenario. It would come at an estimated cost effectiveness of 106 to 111€/ t CO_2eq in the Best Estimate and Ambitious scenarios respectively. Overall, the results obtained in CHAPLIN-TKI and also reflecting part of the work in CHAPLIN XL, showed that for lignin-based asphalt positive scenarios have been identified. It became also clear that this development needs further research and development to optimize the use of lignin in an asphalt binder.

1 Work package 1 Lignin sources

1.1 Objectives & deliverables WP1

In WP1 lignin sourcing the main objective is to generate an up-to-date overview of lignin sources which could be used as substitute for bitumen in an asphalt binder. Both information on local sources as international lignin sources (globally) will be collected. Furthermore characterization of selected lignin sources from the Netherlands will be performed and benchmarking against reference lignins (e.g. Kraft lignin) will be made.

Deliverables
D1.1 Report with overview of lignin sources in the Netherlands (M3)
D1.2 Report on the availability of lignin sources globally (M3)
D1.3 Report with detailed characteristics of selected lignins used in CHAPLIN (M9)

1.2 Methods

To generate the overview of lignin sources in the Netherlands and globally available are based on network information, e.g. via the COST Action CA17128 LignoCOST and literature (Mastrolitti, S. et al. 2021). De deliverables D1.1 and deliverables D1.2 have been integrated in this report under section results.

Compositional analyses of the lignins were determined for important parameters such as purity (lignin content) and impurities (such as carbohydrates, ash, proteins), molar mass distribution and functional groups to judge its suitability for the asphalt binder application. All methods applied were described by Constant et al. (2016).

1.3 Results

1.3.1 Overview lignin sources in NL

In the Netherlands several small scale biorefinery initiatives on lignocellulose fractionation have been started. Some of these processes are run at ton scale such as by Avantium (Dawn process on wood), WEPA (soda process on miscanthus) and Miscancell (soda process on miscanthus). Other initiatives are at lower technology readiness level (TRL) as displayed in the Table 1.1 below. The lignins produced in the Netherlands selected for this project are:

1. HCl hydrolysis lignin from aspen wood by Avantium
2. Steam explosion lignin from oak wood by BBD
3. Starting and processed lignin fractions by Vertoro

Table 1.1 Lignin sources in the Netherlands

Process	Company	Feedstock	Lignin production (kton/y)	Lignin isolation process	Lignin with sulphur	Purity lignin	Products	Current application for lignin
Dawn (hydrochloric acid)	**Avantium (NL)**	**Wood**	**Pilot (1 kton/y)**	**Residual**	**No**	**high**	**Sugars, lignin**	**Energy, Bio-asphalt**
Solvolysis	**Vertoro (NL)**	**Wood, lignin**	**Pilot**	**Own technology**	**No**	**Medium-high**	**Lignin oligomers, residual lignin**	**Marine fuels, PU, resins**
Steam explosion	**BBD (NL)**	**Wood**	**Lab**	**Own technology**	**No**	**Medium-high**	**Cellulose, lignin**	**Bio-asphalt**
Soda extrusion	Miscancell (NL)	Miscanthus	Pilot	Own technology	No	Medium-high	Cellulose, lignin	Bio-asphalt (grasfalt)
Soda	Wepa (NL)	Miscanthus	Pilot	Own technology	No	Medium-high	Cellulose fibres, lignin	Application under study
BIOeCON's G-2 technology	Cellicon (NL)	Wood	Lab	Residual	No	Medium-high	Cellulose, lignin	Under evaluation
Fabiola (organosolv)	ECN/TNO (NL)	Agricultural residues	Lab	Water addition	No	High	Cellulose, furans, lignin	Under evalaution
Soda, acetic acid	WFBR	Agricultural residues	Lab	Own technology	No	High	Cellulose, lignin	Application under study

1.3.2 Overview of lignin sources globally

The best available lignin globally is Kraft lignin produced by several commercial suppliers (see Table 1.2). This Kraft lignin, for this project supplied by Stora Enso, was selected as benchmark lignin and was already used in some demonstration roads in the Netherlands. The global supply of Kraft lignin is about 125 kton per year, but larger amounts are expected in the coming decades. The availability of hydrolysis lignin, from various processes, is about 20 kton/year in 2021, but this amount is expected to increase substantially (lignin producers see Table 1.3). As the process applied by Praj in India is similar to the process used by BBD, we selected steam explosion lignins from wheat and rice straw as interesting lignins for this project.

Table 1.2 Commercial lignin availability globally

Process	Company	Feedstock	Lignin production (kton/y)	Lignin isolation process	Lignin with sulphur	Purity lignin	Products	Current application for lignin
Kraft	**Ingevity (US)**	**Pine**	**60**	**Own technology**	**Y**	**High**	**Cellulose, sulphonatedlignin, purified Kraft lignin (Indulin AT™)**	**After sulphonation in cement, Composites, resins, in batteries**
Kraft	**Stora Enso (FIN)**	**Softwood**	**50**	**Ligno-Boost**	**Y**	**High**	**Cellulose pulp, Kraft lignin (Lineo™)**	**Bio-asphalt, batteries, PF resins, others under evaluation**
Kraft	Domtar (Can)	Softwood	25	Ligno-Boost	Y	High	Cellulose pulp and lignin (BioChoice™)	Evaluation ongoing
Kraft	West Fraser (CAN)	Softwood	10	LignoForce System™	Y	High	Cellulose, Kraft lignin	PF resins for plywood, others under evaluation
Sulphite	Borregaard (NO) TEMBEC (CA, FR, US), Domsjö Fabriker (SE) La Rochette Venizel (FR), Nippon Paper Chemicals (JPN)	Softwood, hardwood	1000	Own technology	Y	Low	High purity cellulose pulp (dissolving pulp), bioethanol, vanillin, lignosulphonates	In cement, asphalt, agrochemicals, resins
Soda	**Greenvalue Ltd (US)**	**Straw/grass**	**5-10**	**LPS system**	**No**	**High**	**Cellulose pulp, soda lignin (Protobind)**	**In resins, cement, antibiotics, feed**

Table 1.3 Lignin production at demonstration and pilot scale globally

Process	Company	Feedstock	Lignin production (kton/y)	Lignin isolation process	Lignin with sulphur	Purity lignin	Products	Current application for lignin
Kraft	Suzano (BR)	Eucalypt wood	Demo (20 kton/y)	LignoBoost	Yes	high	Cellulose, Kraft lignin Ecoligtm	Resins, rubber, thermoplastics
Cellunolix®	St1 (FI)	Wood residues	Demo (15 kton/y)	Own process	No	Medium	Ethanol, lignin	Energy
Kraft	LignoCity/RISE/Nordic Paper (SE)	Wood	Demo (8 kton/y)	Ligno-Boost	Yes	High	Cellulose pulp, lignin	Various under research
Enfinity	**Praj (IN)**	**Agricultural residues (e.g. straw)**	**Demo**	**Own process**	**No**	**Medium**	**2G ethanol, biogas, lignin**	**Various under research**
SUNLIQUID®	Clariant (DE)	Agricultural residues	Demo, building plant (RO)	Own process	No	Medium	Ethanol, lignin	Energy
Steam/acid	SEKAB (SE)	Wood	Demo	Own process	No	Medium	Ethanol, lignin, acetaldehyde	Energy, others under evaluation
Organosolv (formic acid)	Fortum/Chempolis (FI)	Straw, wood	Pilot / building in India	Own process	No	high	Cellulose, lignin	Under evaluation
Sweetwater's Sunburst	GraanulInvest	Wood	Demo building phase	Own process	No	high	Cellulose, lignin	Under evaluation
Own technology	UPM (DE)	Wood	Pilot / plans for demo	Own process	No	unknown	Cellulose, lignin	Under evaluation

1.3.3 Characterization of selected lignins

In Table 1.4 all lignins studied in this WP are displayed. Praj lignins, Avantium lignin and Renmatics lignin can be produced at ton scale, but for the CHAPLIN-TKI project only several kg amounts sufficient for the tests in WP1-3 have been delivered. Stora Enso lignin is available in large quantities for the project.

Table 1.4 Selected lignins

code	supplier	origin	process
PR20	Praj (India)	rice straw	steam explosion
PW20	Praj (India)	wheat straw	steam explosion
SE20	Stora Enso (Finland)	softwood	Kraft
B1O20	BioBased Delta	hardwood oak 1	steam explosion
B4O20	BioBased Delta	hardwood oak 4	steam explosion
VR20	Vertoro / Renmatix	hardwood	SC water hydrolysis#
VH20	Vertoro	high Mw fraction	solvolysis
VO20	Vertoro	oligomers	solvolysis
AV20	Avantium	aspen wood	acid (HCl)

Supercritical water hydrolysis process to fractionate hardwood

Avantium Dawn Technology lignin

The production of HCl lignin takes place at the Avantium Dawn Technology Pilot Plant in Delfzijl, the Netherlands. A schematic overview of the Dawn process is shown in figure 1.1. The Bergius-Rheinou process forms the basis for the Dawn Technology, in this process concentrated hydrochloric acid is used for the hydrolysis of lignocellulosic biomass. In the Dawn pilot plant, this hydrolysis takes place in two steps, in the first steps the biomass is contacted with a 37% HCl solution, resulting in hydrolysis of the hemicellulosic fraction of biomass and the production of a mixed sugar fraction. In the second hydrolysis step, a 42% HCl solution is used, resulting in hydrolysis of the cellulose to form pure glucose. The sugars exit the reactor together with the acid solution, leaving behind the solid lignin fraction of the biomass. The lignin is then washed with water and stored in drums.

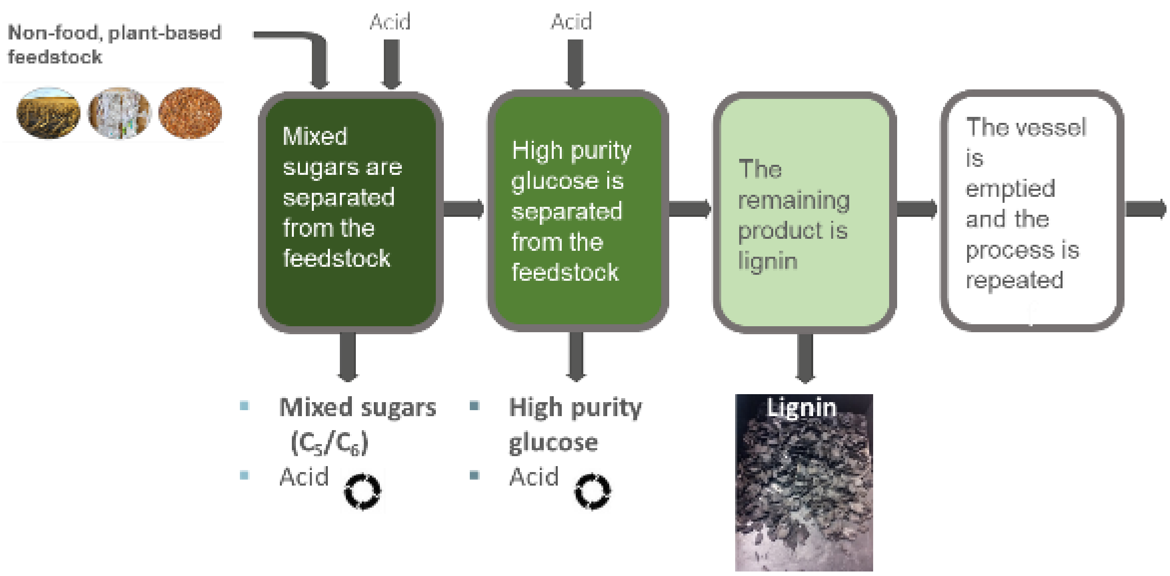

Figure 1.1: Production of HCl lignin in the Dawn Technology process

The Dawn Pilot Plant contains of a series of reactors that operates in continuous batch mode. Each reactor is loaded with 30 kg of wood chips and will eventually produce around 10 kg lignin. For Chaplin-TKI, one reactor load of Aspen lignin was dried and ground for characterization (WP1), modification (WP2) and lab scale asphalt testing (WP3).

Vertoro's GOLDILOCKS® technology

Vertoro has developed a patented Goldilocks® technology, which enables to transform all types of lignin into a solvent-soluble and melt-flowable lignin intermediate with interesting properties for down-processing, called crude lignin oil (CLO). Goldilocks® involves an (acid) thermal solvolysis technology for fractionation of lignin into oligomers. Uniquely, the chemical characteristics of Goldilocks® are insensitive to feedstock variability (i.e. feedstock agnostic), meaning that multiple feedstocks can be sourced simultaneously, bringing down the average cost, and the risk of supply shortages. Goldilocks® process can be applied to both technical lignin and lignocellulosic biomass with only a small adaptation of the process. Technical lignin is subjected to a mild (<200°C) solvolysis with low residence time (<30 minutes) and this effectively fractionates lignin into two distinct product streams, char and Goldilocks®, comprising high and low molecular weight lignin fragments, respectively. The same solvolysis chemistry is employed, but under mildly acidic conditions for lignocellulosic biomass. The acid, typically sulfuric acid, is here required to liberate lignin from the lignocellulosic matrix. Instead of char, now cellulose is the solid residual stream alongside Goldilocks®, which is separated through an additional purification step.
For the developments of this project, Vertoro processed technical lignin sourced from Renmatix. A methanolic crude lignin oil fraction was obtained from Renmatix lignin, by solvolysis in MeOH in a pilot 300L batch reactor available in Brightlands Chemelot Campus. 8.4 kg of dried and grinded Renmatix lignin was loaded in the 300 L reactor and mixed with 126 kg of methanol (1:15 w/w). The solvolysis was carried out at 200 °C, 10 bar of nitrogen gas for 30 minutes. After the end of the reaction, the product mixture was filtered in a pilot filter, and the lignin residue (high molecular weight lignin) was washed with fresh methanol to recover all the Goldilocks® fractions. After the separation step, the lignin oil was further concentrated to a ratio of 1:5 w/w in the reactor. As final step, the concentrated lignin oil was precipitated in water and dried in order to isolate the solid Goldilocks® fractions. These fractions were supplied to partners for downstream developments.

BBD lignin samples

As part of the "Redefinery program" BBD has performed biorefinery experiments at BPF in Delft. As a result of these experiments 2G sugars and lignin samples were obtained for further evaluation. The lignin samples were used for the asphalt experiments in the CHAPLIN-TKI project. The details of the BPF trails are described in a separate report. Since BBD discontinued the Redefinery program no further action has been taken on these samples, they do however provide useful insights into the performance of these hardwood based steam explosion refinery lignin samples.

Praj

The Praj lignin samples have been provided by Praj Industries from India. Praj operates a continuous process pilot facility which was used to create these samples from wheat- and rice-straw (Figure 1.2). See below for the schematic overview of the process. The samples were prepared based on the specification provided to Praj based on our experiences with lignin based asphalt. Based on the promising progress with the pilot facility Praj is working on multiple projects to realize a demo scale biorefinery. Once these are operational larger quantities of this lignin type will become available to the market. As a next step Praj is now preparing a 2 ton sample for the CHAPLIN program because the outcomes of the sample evaluation are very promising.

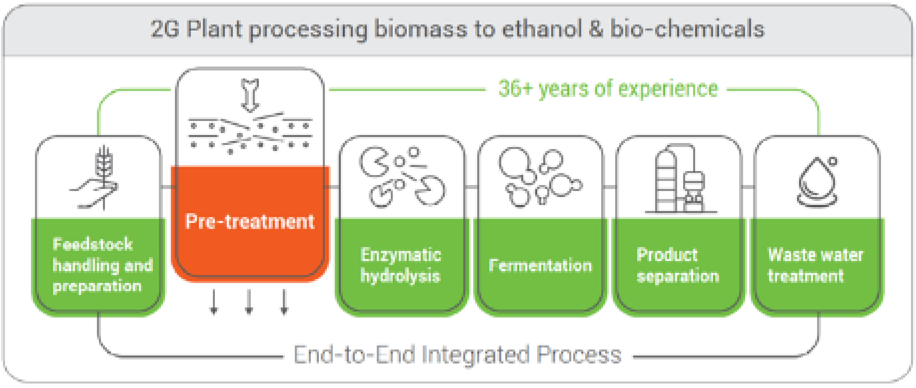

Figure 1.2 Biomass processing to cellulosic ethanol and lignin operated by Praj

The different characteristics of all lignins are presented in Table 1.5. For some lignins the solubility in the solvents used for the characterization of the molar mass and functional groups is rather limited and only reflects the solubilized part of the lignin raw material. Except for steam explosion oak lignin (BBD4) and the high molecular weight fraction of the processed lignin by Vertoro (VH20) all lignins have a high purity over >80%. Based on the characteristics of all lignin displayed in Table 1.5 a selection is made for 4 lignins (Table 1.6), which potentially fulfil the requirements for an asphalt binder and these are further tested in WP2 (modification) and WP3 (asphalt tests).

Table 1.5 Characteristics of various lignins

supplier	source	lignin (%) > 60%	impurities (%) low	moisture (%) < 10%	OH (mmol/g) high	Mw (Da) <5000	PD low	cap high
Praj	rice straw	89	20	7	4.3	5956	7	+
Praj	**wheat straw**	92	13	6	4.5	10390	10	+
SE	**softwood**	94	3	5	6.5	3544	5	+
BBD1	oak	80	16	3	5.1 *	9252 °	10	-
BBD4	oak	62	32	4	4.4 *	12852 °	15	-
V R	**hardwood**	83	14	1	4.6	9436 °	11	+
V H	hardwood	66	31	3	28	2479 °	4	-
V O	hardwood	90	1	2	5.2	2152	4	-
AV	**aspen**	90	4	9	3.8 *	4128 °	6	-

* does not dissolve very well in DMF / pyridine / CDCl3

° does not dissolve very well in 500mM NaOH

\+ potentially high capacity; - low capacity

Da Dalton = g/mol

PD polydispersity (Mw/Mn)

Table 1.6 Selected lignins which fulfill the requirements for application as asphalt binder

supplier	source	lignin (%) > 60%	impurities (%) low	moisture (%) < 10%	OH (mmol/g) high	Mw (Da) <5000	PD low	capacity high
Praj	straw	92	13	6	4.5	10390	10	+[A]
Stora Enso	softwood	94	3	5	6.5	3544	5	+
Vertoro R	hardwood	83	14	1	4.6	9436	11	-[B]
Avantium	aspen	90	4	9	3.8	4128	6	-[C]

A = Lignin produced at ton scale, but in kg amounts available in this project; B=Lignin from Renmatics at ton scale, but in kg amounts available for this project; C=Acid lignin produced at tonnes scale, but from another feedback. This aspen lignin was available at kg scale.

1.4 Discussion, conclusions and recommendations

Based on the characteristics, such as lignin purity, molar mass distribution, functional groups and (potential) availability several lignins are interesting to pursue as part of an asphalt binder. Lignins selected for further study as asphalt binder are Dawn wood lignin, produced by Avantium, steam explosion straw lignin by Praj, hydrolysis lignin by Vertoro and Kraft lignin by Stora Enso. Processed lignin oligomers, produced by Vertoro, will be further evaluated in the extension of this project.

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2 Work package 2 Modification of lignin

2.1 Aim & deliverables

The aim of this work package is to evaluate chemically modified lignins as partial substitutes for bitumen. Through chemical modification it is possible to adjust lignin properties in such a way that the resulting lignin product is more compatible with the chemical structures present in bitumen. This work package has two objectives, 1) Study the effect of chemical modification of lignin on the properties of the asphalt binder blend compared to regular asphalt binder and 2) Study selected modified lignin/bitumen blends in asphalt test blocks. the lignins, that are modified have been selected in work package 1.

2.2 Methods

The chemical modification of the chosen lignins from work package 1 have been carried out according to methods described in patent (WO 2015/137813). The chosen lignins are substituted under alkaline conditions with glycidyl ethers and acetic anhydride. The resulting mixture is dialyzed against tap water and lyophilized. The resulting modified lignins have been used for blending in bitumen with hardness 70/100 (typical hardness bitumen that is used in The Netherlands for road construction). The modifications have been caried out using the following reactants: phenyl glycidyl ether, allyl glycidyl ether, ethyl hexyl glycidyl ether, and acetylation. Several degrees of substitution have been evaluated on lab scale and it turns out that a degree of substitution of around 0.2 calculated on the amount of hydroxyl groups present in lignin seems to be the optimum. The modified lignins have been blended in 70/100 bitumen according to WO 2015/137813. The total acid number (TAN) of the base bitumen is found to have an influence on the blending circumstances: compatibility of the modified lignin with the base bitumen and on the rheological properties of the resulting blends. Bitumen processed from a naphthenic crude source has a relative high total acid number (TAN) when compared to earlier experiments using 70/100 bitumen processed from paraffinic crudes, which has a significantly lower TAN number. However the modified lignins are successfully blended into the naphthenic bitumen at 25% (w/w). Viscoelastic measurements using dynamic shear rheology (DSR) show that the blended lignins in bitumen 70/100 increase the viscosity of the residual blend and that the ethyl hexyl glycidyl modified lignin is able to substitute bitumen at 25% (w/w) with a slight increase in viscosity. Apparently this specific modification enhances the compatibility of the modified lignin with the naphthenic bitumen. This is the first time that this chemically modified lignin is successfully produced. Based on blending experiments the phenyl glycidyl ether lignin have been scaled-up to 2 kg for experiments in asphalt blocks. A second experiment is foreseen once the extension is approved whereby a 2 kg batch of ethyl hexyl glycidyl lignin will be produced and used for lab scale asphalt experiments. The asphalt experiments are performed using the technology described in patent application WO 2019/092278. The reason for this choice is that this technology has already been applied on larger scale both in the asphalt mill and as surface layer on test roads.

2.3 Results

The chemistry obtaining the substituted lignins as described above is successful and is such that up-scaling to pilot and industrial scale feasible is. Repeatability of the chemistry has been shown. For lab purposes the amount of modified lignins have been produced at 300 gram scale. For the asphalt tests a modified lignin (phenyl glycidyl ether) using lignoboost Kraft has been produced at 2 kg scale. The choice for lignoboost Kraft is due to the industrial scale availability for this type of lignin. An extension of this project has been requested and during this period a second modified lignin (ethyl hexyl glycidyl ether) on a 2 kg scale is foreseen which can be tested in lab scale asphalt experiments using the technology described in WO 2019/092278 and in WO 2015/137813 .

Blending of these modified lignins are also successful. Using the blending lab equipment at the bitumen lab in TNO all the blends have been made and analyzed. Rheological behavior show that the blended lignin/bitumen blends show a higher viscosity when compared to the blank bitumen with the exception of the ethyl hexyl glycidyl modified lignin, which almost coincides with the blank(base) naphthenic bitumen. Further all the blends with modified lignin follow the same curve as the blank bitumen indicating predictable viscoelastic behavior when applied in asphalt. The modified lignin however do not show polymer modified bitumen behavior in the naphthenic bitumen. Earlier such behavior has been identified in the paraffinic bitumen (WO 2015/137813). Polarized microscopy show that the modified lignins do show some scale of dispersion (indicating that part of the lignin fraction does not integrate in the bitumen matrix. The dispersion relates to the earlier mentioned increase in viscosity as determined with DSR. Compatibility of lignin or modified lignin also depends on the ratio of components in bitumen. The components in bitumen are characterized via measuring their SARA contents (Saturates, Aromatics, Resins, and Asphaltenes). Since bitumen consists on countless individual components measuring the SARA content is the suitable way of characterizing bitumen.

This work package carried 4 different deliverables. They are discussed individually

D2.1: production of at least 4 different chemically modified lignins on 0.5 kg scale.
During the course of the project it appeared that batches of 300 grams did satisfy the follow up blending and analysis of the lignin/bitumen blends. Therefore this deliverable has been reached.

D2.2: production of at least 4 different blends of chemically modified lignins and bitumen on lab scale.
During the project several modified lignins have been synthesised varying in both degree of substitution as type of substitution. As earlier mentioned a degree of substitution of around 0.2, calculated from the amount of hydroxyl groups present in lignin, seems to be the optimum. These lignins have been blended in bitumen and at least 4 different blends have been produced. Therefore this deliverable has been reached. To assess the compatibility of the base bitumen with lignin, the sources of the base bitumen PEN 70/100 were varied. The consistency of the blend is then correlated with the chemical properties of the base bitumen like its acidity (i.e. total acid number) and the molecular fractions (i.e. SARA). The rheological and morphological properties of these blends are characterized using Dynamic Shear Rheometer (DSR) and optical microscope. The results are presented below.

Total acid number (TAN) of the base bitumen
Acidity of bitumen is determined by the Total Acid Number (TAN) using ASTM D664 – 18en. TAN values of bitumen 70/100 obtained from different sources are presented in Figure 2.1. According to the TAN value Bitumen A is the least acidic bitumen whereas Bitumen D is the most acidic. In the case of phenyl glycidyl ether modified lignin, it is easily mixed with bitumen of lower acidity (TAN < 0.8), but a coagulation during mixing is observed with higher acidity bitumen.

Molecular composition
Molecular fractions SARA are determined using Iatroscan, a thin layer chromatography–flame ionization detector (TLC–FID) technique and the results are presented in Figure 2.2. Composition of bitumen is found to have a correlation with the compatibility of modified lignin to the base bitumen. For phenyl glycidyl ether modification, a high resin content in bitumen facilitated the compatibility of lignin into bitumen.

Micrographs of the lignin- bitumen blends
Compatibility of lignin with different chemical modification and the base bitumen is assessed through microscopic study. Figure 2.3 presents the micrographs of the Kraft- lignin bitumen blends where the lignin is modified with different functional groups. The micrographs show lignin micro-phase dispersed

in a continuous binder phase. The continuous phase is composed of the bitumen and probable soluble components of the lignin blended into it. From the micrographs it is apparent that the chemical modifications didn't result in any additional phase-separation. Ethyl hexyl glycidyl ether shows better dispersion of the lignin phase.

Rheological properties of the blends
25wt.% of modified lignin- bitumen blends are prepared where the chemical modifications and the base lignin are varied. Figure 2.4 presents the viscoelastic response properties of the blends of phenyl glycidyl ether modified lignin where the base lignins are obtained from different processes. The blends show higher shear modulus compared to the reference base bitumen PEN 70/100. This modified lignin is found to act as a viscosity building component as seen in Figure 2.4(a).
All blends show viscoelastic response comparable to pure bitumen at a broad range of frequencies as shown in Figure 2.4(b). At high frequency range, lignin- bitumen blends show more elastic response compared to the reference base bitumen. Modified kraft lignin shows more elasticity at a low frequency range (i.e. high temperature range) and more flexibility at a high frequency (i.e. low temperature range) compared to the base bitumen and also other blends. These properties are characteristic to polymer modified bitumen (PmB) and are desirable as a binder for asphalt applications.
Figure 2.5 presents the viscoelastic response properties of the ethyl hexyl glycidyl ether modified kraft lignin-bitumen blend. Here the modification has introduced a shift towards lower modulus where the master curve of the blend overlaps with the base bitumen: showing more binder-like properties.

D2.3: production of at least 4 different asphalt varieties using the chemically modified lignin/bitumen blends on lab scale.

In this project one type of asphalt using a modified lignin (phenyl glycidyl lignin)/ bitumen blend has been made and characterized. Therefore this deliverable has partly been reached. In the meantime an extension has been requested where a second asphalt using ethyl hexyl glycidyl modified lignin/bitumen will be made and characterized. Therefore this deliverable has partly been reached.

D2.4: testing of lab scale produced asphalt with modified lignin on strength before and after emerging under water (ITSR and SCB results).
Several asphalt mixtures have been tested on strength before and after emerging under water as reported in Chapter 3 (section 3.4). A second mixture on ethyl hexyl glycidyl modified lignin was tested during the extension period by TNO and AKC.

Evaluation of Vertoro lignin at the binder level with the blending technology of TNO
<u>As an additional task to CHAPLIN-WP2, Vertoro lignin was planned to be evaluated as a potential binder for asphalt application. Following activities were planned within the task:</u>

- Preparation of a Vertoro lignin- bitumen blend having 25 wt.% lignin content
- Evaluation of visco-elastic properties by obtaining the mastercurves of the blend using Dynamic Shear Rheometer (DSR)
- Microscopy on the blend to assess the phase behaviour

Preparation of lignin and the blend:
Grinding and milling of lignin
The physical state of Vertoro lignin was solid, mix of powder and granular particles and dried. The lignin-granules needed to be grinded or milled prior to addition into the base bitumen. Initially grinded using mortar and pestle but couldn't result in particle-size that can pass through a 0.5 mm sieve. Later the lignin was milled using a milling device to obtain finer particles. Lignin fraction passing 0.5mm sieve are added into bitumen. Figure 2.6 presents the preparation on lignin and the blending set-up.

Preparation of the blend
Bitumen was pre-heated and the blending was performed at 170°C. Base bitumen of PEN 70/100 was used for this study. A total content of 25 wt.% lignin was added into bitumen. Lignin was added in few steps to facilitate better dispersion and at high shear with the rotational speed ranging from 1000 to

1200 rpm. Then the mixture was blended with high shear mixer at 1200 rpm for 5 min followed by using a low-shear mixer at 225 rpm for 30 min.

Phase separation in the blend
After the blending procedure, the blend was checked for its homogeneity and consistency. The blend was poured into another can through a sieve to check for any sediments or residue. Larger agglomerates were found as retained in the sieve while decanting and there were residual agglomerates in the can as seen in Figure 2.7. This demonstrates an incompatibility of the Vertoro lignin and bitumen resulting in phase separation of lignin and bitumen phases. Due to this phase separation, further rheology (DSR) and microscopy measurements were not carried out.

2.4 Discussion, conclusions and recommendations

This project has shown that the source of the bitumen (both geographically and crude oil refinery process) has a large influence on properties of the blends e.g. with and without a polymer effect. It is expected that in the near future several more dedicated crude oil refineries for the production of bitumen will close. The result will be an ever increasing variation in quality of bitumen becoming available for the asphalt industry. Lignin or modified lignin could play an important role in the future with regard to balance quality of bitumen.

At the end of the project the number of asphalt mixtures did not reach the amount that has been anticipated while writing the project proposal. The reason is that more time has been put into searching for the optimal synthesis, degree of substitution versus rheological behavior. In the end however one type of chemical modification (ethyl hexyl glycidyl ether) does show promising viscoelastic results. This type of modification was tested in lab scale asphalt tests by AKC in March/April 2022 via the extension of the project.

The earlier mentioned relation between the ratio of bitumen components (SARA and TAN) and type of lignin or modified lignin needs to be investigated. The result will be that different lignins will become available for upgrading bitumen to specs suitable for asphalt production.

Next steps in this research is that the current modification technology should be scaled up to pilot scale (e.g. 1 ton) and that at the location of bitumen blenders (e.g. Latexfalt) pilot blends should be produced for tests stretches for road top layers.

With regard to the technology described in WO 2015/37813 first experiments on top asphalt layers are foreseen. With regard to the technology described in WO 2019/092278 a next step would be to construct a 2 km road asphalt top layer for a more extensive testing of reduced fuel consumption and noise reduction. Also this technology should be upgraded to a TRL level of 9 and introduced in other countries.

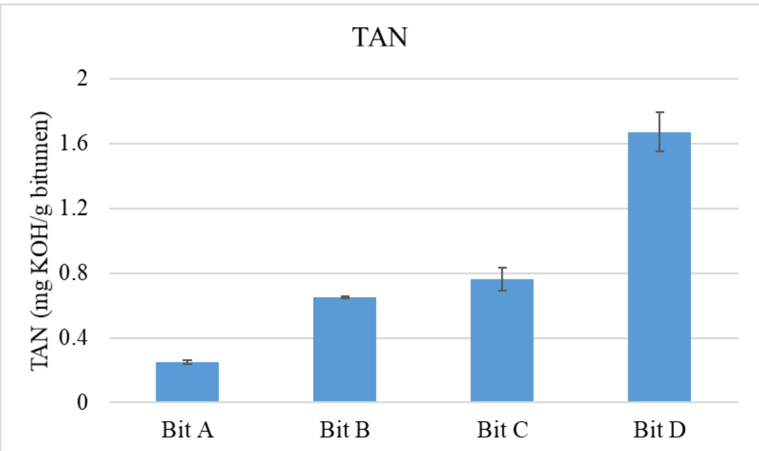


Figure 2.1: Acidity of 70/100 bitumen from different sources.

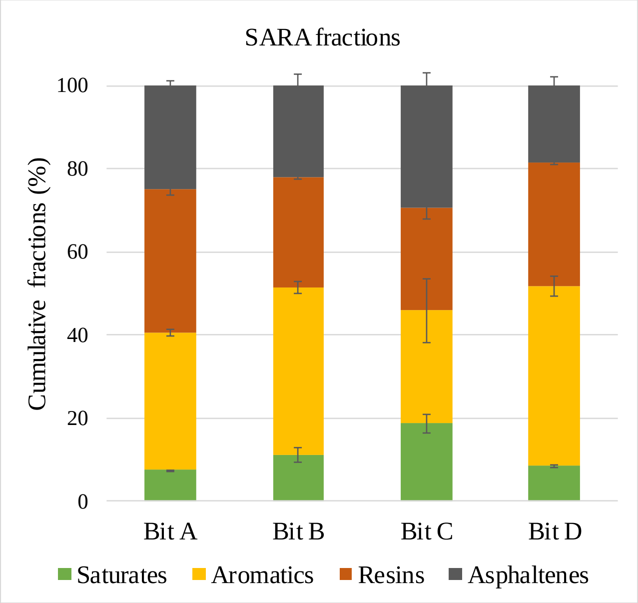


Figure 2.2: SARA molecular fractions of 70/100 bitumen from different sources.

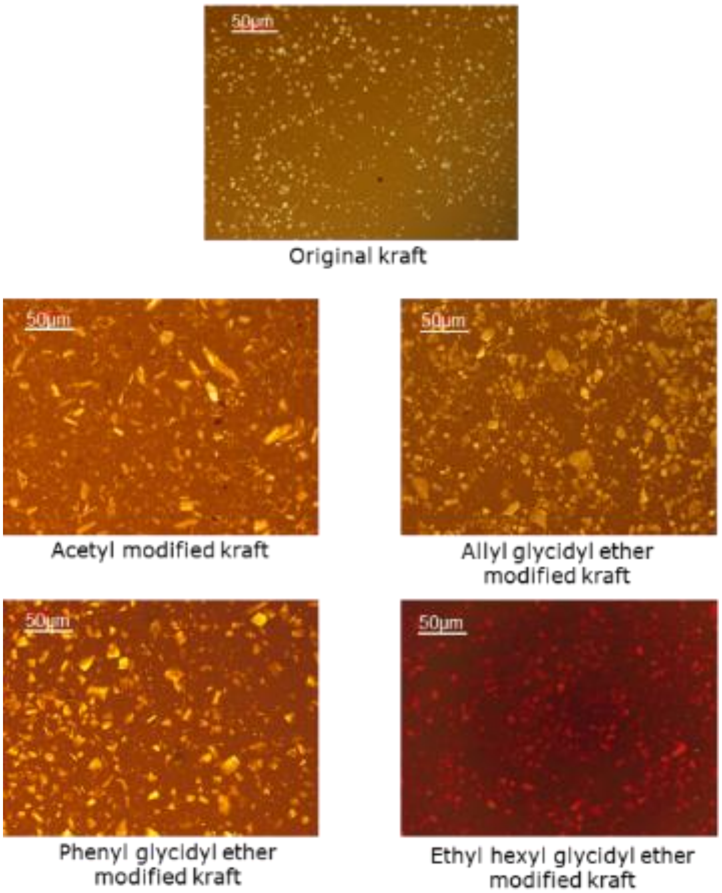
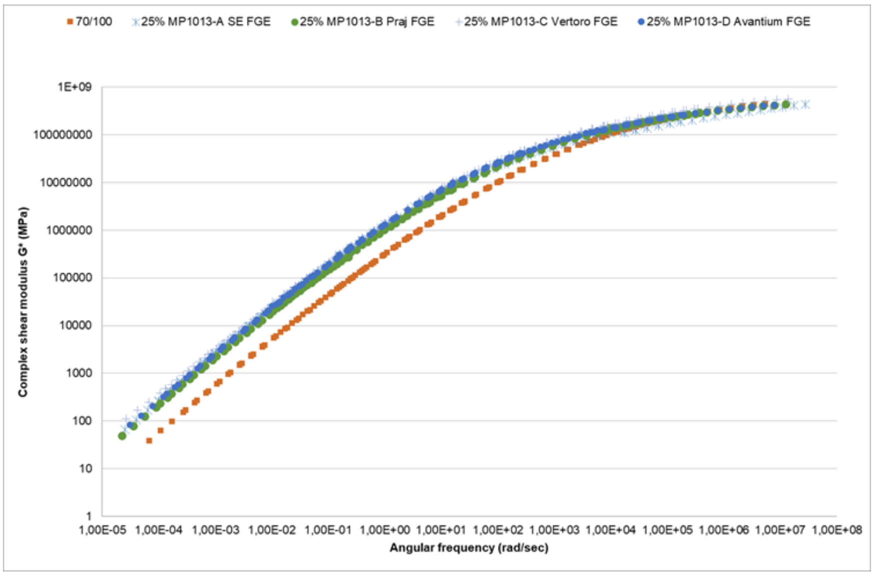


Figure 2.3: Micrographs of 25 wt. % kraft lignin-bitumen blends with different modifications.

(a)



(b)

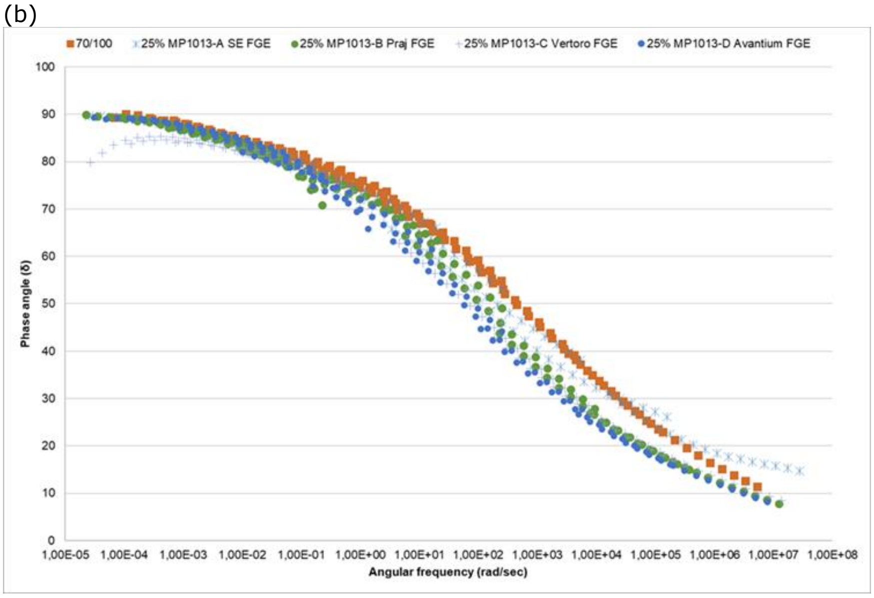
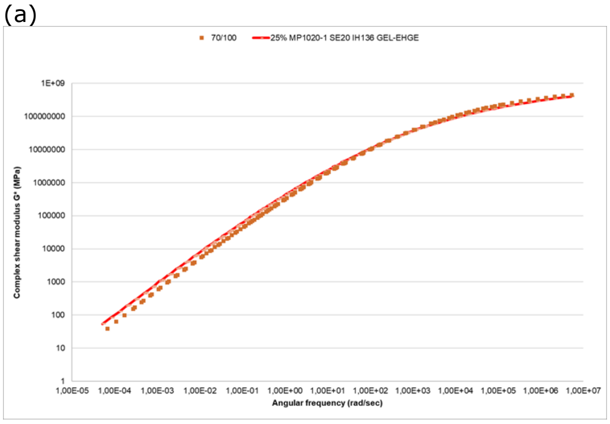


Figure 2.4: Mastercurves of 25wt.% phenyl glycidyl ether modified lignin-bitumen blends as a function of frequency. (a) Complex shear modulus (b) Phase angle.

(a)



(b)

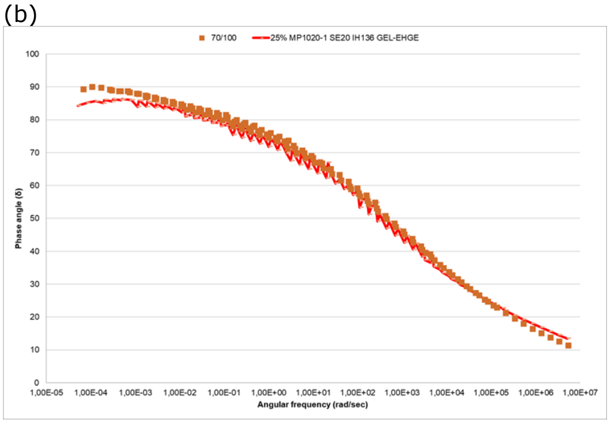


Figure 2.5: Mastercurves of 25wt.% modified ethyl hexyl glycidyl ether modified kraft lignin-bitumen blend as a function of frequency. (a) Complex shear modulus (b) Phase angle.

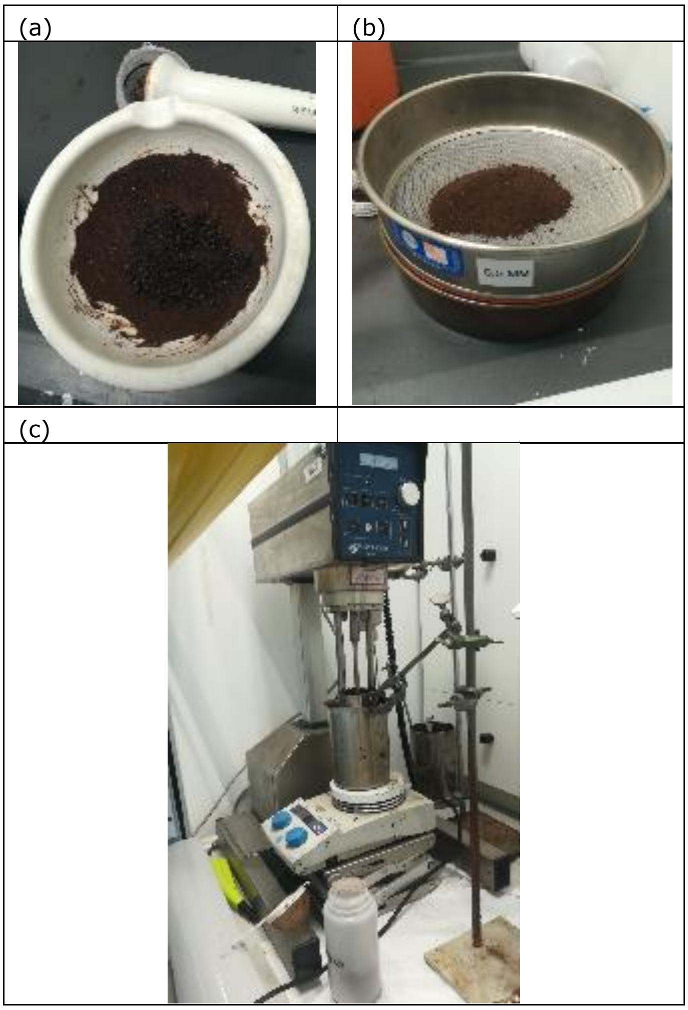


Figure 2.6: Lignin and blend preparation. (a) Granular lignin is initially grinded using a mortar and pestle, (b) Lignin particles passing 0.5mm sieve are added to bitumen, (c) High shear mixer assembly that was used for blending in combination with a low shear mixing.

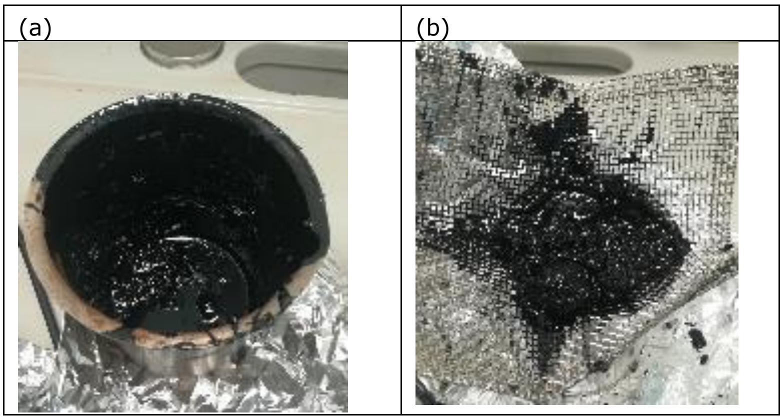


Figure 2.7: Phase separation in the Vertoro-lignin blend. (a) Residue in the can after decanting of the blend, (b) Agglomerates as sieve residue.

2.5 References

Slaghek, Vliet van, Giezen, Haaksman (2015) Bitumen composition, WO2015/137813
Landa, Gosselink (2019) Lignin-based Bio-asfalt, WO2019/092278

3 Work package 3 Asphalt testing on labscale

3.1 Inleiding

De werkzaamheden die zijn uitgevoerd in dit specifieke werkpakket 3 heeft zijn voorbereiding gevonden in twee voorgaande trajecten; te weten:

- de eerste ervaringen die opgedaan zijn in de praktijk in samenwerking met de WFBR (penvoerder) en H4A (uitvoerder) vanaf 2014.
- en een verdiepend onderzoek in het kader van een CHAPLIN XL DEI+ subsidie traject welke ook in 2020 gestart is met een looptijd van anderhalf jaar (penvoerder Universiteit Utrecht).

Dit project gaat verder in de ontwikkeling met de in het voorgaande traject opgedane ervaring in SMA asfaltdeklagen vanaf 2014. Hierbij zal ook de toepassing van Lignine in een dichte AC Surf deklaag worden ontwikkeld. Hiervoor moest het mengselontwerp worden aangepast.

Andere aspecten zoals de herbruikbaarheid (recycleerbaarheid van lignine houdend asfaltgranulaat), ARBO en milieuzaken op met name formaldehyde en de algemene technische prestaties van deze mengsels zullen worden getest op laboratorium schaal. Dit ter voorbereiding op de productie (dosering, menging en opslag), uitvoering (transport en verwerking) en begeleiding met temperatuurhomogeniteit, ontmengingsgevoeligheid en verdichting) van de proefvakken in WP4.

3.2 Doelstellingen en deliverables van WP 3

Taak 3.1: Het testen van verschillende lignines die door de partners van dit project zullen worden aangeleverd.

Ze zullen worden getest in de "lakmoesproef" (geschiktheidsonderzoek in ZOAB) en voor verdere toepassing in SMA. Mix Design, dosering en mengvolgorde en de optimale mengtijd en temperatuur zullen worden vastgesteld. ITSR voor de watergevoeligheid zal getest worden voor de duurzaamheid en mechanische treksterkte.
Tevens zal er uit WP2 een gemodificeerd lignine worden verkregen en getest in vergelijking met de referentie lignine waaruit het gemaakt is. Dit zal worden toegepast is een AC Surf deklaag mengsel en getest op onder andere op levensduur.
De beoogde deliverables zijn gehaald waarbij respectievelijk de volgende lignine soorten en bronnen zijn verkregen en getest:

- Stora Enso als langjarige lignine referentie via H4A
- Gemodificeerde Stora Enso (SE20 IH181GEL-Ph: ge-etherificeerde kwaliteit) Via WFBR
- Gemodificeerde Stora Enso (SE21 IH256 GEL-EHGE: ge-etherificeerde kwaliteit 2) via WFBR
- Praj lignine gemaakt Wheat straw resp. Rice straw via BioBased Delta
- Vertoro Lignine olie: is niet geleverd voor testen
- Lignine geproduceerd uit espenhout (Aspen) in de Dawn Technology pilot plant van Avantium. (Voor Chaplin XL proefvak Siddeburen is dennenhout (Pine) gebruikt).
- De espenlignine van Avantium voldoet niet aan de minimale specificaties, de reden hiervan en het verschil met dennenlignine moet verder worden onderzocht.

Taak 3.2: Het uitvoeren van ARBO gerelateerde emissies bij de bereiding van de laboratorium mengsels en proefstukken.

Bij de bereiding van de mengsels en proefstukken van Taak 3.1 zullen er formaldehyde metingen worden verricht waarbij de concentraties gemeten worden bij de menging, opslag en verwerking van het asfalt tot proefstukken. De dimensies zullen op pilotschaal worden vastgesteld overeenkomstig de opschaling naar de verschillende praktijksituaties op grote industriële schaal. Dit alles ter voorbereiding op de uitvoering van de proefproducties en wegvakken in WP4.

De beoogde deliverables zijn ook hier behaald. De beoogde lignines zijn allen getest op formaldehyde emissie op lab en pilot schaal. Dezelfde test procedure is aangehouden zoals gerapporteerd in het CHAPLIN XL DEI+ project.

Uit deze voorgaande onderzoeksprogramma's is vast gesteld dat formaldehyde mogelijk vrij kan komen bij de toepassing van lignine in asfalt. Dit is dus de kritische parameter waarop gefocusseerd is bij de productie- en verwerking van asfalt. De geurproblematiek die duidelijk wordt ervaren bij het toepassen van lignine houdend asfalt is dat wat men waarneemt en hinder van ondervindt. Met een draagbare formaldehydemeter zijn de producties in het laboratorium al vooraf gesimuleerd en herhaald in de praktijk bij de grootschalige asfalt productie en de verwerking in proefvakken welke door ons zijn begeleidt en daadwerkelijk gemeten kunnen worden.

Met het "managen" van de temperatuur van het asfaltmengsel en dit voldoende laag te houden (niet warmer dan strikt genomen noodzakelijk is voor de verwerking; max. 150°C) kan de geurhinder sterk beperkt worden en de formaldehyde concentratie ruim onder de grenswaarde op de werkvloer gehouden worden.

De emissies bij de eerste productie runs van het DAWN proces van Avantium gaf nog teveel ontsluitingsmiddel damp wat hinderlijk is voor de omgeving en ook terug te zien was in de watergevoeligheid van het asfaltmengsel dat hiermee bereid was.

De vervolgstappen zijn vervolgens doorgevoerd en onderdeel geworden van het andere CHAPLIN XL DEI+ project alwaar het ook in een proefvak toegepast is.

Taak 3.3: De recycleerbaarheid zal worden onderzocht en aangetoond op asfaltgranulaat verkregen uit monsters uit WP4.

In samenwerking met TNO zal worden vastgesteld wat de veroudering, reologie en de blendverhoudingen kunnen zijn op de geëxtraheerde bindmiddelen uit dit recyclaat. Using traditional extraction methods such as dichloromethane it is known that not all of the lignin dissolves in dichloromethane. Therefore the current method is not able to evaluate all of the bitumen/lignin binder. This fact should be taken into account when evaluating the life span results.

De beoogde deliverable is behaald waarbij is voldaan aan de huidige regelgeving die maximaal 30% asfaltrecycling toestaat in een AC Surf mengsel. Het asfaltgranulaat wat is toegepast betreft lignine houdend SMA uit de Wervenweg te Zeeland aangeleverd door H4A (proefvak gerealiseerd in 2015).

Het mengsel voldoet aan alle relevante functionele eigenschappen die ook aan regulier asfalt gesteld worden en is daarmee toepasbaar om in de toekomst weer hergebruikt te kunnen blijven worden.

In het andere CHAPLIN DEI+ project zijn de grenzen van opschaling van het lignine gehalte nog verder onderzocht waarbij naast Lignine houdend asfaltgranulaat ook lignine aan het nieuwe bindmiddel is toegevoegd.

In dit project is op lab/pilotschaal aangetoond dat het technisch mogelijk is om lignine houdend asfalt te kunnen recyclen. In het CHAPLIN DEI+ project is het ook daadwerkelijk toegepast op industriële schaal op Field/Lab niveau wat de conclusie van herbruikbaarheid in het lab bevestigd.

Taak 3.4: De opschaalbaarheid van de procescondities vanuit de lab/pilotschaal zal worden geëvalueerd voor de volgende aspecten:

- Dosering en mengvolgorde
- Opslag condities
- Transport naar het project

- Homogeniteit
- Monitoring van Temperatuur, afkoeling, verwerking met spreidmachine en verdichting met de afwerkbalk, walsen (met gebruikmaking van de zgn. ASPARI technieken)

De beoogde deliverables zijn met leermomenten die tijdens de uitvoering van het traject doorlopen zijn zeker gehaald. Het is duidelijk geworden dat de dosering van lignine in het asfaltproces maatwerk is en op het juiste moment en tijdstip moet gebeuren. Per project zal worden stilgestaan bij de processing en ervaringen.

3.3 Werkwijze

Alle mogelijke asfaltproeven die normaal volgens geharmoniseerde Europese normen voor asfalt uitgevoerd worden, zijn ook toegepast voor lignine houdende asfaltmengsel ondanks het feit dat er eisen zijn gesteld aan de toepasbaarheid van deze asfalt en bitumennormen voor afwijkende materialen qua soort en oorsprong. Dit moet dan ook bij de interpretatie van de data altijd in het achterhoofd worden gehouden. Asfalt is gedefinieerd als een mengsel van grove en (zeer) fijne mineralen met bitumen (ook weer gedefinieerd) van (fossiele) aardolie herkomst.

De volgende Europese Normen zijn van toepassing op deze lab studies:

AC surf/Bin/Base:

NEN-EN 13108-1;	Materiaalspecificaties - Deel 1: asfaltbeton
NEN-EN 13108-20;	Materiaalspecificaties - Deel 20: Typeonderzoek
NEN-EN 12697-8;	Bepaling van het gehalte aan poriën in bitumineuze materialen
NEN-EN 12697-12;	Bepaling van de watergevoeligheid
NEN-EN 12697-23;	Bepaling van de splijttreksterkte van bitumineuze proefstukken
NEN-EN 12697-24;	Weerstand tegen vermoeiing
NEN-EN 12697-25;	Cyclische drukproef, weerstand tegen permanente vervorming
NEN-EN 12697-26;	Stugheid
NEN-EN 12697-31;	Proefstukken verdicht met een gyratorverdichter
NEN-EN 12697-33;	Proefplaat verdichting
NEN-EN 12697-35;	Mengen in het laboratorium

SMA en ZOAB:

NEN-EN 13108-5;	Materiaalspecificaties - Deel 5: Steenmastiekasfalt
NEN-EN 13108-7;	Materiaalspecificaties - ZOAB
NEN-EN 13108-20;	Materiaalspecificaties - Deel 20: Typeonderzoek
NEN-EN 12697-8;	Bepaling van het gehalte aan poriën in bitumineuze materialen
NEN-EN 12697-12;	Bepaling van de watergevoeligheid
NEN-EN 12697-23;	Bepaling van de splijttreksterkte van bitumineuze proefstukken
NEN-EN 12697-31;	Proefstukken verdicht met een gyratorverdichter
NEN-EN 12697-35;	Mengen in het laboratorium

Daarnaast zijn de in de Standaard RAW bepalingen 2020, Proef 62, opgenomen afwijkingen en/of aanvullingen op boven genoemde normen aangehouden. Hierbij is ook de vrijheid genomen om in het geval van Lignine als vervanger voor bitumen en/of vulstof hiervan af te wijken.

3.4 Resultaten en discussie

Binnen dit werkpakket 3 zijn de verschillende taken geformuleerd die hier meer in detail worden beschreven.

Taak 3.1 Het testen van verschillende lignine monsters op lab schaal die door de partners van dit project zullen worden aangeleverd.
Dit zijn respectievelijk:

- Stora Enso als Ligno Boost referentie
- Praj (met wheatstraw en ricestraw)
- Avantium met Dawn aspenwood
- Gemodificeerde (chemisch) Stora Enso

In de volgende deklaagmengsels:

- SMA en AC Surf met voorafgaand de ZOAB "lakmoes" geschiktheidstest.

De keuze voor deze lignine soorten zijn bepaald op basis van beschikbaarheid en volume op industrieel- en pilotschaal niveau (zie WP1). Zo is de keuze voor de meest relevante Kraft lignine gevallen op het product van Stora Enso, geleverd via H4A. Dit product is ruim voorhanden en het experiment betrof vooral de toepassing in SMA en AC Surf mengsels in combinatie met recycling van asfalt en lignine asfalt in zogenaamde PR mengsels. Deze soort geldt min of meer als inmiddels vaste referentie voor de meer experimentele nieuwe lignines.

Deze lignoboost lignine van Stora Enso is door de WFBR en TNO tevens geselecteerd om een chemische modificatie op uit te voeren. Dit is dus één van de experimenten van dit traject.

Verder is het Avantium lignine product uit hun zogenaamde "Dawn" proces ter beschikking gesteld wat in hun lab / pilot plant is geproduceerd gedurende de looptijd van dit traject. Dit is dus een experimenteel lignine uit aspen hout wat op zeer beperkte schaal verkrijgbaar kwam. In dit project is de eerste variant getest welke uiteindelijk niet voldeed maar wel de basis was onder optimalisaties qua zuiverheid en korrelfijnheid qua maling.

BioBased Delta is als WP leider van WP4 met Praj uit India aan de slag te gaan om ook van hun proeffabriek de nodige soorten lignine te verkrijgen. Praj heeft één type hydrolyse proces waarin twee feedstock stromen worden verwerkt. Dit zijn de zogenaamde wheat straw en rice straw producten. Beide lignines zijn getest en voldoen aan de gestelde eisen maar men geeft de voorkeur aan het rice straw product. Dit materiaal is echter niet meer op tijd geleverd voor toepassing in een proefvak (WP4).

De asfaltonderzoeken op deze referentie en experimentele asfalt/lignine mengsels zijn allen samengevat in een "long list" waarbij de uitkomsten vaak aanleiding gaven voor aanpassing van de receptuur en of vervangingsgraad van bitumen. Deze "long list" staat in de volgende tabel opgesomd en zal in een bijlage qua data verder in detail worden gerapporteerd. Van deze long list is een keuze gemaakt voor toepassing in proefvakken naar rato van toepasbaarheid voor aard en type van de wegverharding. Van deze toegepaste AC Surf mengsels is opgesteld, waarop de asfaltproducent normaal gesproken zijn CE markering baseert.

Deze verkorte verslagen, opgesteld door AKC, zullen als bijlage worden toegevoegd:

- Annex 1 Rapportage M1 tm M4 ZOAB 16 Lignine
- Annex 2 Rapportage M7 tm M12 SMA 11 Lignine
- Annex 3 Rapportage M13 tm M15 AC 11 Surf, 30% PR, Lignine

Tabel 3.1 ZOAB test monsters voor ITSR metingen (Annex 1 more details)

Nr:	**Type mengsel**	**Welke soort**	**PR**	**% bitumen vervanging**	**ITSR (MPa en %)**		
			%	Hoog/medium/laag	Dry	Wet	Ratio
1	ZOAB 16	Stora Enso	0	Hoog	1,09	0,92	84
2	ZOAB 16	Praj Wheat	0	Hoog	1,00	0,81	81
3	ZOAB 16	Praj Rice	0	Hoog	1,12	0,95	85
4*	ZOAB 16	Dawn Aspen	0	Hoog	1,18	0,57	**48**
5	-	Gemod. S.Enso(1)	0	Not Available SMA Priority	-	-	-
6	-	Gemod. S.Enso(2)	0	Not Available SMA Priority	-	-	-

*) Het eerste monster Aspen van het Dawn proces van Avantium voldoet niet aan de gewenste waarde (min 80% ITSR waarde) en is dus aanleiding geweest voor verbetering.

Tabel 3.2 SMA test monsters voor ITSR metingen (Annex 2 more details)

Nr:	Type mengsel	Welke soort	PR gehalte	% bitumen vervanging	ITSR (MPa en %)		
			%	Hoog/medium/laag	Dry	Wet	Ratio
7a	SMA	Stora Enso	0	Medium (2%)	1,4	1,16	83
7b	SMA	Stora Enso	0	Hoog (3%)	0,82	0,75	91
8	SMA	Praj Wheat	0	Hoog (>3%)	1,41	1,33	94
9	SMA	Praj Rice	0	Hoog (>3%)	2,14	1,86	87
10	SMA	Dawn Aspen	0	Hoog (>3%)	1,09	0,60	**55**
11	SMA	Gemod. S.Enso	0	Medium (2%)	1,42	1,21	85
12	SMA	Gemod. S.Enso	0	Hoog (3%)	0,82	0,72	88

*) Het SMA mengsel (10) met het eerste monster Aspen van het Dawn proces van Avantium voldoet niet aan de gewenste waarde (min 80%) en is dus ook in dit praktijk mengselaanleiding geweest voor verbetering. De lakmoesproef heeft dus een goede voorspellende waarde.

Tabel 3.3 AC Surf test monsters voor ITSR en functionele metingen (Annex 3 more details)

Nr:	Type mengsel	Welke soort	PR gehalte	% bitumen vervanging	ITSR (MPa en %)		
			%	Hoog/medium/laag	Dry	Wet	Ratio
13	AC Surf	Stora Enso	30	Medium	2,88	2,65	92
14**	AC Surf	Stora Enso	30% lignine	Hoog	2,24	1,56	70
15	AC Surf	Praj Rice	30	Medium	X,xx	X,xx	xx
					Functionele eigenschap		
					Smix	**ε6**	**Fcmax**
13	AC Surf	Stora Enso	30	Medium	5537 MPa	131 µm/m	0,2
14**	AC Surf	Stora Enso	30% lignine	Hoog	7058 MPa	**79** µm/m	0,2
15	AC Surf	Praj Rice	30	Medium	5500-6000	± 150 µm/m	

**) Mengsel 14 met (te) hoog % BioBased vervanging voldoet niet aan de min. Eis voor de vermoeiing (100 µm/m)

<u>Opsomming van de behaalde deliverables en mogelijke extra output:</u>

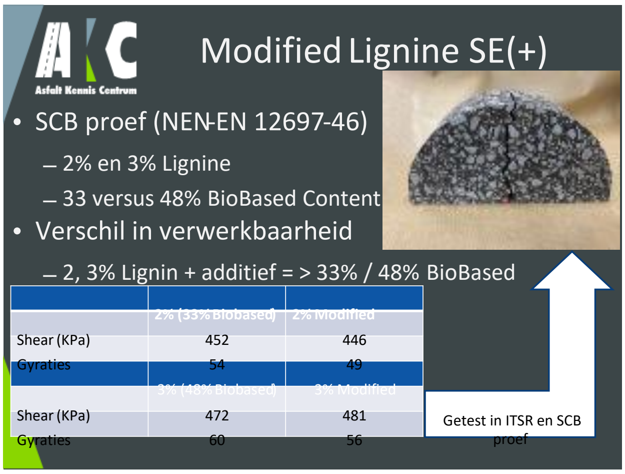
Met de kennis die we in het verleden hebben opgedaan is het mogelijk gebleken om max. 50% van het bindmiddel te gaan vervangen door llignine aangevuld met BioBased LynPave olie. Deze ervaring is vooral opgedaan in de SMA recepturen reeks. Deze relatief veilige SMA mengsels (met overmaat aan bindmiddel t.o.v. AC Surf mengsels) zijn vooral toegepast in het Chaplin XL (DEI+) project.

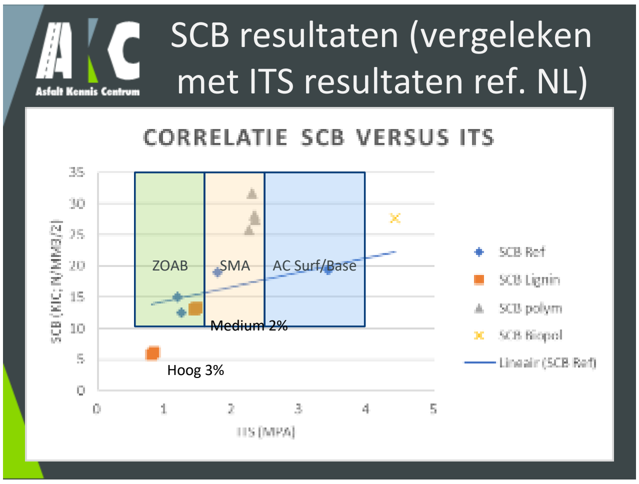
In dit project is de focus verlegd naar een schralere AC Surf deklaag met standaard 30% recycling asfaltgranulaat en een lager bitumengehalte. Deze mengsels dienen voor het Functionele Ontwerp (Mix Design) volledig getest en geschikt te zijn op nog 3 extra prestatie aspecten (Stijfheid, weerstand tegen vermoeiing en weerstand tegen permanente spoorvorming). In dit mengseltype is dan ook de recycleerbaarheid van lignine houdend asfalt getest als onderdeel van taak 3.3.

Chemische Modificatie van (Stora Enso) Lignine werd uitgevoerd door WFBR in WP2 en dit materiaal is ter beschikking gesteld aan AKC teneinde de meerwaarde hiervan in asfalt vast te stellen. Er is een extra deliverable verkregen door zowel de niet gemodificeerde als de chemisch gemodificeerde Stora Enso in twee doseringen te onderzoeken. De toegevoegde waarde van de chemische modificatie vertaald zich niet in significant beter technisch gedrag. Ook hier blijkt de medium dosering het maximaal haalbare en de treksterkte is met slechts 1,5% verbeterd.

Om dit goed te onderzoeken is een meer discriminerende treksterkte proef toegepast i.p.v. de directe Treksterkte met de ITSR proef. Dit betreft de zogenaamde SCB proef waarbij een “halve maan” vorm proefstuksegment wordt belast op doorbuiging.

De extra deliverable welke gehaald is, uit zich in een benchmarking met ZOAB en AC Surf waardes uit de literatuur. (zie PPT presentatie en volgende plaatjes)





Taak 3.2 Onderzoek naar emissies van formaldehyde bij de bereiding van asfalt met de verschillende standaard en experimentele lignines (details zie Annex 4).

Uit eerder ARBO onderzoek door H4A (zie bijlage H4A arbeidshygiënist) is gebleken dat bij de verwerking van lignine houdend asfalt formaldehyde kan vrijkomen. Het was op dat moment onduidelijk waar dit vandaan komt en heeft toen geleidt tot een uitgebreid laboratorium onderzoek waarbij diverse lignines zijn opgewarmd als puur product in een laboratoriumopstelling waarbij een vaste hoeveelheid in een zogenaamde rotavapor (ronddraaiende glazen kolf welke in een oliebad gecontroleerd in temperatuur kan worden opgevoerd). Er worden dan ook maalkogels toegevoegd om te voorkomen dat het lignine in de smeltfase gaat samenklonteren. Via de hals van de kolf kan dan een gassonde worden ingebracht die de vrijkomende damp opzuigt naar het meetapparaat.

Op die manier is vast gesteld dat er verschil zit in lignine soorten en er soms meer of minder wordt gemeten. Met name bij iets verhoogde vochtgehaltes (paar procent waarbij het ligninepoeder nog steeds droog aanvoelt) komt er meer formaldehyde vrij. (zie bijlage lab studie emissies ARBO)

Uit literatuuronderzoek blijkt dat formaldehyde een afbraakproduct is wat vrijkomt bij de dehydrogenering van methanol. Er is methanol in lignine producten aanwezig via de methoxy groepen. Of in fossiel bitumen methanol en of formaldehyde zit is onbekend maar er is wel een grote vochtvorming en afscheiding waargenomen bij de productie en opslag van lignine houdend asfalt.

Uit het onderzoek is ook gebleken dat de temperatuur van lignine houdend asfalt bepalend is voor de mate van geurhinder en ook formaldehyde vorming. Er wordt door ons een geadviseerde max.

temperatuur van 150°C aangehouden. Bij 130°C is het asfalt in combinatie met een BioBased verjongingsmiddel LynPave olie voldoende "soepel" om goed verwerkt en verdicht te kunnen worden.

In de ARBO studie voor dit project op laboratorium/pilot schaal op de nieuwe experimentele lignines zijn dezelfde meetprotocollen gebruikt. Daarbij is onder alle omstandigheden de formaldehyde concentratie nooit boven de grenswaarde uit gekomen. Uitzondering hierop is de meting direct boven de menger voor de standaard Stora Enso lignine en het lignine uit de eerste batch van het Dawn proces van Avantium.

De grenswaarde voor formaldehyde is door Nederland vast gesteld op max. 0,15 mg/m^3 voor een 8 uurs gemiddelde met 0,5 mg/m^3 als 15 min. Piek. (T.o.v. andere Europese landen is dit extreem laag. (Fin,Zwe,Zwi: 0,37 – Nor: 0,6 en UK: 2,5 mg/m^3).

Echter een arbeidshygiënist zal altijd de 10% waarde van de grenswaarde aanhouden als een veilige werkomgeving (zoals al eerder gezegd zijn er niet zo heel veel slechtere condities nodig om de dampconcentratie snel te doen laten stijgen.). Daarom is er een uitgebreide voorstudie verricht bij de mengselbereidingen in het laboratorium waarbij de concentratie is gemeten op verschillende locaties tijdens de proef. Met een pilot opstelling is de situatie nagebootst op verschillende andere plekken in het productie proces bij een asfaltinstallatie. De "headspace" / massa (lucht/massa) verhouding is hierbij geometrisch gevolgd. Zo zijn er metingen verricht op de volgende locaties:

- boven de menger (als afgesloten ruimte)
- in de opslag silo (met een nagenoeg geheel met asfaltmassa gevulde afgesloten ruimte)
- In de binnenruimte van de geluidsomkasting van een asfaltcentrale
- Bij de verwerking van het asfalt met de hand in een mal van 0,5 m^2 en het walsen ervan.
- (Zie tabel Bijlage)

Deze ARBO deliverable is dus in zijn geheel gehaald en heeft gezorgd voor een veilige werkomgeving en een sterk verminderde geurhinder. Als extra output kan gesteld worden dat bij de verificatie metingen aan asfalt ZONDER lignine ook met tijd en wijle dezelfde concentraties aan formaldehyde gemeten zijn. Ook fossiel bitumen is net als lignine een organische stof waarin reactieve groepen kunnen voorkomen die eerder genoemde dehydrogeneringsreacties kunnen vertonen en (form)aldehydes kunnen vormen.

Als extra output kan ook de zoutzuur damp worden genoemd wat voor Avantium de reden was om het proces nog eens te gaan fine tunen voor de grotere producties die in CHAPLIN XL Dei+ zijn gebruikt.

In Annex 4 "ARBO metingen Formaldehyde" de gedetailleerde metingen aan formaldehyde zijn gerapporteerd door AKC.

<u>Taak 3.3 Het testen van de recycleerbaarheid van Lignine houdend oud asfalt.</u>

Er zijn eerder al drie mix design recepturen ontwikkeld en getest in een AC Surf 11 met 30% standaard hergebruik van asfalt (vanwege circulariteit en economie is dit DE standaard in Nederland voor AC Surf mengsels). De grenzen van wat maximaal haalbaar is als BioBased bitumenvervanging is onderzocht en uit een eerste te hoog gegrepen gehalte is een asfalt receptuur ontworpen en getest welke het maximaal haalbare blijkt te zijn. (30% bitumenvervanging door lignine i.p.v. 50%)

Ook hergebruik van lignine houdend oud asfaltgranulaat als recyclemateriaal is getest in deze geoptimaliseerde samenstelling maar ook dat kent zijn beperkingen.

Hiervoor is door H4A in het kader van WP4 lignine houdend freesmateriaal uit een oud wegdek gehaald (demonstratieweg neergelegd in 2015) en ter beschikking gesteld aan AKC. Dit materiaal is onderzocht op veroudering van het toen gebruikte bitumen en qua reologie gecorrigeerd met biobased LynPave olie waarmee een blendlijn is gemaakt met het geëxtraheerde bindmiddel uit het lignine houdende asfaltgranulaat.

Vanwege de voortgang van beide projecten (chaplin-TKI en Chaplin XL) is dit materiaal eerst getest in een onder/tussenlaag mengsel in het kader van beide projecten. Dit materiaal functioneerde goed in een schraal tussenlaag mengsel met een dosering van 25%.

In een bitumenrijkere AC deklaag is meer ruimte voor lignine en daarbij is een mengsel op laboratorium schaal bereidt met zowel lignine in het asfaltgranulaat alsmede lignine in het nieuw toe te voegen bitumen. Dit mengsel haalde uiteindelijk niet de gewenste functionele eigenschappen en daarmee is in feite een extra deliverable gerealiseerd door de maximum vervangingsgraad te kunnen benoemen.

Ook hier is 50% BioBased verdeeld over oude en nieuwe grondstoffen dus de maximum limiet.

<u>Taak 3.4 De opschaalbaarheid van de procescondities vanuit de lab/pilotschaal zal worden geëvalueerd voor de volgende aspecten:</u>

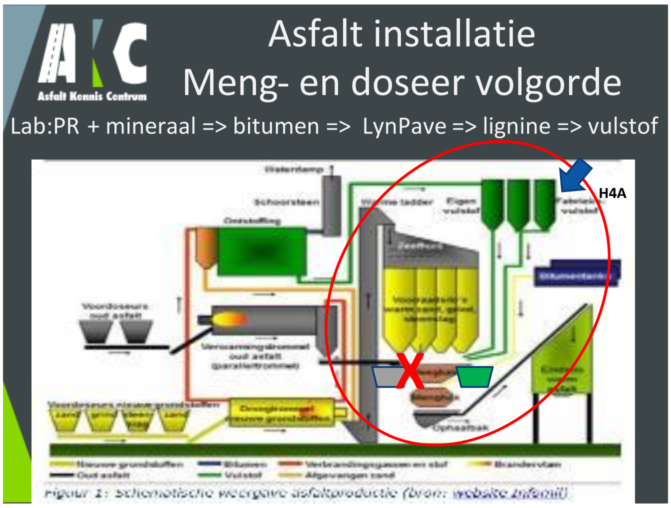
- Dosering en mengvolgorde
- Opslag condities
- Transport naar het project
- Homogeniteit
- Monitoring van Temperatuur, afkoeling, verwerking met spreidmachine en verdichting met de afwerkbalk, walsen (met gebruikmaking van de zgn. ASPARI technieken)

<u>Dosering en Mengvolgorde:</u>

De dosering van lignine gebeurt in het geval van de AKC/WFBR technologie (WO 2019/092278) in het asfaltbereidingsproces in de menger van de asfaltinstallatie. Dit is in eerste instantie gebeurt met Plastic smeltzakken welke vooraf "ingepakt" moeten worden bij een speciaal verpakkingsbedrijf voor chemicaliën. Vanwege de fijnheid en stofvorming (veiligheid) wil men deze service niet meer leveren en is er een alternatieve verpakkingsmethode ontwikkeld met zogenaamde "kunstmestzakken". Deze kunnen echter maar voor een beperkt deel gevuld worden. Al met al geen ideale situatie en voor H4A aanleiding geweest om te investeren in een silotransport auto voor vulstoffen die aangekocht en aangepast is. De werken die hiermee uitgevoerd zijn vertonen een betere homogeniteit en de juiste doseer volgorde kan veel makkelijker gevarieerd worden qua moment en plaats in het proces. Echter niet alle asfaltinstallaties kunnen of willen hiermee werken vanwege de vrees voor stofexplosies en blijven liever bij smeltzakken dosering vooralsnog.

Een alternatieve doseermethode met los gestort materiaal in emmers (op kleine schaal gebeurt het zo ook in een laboratorium menger) heeft laten zien dat stofvorming en doseersnelheid deze methode op industriële schaal onbruikbaar maken. Een proefproductie in de installatie Bovenveld van NTP liet zien dat de homogeniteit en goede menging niet bereikt kon worden door de te langzame dosering (stofvorming en zichtprobleem) met dito te lange mengtijd van het mengsel in de menger. Dit asfalt is afgekeurd voor transport naar het project wat vervolgens is aangelegd met alleen het normale referentie mengsel. Deze extra deliverable laat zien dat deze vereenvoudigde methode dus echt niet kan en dit pad dient derhalve afgesloten te worden.

Een andere belangrijke voorwaarde voor de doseer volgorde blijkt het zogenaamde "nat" inmengen. Dit betekent dat het opgewarmde mineraal als eerste omhuld moet worden met het benodigde fossiele bitumen en LynPave olie. Daarna volgt bij voorkeur vóór de vulstofdosering de lignine dosering (met smeltzakken of silo auto dosering). Op die manier is er een maximale benatting en insmelting van het lignine mogelijk. Daarna volgt de benodigde resterende vulstoffractie. Deze mengvolgorde blijkt cruciaal voor een goede homogeniteit en opneming van het lignine in het bitumineuze bindmiddel.



- Opslag condities

Bij de opslag van lignine houdend asfalt in een afgesloten silo van de asfaltinstallatie kan weinig fout gaan t.o.v. normaal asfalt. Afkoeling en oxidatie wordt voorkomen door maximale isolatie en afsluiting. Echter het is wel mogelijk dat de mogelijk optredende dehydrogeneringsreactie extra formaldehyde en waterdamp vrij geeft als reactie producten. Deze zijn dan ook op lab/pilotschaal en industriële schaal gemonitord.

- Transport naar het project

Dit gaat niet anders als bij normaal asfalt met geïsoleerde en afgestoten laadbakken. Enige temperatuur terugval hangt af van rijsnelheid, afstand, wachttijd, windsnelheid en buitentemperatuur. Bij het verlaten van de vrachtauto bij de asfaltcentrale wordt de temperatuur indirect gemeten en gerapporteerd en bij aankomst en storten in de hopper van de asfaltspreidmachine en uiteindelijk onder de verdeelbalk achter de spreidmachine “pikt” de ASPARI thermocamera weer de dan “afgekoelde” temperatuur op.

- Homogeniteit

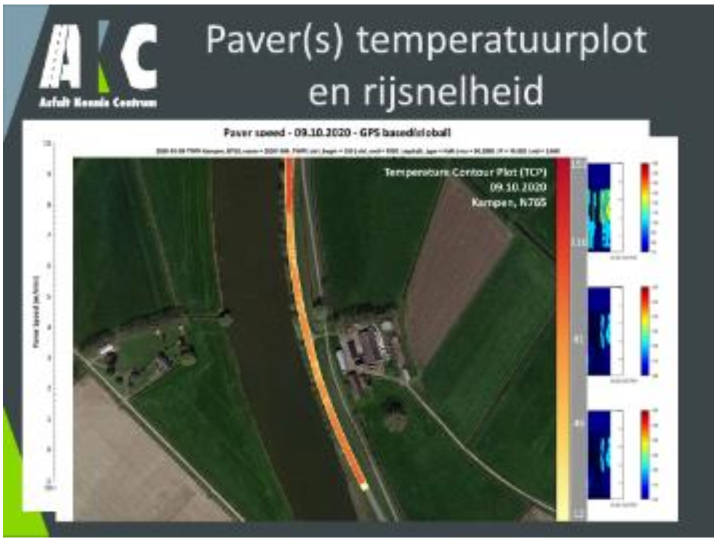
In een enkel geval is vanwege de grote breedte van de weg, wat twee naast elkaar werkende asfaltspreidmachines noodzakekijk maakt om naadloos te kunnen draaien, een zogenaamde shuttle buggy ingezet. Hierin wordt de batch van de vrachtauto gestort en vanuit die hopper wordt het materiaal nog een keer “over de kop” gemengd en getransporteerd naar beide spreidmachines. Er vindt dus vooral opmenging plaats van afgekoelde gedeeltes. Er vindt nauwelijks homogenisatie op micro niveau plaats wat soms gedacht wordt. M.b.v. deze tussen opslag kunnen de vrachtauto’s sneller lossen en het werkvak verlaten om de volgende vrachtauto weer te laten manoeuvreren zonder dat de spreidmachines “leeg vallen”. Zodoende is de kans op stopplekken die moeilijk te verdichten zijn door afkoeling het laagst.

Het temperatuur verlaagde lignine asfalt is echter geen HMA (Hot Mix Asphalt) zoals men in de USA gewend is met temperaturen van ver boven de 170°C. De machine had dus wel wat problemen met de doseer- en transportinrichting. Dit is dus een extra deliverable als uiterste minimum temperatuur waarbij deze machines dus nog inzetbaar zijn.

Daarnaast kunnen ze binnenstedelijk nauwelijks ingezet worden vanwege hun hoogte en brede afmetingen (zie volgende illustratie).

- Monitoring van Temperatuur, afkoeling, verwerking met spreidmachine en verdichting met de afwerkbalk, walsen (met gebruikmaking van de zgn. ASPARI technieken)

Met de genoemde ASPARI techniek (Universiteit Twente) is het gehele “Paving” proces realtime gemonitord en vast gelegd met “Big Data” Dit betreft het logistieke proces vanaf de belading bij de asfaltinstallatie van de vrachtauto’s tot aan het spreiden van het asfalt en afwalsen van de aangebrachte deklaag. Hierbij worden voertuigbewegingen gevolgd met GPS systemen zodat plaats en tijd en snelheden worden vastgelegd om later te kunnen checken op stopplekken, rijsnelheid van de pavers (wat van belang is voor de voorverdichting) en aantal en locatie in het dwars- en langsprofiel van de verschillende walsen. Verder wordt de temperatuur gemeten in dwarsraaien van steeds 1 decimeter zodat er een temperatuurplot gemaakt kan worden direct achter de afwerkbalk van de paver. Hot- en Cold spots kunnen zo herkent worden waarbij de verschillen kunnen worden geïnterpreteerd met de locatie gegevens in de toekomst van eventueel optredende schadebeelden. Zie volgende illustraties.



3.5 Discussie, conclusies en aanbevelingen

Several technical lignins, from the Dutch biorefinery processes (executed by Avantium, Vertoro, BBD) were evaluated as partial substitute for bitumen. Most lignins are suitable for this asphalt applications as measured on asphalt level. Health and safety aspects in the production and pavement of lignin-based asphalt can be realised. Release of volatiles such as formaldehyde do not exceed the threshold value for a safe working environment. With the so-called ASPARI technique is the pavement of a demonstration road realtime monitored for the first time. This is a technique which is helpful for the further development of the technology.

4 Work package 4 Demonstration roads

4.1 Objectives & deliverables of WP 4

The main objectives of this work package is to construct new stretches of roads using commercially available lignin in the two municipalities designated (Bergen op Zoom and Wageningen) with the aim of monitoring from production of asphalt, construction of the roads until the end of the project. During the project it became clear that due to manpower capacity the municipality of Wageningen could not accommodate a test strip and we worked successfully with the municipality of Altena and the province of Gelderland instead.

4.2 Methods

Task 4.1 Determine types of lignin based bitumen
These activities are described in more detail in WP 2 and 3. As a result we have worked with commercially available Lignoboost Kraft lignin in both test strips.

Task 4.2 Risk analyses and monitoring of risks
Based on the results of task 4.1, an overview has been made of the lignins that are suitable for use within road test sections.

Subsequently, the availability was assessed and a choice was made for the lignin type to be used.

Based on the type pavement of the test section, AKC prescribed a mixture that had been tested in the laboratory beforehand and the results of which were positive.

AKC supervised the production process to ensure that this was carried out according to plan. An important measure in this regard was to conduct a test run before proceeding to actual production. By having 1 party (AKC) supervise the production, the lessons learned during previous productions were automatically transferred to the next one, ensuring continuous optimization and further reducing the risks.

The Corona pandemic has run like a red thread through the project. It was very difficult to actively manage this because the regulations and the expectation of their development were constantly subject to change. That is why it has been decided to always have a fallback scenario for various matters.

During the implementation of the project, various meetings were organized at work package leader and project participants level, during which, in addition to progress, the risks were also discussed and how these risks could be reduced/removed together. What was discussed during these meetings is recorded in minutes of meeting.

The principles of the RISMAN method have been applied throughout this risk management process. However, deviating from the original project plan, no risk log was kept as this was deemed unnecessary considering all the other procedures in place.

Task 4.3 Monitoring the execution of road sections
It was not possible for both the municipality of Bergen op Zoom and Wageningen to realize a test section in 2019 and 2020. This is one of the reasons why we looked for other options. This was done by bringing the CHAPLIN-TKI project to the attention of the various road owners in the Netherlands. To this end, we have approached the CBBD network and also BouwCirculair, which has a broad network to which we have pitched the project.

Additional talks have been held with interested road owners. The topics below were discussed, among others:
- Warranties and risks;
- Tender and award;
- Monitoring;
- (Additional) costs.

For the monitoring, the agreements below have been made with the road owners and the contractor about what and by whom matters should be monitored.

Production phase
- Type Test ITSR en HR (AKC)
- Trial production (few batches) (AKC)
- Emission measurements (AKC in case necessary)

Construction phase
- layer thickness: NEN-EN 12697-36 (contractor)
- degree of compaction: NEN-EN 12697-6, (Nuclear density measurements and any drill cores) (contractor)
- composition and cavity space: NEN-EN 12697-8 (contractor)
- ASPARI measurements (contractor ***see results in WP3***)
- Skid resistance measurement (contractor)
- braking deceleration (contractor)
- emissie metingen (project task 5.4 AKC)

Usage phase
- 1st visual inspection after 6 months (project task 5.3 contractor);
- durability binder (project task 5.3 TNO)
- visual inspection 1st/year (road owner)

Task 4.4: Recycling of current lignin/bitumen based asphalt.
These activities are described in more detail in WP3.

Task 4.5 Drafting full OVERALL CHAPLIN road section testing program (M22-24)
At the end of 2020 and 2021, meetings were organized with the road authorities who are connected and who have shown serious interest in the CHAPLIN program. In addition, meetings were organized in 2020 and 2021 with the participants (lignin producers, bitumen suppliers, asphalt producers and contractors, road owners and knowledge institutions) in the CHAPLIN program.
During these meetings, the various parties were informed about the development of the project and their input was asked about what still needs to be done to further develop this technology as well as to get it widely accepted in the market.

4.3 Results

Evaluation report of the (re)constructed road sections
For the realization of test sections, talks were held with approximately 10 road owners about the realization of a road section with lignin-containing asphalt. Of these 10 road owners, there were ultimately 3 road owners for whom we were able to realize a test section for this project during the project period. This information regarding these test sections is further elaborated below. With regard to the work carried out and the results with regard to the monitoring, the monitoring of the test sections realized during the project is explained in more detail in WP5. It should be noted here that monitoring during the use phase of these road sections falls outside the project duration.

Province Gelderland (fietspad N315 near Neede)

Surf layer: AC 11 Surf (approx. 75 ton)

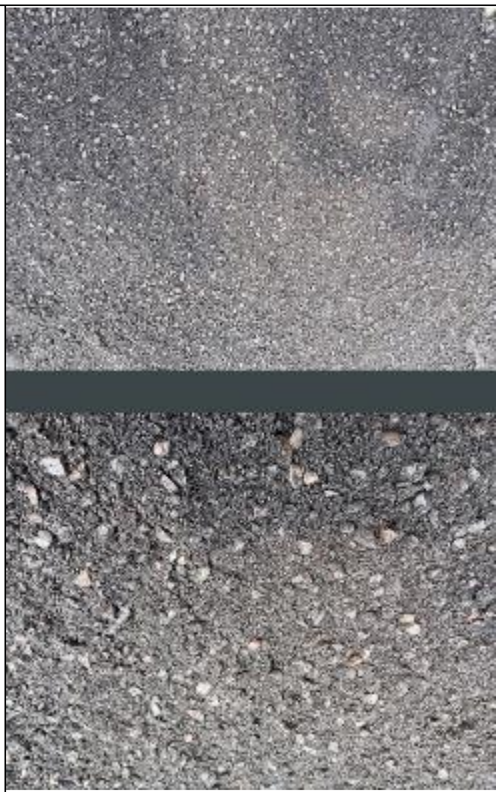
- Performance class : DL-B

Grondstoffen:

- 30% PR (surf layer material)
- Coarse aggregate : Morene
- Filler: Wigras 40K
- Coarse aggregate : Morene
- Filler: Wigras 40K
- Lignine: 30% (substitution bitumen)

Kraftproces Stora Enso

- Bitumen: 40/60
- Additive: 0,3% LynPave oil.



Particularities:

- The test section and reference section would be constructed by NTP;
- The lignin was manually added to the asphalt mixture by means of melting bags..
- Manual repacking of lignin in melting bags caused the necessary problems because some copackers did not want to carry out the work because of the fine dust that is released. This is because particulate matter (OSH) brings safety and pollution risks with it. By dosing the lignin in melting bags, the chance of errors in dosing is greater;
- During the production process, the lignins were not dosed correctly, as a result of which the asphalt mixture produced did not meet the required quality requirements, based on which it was decided not to use it on the bike path. (The mixing order of the asphalt mixture, just like with cooking, it is crucial to add lignin that it is done in the right order, otherwise you will get lumps and less binding will take place. The correct order in this is:
    1. PR+minerals;
    2. Bitumen;
    3. LynPave;
    4. Lignin;
    5. Filler.)

Municipality Bergen op Zoom (Klutsdorpseweg Lepelstraat)

Surf layer: AC 11 Surf (approx. 85 ton)

- Performance class : DL-B

Grondstoffen:

- 30% PR (surf layer material)
- Coarse aggregate : Morene
- Filler: Wigras 40K
- Coarse aggregate : Morene
- Filler: Wigras 40K
- Lignine: 30% (substitution bitumen)

Kraftproces Stora Enso

- Bitumen: 40/60
- Additive: 0,3% LynPave oil.

Particularities:

- Prior to the realization of this road section, H4A constructed a test piece with the same mixture on the cycle path on Europaweg;
- The lignins were added to the asphalt mixture by means of an automatic dosing system (silo truck).
- It was agreed to realize the test section with hydrolysis lignin Praj. Due to the impact of the Corona pandemic in India, it was not possible to get this lignin to the Netherlands in time.
- Both the test section and the reference section are realized by H4A.

Publicity: (see WP7)

Municipality Altena (Midgraaf Almkerk)

Surf layer: AC 11 Surf (approx. 93 ton) • Performance class : DL-B Grondstoffen: • 30% PR (surf layer material) • Coarse aggregate : Morene • Filler: Wigras 40K • Coarse aggregate : Morene • Filler: Wigras 40K • Lignine: 30% (substitution bitumen) Kraftproces Stora Enso • Bitumen: 40/60 • Additive: 0,3% LynPave oil.	

Particularities:

- It was initially planned to realize the test section with hydrolysis lignin of Praj from India. Due to the impact of the Corona pandemic in India, it was not possible to get this lignin to the Netherlands in time.
- The test section was constructed by NTP and the reference section was constructed by a contractor outside the project and the CHAPLIN program.
- The lignoboost Kraft lignin was manually added to the asphalt mixture by means of melting bags.
- Some parts of the road show some irregularities and a more detailed study is needed to elucidate the exact reason for this.

Publicity: (see WP7)

4.4 Discussion, conclusions and recommendations

It can be concluded that the realization of two test roads has been carried out successfully. The road in Almkerk however needs some more investigation to elucidate the reason for some irregularities in the top layer.
Additionally one test strip failed which has led to very valuable lessons learned such as:

- the importance of the correct mix order.
- the importance of automatic dosing system.

In addition, the first contours have been set for the further development of lignin-containing asphalt. The risks were identified and discussed during the various meetings.

5 Work package 5 Monitoring of asphalt

5.1 Aim & deliverables WP 5

WP-5 focuses on assessing durability and monitoring performance of the road test sections. TNO will be the lead partner in this work package and H4A, NTP, AKC, DURA Vermeer will participate actively by supporting testing at the lignin-bitumen blend, asphalt production levels. For the performance monitoring of lignin based asphalt test sections, the partners will support extraction of asphalt cores and making the test section available for monitoring and inspections.

Specific objectives:

- Assessing performance of materials in relation to durability at both binder and asphalt scale where tests will be performed on extracted asphalt cores both for the new roads constructed and the already existing roads.
- Inspection of test sections for possible failure modes: ravelling, cracking, rutting etc.;
- Monitoring performance of the old & new test sections of surface layers: stone mastic asphalt (SMA), porous asphalt (PA);
- Monitoring emissions and possible health related hazards with respect to lignin sources.

5.2 Methods

Task 5.1: Assessment of the extracted binder performance

This task is dedicated to the durability of the Kraft lignin based asphalt binder. H4A has provided asphalt cores from the old and new test sections by extracting asphalt specimens from the roads. TNO has extracted binders from the asphalt specimens (both lignin and reference asphalt) and evaluated the performance of the recovered binders. **The binder was extracted with dichloromethane. It is known that not all of the lignin dissolves in dichloromethane. Therefore the current method is not able to evaluate all of the bitumen/lignin binder. This fact should be taken into account when evaluating the life span results.** The following approach was taken to assess the properties of the extracted binders.

- To perform rheological assessment of the binders using Dynamic Shear Rheometer (DSR)
- To perform chemical characterization of the binders
- Fourier-transform infrared spectroscopy (FTIR)
- Gel permeation chromatography (GPC)
- To assess the functionality of lignin:
- Ash content
- FTIR on both recovered filler & binder

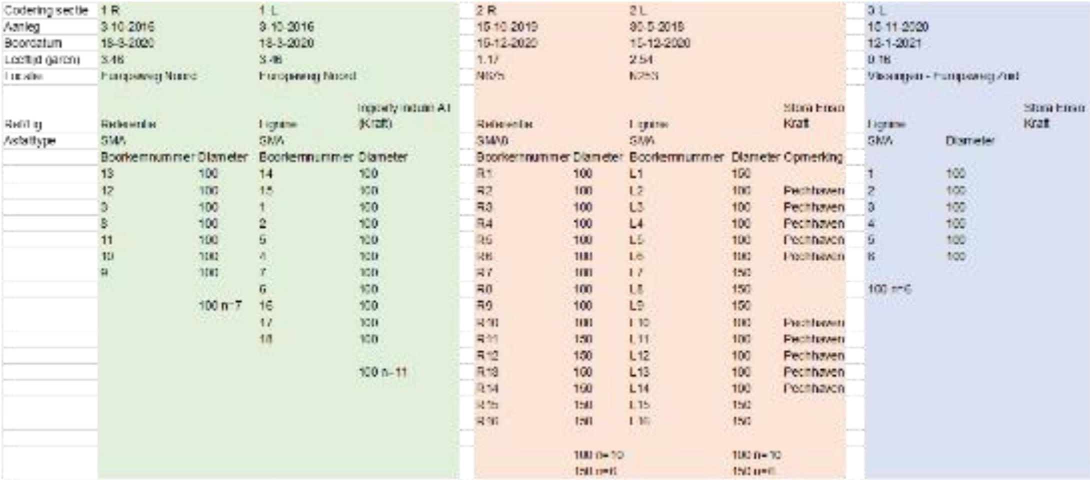
Task 5.2: Assessment of asphalt performance

This task is dedicated to evaluate the moisture damage of 8 asphalt core samples taken at different time intervals. Freeze-thaw tests will be performed in extreme temperature cycles to reveal the response of lignin based asphalt in moisture-saturated conditions. The following approach was taken: To perform Freeze-thaw tests on asphalt specimens to evaluate moisture susceptibility while exposed to thermal cycles. To perform Thermal Stress Restrained Specimen Test (TSRST) on asphalt specimens to assess performance at low temperature:

The specimens were prepared by sawing the top layer from the asphalt cores followed by measuring the dimension and density.
Freeze-thaw: Dry and wet conditioning and Indirect Tensile Test (ITT) has been performed.

Thermal Stress Restrained Specimen Test (TSRST): TSRST couldn't be performed due to the limitation of the sample size which doesn't meet the minimum dimension criteria of the test.
A thin section of asphalt was prepared to perform microscopy on the bulk of asphalt concrete Freeze-thaw conditioned samples were used for the microscopy.

The overview of the test sections are given below in Figure 5.1.



Codering sectie	1 R		1 L			2 R		2 L			3 L		
Aanleg	3-10-2016		3-10-2016			15-10-2019		30-5-2018			10-11-2020		
Boordatum	18-3-2020		18-3-2020			15-12-2020		15-12-2020			12-1-2021		
Leeftijd (jaren)	3.46		3.46			1.17		2.54			0.16		
Locatie	Europaweg Noord		Europaweg Noord			N675		N253			Vlissingen - Europaweg Zuid		
Richting	Referentie		Lignine		Ingevity indulin AT (Kraft)	Referentie		Lignine		Stora Enso Kraft	Lignine		Stora Enso Kraft
Asfalttype	SMA		SMA			SMA8		SMA			SMA	Diameter	
	Boorkernnummer	Diameter	Boorkernnummer	Diameter		Boorkernnummer	Diameter	Boorkernnummer	Diameter	Opmerking			
	13	100	14	100		R1	100	L1	150		1	100	
	12	100	15	100		R2	100	L2	100	Pechhaven	2	100	
	3	100	1	100		R3	100	L3	100	Pechhaven	3	100	
	8	100	2	100		R4	100	L4	100	Pechhaven	4	100	
	11	100	5	100		R5	100	L5	100	Pechhaven	5	100	
	10	100	4	100		R6	100	L6	100	Pechhaven	6	100	
	9	100	7	100		R7	100	L7	150				
			6	100		R8	100	L8	150		100 n=6		
		100 n=7	16	100		R9	100	L9	150				
			17	100		R10	100	L10	100	Pechhaven			
			18	100		R11	150	L11	100	Pechhaven			
						R12	150	L12	100	Pechhaven			
				100 n=11		R13	100	L13	100	Pechhaven			
						R14	150	L14	100	Pechhaven			
						R15	150	L15	150				
						R16	150	L16	150				
							100 n=10		100 n=10				
							150 n=6		150 n=6				

Figure 5.1 Overview of the extracted asphalt core specimens from the selected test sections.

Task 5.3 Visual inspection of current and novel test sections.
DURA, H4A, NTP will perform visual inspections of current and novel test road sections every 6 months which will complement the asphalt performance tests of the asphalt cores.

Task 5.4 Monitoring health and safety aspects.
Health and safety related aspects of lignin during handling and lignin based asphalt production will be monitored by H4A/AKC. Air quality and possible inhalation hazards will be evaluated while handling lignin powders or fine particulates. Odour and emissions will be probed in-situ during production of asphalt, transportation and operations. The extent of toxicity from the resulted emissions and its impact on health and safety will be evaluated. From the first phase of evaluation, if required, structured monitoring of possible emissions during service life of new lignin based asphalt sections can be performed. Results on emissions of formaldehyde were presented in WP3 by AKC.

5.3 Results

D5.1: Assessment of the extracted binder performance
The extraction test was performed on 3 lignin asphalt cores and 2 reference asphalt cores. All components of the asphalt loose mix were collected after extraction and weighed at room

temperature. The total soluble binder is obtained after being extracted by the solvent with successive distillation to remove the solvents.

GPC- molecular weight distribution
In this section paper presents a laboratory study in which Gel permeation chromatography (GPC) was used to determine the molecular weight distribution of the extracted lignin-SMA binder and reference SMA. The extracted binder from lignin-SMA is PEN 70/100 bitumen combined with the extract of the part of lignin while using dichloromethane (CH_2Cl_2) as a solvent for extraction of the binder. The extracted binder from reference SMA is pure PEN 70/100 bitumen where same extraction procedure was used. GPC analysis show that the oxidation mechanism a.k.a. increases the average molecular weight ageing but the aging impact on the change in molecular weight of the soluble lignin part of the binder is less known.

FTIR on the extracted binder
FTIR is a promising tool to detect the presence of lignin through the chemical signature of functional groups. For this purpose the infrared spectrum of both binders and fillers extracted from the lignin-SMA and reference- SMA are measured. The chemical signature of lignin and linseed oil are clearly visible. When the binder is ageing due to oxidation two specific components will increase in the FT-IR spectrum. Both the carbonyls as the sulfoxides, from the sulfur components present in bitumen, are detected. The degree of change in carbonyl and sulfoxide functional groups are characteristic to the level of oxidation in bitumen. The reference binders show that the peak height of both carbonyl and sulfoxide increase with the increase of service life of the pavement. Lignin containing binders show broadening of the peaks at the sulfoxide band compared to the reference binders.

Microscopy of recovered binder and asphalt thin slices
Optical microscopy has been performed on the recovered binder from the lignin- SMA asphalt specimens. The micrographs of the binders show traces of lignin particles in the microscopic images indicating that some of the lignin is present as particles in the asphalt binder. This is confirmed by microscopy of the asphalt thin slices.

Viscoelastic properties of the recovered binder
A rheological database is available at TNO from an earlier research on asphalt binder, where the binders were extracted from porous asphalts specimens obtained from a series of road sections. The asphalt cores for the database have been drilled from the asphalt pavements at regular intervals over the years. The rheological properties of the extracted binders were then studied using the Dynamic Shear Rheometer (DSR). The complex moduli and phase angles were measured at different temperatures and loading frequencies.
In this current context of evaluation of durability of the extracted binder in this project, this TNO dataset is used as a background knowledge to link between binder rheology to service life.
Figure 5.2 presents the black-space data sets in which rheology of the extracted binder at 20°C and 10 rad/s obtained in WP-5 are also plotted. The binders extracted from the lignin-asphalt specimens are as seen as orange markers and green markers are showing the rheology of the binders that are extracted from the reference SMA. While comparing the data, we should keep note of two aspects. The asphalt specimens that we are studying in CHAPLIN WP5 are stone mastic asphalt (SMA), whereas the TNO black-space dataset is built on the binders extracted from porous asphalt (PA). Due to the less porosity of SMA compared to PA, ageing of the binder in SMA is expected to be slower compared to that of PA. Another difference is that the binders extracted from lignin SMA using dichloromethane (i.e. CH_2Cl_2) contains bitumen with soluble lignoboost Kraft lignin whereas other binders are extracted bitumen only.

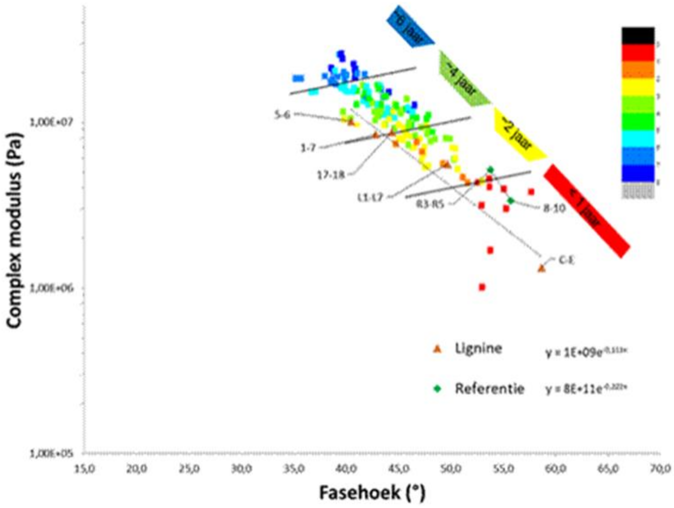


Figure 5.2: The field rheological data at 20°C and 10 rad/s with pavement service life in years (colour coded).

Insights in relation to durability of the lignin-asphalt on the basis of black-space data compared to porous asphalt (PA) are given below:

- Lignin-SMA:

Lignin-SMA binders shows 1 to 4 years of field ageing from the black-space reference of porous asphalt binders which resembles the age of the asphalt. From the data, it commonly shows that ageing of lignin SMA is comparable to the ageing of PA.

- Reference SMA:

SMA is less susceptible to ageing, hence more durable compared to porous asphalt but the results show a more diverse ageing. This deviation can be attributed to the difference in sources of the bitumen used in the reference sections. Variations in quality in bitumen, used over the years, is an additional parameter which influences the rate of aging.

D5.2: Assessment of asphalt performance

Influence of density in strength properties

Density of asphalt influences the strength properties. To manifest this, lignin-asphalt specimens are studied and the indirect tensile strength properties are shown with increasing asphalt density and the results are shown in Figure 5.3. In freeze-thaw conditioning, the strength increases with increasing density. A selection of data after omitting the outliers are taken into consideration to calculate the average strength. The selected data points are marked in Figure5.3.

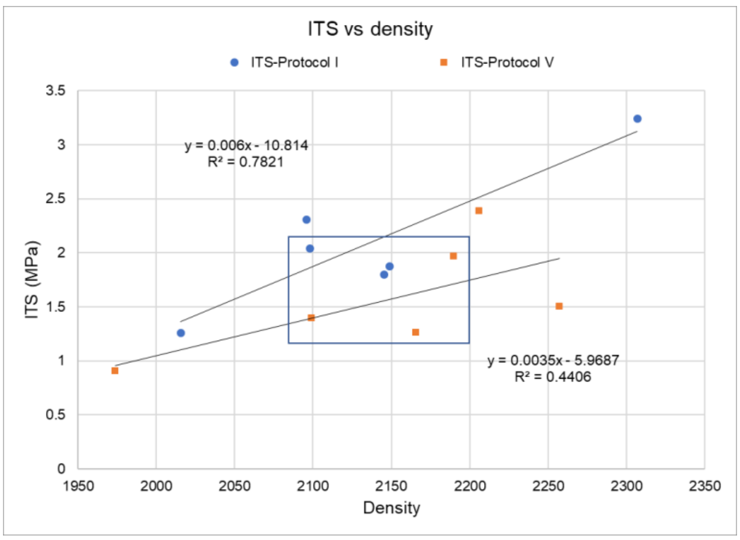


Figure 5.3: Influence of density in indirect tensile strength of the lignin asphalt

Indirect Tensile Strength (ITS)
Values are measured at 5 °C after dry conditioning, ITS measured for reference and lignin- asphalt specimens show increasing strength with service life. Whereas, lignin asphalt shows lower strength compared to the reference asphalt. This development of strength is related to the hardening of asphalt as a result of the ageing of bitumen. Here, due to ageing, the strength of asphalt initially builds up but later under the influence of weather conditions and mechanical loading, asphalt further tend to fail with time.

ITSR after Freeze-Thaw conditioning
At early service life, ITSR (Indirect Tensile Strength Ratio) values are higher in asphalt and with increasing service life the value decreases. ITSR value in lignin-asphalt decreases faster compared to the reference asphalt. It shows a delta in service life of approximately 1.5 years between lignin and the reference asphalt where the average service life span of a reference SMA asphalt is 15 years. Here, in this study we have limited data points to have a trend line on change in ITSR as progressing service life.

5.4 Discussion, conclusions, recommendations

This work package has shown that lignoboost Kraft lignin is well distributed in the continuous bitumen phase of the binder. Also some of the lignin is present as intrinsic particles. Compared to the data base of TNO regarding porous asphalt there seems to be a similarity in ageing of the binder systems. However since over the years a variety of different bitumen sources (both regionally as producer) have been used comparison between the collected samples and with the porous asphalt data of TNO a prolonged monitoring of the development of the binder is necessary for evaluating of the lignin/bitumen binder on its merits.

Collection of the core asphalt samples took far more time than anticipated due to availability of the road constructors, road authorities and safety measurements that are necessary when asphalt cores are drilled from the road next to heavy traffic.

6 Work package 6 Techno-economic and Life cycle assessment of lignin-based asphalt

6.1 Objectives & deliverables WP 6

The main objectives of WP6 were to determine the environmental and techno-economic performance of asphalt with various lignin additions, and explore the possible deployment after reaching TRL 9 in the Netherlands. To meet these objectives, the work in WP6 was organized in the following three tasks:

- **Task 6.1 Life-cycle assessment (LCA)** determined the full environmental impacts by carrying out a comprehensive life cycle assessment for production and end-of-life phase including climate change and at least 10 other impact categories;
- **Task 6.2 Techno-economic assessment (TEA)** carried out a techno-economic assessment, comparing the production costs of lignin-based asphalt with 100% bitumen based reference, assess potential for cost reductions and – linked to the first objective – determine the possibility for a premium for lignin-based asphalt;
- **Task 6.3 Roadmap for deployment** determined the market potential for lignin-based asphalt until 2030 and beyond and actions needed to full commercial implementation.

6.2 Methods

Task 6.1 Life-cycle assessment
First, a comprehensive review of 42 LCA studies of lignin and lignin-based products was conducted to identify methodological issues, lessons learnt and provide recommendations on environmental claims about lignin and lignin-based products. Secondly, a cradle-to-grave LCA of various types of asphalt top-layers using kraft lignin was conducted. The method was mainly based on the Dutch Product Category Rules (NL-PCRs) for asphalts. The impact categories followed SBK bepalingsmethode 3.0, with the weighting factors followed the Environmental Cost Indicator (ECI) method, also known as Milieu Kosten Indicator (MKI). The results of the LCA provide insight in the potential environmental impact of lignin based asphalt top-layers over their entire life cycle while the MKI score reflects on the associated environmental costs (or shadow costs). The input data for lignoboost Kraft lignin from Kraft pulp was based on the insights of the critical review. Input data for asphalt production and pavement were partly based on inputs from industrial project partners of Chaplin-TKI including Asfalt Kennis Centrum (AKC), Holding de Vier Ambachten (H4A), and Roelofsgroep.

Task 6.2 Techno-economic assessment
A detailed techno-economic assessment (TEA) was conducted to analyse and compare the economic performance of producing and paving the top layer of bitumen- and lignin-based asphalts. The economic assessment includes six different sources of lignin that are already available at commercial scale (kraft lignin) or that will likely become available in the future (lignosulfonates, soda, hydrolysis, Organosolv and steam explosion). The study includes the three most applied types of asphalt top-layers in the Netherlands: Stone Mastic Asphalt (SMA), Asphalt Concrete (AC) and Porous Asphalt (ZOAB in Dutch). Future cost projections were made based on expected improvements in asphalt production processes and the expected market development of biorefineries and associated supply of lignin. Similar to the LCA, the TEA was developed in close collaboration with the industrial project partners of Chaplin-TKI.

Task 6.3 Roadmap for deployment
To assess the potential future impact of scale up possibilities of lignin-based asphalt in the Netherlands, four scenarios were developed that vary over two axis: 1) financial support to cover the price difference between conventional and lignin based asphalt, and 2) the adoption rate of innovative solutions in the road construction sector. The four scenarios cover the period 2020 – 2050 and are shown in Figure 6.1. The *No Support* and *Baseline* scenario are almost identical because lignin based asphalt only becomes cost-competitive when financial support is provided. The *Best Estimate (BE)* scenario assumes a that 50% of the SMA and AC will be lignin-based by 2050, and lignin based ZOAB will have a market share of 25% by 2050. The *Ambitious (A)* scenario assumes that 100% of the SMA and AC will be lignin-based in 2050, and lignin based ZOAB will have a market share of 50% by 2050. The results of the scenarios include annual bitumen and lignin requirement and annual cost and GHG savings.

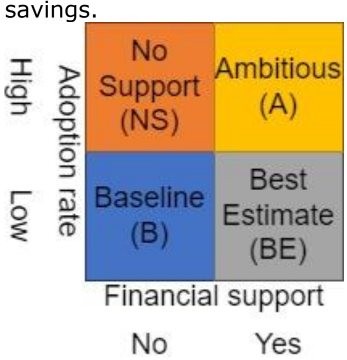


Figure 6.1: Future scenarios

6.3 Results

Task 6.1 (life-cycle assessment)
The results of the comprehensive review of LCA studies of lignin and lignin-based products and results of the LCA are published in scientific journal publications (Moretti et al. 2021, 2022). Both publications are open access which means that they can be accessed globally without restrictions. As an example of the results, the results of lignin based SMA are shown in Figure 6.2 (climate change) and Figure 6.3 (MKI score). Other types of asphalt and environmental impact categories are presented in the publication (Moretti et al. 2022). The results show that the climate change impact of lignin-based asphalt in top-layers is between 30 to 75% lower than conventional asphalts. The large range is mainly caused by the fuel used to replace lignin in kraft pulp mills. If natural gas (NG) is used to replace lignin in kraft mills, the carbon footprint of lignin based asphalt is substantially higher compared to lignin replaced by low-grade biomass fuels in kraft mills (BIOM). Besides climate change, other relevant environmental impacts are marine aquatic ecotoxicity, human toxicity, eutrophication and acidification that are, in some cases, higher for lignin-based asphalt compared to conventional asphalt. These trade-offs result in a less profound difference between lignin based asphalt and conventional asphalt in MKI scores in comparison to climate change.

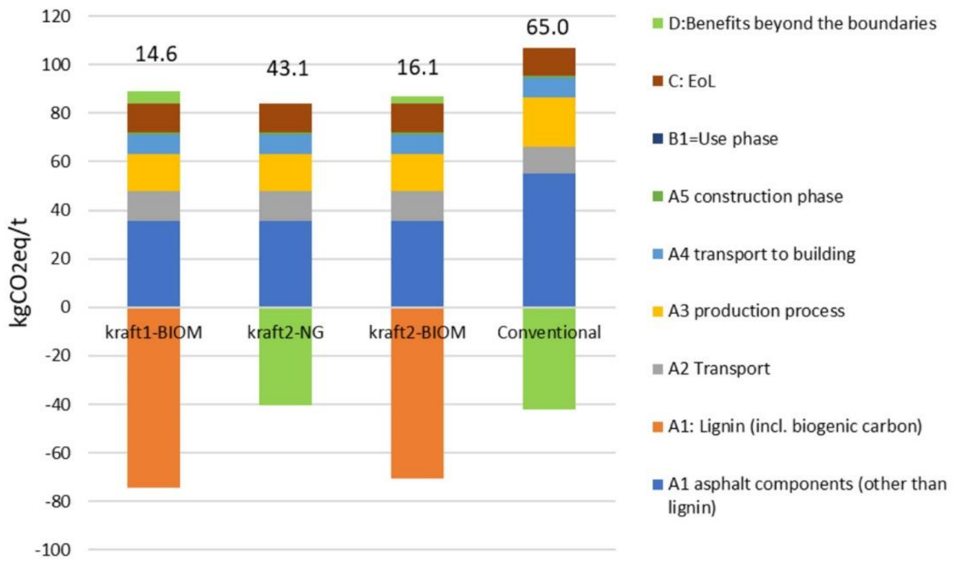


Figure 6.2 Cradle-to-grave climate change impact of 1 tonne SMA top-layer asphalt compared to conventional asphalt (Moretti et al. 2022)

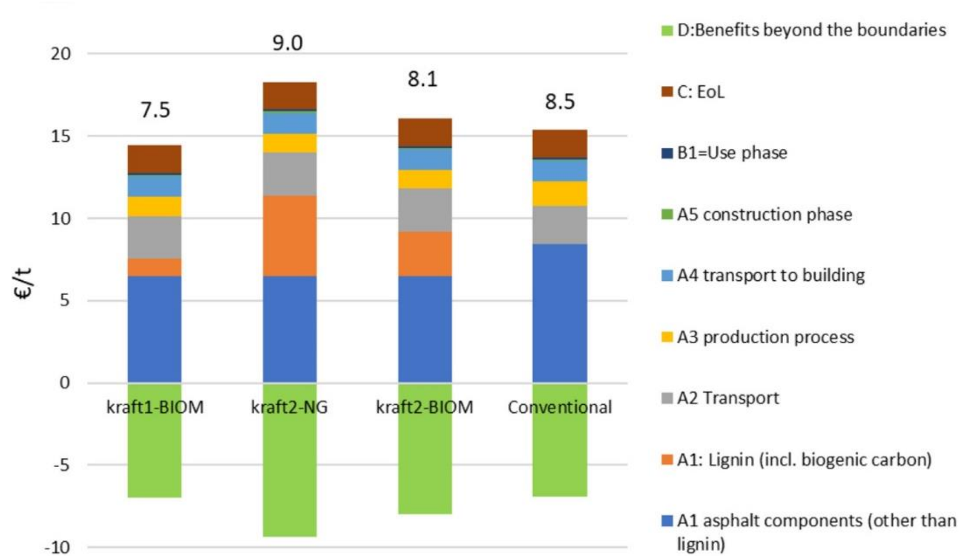


Figure 6.3 MKI score of 1 tonne SMA top-layer asphalt compared to conventional asphalt (Moretti et al. 2022)

Task 6.2 (techno-economic assessment) and Task 6.3 (Roadmap for deployment)

The current production and pavement cost of lignin-based asphalt are calculated to be 7 €/t to 17 €/t higher compared to conventional asphalt. However, future lignin-based asphalt, mainly SMA, could reach cost-price parity with bitumen-based asphalt in the future, but at least beyond 2035 as shown in Figure 6.4. When financial support is provided to cover the price difference between conventional and lignin-based asphalt, these markets could develop with lignin demand increasing to 61 -121 kt/y by 2050 according to the scenarios. These volumes are in range with today's global commercial production of lignin at 125 kt/y. However, it is only a minor fraction of the total amount of Kraft lignin extracted (as black liquor) and currently used for internal energy for running the pulping and paper making process. Furthermore, many other types of lignin are expected to come available in the future that could supply to new bio-based markets including asphalt, in particular from biorefineries. The annual required additional cost compared to baseline for lignin-based asphalt increase to M€ 7.5 (BE) and M€15 (A) by 2050 (Figure 6.5). A rough estimate of avoided GHG emissions indicates savings that could increase to between 85 (BE) and 170 (A) kt CO_2eq/y by 2050, compared to 550 kt CO_2eq/y for the asphalt sector today[1].

[1] Korevaar, Gijsbert, and Kornelis Blok (2018). "Op weg naar een klimaatneutrale infrasector in Nederland"

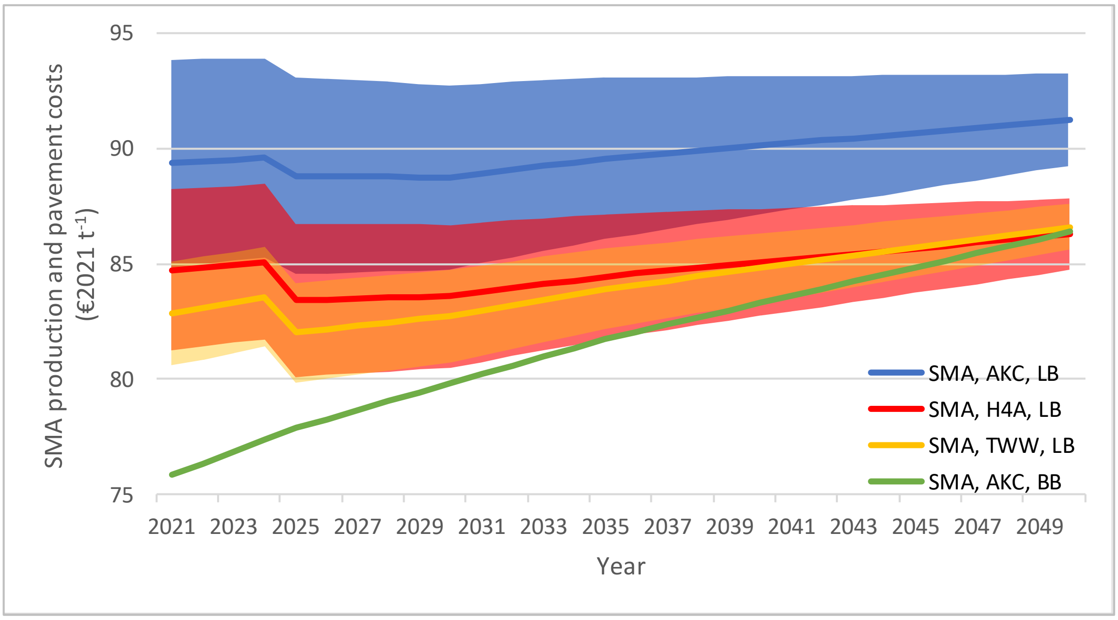


Figure 6.4 SMA top layer production and pavement cost development over time (€/tonne)

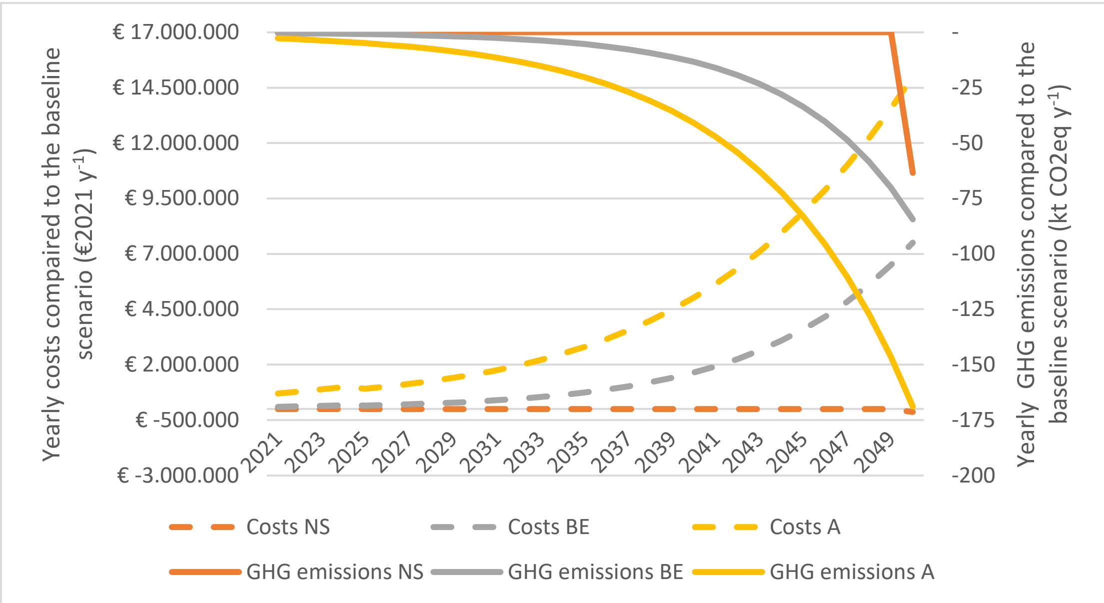


Figure 6.5 Annual costs and GHG emissions of the scenarios compared to the baseline scenario with conventional asphalt top-layers.

6.4 Discussion, conclusions and recommendations

The results of WP6 have demonstrated that significant GHG savings can be achieved if bitumen binders in asphalt are replaced with lignin. The GHG footprint of lignin-based asphalt in top-layers is between 30 to 75% lower than conventional asphalts. High GHG savings can only be achieved if lignin, that is currently used for internal energy supply, is replaced with renewable energy sources such as low-grade biomass. Other environmental impact categories show a mixed picture. This is also the reason that the difference between lignin-based asphalt and conventional asphalt is smaller for the in MKI score that aggregates the weighted impact of 11 impact categories based on its shadow price. The current production and pavement cost of lignin-based asphalt are calculated to be 7 €/t to 17 €/t higher compared to conventional asphalt. The net difference in MKI score between lignin-based and conventional asphalt top-layers is small, and does, even in the best case, not make up for today's difference in production and pavement cost.

The future scenarios are exclusively developed for surface layers in accordance with the scope of Chaplin-TKI. However lignin could also be applied to base and bind layers. The overall indicative GHG savings potential is significant: 84 kt CO_2eq/yr in Best Estimate to 170 kt CO_2eq/yr in Ambitious scenario by 2050 compared to the Baseline scenario. It would come at an estimated cost effectiveness of 106 to 111€/ t CO_2eq in the Best Estimate and Ambitious scenarios respectively[2].

The future GHG savings of lignin-based asphalt are only indicative because they calculated based on the current environmental performance as presented in Moretti et al (2022). Future research should also take other measures into account to reduce the GHG footprint and increase the circularity of the road construction sector and assess how lignin could be effectively integrated in combination with other measures. For example, rejuvenators could improve the durability of asphalt layers and increase recycling rates. Furthermore, the impact of alternative production processes or low-carbon heat and energy sources in asphalt production are relevant to assess in combination with lignin-based binder applications.

Finally, future research should focus on the possibilities to deploy biobased asphalt in the remainder of EU (and beyond), increase the share of bio-based binders, and aim to lower the additional cost. Also, the hotspots causing impacts in categories such as toxicity, eutrophication and acidification should be further investigated and addressed. However, these are mainly related to the lignin production, and as such outside the realm of control of the asphalt sector.

6.5 References

The results of WP6 are published in two open access scientific journal publications and a public report:
Scientific journal publications (open access):

- Moretti, C., Corona, B., Hoefnagels, R., Vural-Gürsel, I., Gosselink, R. and Junginger, M., 2021. Review of life cycle assessments of lignin and derived products: Lessons learned. *Science of the Total Environment*, p.144656.
- Moretti, C., Corona, B., Hoefnagels, R., van Veen, M., Vural-Gürsel, I., Strating, T., Gosselink, R. and Junginger, M., 2022. Kraft lignin as a bio-based ingredient for Dutch asphalts: An attributional LCA. *Science of the Total Environment*, *806*, p.150316.

Public report:

- Marco van Veen, Ric Hoefnagels, Christian Moretti, Iris Vural Gürsel, Richard Gosselink, Martin Junginger. Lignin based asphalt in the Netherlands –An outlook to 2030 and beyond, Report of WP6.2 Techno-economic assessment (TEA) and WP6.3 Roadmap for deployment. Public report, 2022.

[2] Calculated based on the net difference in cumulative cost and GHG savings over the period 2020 – 2050 between the Baseline and Best Estimate or Ambitious scenario. Details are available in Veen et al. (2022).

7 Work package 7 Project management, dissemination and roadmap

7.1 Objectives & deliverables WP7

The main objectives of WP7 were:

- To execute the scientific and content management of the project
- To organise both the external project communication and internal/external dissemination of project results
- To create a plan for the exploitation of the CHAPLIN technology. Since already two patent application have been filed new results will be evaluated for additional patent filing.

Deliverables:
D7.1: Annual report (M12)
D7.2: Final report ultimately 13 weeks after finishing project.
D7.3: Project leaflet at project start and by the end of the project
D7.4: TKI/BBEG Event contributions
D7.5: Conference Paper/Poster
D7.6: National stakeholder workshop
D7.7: Exploitation CHAPLIN results

7.2 Description of work

Task 7.1. Project Management Activities (M1-M24)
Activities were:

- A kick off meeting was organized by WFBR in Wageningen
- Monitoring the progress of the project: every 6 month a consortium meeting was organized. In between WP leader meetings were organized. Additionally smaller group meetings related to a WP or specific task were organized.
- An annual progress report was prepared and send to RVO by WFBR with input of all the partners
- A final report (task 7.2) will be submitted longer than 13 weeks after ending of the project. Also the financial report and the final grant fixation request will be send to RVO within 13 weeks after the end of the project by WFBR with input of all the partners.
- For the consortium members a dedicated team site was organized and hosted by WFBR.
- WFBR will make a communication note at the start of the project about the scope and deliverables of the CHAPLIN-NL project which will be sent to relevant stakeholders.

Task 7.3 Dissemination Activities (M1-M24)

- Major project results will be submitted for an oral lecture at a suitable international congress/symposium.
- Contributions to the TKI-BBEG events in 2020 and 2021 were not made as these events were not organized (D7.4).
- Four national stakeholder workshops named CHAPLIN Days were organized.

Task 7.4: Exploitation of CHAPLIN results (M12-M24)

- A CHAPLIN technology exploitation plan will be prepared by BBD taking the already filed patent applications into account. Reports for the customer (RVO/TKI BBEG), project communication materials, dissemination activities, and the results of the exploitation of the CHAPLIN technology will be reported together with the follow-up activities.

7.3 Results

Task 7.1. Project Management Activities (M1-M24)

The following meetings were held with the consortium:

- Kick off meeting, 24 February 2020, Wageningen (physical)
- 1st Progress meeting, 8 June 2020 online
- 2nd Progress meeting, 26 November 2020 online
- 3rd Progress meeting, 31 March 2021 online
- 4th Progress meeting, 29 September 2021 online
- Final meeting, 8 December 2021 online
- WP leaders meeting, 2 March 2021 online
- WP leaders meeting, 23 June 2021 online
- Several smaller group meetings for WP or specific tasks/topics including the extension of the project in 2022
- All information shared at the above mentioned meetings can be found on the team site of CHAPLIN TKI.

Task 7.3 Dissemination Activities (M1-M24)

The start of the project has been communicated via the WFBR website:
https://www.wur.nl/nl/nieuws/Bio-asfalt-op-basis-van-lignine-krijgt-flinke-zet-in-de-rug.htm
https://www.wur.nl/nl/project/TKI-project-CHAPLIN-Collaboration-in-aspHalt-APplications-with-LIgniN.htm

Publications of peer reviewed papers in International open-access journals:

Moretti, C., Corona, B., Hoefnagels, R., van Veen, M., Vural-Gürsel, I., Strating, T., Gosselink, R., Junginger, M. (2022) Kraft lignin as a bio-based ingredient for Dutch asphalts: An attributional LCA. Science of the Total Environment, 806, 150316. DOI: 10.1016/j.scitotenv.2021.150316 (related to WP6).
Moretti, C., Corona, B., Hoefnagels, R., Vural-Gürsel, I., Gosselink, R., Junginger, M. (2021) Review of life cycle assessments of lignin and derived products: Lessons learned. Science of the Total Environment, 770, 144656. DOI: 10.1016/j.scitotenv.2020.144656 (related to WP6).
Moretti, C., Hoefnagels, R., van Veen, M., Corona, B., Obydenkova, S., Russel, S., Vural-Gürsel, I., Jongerius, A., Junginger, M., Using lignin from local biorefineries for asphalts: LCA case study for the Netherlands. Journal of Cleaner Production (*accepted 19 Febr. 2022*).

Publication(s) in preparation:

Sayeda Nahar, Ted M. Slaghek, Dave van Vliet, Ingrid K. Haaksman, Richard J.A. Gosselink, Mutual compatibility aspects of (modified) lignin with bitumen as asphalt binder (related to WP2) *under review September 2022*.

WOW juryprijs:
Beide CHAPLIN projecten (TKI BBEG en XL DEI+) werden genomineerd voor de WOW prijs. Naast de opname van de WOW-podcast voor de WOW-dag 2021, werd de WOW juryprijs toegekend. Op 5 November 2021 is op de nationale WOW-dag het CHAPLIN project gekozen als prijswinnaar door de vakjury, hetgeen tot extra aandacht in de pers heeft geleid. De jury benadrukte de zeer brede en succesvolle samenwerking tussen diverse partijen, de grote innovatie van biobased asfalt, de duurzaamheid van de oplossing en tevens de schaalbaarheid en het feit dat hiermee een significante

bijdrage kan worden geleverd aan het tegengaan van klimaatverandering en het verduurzamen van de wegenbouw.

D7.5 Conference papers:

Minimising environmental impacts while maximising resource circularity: the case of biobased asphalt. Circular@WUR Conference, 6 – 8 December 2021 (postponed to 2022). Corona B, Moretti C, Hoefnagels R, van Veen M, Vural-Gürsel I, Junginger M
Exhibition (EUBCE) in May 2021 with presentation by Joop Groen at the Industry day
Climate change hotspots and consequences from kraft lignin valorization: a matter of methodological choices. Moretti C., Corona B., Junginger M. & Hoefnagels R. visual presentation at EUBCE 2021
Jeroen Besamusca, Paul Landa, Rop Zoetemeyer, Richard Gosselink, Bram Lommers, Martin Junginger, Martijn Verschuren, The use of lignin as bio-binder in asphalt applications, Conference Paper, June 2021 7th Eurasphalt & Eurobitume Congress (online)
Jeroen Besamusca, Paul Landa, Rop Zoetemeyer, Richard Gosselink, Bram Lommers, Martin Junginger, Martijn Verschuren, The use of lignin as bio-binder in asphalt applications, Proceedings of the 7th Eurasphalt & Eurobitume Congress v1.0, Madrid May 2020 (online), first published 1st July 2020, ISBN: 9789080288461

Report:
Type: English Report
Title: Comparing the production, life cycle costs and environmental life cycle costs of bitumen-based asphalt with 2nd generation biorefinery lignin-based asphalt. van Veen, M., Moretti, C., Corona B., Hoefnagels, R., Junginger, M., Vural-Gürsel, I., Jongerius, A., Russell, S. , and Obydenkova, S.

D7.6 The results of CHAPLIN TKI have been presented by the coordinator at the following events:

- CHAPLIN expeditions days for stakeholders and consortium: 4 March 2020, 7 October 2020, 8 June 2021, 15 December 2021
- CHAPLIN stakeholder meeting 16 December 2020

The realization of the demonstration roads, reported in WP4, have been disseminated extensively as can be found hereafter:

Municipality Bergen op Zoom (Klutsdorpseweg Lepelstraat), 7 October 2021

- https://www.bndestem.nl/bergen-op-zoom/br-groen-asfalt-voor-klutsdorpseweg-bij-lepelstraat-een-klimaatvriendelijk-alternatief~afb9afa3/
- https://www.ad.nl/bergen-op-zoom/br-groen-asfalt-voor-klutsdorpseweg-bij-lepelstraat-een-klimaatvriendelijk-alternatief~afb9afa3/
- https://www.zuidwestupdate.nl/nieuws/innovatief-asfalt-op-klutsdorpseweg-in-lepelstraat-net-zo-goed-en-het-liefst-nog-beter/
- https://www.halsterse-zuidwestkrant.nl/nieuws/algemeen/22234/uniek-asfalt-op-klutsdorpseweg
- https://www.duurzaambergenopzoom.nl/2021/10/13/groen-asfalt-in-bergen-op-zoom/
- https://www.internetbode.nl/regio/bergen-op-zoom/346693/unieke-weg-aangelegd-in-lepelstraat-klimaatvriendelijk-alternat
- https://www.linkedin.com/feed/update/urn:li:activity:6851913331188539392/
- https://www.linkedin.com/feed/update/urn:li:activity:6851904189988143104/

Municipality Altena (Midgraaf Almkerk), 1 November 2021

- https://www.stichtingmilieunet.nl/andersbekekenblog/duurzaam/chaplin-weg-van-bio-asfalt-in-almkerk-gemeente-altena-25-proefstroken-in-nederland.html
- https://www.altenatv.nl/2021/11/02/bio-asfalt-aangebracht-aan-de-midgraaf-in-almkerk/
- https://www.ad.nl/rivierenland/rijden-over-een-snelweg-gemaakt-van-planten-en-bomen-almkerk-krijgt-een-stuk-weg-van-bio-asfalt~a778066e/

- https://www.bndestem.nl/rivierenland/rijden-over-een-snelweg-gemaakt-van-planten-en-bomen-almkerk-krijgt-een-stuk-weg-van-bio-asfalt~a778066e/?cb=0907b34ed56c6b3bfd67cb356ef51aff&auth_rd=1
- https://www.altena.net/nieuws/gemeente-legt-proefstrook-aan-op-midgraaf-met-bioasfalt-2021-11-01
- https://drimble.nl/regio/noord-brabant/waalwijk/80224883/bio-asfalt-aangebracht-aan-de-midgraaf-in-almkerk.html
- https://altena.nieuws.nl/gemeente/985/uniek-stuk-biobased-weg-in-almkerk/
- https://www.hetkontakt.nl/regio/heusdenenaltena/294733/bio-asfalt-in-altena
- https://www.streek.nu/altena/uniek-stuk-biobased-weg-in-almkerk/

7.3.1 Exploitation of CHAPLIN results & CHAPLIN roadmap for further testing of transects

Based on input from all participants in the CHAPLIN Expedition day, which digitally took place on December 15, 2021, the following priorities have been ranked for matters in their opinion that still need to be investigated/addressed after the project (in brackets the priority score):

1. Lignin quality vs bitumen and asphalt quality, variability (54%)
2. Measuring and monitoring test strips (54%)
3. 100% biobased asphalt technology (39%)
4. Environment and health (29%)
5. Recyclability (29%)
6. Bring first formulations to TRL 9 (29%)
7. Total cost optimization (25%)
8. Improved formulation, premium properties (25%)
9. CO_2, GHG emissions reduction (21%)
10. Lignin volume suppliers and market (4%)

In addition to identifying the issues that are desirable for further investigation and prioritization, the participants in the CHAPLIN program were also asked whether they would like to participate in the program after 2021. To date, most parties have confirmed that they will continue to participate. Preparations are being carried out for a CHAPLIN EU project and in the Growth fund proposal "Materialen-NL" there is a section on biobased asphalt where CHAPLIN is specifically mentioned.

The challenge of making 100% biobased asphalt has already been taken up by some participants in another TKI AF project called "Fully biobased binder for asphalt concrete".

Along with Covid-19 related issues, during scale-up Praj ran into problems related to logistics. Now they are closer to a solution and they hope to finish the production of 2 ton of hydrolysis lignin by mid-April 2022.

Agreements have been made with BouwCirculair to set up a testing ground (a cluster of projects around one circular innovation/theme in construction in order to validate it) for lignin-containing asphalt. The first steps have been taken by drawing up a draft protocol. This protocol shows the generic way in which Living Lab projects are assessed. The objective of the protocol is to guarantee the quality of the monitoring and assessment of the Living Lab projects. Based on the provided substantiation of the project performance, it can be determined whether the innovation can achieve comparable performance compared to traditional implementation.

8 Conclusions and recommendations

Several technical lignins, from the Dutch biorefinery processes (executed by Avantium, Vertoro, BBD) were evaluated as partial substitute for bitumen. Most lignins are suitable for this applications. The process of the BBD was similar to the hydrolysis process of Praj and these lignins passed the requirements for application on the road. However, demonstration of this lignin could not be performed within this project as a delay of the delivery from India was faced. An Avantium lignin was successfully tested in a demo road in the CHAPLIN XL project. The order of addition in the asphalt mill for the realisation of a suitable asphalt mixture is critical and more lessons were learned from this.

Health and safety aspects in the production and pavement of lignin-based asphalt can be realised. Release of volatiles such as formaldehyde do not exceed the threshold value for a safe working environment.

Two demonstration roads have been successfully paved in this project, e.g. in Altena and in Bergen op Zoom. Together with the other CHAPLIN project more demonstration roads have been realised. The demonstration road in Gelderland was not successful as the mixing procedure in the asphalt mill was not optimal, which gave a lot of learning aspects.

Monitoring of the currently paved demonstration roads needs to be continued in order to have complete insight into the durability of the lignin/bitumen binder in the user phase. Based on a limited set of data samples, the expected life time of lignin-based asphalt is somewhat shorter (ca. 1.5 year) than for asphalt with a conventional bitumen binder.

Also transferring the technology described in WO 2019/092278 should be incorporated in a 2 km road for a more extensive testing of e.g. reduced fuel consumption and noise reduction. These activities should lead to a TRL 9 level of the WO 2019/092278 technology. The next step would be to introduce the technologies in other countries via an e.g. EU project and collaborations with companies in other climatologically different regions.

The sustainability of lignin-based asphalt compared to traditional asphalt is more favorable due to the large reduction of carbon dioxide emissions (35-70%). The current production and pavement cost of lignin-based asphalt are calculated to be 7 €/t to 17 €/t higher compared to conventional asphalt. The net difference in MKI score between lignin-based and conventional asphalt top-layers is small, and does, even in the best case, not make up for today’s difference in production and pavement cost.

The future scenarios are exclusively developed for surface layers in accordance with the scope of Chaplin-TKI. The overall indicative GHG savings potential is significant: 84 kt CO2eq/yr in Best Estimate to 170 kt CO2eq/yr in Ambitious scenario by 2050 compared to the Baseline scenario. It would come at an estimated cost effectiveness of 106 to 111€/ t CO2eq in the Best Estimate and Ambitious scenarios respectively.

Overall, the results obtained in CHAPLIN-TKI and also reflecting part of the work in CHAPLIN XL, showed that for lignin-based asphalt positive scenarios have been identified. It became also clear that this development needs further research and development to optimize the use of lignin in an asphalt binder. Finally, future research should focus on the possibilities of deploying biobased asphalt in the remainder of EU (and beyond), increasing the share of bio-based binders, and aim to lower the additional cost. Also, the hotspots causing impacts in categories such as toxicity, eutrophication and acidification should be further investigated. However, these are mainly related to the lignin production, and as such, outside the realm of control of the asphalt sector.

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9 Annexes

- Annex 1 Rapportage M1 tm M4 ZOAB 16 Lignine (will not be disclosed)
- Annex 2 Rapportage M7 tm M12 SMA 11 Lignine (will not be disclosed)
- Annex 3 Rapportage M13 tm M15 AC 11 Surf, 30% PR, Lignine (will not be disclosed)
- Annex 4 ARBO metingen Formaldehyde