Project Number: 5-21295.00

Bitumen Alternatives

31 August 2021

150 YEARS IN AOTEAROA

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Document Details:

Date: 8 March 2022
Reference: 5-21295.00
Status: Final

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Document History and Status

Revision	Date	Author	Reviewed by	Approved by	Status
1	20 May 2021	Lia van den Kerkhof, Phil Herrington	Phil Herrington		Draft
2	29 June 2021	Lia van den Kerkhof, Phil Herrington	Jeremy Wu		Draft Final
3	31 August 2021	Lia van den Kerkhof, Phil Herrington	Jeremy Wu	Wendy Turvey	Final
4	8 March 2022	Lia van den Kerkhof, Phil Herrington	Jeremy Wu	Wendy Turvey	Final

Revision Details

Revision	Details
1	Technical edits
2	Technical edits
3	Minor referencing and grammar editing.
4	Minor correction – page 5

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Disclaimers and Limitations

This report ('**Report**') has been prepared by WSP exclusively for Waka Kotahi New Zealand Transport Agency ('**Client**') in relation to alternatives to bitumen ('**Purpose**') and in accordance with the Independent Professional Advisors (IPA) Contract NO 17 – 286 dated 23rd March 2021. The findings in this Report are based on literature collected by the project team. WSP accepts no liability whatsoever for any reliance on or use of this Report, in whole or in part, for any use or purpose other than the Purpose or any use or reliance on the Report by any third party.

1 Introduction

This review investigates potential alternatives to petroleum bitumen for construction of road surfacings in New Zealand. The review provides a high level overview of available materials and materials in development. Factors considered, where information is available, include likely costs, availability/supply, performance, limitations, environmental implications, and carbon footprint.

Bitumen is the residue remaining from the distillation of crude oil to make petrol, kerosene and other petroleum products. Approximately 110 million tonnes of bitumen is produced annually worldwide- most of which is used in road construction applications. About 150-170,000 tonnes is used annually in New Zealand, and this is now all directly imported into New Zealand. However, bitumen supply and price is closely tied to that of crude oil. Changes in Government policy internationally in response to climate change effects are leading to growth of renewable (or nuclear) power generation and the move to electric vehicles. These changes are likely to ultimately result in reduced crude oil refining and hence bitumen production, leading to escalating prices.

An alternative material for road surfacing construction could help insulate New Zealand from future price fluctuations. A locally produced bio-mass derived material may also help reduce the carbon footprint of road construction with the surfacing becoming a mechanism for carbon sequestration.

2 Bitumen used for road construction

2.1 Sources and origins

Bitumen used for road construction is obtained as a by-product of the refining of crude oil (petroleum). The crude oil is mixed with water to remove salts then heated to distil the lighter fractions (petrol, kerosene etc.). Softer grades of bitumen (such as the 180-200 penetration grade used for chip sealing in New Zealand) are produced as the residue from distillation at temperatures up to 350 °C and at pressures of less than one mbar. Harder grades are produced by air-blowing (oxidation) or by mixing with residue from solvent de-asphalting processes applied to the vacuum residues (Speight, 1999).

Crude oil varies in composition depending on the geographical source but bitumen typically comprises less than 15% of crude oil by weight.

2.2 New Zealand bitumen requirements and supply

New Zealand uses approximately 150,000-170,000 tonnes of bitumen annually for asphalt and chip sealing. There are numerous other commercial uses for bitumen internationally (Hunter et al., 2015) including sealants, and roofing products but manufacturing for these applications is negligible in New Zealand and such products are imported already made up.

From the 1970's up until the mid-1990's all New Zealand bitumen was produced at the New Zealand Refining Company (NZRC) Marsden Point Refinery and from a limited slate of middle eastern crudes (in particular Safaniya). Semi-regular imports of bitumen began in the 1990's of bitumen from Venezuelan and Asian crude sources.

Until January 2021 about two thirds of the New Zealand bitumen supply was still manufactured at Marsden Point. The refinery produced 180-200 and 40-50 penetration grades to meet the Waka Kotahi New Zealand Transport Agency (WK-NZTA) M01 specification on behalf of Z Energy Ltd. The two grades were then blended to produce the intermediate grades. The 180-200 grade was produced by vacuum distillation and the 40-50 grade manufactured using air blowing and solvent precipitated asphalt. The Marsden point refinery has now ceased production of bitumen which is now imported (Z Energy is the main importer).

2.3 Implications of a bitumen supply shortfall

New Zealand has a combined bitumen storage capacity of about 120,000 tonnes (Prendergast, 2021). This constitutes about 9 months' supply. In practice our effective capacity is less than this as multiple grades of bitumen are required. Bitumen shipments require specialised freight vessels and the import lead time is in the order of several months. The size of shipments (typically 5-20,000 tonnes) is limited by current storage and distribution capacity. A break in this supply chain of even one or two shipments would thus have significant consequences for the contracting industry. In the longer term obviously the performance of the network would also suffer from reduced maintenance.

2.4 Decreasing global demand for crude oil

It is important to note that bitumen is effectively extracted from crude oil – not manufactured. Numerous processes have been developed with the goal of producing lighter (more volatile) fuel fractions from crude oil (e.g. cracking, hydrocracking) but there are no processes available to convert lighter fractions to usable roading grade bitumens. Based on US refinery production, currently 75% of crude oil is used for transport and heating fuels (Administration, 2021).

If global demand for transport and heating fuels decreases significantly, as is predicted over the next 30 years, the economics of oil refining will change which has important implications for bitumen supply and cost.

In OECD countries demand is predicted to drop by 17% by 2045 (IEA, 2020). Even BP (BP, 2021) and (OPEC, 2020) predict global demand to peak in 2030-2040 and decrease by 5-70% by 2050.

A reduction in crude oil demand and production is likely for several reasons: replacement of petrol driven vehicles with electric vehicles, more use of renewable energy generation, reduction of single use plastics and improved efficiency standards for internal combustion engines. At the same time as oil production is predicted to decline, the demand for bitumen is projected to increase, particularly in Asia. The most likely outcomes from this scenario are increased costs of production, bitumen shortages, and a significant cost increase for bitumen in New Zealand.

2.5 Carbon footprint

Bitumen production has a significant carbon footprint. Now that the NZRC refinery has stopped bitumen production, all bitumen is imported to New Zealand as a refined product. This may then be further modified by blending or addition of polymers or other additives to bring it "on-grade" in terms of the NZTA M01 and M01-A specifications. The key steps are involved are:

- Crude oil extraction and preparation for shipment
- Crude oil transport to a refinery (may involve shipping and long-distance pipeline transfer)
- Refining and processing into bitumen and storage
- Transport to New Zealand by sea
- Storage and possible further processing in New Zealand to produce an on-grade bitumen

The CO_2e produced per tonne of bitumen will obviously vary depending on the crude oil source, refinery location, ship capacity etc. New Zealand is at a disadvantage in that we are a very remote and small market (limiting the size of shipping). A number of studies have been undertaken to assess emissions from bitumen manufacture and which are discussed below, note that the figures quoted are operational and do not include emissions resulting from construction of refinery and other necessary infrastructure. Most of the energy needed in the supply chain is obtained by burning diesel or other fossil fuels- little from renewable energy sources.

Downer (2013) undertook a carbon footprint study comparing the life-cycle emissions of hot cut-back bitumen (i.e. with added kerosene) to that of bitumen emulsions in New Zealand. A figure of 438.5 kg CO2e per tonne was quoted for bitumen raw material acquisition, but this figure includes a contribution for kerosene and adhesion agent (probably about 4% of the total mass). Although not stated it is likely that the calculations carried out in 2013 assumed bitumen supply from the NZRC refinery that was then shipped around New Zealand by sea. In the current situation significant additional shipping distances may be involved in transporting crude oil to an overseas refinery and thence to New Zealand so the actual figure is likely to be higher than that calculated.

Blaauw et al. have undertaken a study for bitumen used in South Africa (Blaauw et al., 2020). Crude oil is shipped to South Africa and refined either at coastal refineries or piped to inland refineries. Figures of 221.89 and 233.36 kg CO_2e per tonne were reported inland and coastal plants respectively.

The European Bitumen Association have produced a detailed study on CO2e produced for bitumen supply in Europe. As in South Africa some crude oil is supplied by sea and some by pipeline (from the former Soviet Union). An overall figure of 149.6 kg CO_2e per tonne was calculated (this increased to 207.5 kg CO_2e per tonne if emissions from infrastructure are included).

In the US and Canada about 85% of crude oil refined is produced locally and of that about 44% is from unconventional sources , mainly shale oil (Asphalt Institute and Thinkstep, 2019, US Energy Information Administration, 2020).

A study conducted by the US Asphalt Institute (an industry organisation) calculated a value of 637 kg CO_2e per ton. The bulk of this was due to crude oil extraction and was high due to the significant amount of shale oil extracted. Extraction of non-conventional, synthetic crude oils requires significant additional energy inputs compared to conventional sources, (Charpentier et al., 2009).

Given that New Zealand uses about 160,000 tonnes of bitumen per annum and using the range of the values above then the total CO_2e emitted as a result would range from about 24,000 tonnes up to 100,000 tonnes. To put this in perspective New Zealand's total net CO_2e emissions were 78 million tonnes in 2018 (Statistics NZ, 2020).

3 Bitumen alternatives – requirements

Ideally, a material suitable as a complete, sustainable and secure replacement for imported petroleum bitumen (or significant (>20%) partial replacement) would need to meet the following requirements:

- Result in similar or lower life-cycle costs than those achieved with bituminous surfacings.
- Be able to be produced, preferably in New Zealand, in sufficient volume and with a sustainable supply chain in the long-term (i.e. tens to hundreds of thousands of tonnes p.a.)
- Have an equivalent or preferably lower carbon foot-print (i.e. it should not add to New Zealand's net CO_2e emissions).
- It should not leach or emit through volatilisation, environmentally harmful species.
- It should be recyclable (i.e. reusable as a road surfacing binder).
- Have similar or better physical properties and durability than bitumen. It must be water insoluble, non-biodegradable over the life of the surfacing.
- Be compatible with existing bitumen handling and surfacing construction plant.
- Be non-toxic or hazardous to workers or the public over the entire life of the surfacing

4 Bitumen modification at low concentrations

There is a very substantial literature dealing with addition of various virgin and recycled or waste materials or natural products as a means of modifying the physical properties of bitumen and /or a means of disposing of a given material.

Whilst potential benefits to physical properties or durability are sometimes demonstrated these applications do not appear to offer a realistic path to development of a bitumen alternative in the foreseeable future or even for substantial bitumen substitution, as the useable concentrations are usually much less than 10%.

Examples are the use of coffee grounds (Zofka and Yut, 2012), cigarette butts ((Rahman et al., 2020), cashew nut shell oil (Albrecht Supply Concepts, 2021), poppy capsule pulp oil (Gürer et al., 2020), waste cooking oil (El-Shorbagy et al., 2019, Asli et al., 2012), Damar resin (Verma and Chauhan, 2019), sargassum (Salazar-Cruz et al., 2021) and colophony (Putri et al., 2020). Many such applications are aimed at disposing of waste from relatively small scale, geographically localised, specialised industrial or agricultural processes and the probability of a suitable supply chain being developed capable of delivering large volumes, is highly unlikely.

5 Bitumen alternatives – non-renewable materials

5.1 Coal Tar

Coal tar is produced from the destructive distillation (carbonisation) of coal (in the absence of air). It was widely used for road construction internationally and in New Zealand up until the 1970's but is typically more temperature sensitive than bitumen, making it less desirable for road surfacings (Depree and Frobel, 2009, Road Research Laboratory, 1962). Internationally, coal tar still has application in some specialist surfacings due to its resistance to fuels.

The chief drawback with possible greater use of coal tar for road surfacings is its toxicity. Coal tar is carcinogenic due to high concentrations of polyaromatic hydrocarbons (Metre et al., 2009, Jang et al., 2018). Polyaromatic hydrocarbons, phenolics and other harmful chemicals from coal tar road surfacings are known to leach into the environment (Depree and Frobel, 2009).

Historically coal tar was produced largely as a by-product of the manufacture of coke and coal gas for home heating and lighting, it is not currently manufactured in New Zealand and internationally is no longer used to any extent for road construction. Globally about 20 million tonnes is still manufactured annually, principally in China, for a variety of industrial applications.

5.2 Concrete

Internationally concrete is widely used to construct both road pavements and surfacings but overall the percentage is still small compared to bituminous materials. In the US approximately 17% of the highway network has a concrete base but much of this has a bituminous surfacing (Horvath and Hendrickson, 1998, Zapata and Gambatese, 2005). In New Zealand concrete has not been used to any extent on the road network and the specialised plant and expertise required for concrete road design and construction is largely absent in the sector.

Modern concrete road construction techniques include continuously reinforced concrete roads which eliminate the noise/vibration and joint sealing problems of jointed surfaces. Concrete roads are typically considered to be very expensive and disruptive to construct but have longer lifetimes and reduced maintenance compared to asphalt pavements/surfacings. If that is the case then an argument can be made that concrete roads may be more cost effective than asphalt mix (not chip seal) in some cases (Infometrics, 2020). However, the relative merits of concrete roads in terms of performance (e.g. maintenance of skid resistance) and lifetime are still the subject of debate and performance on the New Zealand road network unknown. Studies in the US, provide contradictory findings regarding comparative lifetimes of concrete and asphalt roadways (Zapata and Gambatese, 2005).

Noise generation from concrete road surfacings is also traditionally seen as a drawback and would be difficult to reconcile with the NZTA's emphasis on the use of OGPA surfacings for noise reduction (Gribble, 2018).

The CO_2 emissions from Portland cement manufacture (and steel for reinforced concrete) are also often seen as a disadvantage for concrete road construction. Carbon dioxide is produced both by the chemical reaction to produce Portland cement and also by the fuel needed to produce the heat required for the process. Values of about 500 to 1000 kg CO_2e per tonne are typical for cement manufacture internationally (Barcelo et al., 2014, Hammond and Jones, 2011). These values are higher but not too dissimilar to those discussed above for bitumen.

In their Environmental Product Declaration, New Zealand's largest concrete manufacturer Firth Industries, state that the CO2e emissions for concrete manufacture ranges from 199 to 495 kg m^{-3} or about 83 to 206 kg per tonne (assuming a concrete density of about 2.4 tonnes m^{-3}). An equivalent figure of 38 kg CO_2e per tonne was reported for typical hot mix asphalt production (5% bitumen) in New Zealand based on data provided by Fulton Hogan Ltd (Ball, 2010). The latter figure is comparable to a more recent figure of 43 kg CO_2e per tonne calculated from data provided by Ma et al. (Ma et al., 2016) for a case study of highway in China (that utilised a component of polymer modified binder with a higher carbon footprint than conventional bitumen).

The carbon footprint of constructed roadways using concrete and asphalt mix have been compared in a number of studies (Espinoza et al., 2019, Infometrics, 2020, Brown, 2009). Such calculations depend on a wide range of factors beyond material inputs- principally pavement design (layer thicknesses etc.) design life and maintenance assumptions. Although the production of concrete produces significantly more CO_2e per tonne, the results show in some cases concrete roads can have a lower or equivalent carbon footprint than asphalt mix.

Potentially, options such as using fly ash or natural pozzolans from volcanic ash (readily available in New Zealand) to partially replace cement or the use tyre derived fuel (TDF) from waste tyres (about 30% renewable natural rubber) exist to lower the carbon footprint of concrete (Thinkstep, 2019).

From a technical perspective, concrete is a practical alternative to bituminous road surfacings, even if the relative merits in terms of performance in terms of noise and skid resistance are still the subject of debate. The relative carbon footprint of the materials is difficult to assess without a more detailed study of likely construction scenarios specific to New Zealand.

5.3 Sulphur

Globally, tens of millions of tonnes of elemental sulphur are produced annually. Most comes as a by-product of crude oil refining and natural gas processing but substantial amounts are produced during the smelting of metals.

Sulphur melts at about 115 °C and exists in various polymeric forms (allotropes). Its interaction and reaction with bitumen is complex. Replacement of up to 10-40% of bitumen with sulphur produces blends that have generally improved properties but suffer from phase separation and (toxic) hydrogen sulphide gas generation at mixing temperatures (Petrossi et al., 1972, De Filippis et al., 1998, Al-Dobouni et al., 2008, Gul et al., 2021, Gedik and Lav, 2016).

Sulphur has also been explored as a total replacement for bitumen in asphalt mixes. In order to produce a workable product, the sulphur was plasticised by reaction with unsaturated hydrocarbons such as dipentene. Successful field trials were conducted in the 1970's using a formulation ("Sulphlex") with 70% sulphur and 30% plasticiser compounds (Sakib et al., 2021, Little, 1986).

The use of sulphur as a bitumen extender or as a total replacement for bitumen in asphalt mixes has been investigated extensively since the 1970's due to its generally low cost relative to bitumen; currently about 20% that of bitumen (Sakib et al., 2021, Gedik and Lav, 2016). However, commercial application of these technologies has been minimal. This is likely due to the practical problems particularly with hydrogen sulphide generation. Shell produce a "Thiopave" binder which is a sulphur extended bitumen. Properties of that material have been discussed by (Urquhart, 2012, Urquhart and Malone, 2013). Given that the bulk of sulphur production is as a by-product of fossil fuel refining, the long-term supply and cost of sulphur is also uncertain.

5.4 Shale oil/oil sand bitumens

Large deposits of oil shale and "tar "sands exist world-wide. Hein et al. estimate that approximately 360 billion tonnes of recoverable oil exists (Hein, 2017). The Athabasca deposits in Canada being the most well-known and extensive (Masliyah et al., 2004). Such deposits consist of sedimentary rock or sands impregnated with organic materials ranging from very heavy oils to kerogen (solid). Open cast mining is used to extract the raw material and transport fuels and other products equivalent to those from refining of petroleum can be recovered by various processes. The characteristics of heavy bituminous fractions from these deposits or materials arising from processing and refining of the oils as roading bitumens has been investigated by a number of authors (Ishai et al., 1996, Thomas et al., 1996, Barrett et al., 1990, Petersen, 1988). Although properties vary widely depending on the shale oil source, generally shale oil bitumens have been found to be suitable as road surfacing bitumens either alone or as a blend with petroleum bitumen. Bitumens derived from shale oil have long been used extensively in Russia and Estonia for roading applications (Meshin and Purre, 1993).

Although New Zealand possesses deposits of shale oil in the South Island e.g. Orepuki on Southland's south coast, the open cast mining of this material is unlikely to be environmentally acceptable.

In a similar manner to petroleum, the economics of shale oil bitumen production are likely to be adversely affected by an overall reduction in demand for fossil fuels.

5.5 Peat

Peat consists of organic fibrous material in various stages of decomposition usually at the ground level but often extending metres below the surface. Historically, once dried it has been used as a heating fuel. It is possible to extract organic material from the peat (up to about 9% of the dry weight) which has been investigated as a partial substitute for petroleum bitumen (Leahy et al., 1990a). The extracted material is chemically very different from petroleum bitumen and has a high concentration of waxy esters. These react during aging of the material suggesting that the acceptable concentration of peat bitumen in petroleum bitumen would be less than 17% (Cavalier and Chornet, 1978, Leahy et al., 1990b). Peat bitumen has not apparently been used commercially as a bitumen substitute.

New Zealand has large areas of peatland- in particular in the Waikato. Peatland is generally highly productive as agricultural land and open cast mining of New Zealand peatland for bitumen production is a highly unlikely scenario.

5.6 Natural bitumen

Naturally occurring surface deposits of solid bituminous materials exist which are mined and used for various applications. These materials are known by a variety names such as rock asphalt, asphaltites and asphaltoids (Shepherd, 1964, Speight, 1999). They differ in terms of their chemical composition and physical properties but are typically high softening point solids infused into/with mineral material. The most significant for road surfacing applications are discussed below.

5.6.1 Asphaltites

Deposits of natural, solid, bituminous materials collectively known as asphaltites exist in a number of locations, e.g. Gilsonite (USA and Iran), Grahamite (USA), Asbuton (Indonesia) and Selenizza asphaltite (Albania). These materials are mined and processed into powders (see Figure 1). Although chemically and rheologically distinct from refined bitumen they are compatible and are usually used in small percentages as modifiers to stiffen soft grades of bitumen (Ameri et al., 2018, Nciri et al., 2014, Hunter et al., 2015). They are too stiff to use as standalone surfacing binders.

5.6.2 Lake Bitumen

As the name suggests this material is found in lakes formed by natural seeps. The largest is in Trinidad has an area of about 35 ha, is 90 m deep and contains about 10 million tonnes of material (see Figure 2). The bitumen is recovered using excavators and refined to remove water and large particles of foreign matter. The finished product is too hard to use as a standalone surfacing binder (it has about 36% mineral matter) and is used to stiffen softer grades of refined bitumen (Liao et al., 2014, Hunter et al., 2015).

Figure 1: Gilsonite powder (*source*: Nicri et al. 2014)

Figure 2: Trinidad Lake Asphalt (*source*: Nicri et al. 2014)

6 Bitumen alternatives – renewable materials

The term bio-bitumen or bio-asphalt (in the US) is used very loosely in the technical and industry literature. It is used to refer to bitumen to which a natural product of some type has been added even in very low concentrations or alternatively a binder manufactured entirely or largely from renewable materials. The renewable materials employed vary widely and are often poorly defined in terms of their origins and properties. The properties of the resulting "bio-bitumen" also vary wildly and they cannot be considered as a single entity.

6.1 Lignin

Lignin is a complex high molecular weight polymeric material found in plant material. Trees typically contain 20-30% Lignin which acts to provide structural support to the plant. Approximately 50-70 million tons of lignin are produced annually worldwide primarily as a by-product of the pulp and paper industry (Khandelwal, 2019, Gosselink, 2011). A small proportion of lignin (about 2%) is used in a wide variety of industrial applications such as coatings and plastics but the bulk is used as a fuel (Tokede et al., 2020, Gosselink, 2011).

The chemical composition, molecular weight and structure and properties of lignin varies depending on the plant source and the extraction process used. The latter include the Kraft, sulphite, soda-anthraquinone and organosolv processes, such as Alcell and Organocell (Abdelaziz et al., 2016). Most lignin is produced via the Kraft and sulphite processes. Kraft lignin is a non-fusable powder insoluble in organic solvents. The sulphite process gives rise to lignosulfanate powders which are also insoluble in organic solvents (but are water soluble). Both the Kraft and sulphite processes result in lignin with about 1-6% sulphur contents. The Organosolv type processes use solvents to extract the lignin, the product is free of sulphur and is soluble in organic solvents but is only produced in pilot scale quantities compared to the other products. Solvent solubility is a key feature indicating the way in which lignin will interact with bitumen. Organosolve lignins will also melt and flow on heating at bitumen handling temperatures (Vliet et al., 2016).

Lignin has been investigated widely as a bitumen modifier and extender over several decades ((Sundstrom et al., 1983, Vliet et al., 2016, Xie et al., 2017). Lignin is high in phenolic compounds which have been shown to have some anti-oxidant effect when added to bitumen (Xu et al., 2017, Pan et al., 2012, Arafat et al., 2019). Studies on bitumen blends with up to 25 % lignin show that the lignin has a stiffening effect at high temperatures (Vliet et al., 2016). This may require an increase in mixing and compaction temperature (Wu et al., 2021a) but could be offset by using a softer grade of bitumen for blending. Lignin-bitumen blends have been shown to perform satisfactorily although fatigue life may be adversely affected (Zhang et al., 2019, Xu et al., 2017, Norgbey et al., 2020). Given the high molecular weight it is unlikely that lignin completely dissolves in bitumen and is likely to be present as a separate phase with some swelling through absorption of bitumen species. Van Vliet found that up to 25% of various lignin types could be simply blended with bitumen without difficulty and modified the binder properties beneficially in a manner similar to that seen with conventional polymer modification (Vliet et al., 2016).

Trials using bitumen in asphalt surfacings with 50% lignin have recently been constructed in the Netherlands (Besamusca et al., 2020). Khandelwal (Khandelwal, 2019) conducted a detailed life cycle assessment (LCA) to assess the CO_2e emissions from use of a 50% lignin/bitumen binder in a constructed porous asphalt surface in the Netherlands. A reduction of 10% in CO_2e emissions would be achieved compared to conventional bitumen porous asphalt even assuming natural gas was used to replace the energy lost from not burning the lignin. The reduction would be about 75% if wood chip was used as a replacement fuel.

In New Zealand about 1 million dry tonnes of pulp are produced for sale annually (MPI, 2021) giving potentially about 1.8 million tonnes of black liquor dry solids (MBIE, 2016), about 600,000 tonnes of which is lignin. This is substantially greater than New Zealand's total bitumen demand. However most of this material is used as a fuel to power the plant so if a significant amount of the lignin was recovered and diverted to road construction an alternative fuel source would be needed.

In the study by van Vliet, various types of lignin were investigated and chemical modification of the lignin to reduce the concentration of hydroxyl groups and increase hydrophobicity, resulted in better compatibility with the bitumen (Vliet et al., 2016). Similar studies have been carried out by (Xie et al., 2017). Although yet to be fully demonstrated, in principal following this approach chemical modification may produce a thermoplastic material that could be used as a 100% substitute for bitumen (Saito et al., 2012). Incorporation (encapsulation) of lignin in bitumen removes to a large extent the problem of biodegradability by preventing contact with water but this would need to be addressed in a 100% lignin based bitumen substitute.

The cost of technical Kraft lignin (i.e. relatively pure material) is about US$250 per tonne (about NZ$360) in the US (Abbati de Assis et al., 2018, Culbertson, 2017).

6.2 Thermolytic process oils (pyrolysis, hydrothermal liquefaction)

Wood (or other plant material) when heated to above 400°C in the absence of oxygen, breaks down to form an oil. The process is known as pyrolysis. Numerous variants of this technique exist such as hydrothermal liquefaction which is a related high temperature, high pressure process for conversion of high-moisture content biomass ((Jarvis et al., 2017, Dimitriadis and Bezergianni, 2017). Other Internationally commercially operated plants exist to manufacture pyrolysis oils which have a variety of industrial uses but primarily as transport or heating fuels or chemical feedstocks (Mohan et al., 2006). These applications have also been the focus of most of the research in the field. In New Zealand small plants have operated occasionally but not at any scale, for example pyrolysis was used to process timber waste following the Christchurch earthquake to avoid sending it to landfill.

6.2.1 Woody biomass

Typically, depending on the feedstock and conditions used, fast pyrolysis processes using plant biomass produce about 40-60% organic oil, 15-25% char (see below) and about 25-35% water and fixed gases, e.g. carbon monoxide, carbon dioxide etc. (Bridgwater, 2012, Czernik and Bridgwater, 2004, Salehi et al., 2011) Pyrolysis oils are highly acidic and complex mixtures of low viscosity, but usually contain a sub-fraction of polymerised high viscosity tar material, confusingly this is sometimes referred to as "pyrolytic lignin" as some of it derives from lignin degradation products, though its origin and chemical composition bear little relation to true lignin.

Research over the past 40 years has demonstrated pyrolysis oils can be further treated to produce viscous materials similar in appearance to bitumen and with properties potentially suitable for road construction (Butte et al., 1980). Wood pyrolysis is an attractive route for a sustainable replacement for bitumen because of the large amount of waste plant material potentially available (e.g. from pine plantation leavings, sawdust). Researchers have published work on the use of wood pyrolysis oils as extenders or complete substitutes for bitumen in asphalt mixes (Raouf and Williams, 2010a, Raouf and Williams, 2010b, Peralta et al., 2012, Yang et al., 2015). Zhang et al. (2018b) found for example that up to 15% of a sawdust pyrolysis bio-oil produced a satisfactory bitumen (but only after addition of SBS polymer). Work on the properties of pyrolysis oil modified bitumens has recently been reviewed by Su et al. (2018) and Wang et al. (2020a). The published data appears promising but there are as yet no commercially available products. A field trial of a bicycle path was constructed in 2010 using a 6% pyrolysis oil concentration in bitumen (ISU, 2010) but no other trials appear to have to been carried out and progress has not been reported.

Chemically, because the 'bio-bitumens' and bio-oils produced by pyrolysis are derived mainly from cellulose, hemicellulose and lignin (the major components of wood) they are very high in oxygen (about 30-40 wt%) and are not very stable at high bitumen handling and application temperatures (Zhang et al., 2018a). When mixed with conventional bitumens the bio-bitumen materials are not completely soluble in petroleum bitumen and on cooling exist as suspended particulates. These characteristics are not necessarily detrimental in asphalt mixes made with a mixture of bitumen and bio-bitumen, but excessive hardening is undesirable if bio-bitumen is the only component of the binder.

Pyrolysis oil fractions potentially useful for bitumen substitution constitute only a small fraction of the pyrolysis products produced. Pyrolysis of plant biomass as a process to produce bitumen alone would be a highly inefficient process. Additional research is needed to determine if conditions can be manipulated to maximise useful product formation.

6.2.2 Low oxygen content feedstocks

The pyrolysis of animal manure and in particular swine manure has also been investigated as a means of making a bitumen alternative (Fini et al., 2012, Fini et al., 2011, Liu et al., 2020, Wang et al., 2020b, Wang et al., 2021, Pahlavan et al., 2021, Hosseinnezhad et al., 2019). The chemical compounds in swine manure (e.g. fats), have a much lower percentage of oxygen than plant materials and so are less likely to suffer from the stability (polymerisation), problems exhibited by wood derived pyrolysis oils. Similarly the pyrolysis of municipal waste was found to give a high viscosity, waxy, oil, suitable as a partial bitumen substitute but only in 11% yield (Yang et al., 2018).

6.2.3 Bio-char and charcoal

Bio-char is produced during the pyrolysis of woody biomass at temperatures of 400-500 °C. It is a solid non-fusible powder of complex and poorly understood structure. The particle size of bio-char is similar to that of the pyrolyised biomass it is dervied from but can be easily crushed to a fine dust.

Research has been conducted on adding bio-char to bitumen at low concentration as a partial substutute (Zhang et al., 2018a). Zhao et al. found that 4%-10% bio-char with a particle size of less than 150 microns had a benefical effect on the high temperature deformation of bitumen without significant adverse effects on fatigue resistance (Zhao et al., 2014). Similar results were obtained by using a 6% loading (Hu et al., 2021). At 5% concentration bio-char was found to have some effect on oxidation resitance of bitumen under UV radiation (Rajib et al., 2021). Renaldo et al. (2015) also found that resistance to oxidation (Rolling Thin Film Oven procedure) was improved at 6% bio-char concentration.

Charcoal is similar to bio-char in that it is produced by wood pyrolysis but carried out at much lower temperatures and over longer times. Charcoal has also been investigated as a bitumen modifier at concentrations up to 15% with beneficial effects (Chebil et al., 2000).

6.3 Tall oil pitch and other pine resins

Tall oil pitch, is a stiff, brown, sticky, water insoluble resin, extracted from pine trees as a by-product of pulp and paper manufacture. The material consists mainly of a complex mixture of hydrocarbons, sterols with small amounts resin acids and fatty acids (Drew and Propst, 1981). These are very weak organic acids and the material is not corrosive.

Research has demonstrated that tall oil pitch is generally satisfactory, up to about 20%, as a potential material for partial bitumen replacement. (Ball et al., 1993, Bearsley and Haverkamp, 2007a, Bearsley and Haverkamp, 2007b, Peltonen, 1992, Wu et al., 2004). Dust suppressant and stabilisation products for road surfacings based on tall oil pitch are also commercially available.

The viscosity of tall oil pitch is usually lower than that of normal bitumen grades and it cannot be used as a complete bitumen substitute without modification to improve its rheology. A commercially available product (Biophalt) made by Eiffage Construction in France consists of tall oil pitch, other natural resins and up to 20% petrochemical based polymer (Pouget and Loup, 2013, del Barco Carrión et al., 2017). As with the Vegecol, Floraphalte and Sequoia products, discussed below, Biophalt is intended as a colourable binder for cycle ways and pedestrian areas rather than normal road construction.

There is considerable potential for development of new tall oil pitch based products for road surfacing applications but the quantities available world-wide are limited relative to the volume of bitumen used. Globally about 1.2 M tonnes is produced annually (Konwar et al., 2018, Abraham and Höfer, 2012). In New Zealand tall oil pitch is produced by Lawter (NZ) Ltd in Tauranga. Approximately half of the New Zealand production is used as a fuel and the rest is sold for various industrial applications. Annual production is only about 3000 tonnes and is limited by the volume of wood pulp manufactured in New Zealand (Villella, 2020).

Various other types of wood resins and fatty acid fractions are produced during manufacture of wood turpentine and other products. In many cases these have been investigated as bitumen modifiers but only at concentrations of a few percent (e.g. Grilli et al. (2019), Verma and Chauhan (2019), Putri et al. (2020)). Espinosa et al. (2021) studied an unspecified "pine wood resin" and "terpolymer" composite binder as a complete bitumen substitute. Laboratory results were promising and a full scale highway field trial of a dense grade asphalt mix was constructed. The bio-bitumen asphalt performance was good after three years of monitoring.

6.4 Microalgae

Microalgae, specially grown in large scale tanks are being actively investigated as a source of hydrocarbon transport fuels and bitumen alternatives (Qari et al., 2017). The fatty acids or other metabolites produced by the algae can be modified by various processes into thermoplastic materials with suitable rheological properties for extending or replacing bitumen (Audo et al., 2011, Chailleux et al., 2015, Audo et al., 2015). Development of these binders is still at the research stage.

6.5 Polymerised vegetable oils and related products

Vegetable oils such as soybean or sunflower oil have long been used in small quantities to modify the properties of bitumen (Chen et al., 2014, Hugener et al., 2014, Seidel and Haddock, 2012, Król et al., 2016). Cooking oils have also been investigated (El-Shorbagy et al., 2019, Asli et al., 2012, Gong et al., 2016, Sun et al., 2016a, Sun et al., 2016b). Azahar et al. investigated the effects of incorporation into bitumen of up to 8% concentration of residues from the conversion of waste cooking oil to bio-diesel (Azahar et al., 2016).

In New Zealand vegetable oils (e.g. soyabean oil) have been used in the commercial manufacture of sealing grade binders by simple blending of imported harder grades of bitumen with the oils but only at levels of up to 5-8% by weight. The resulting products were stable and met all requirements of the New Zealand bitumen specification. Similarly tung oil at up to 8% has been investigated as a means of rejuvenating the properties of aged bitumen (Yan et al., 2021, Yan et al., 2020).

The viscosity of vegetable oils is much lower than that of bitumen so that polymerisation or other treatment is required if the oil is to be used as partial or full replacement for bitumen. Vegetable oils are relatively easy to polymerise and have long been used in paints and other products for this reason (e.g. linseed oil putty). Polymerised waste cooking oils (typically canola or soyabean oil), have been investigated as a bitumen extender at 30% and 60% concentration (Wen et al., 2013). At the concentrations used the resulting bitumen and asphalt mix properties were adversely affected.

Li et al. added 20% concentration of (mainly) esterified fatty acids solvent extracted from an unspecified seaweed species to an SBS polymer bitumen (Li et al., 2021). The seaweed extracted material resulted in improved binder properties through its swelling of the SBS phase.

Polymerised vegetable oils form an important component in most of the commercially available products which are discussed below.

Polymeric materials with rheological properties similar to those of bitumen produced from low molecular weight ethyl acrylate, methyl acrylate and butyl acrylate monomers have been synthesised (Airey et al., 2008, Airey et al., 2011, Airey et al., 2016). The authors believe that in the future these monomers could potentially be sourced from renewable triglyceride oils and carbohydrate sources. The materials produced were found promising as partial or in some cases complete substitutes for bitumen.

6.6 Commercially available products

There are a number of commercially available bio-bitumen products on the market consisting wholly or largely of renewable plant materials, in particular vegetable oils.

6.6.1 Vegecol

Vegecol is the brand name of a bitumen alternative product manufactured by Colas in France (Colas, 2021). Chemically the product is manufactured from unspecified constituents of vegetable origin (Ballie and Delcroix, 2008), mainly a resin obtained from pine trees and an oil obtained from oleaginous plants (Colas, 2021). The product does not contain petrochemical derived materials. As the product cures with time (the viscosity increases) it is likely that the product is based on vegetable oils that polymerise through autoxidation.

It is a low carbon product, requiring lower manufacturing temperatures for warm asphalt mixes, lowering greenhouse gas emissions compared to conventional hot-mix asphalt. It is also a carbon sink, as it is more than 80% plant-based and plants fix a greater amount of carbon during their growth than is used to manufacture the binder.

Vegecol is semi-transparent and has found some application in specialist coloured asphalt surfacings for cycle ways and pedestrian areas. To date Vegecol has only been used in asphalt mix applications not chip sealing (Colas, 2021), and has recently been applied as an asphalt mix for school footpaths in Vedène, France. By 2007 it was reported that 450 projects had been completed using nearly 2700 tonnes of material (Croteau et al., 2009). Few studies on the properties of asphalt mix have been reported. Good early life performance has been reported but with some concerns over premature hardening which could result in cracking (Chailleux et al., 2015).

Vegecol has properties similar to that of New Zealand 130-150 and 180-200 penetration grade bitumens, a detailed report on the physical properties of the material and its potential application in Australia has been conducted (Urquhart, 2012, Urquhart and Malone, 2013). The authors of those studies concluded that although Vegecol could potentially be used for chip sealing, that would not currently be possible given that the manufacturers control use of the material and require input into the design of potential applications. Design requirements for surfacings using Vegecol would preclude use in chip seals.

6.6.2 Floraphalte

Floraphalte is a colourless vegetable oil and natural resin based binder similar to Vegecol and made by Shell in Europe (Shell, 2016). Flouraphalte contains at least 90% renewable plant based material and a (petrochemical based), polymer compound. The properties of the binder in asphalt mix have been reported (Pouget and Loup, 2013), and, as for Vegecol, early life performance is predicted to be satisfactory but continued curing has been highlighted as a potential problem (Chailleux et al., 2015).

The physical properties of the material (Shell, 2016, Urquhart and Malone, 2013), are such that it would probably meet the NZTA specification for 130-150 grade bitumen used in chip seals (penetration of 100-150 and viscosity at 60° of about 120 Pas). It is however marketed as a binder for asphalt mixes used on cycle ways, parks and pedestrian areas, it is not designed for use in road surfacings.

6.6.3 Sequoia

This is another vegetable oil and natural resin based binder manufactured by Eurovia in France that also contains 5-10% of polymer (Chailleux et al., 2015, Eurovia, 2021). It has a penetration range of 110-140, so has a consistency similar to that of a 130-150 NZTA penetration grade bitumen. As with Vegecol and Floraphalte the intended application of Sequoia is for coloured asphalt surfacings in cycle ways, pedestrian areas and other low stress applications - not general road surfacing.

6.6.4 Eco-Biopave™ GEO 320

Eco-pave™ is a product made from non-petroleum renewable natural waste material manufactured from low molecular weight waste and biomass materials including lignin, cellulose, sugar, molasses, vinasses, natural tree and gum resins, natural latex rubber and vegetable oils (Newman et al., 2012). It is designed for footpaths, driveways, bicycle paths, tennis courts, car parks, roads and highways. It is available in many colours including high-visibility and light-reflective pavements. However, it is unclear as to whether this product is still currently in production.

6.6.5 Instant Asphalt

"Instant asphalt" from Albrecht Industries in South Africa is a 2-part product consisting of asphaltene-containing powder and a liquid "maltene" phase produced from waste cashew nut shells (Albrecht Supply Concepts, 2021). The materials can be transported to site, then mixed and heated, removing the need for heating in transport and lowering its carbon footprint and greenhouse gas emissions. This enables the material also to be used in remote locations that are not easily accessible (Albrecht Supply Concepts, 2021).

6.6.6 Sumac Eco

A French product, Surmac Eco, uses vegetable oils to form a polymer matrix in bitumen to improve its properties (Latexfalt, 2016).

6.6.7 Biophalt

A French company, Eiffage Route, market a product called "Biophalt" (Lahouazi, 2021). This is an asphalt mix product consisting of about 30% recycled asphalt pavement (RAP) with an organic binder from the pulp and paper industry (possibly tall oil pitch) and polymers. The product is designed for use as a petroleum bitumen substitute in roadways. Testing with the material showed that satisfactory, and in some cases improved, asphalt mix properties could be obtained (del Barco Carrión et al., 2017, del Barco Carrión et al., 2019).

7 Recycled materials

7.1 Waste tyre rubber

Approximately 74,000 tonnes of waste tyres are generated in New Zealand annually (3R Group Ltd, 2020) Internationally rubber from waste tyres has been used to modify bitumen properties using concentrations of up to 25% which constitutes a significant partial bitumen substitution. The issues surrounding the use of waste tyre rubber in road surfacings in New Zealand have been discussed in detail in a recent NZTA report (Wu et al., 2021b). A key technical obstacle to uptake of the technology has arisen given NZTA's decision to mandate the use of bitumen emulsions for chip sealing work on the State Highway network (WK-NZTA, 2021) Currently technologies for emulsification of waste tyre rubber modified bitumen are still in the developmental stages and it can currently only be hot-applied, which would prevent widespread use in chip sealing applications in New Zealand.

7.2 Plastics

The addition of waste, post-consumer plastics to bitumen has received much attention in recent years (Austroads, 2019). A number of commercial products are marketed such as "Plastiphalt" by Fulton Hogan Ltd in New Zealand (Fulton Hogan, 2021). Downer Ltd also manufacture a "Reconophalt" asphalt mix containing waste plastics (Downer, 2019).

In road surfacing research, application levels of up to 15% waste plastic or higher are often explored (Wu, 2021) but levels of only up to 5% of the binder are typically used in commercial products (Wu, 2021, Mishra and Gupta, 2018, Austroads, 2020). Levels of 6-8% of the binder are specified in Indian guidelines (IRC, 2013). Given the costs associated with sorting waste plastics and the relatively low levels of plastics used, adoption is normally justified based on improving engineering properties rather than benefits from material substitution.

8 Recycled bitumen

8.1 Asphalt mix

Large tonnages of asphalt mix (and the bitumen it contains) is recycled both in New Zealand and internationally. The NZTA M10 specification allows up to 15% recycled asphalt pavement (RAP) as a matter of course and higher levels on application. The substitution of bitumen in the new asphalt mix is often not quite 15% as not all bitumen in the RAP is considered active (i.e. it has hardened too much through oxidation). Rejuvenating oils (usually petrochemically derived) are also usually added at 5-10 wt% of the binder to soften the aged bitumen. Recent reviews of best practice and mix design procedures are available (Austroads, 2015, Austroads, 2016, NAPA, 2012).

The practical limit for RAP content in hot mix asphalt is usually about 40 to 50% due to mixing constraints and the need to heat large quantities of ambient temperature RAP (i.e. without direct contact with the flame). However new generation asphalt plants with High Recycling Technology (HRT), are now capable of handling 80-90% RAP.

8.2 Chip seals recycling

New Zealand has approximately 65,000 km of sealed roads (MoT, 2021) and the bulk of the 150-170,000 tonnes of bitumen used annually is used to maintain them (there are some other minor industrial applications for bitumen). New Zealand is somewhat atypical to other countries in our practice of developing multiple seal layers sometimes over decades. Seals with 5-7 or more layers are not uncommon. Over time it is often noted that the lives of subsequent layers shorten as the top-layer seals begin to flush within only a few years, well below the theoretical design life. These layers are deemed “unstable” and the only treatment is either an overlay of base course or milling and disposal of the seal layers. Alternatively sometimes the seal layer is broken up and mixed with the underlying pavement, then recompacted (Gray and Hart, 2003, Harrow, 2008). These treatments all result in a lost opportunity to reuse the bitumen.

As 88% of the sealed road network is chip seal, then assuming a 5m wide carriageway and 3 layers of single coat chip seal surface (at 1.5 L/m^2 residual application rate) there is 1,128,600,000 L of bitumen (1.1 million tonnes) already in the country’s road assets. This equates to $846 million worth of bitumen (assuming a $750 per metric tonne of bitumen) as part of the road asset inventory.

There appear to be considerable potential benefits from the recycling of this bitumen but this is not an area of current research.

9 Summary and conclusions

The relative merits of the most promising potential bitumen alternatives for road surfacing materials are summarised in Table 1.

Table 1: Qualitative comparison of promising potential alternatives to bitumen for road surfacings in New Zealand (✓: applies, ✓✓: strongly applies, ✗ does not apply, ?: unknown / maybe)

Material	Usable without chemical modification	Progressed beyond laboratory or testing stage	Potentially could be locally sourced & in sufficient volumes	Raw material commercially available & with established manufacturing infrastructure	Satisfactory engineering performance demonstrated	Compatible with existing NZ surfacing construction plant and methods	Offers short term solution	Likely carbon footprint relative to bituminous surfacings	Renewable resource with long term sustainability
Cement (concrete)	✓✓	✓✓	✓✓	✓✓	✓	✗	✓	✗?[c]	✗
Tall oil pitch	✗[a]	✗	✗	✓✓	✓[b]	✓✓	✓	✓	✓✓
Lignin	✗[a]	✗	✓✓	✓✓	?	✓✓	✗	✓	✓✓
Thermolysis oils	✗	✗	✗	✗	?	✓✓	✗	✓	✓✓
Proprietary vegetable oil based binders	✓	✓	✓	✗	?	✓✓	✗	✓	✓✓

(a) Modification required if to be used as a complete substitute for bitumen. (b) As a partial bitumen substitute. (c) Depends strongly on project scale.

9.1 Shorter term solutions

In the immediate term the only alternatives to importing bitumen for road surfacings are to use concrete or to recycle bitumen already in the road network and in particular chip seals.

9.1.1 Concrete

Currently the only existing, practical alternative to bitumen for road surfacings is to use concrete. Concrete road technology in terms of materials and construction plant is well established and has a long track record but still accounts for only a small proportion of road construction world- wide. The pros and cons of concrete road in terms of engineering properties (useful life, skid resistance), user experience (noise, delays), economics and environmental effects are still the subject of much debate. Only a few kilometres of concrete roads have been constructed in New Zealand (and mainly as pavements below a bituminous surfacing) and a move in this direction would require significant investments in plant and training by the road contracting industry. A large-scale change from bituminous surfacings to concrete would not be a seamless transition but require a complete step-change in the way our roads are designed, constructed and specified. Concrete may also only be economically viable for highly trafficked roads (e.g. motorways) whereas the bulk of our sealed road network is low trafficked and often narrow and windy and with flexible unbound pavements. The cost effectiveness of concrete roads obviously depends strongly on the ultimate cost of bitumen.

The carbon footprint of concrete roads has been studied at length and in some cases use of concrete would not result in significant increases in carbon footprint relative to alternative asphalt mix solutions. This is unlikely to be the case though for the bulk of New Zealand's lightly trafficked chip seal network. Ultimately the carbon footprint of the constructed concrete road surfacing/pavement depends very strongly on the assumed life of the surfacing and other maintenance requirements which in the New Zealand context, remain speculative.

9.1.2 RAP and recycled chip seals

Techniques for recycling asphalt mix are well established in New Zealand. The proportion of recycled material is generally 15-20% but internationally there is much research aimed at increasing the percentages used. Most bitumen in New Zealand though is used in chip seals, for which there is currently no existing recycling technology. The options and research needs to enable the recycling of chip seals is currently the subject of a separate NZTA study.

Ultimately any recycling system is limited by the fact that it is unlikely to be 100% efficient (i.e. there will be material losses) and there will be a limit to the number of recycling- cycles that are possible before material properties degrade.

9.2 Longer term solutions

A binder developed from a renewable, biomass resource is the best long term alternative to use of petroleum bitumen. This also offers potential to significantly reduce the carbon footprint of road construction and even provide net benefits through carbon sequestration in the road surfacing. The two most promising options based on current research are discussed below.

9.2.1 Vegetable oil based binders

A number of commercially available binders based largely on vegetable oils are available and are discussed above. These materials have been in existence for some time but have not gained any significant traction in general road surface construction. They are currently produced in only very small tonnages and occupy a niche market largely as clear binders for coloured surfacings, cycle ways etc. The long term potential for these materials is difficult to assess without more information of their performance, durability and suitability for significant scaling up of production.

A reservation often expressed concerning with use of vegetable oils for biofuels, which also applies to bitumen, is that such uses will increase the price of foodstuffs. Globally approximately 200 million tonnes of vegetable oils are produced annually (El-Hamidi and Zaher, 2018). In theory therefore large-scale use of vegetable to replace the 100 million tonnes of bitumen used annually could thus have a significant impact and a large increase in production would be needed to offset the effects.

9.2.2 Lignin

Although much research is still needed, lignin is attractive as a potential bitumen alternative because it is already produced in very large quantities. Bitumen could potentially be manufactured without the need for development of a completely new large scale industrial activity to produce the raw materials, as would be the case for example, with pyrolysis oils. Bitumen could be produced as a by-product of the pulp and paper industry as it is currently a by-product of petroleum fuels production. New Zealand produces more than sufficient lignin for road construction purposes so that we could be self- sufficient in locally produced bitumen.

Chemically, lignin provides in theory, ample opportunity for simple modification to a thermoplastic material that could be used as a bitumen extender or complete replacement and that could be used with existing road surfacing construction plant designed for petroleum bitumen.

10 Recommendations

10.1 Chip seal recycling

Further investigation into the feasibility of recycling chip seal materials both as millings and through bitumen and aggregate recovery should be carried out. The research needs to specifically address the behaviour of chip seal millings during storage and grading to identify potential handling problems. The properties of mixes made partially and wholly from chip seal millings need to be determined. As a first step the effects of using chip seal millings at up to 15% (as is the case with RAP) should be investigated.

Potential options for bitumen recovery need to be evaluated in detail and assessed in terms of efficiency and likely practicality of scale up.

10.2 Lignin

Currently the use of lignin or chemically modified lignin appears to be the most promising starting material for development of a practical, renewable alternative to bitumen. Work is needed to properly understand the behaviour of lignin-bitumen compositions and to formulate research strategies for development of a 100% lignin based binder.

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