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Waste Materials as Extender of Bituminous Binder: A Review

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ABSTRACT

Bitumen is a hydrocarbon material which can be largely obtained from crude petroleum distillation. It used widely in flexible pavement construction as a binder that binds the aggregate. Flexible pavements make up around 95% of the pavement in entire world. The quality of the materials used in the flexible pavement is essential for its performance. The modification of bituminous binders using modifiers is applied to improve the quality and properties of bitumen. Due to the limitation of crude oil, higher price, and impact of bitumen on the environment, an efficient, cost-effective, and environmentally friendly modifier for the modification of bituminous binders is vital in road industry. This paper aims to provides a review and propose the waste materials that can be used as modifier or bitumen extender. Among these waste materials, silicon, rubber, cooking oil (mix with other modifiers) lignin and calcium lignosulfonate shown a promising results as bitumen extender. Furthermore, the cost analysis of the modified bituminous binders was also look upon. The findings from this study recommend lignin among the extender being compared. Lignin can be mixed directly in the bitumen with a minimal process involved. Moreover, lignin is a bio-based waste materials and the second most abundant organic material on earth, after cellulose. With 25% replacement of bitumen, lignin reduces the consumption of non-renewable bitumen and has a great potential to be used in pavement construction.

Keywords: Bitumen; waste materials; bituminous mixture; bitumen extender

INTRODUCTION

Bituminous pavements are the most common pavement in Malaysia as well as the entire world. One of the essential materials of bituminous pavement is bitumen, a non-renewable material, primarily obtained from crude petroleum distillation. The price of bitumen depends on the price of crude oil. Over the years, an increment in price has been observed (Aziz et al. 2015).

Besides being a non-renewable material, the petrochemical industry is becoming more efficient in cracking longer chain hydrocarbons (bitumen) into shorter chain hydrocarbons, a high value product than bitumen (van Vliet et al. 2017). Hence this will hinder the supply of bitumen in the future.

The construction of flexible pavement requires heating the bitumen. At high temperature ranging 165 - 200°C, the heating process will release a greenhouse gas (Meijer et al. 2018). Furthermore, increase in traffic volumes, loading, rainfall, and temperature variations leads to pre-mature road failure and higher maintenance.

Therefore, the challenges face by the bitumen industry is to find an alternative binder that is sustainable and can partially replace bitumen while maintaining and/or improving the properties of the binder and pavement. To overcome these challenges, modified bitumen has been introduced by enhancing the properties of conventional bitumen using modifiers, thus lead to improvement in pavement performance.

A modifier that can be incorporated in bitumen more than 10% can be regarded as an extender (Aziz et al. 2015; Wu et al. 2021). One of the modifiers that can be bitumen extender is sulphur. Sulphur can be added at high concentrations approximately 12-18% and gives a significant improvement on the properties of bitumen (Hunter et al. 2000). However, at a temperature higher than 150°C, the reaction between bitumen and sulphur will release a toxic hydrogen sulphide gas. The sulphur also needs to be modified to be stable, economical, and environmentally safe (Souaya et al. 2015; Hunter et al. 2000).

Several studies have been attempted to examine the viability of waste materials to be used in bitumen and bituminous mix for instance rubber (Liu et al. 2018; Wang et al. 2020; Azadedel & Solatifar 2022), plastic (Ahmedzade et al. 2015; White & Magee 2019; Nizamuddin et al. 2020; Ponnada & Vamshi Krishna 2020; Mashaan et al. 2021; Dadwal et al. 2022), silicon (Saeed et al. 2019), cooking oil (Khan et al. 2019; Gokalp & Uz, 2019), lignin (Wang & Derewecki, 2013; Xu et al. 2017; Van Vliet et al. 2017; Batista et al. 2018; Arafat et al. 2019; Perez et al. 2019; Zhang et al. 2019; Zhang et al. 2020; Norgbey et al. 2020; Gao et al. 2020; Zahedi et al. 2020; Yu et al. 2021; Xu et al. 2021; Wu et al. 2021; Zhang et al. 2021; Ren et al. 2021), calcium lignosulfonate (Fatemi et al. 2021, 2022) and cast iron (Kalpana et al. 2020). Utilizing waste materials in bitumen is believed to be an effective solution since it is cost-effective, and can improves road performance while also assisting in the recycling and disposal of waste products (Aziz et al. 2015).

The performance of modifier or extender can be assessed by physical properties, rheological properties, workability, rutting, and cracking resistance of the modified bitumen. Based on previous studies, the addition of waste materials has shown an improvement in the properties of bitumen. Table 1 shows the studies on the waste materials as bitumen modifier or extender. Shown in the table, waste materials such as rubber, silicon, lignin and calcium lignosulfonate can be incorporated at higher content in bitumen. Therefore, these waste materials can act as a dual role, modifier, and extender.

This paper provides a review on the waste materials that can serve as bitumen extenders. The next section will review on the materials, methods, and performance of these waste materials as bitumen extender. This will follow by a cost-effective analysis of the modified bituminous mixture with waste materials. A conclusion was given based on the findings.

TABLE 1. Investigation on waste materials as bitumen modifier or extender

No	Reference	Waste Materials	Trial Content (%)	Role in Bitumen	
				Modifier	Extender
1	Liu et al. (2018); Wang et al. (2020); Azadedel & Solatifar (2022)	Rubber	5 – 22	x	x
2	Ahmedzade et al. (2015); White & Magee (2019); Nizamuddin et al. (2020), Ponnada & Vamshi Krishna (2020); Mashaan et al. (2021); Dadwal et al. (2022)	Plastic	2.5 – 20	x	
3	Saeed et al. (2019)	Silicon	10 – 50	x	x
4	Khan et al. (2019); Gokalp & Uz (2019)	Cooking oil	2 – 20	x	
5	Wang & Derewecki (2013); Xu et al. (2017); Van Vliet et al. (2017); Batista et al. (2018); Arafat et al. (2019); Perez et al. (2019); Zhang et al. (2019); Zhang et al. (2020); Norgbey et al. (2020); Gao et al. (2020); Zahedi et al. (2020); Yu et al. (2021); Xu et al. (2021); Wu et al. (2021); Zhang et al. (2021); Ren et al. (2021)	Lignin	1 – 60	x	x
6	Fatemi et al. (2021, 2022)	Calcium Lignosulfonate	5 – 20	x	x
7	Kalpana et al. (2020)	Cast Iron	4 –10	x	

WASTE MATERIALS AS BITUMEN EXTENDER

MODIFIERS

Van Vliet et al. (2017) assessed the use of lignin as partial substitutes for bitumen. Four types of lignin have been included namely Organosolv, Klason, Kraft and SuperHeated Steam (SHS). SHS is a lignin obtained through mild extraction process of wood and straw. Each type of lignin was blended with 70/100 bitumen grade at 25%. The blends were evaluated based on rheological performance using dynamic shear rheometer (DSR). The findings show that all types of lignin found to be suitable to blend with bitumen. Excluding Klason lignin, addition of lignin shows a significant effect by increase in stiffness. Modification with Klason lignin did not change the viscoelastic properties of bitumen (comparable to the base binder properties) while other types of lignin behave like a polymer modified bitumen.

Pérez et al. (2019) and Pérez et al. (2020) has attempted to utilized industrial waste containing a vegetal lignin for partial substitution of the bitumen in bituminous mixtures. The industrial waste is from the wood hardboards manufacture was used without any conversion, mixed directly into conventional bitumen (B50/70 penetration grade). Different percentages of bitumen substitution by liquid waste containing lignin were prepared and tested: 0% (control mix), 5%, 10%, 20%, 40% and 60% by weight of bitumen. The penetration and softening point increase as the percentage of lignin increase. From the findings, it is suggested that the mixtures with 20% of industrial waste showed the best results for medium and low traffic roads.

Concern with the disposable of silicon mobile phone covers, Saeed et al. (2019) utilized the silicon waste as a partial bitumen replacement. The silicone waste was mixed with bitumen grade 60/70 at 10%, 20%, 30%, 40%, and 50%. Based on the stability and flow values, the study suggested replacing bitumen with up to 40% of silicon waste is likely feasible for asphalt pavements.

Three types of waste from different sources have been utilized in an attempt to reduce the bitumen usage namely waste cooking oil (WCO), tires rubber powder (TRP) and bagasse ash (BA) obtained from restaurants, tire industries and sugar mills, respectively (Khan et al. 2019). Based on the penetration and softening point values, three set of trials have been made to obtain the optimum content or suitable ratios of waste cooking oil, tires rubber powder and bagasse ash to be added in bitumen grade 60/70. The optimum content of these waste materials that meet the specifications of bitumen is 5% WCO and 15% TRP (with 8% BA) which replaced 20% of the bitumen. It was found that the modified bitumen shows an improve rheological properties compared to base binder.

Wu et al. (2021) investigated the bitumen performance with a single high content (25% by bitumen mass) of klason lignin powder as an alternative extender. The wood lignin was extracted from the waste of paper manufacture industry and incorporated into bitumen penetration grade 70. The findings concluded that with a right procedure, lignin could be a suitable extender for the bitumen.

Fatemi et al. (2021, 2022) studied the effects of Calcium Lignosulfonate (CLS) on the bitumen ageing properties and bituminous mixture performance. In this study, four percentages of CLS powder at 5%, 10%, 15%, and 20% by weight of bitumen were added to 60/70 penetration grade bitumen. Based on the results, incorporating 15% of CLS in bitumen achieved the highest aging and rutting resistance of bitumen.

An industrial waste namely cast iron in powder form has been investigated as a bitumen modifier by assessing the bitumen conventional properties such as penetration, softening point, ductility, and viscosity (Kalpana et al. 2020). The addition of cast iron in bitumen increases the penetration value while decreasing the softening point value. The ductility shows no significant changes while the viscosity of cast iron modified bitumen increases compared to base bitumen.

Nizamuddin et al. (2020) examines the use of a common and widely accessible soft plastic, recycled linear low-density polyethylene (R-LLDPE), for bitumen modification. The physical, chemical, rheological, and thermal properties of base bitumen (50/70 penetration grade) and R-LLDPE modified bitumen (replacement of 3%, 6%, 9%, and 12% by weight of the bitumen) were examined to assess their usability in the road pavement. Based on the findings, the suggested content of R-LLDPE is 3% - 6%, where 6% can only be utilized for tropical regions.

Higher content of plastic waste has been attempted at 10%, 12.5%, 15%, 17.5 %, and 20% as bitumen substitution (Dadwal et al. 2022). The study findings, however, limit the 10% plastic content as an acceptable bitumen substitute.

Modifiers such as rubber, plastic, silicon, lignin, and calcium lignosulfonate have comparable effects on bitumen where the addition of these modifiers in bitumen increased its stiffness by decreasing the penetration value while increasing the softening point and viscosity. Except for plastic, these modifiers can be incorporated more than 10% in bitumen. Cooking oil, and cast iron, on the other hand, increased the penetration value and reduced the softening point.

MIXING METHOD

Various mixing methods have been applied in incorporating waste materials into bitumen to obtain a homogenous mix. Van Vliet et al. (2017) initially performed the mixing of organosolv lignin and bitumen by hand. Blending at a higher percentage (25%) of organosolv lignin in bitumen formed coagulation which is unfavourable if done by hand. Modification in blending procedure by adding the lignin using a sieve to achieve even distribution of lignin on the bitumen surface and using a low shear mixer was carried out to avoid coagulation and obtaining a homogeneous mix. The mixing temperature at 155°C with no specified time and speed.

Pérez et al. (2019) mixed directly an industrial liquid waste wood extracts, mainly from eucalyptus, from the manufacture of hard fibres boards with 50/70 penetration grade bitumen. The mixing temperature was 170°C and blended manually for 10 minutes. Saeed et al. (2019) shredded the waste silicon cover prior mixed in hot bitumen at 160–170°C with no specified time and speed.

Khan et al. (2019) prepared the sample by added the tire rubber powder to hot bitumen at the temperature of 150°C for 30 minutes at 900 rpm. Following that, waste cooking oil and bagasse ash were added, and the temperature was reduced to 135°C to avoid the waste cooking oil evaporation at high temperatures.

In another study using powder lignin, the lignin was pre-heated at 105°C for two hours before the mixing process (Wu et al. 2021). The mixing was carried out using a high-speed shear mixer for 45 minutes at 1200 rpm and a temperature of 160 – 168°C.

The high-shear mixer was used to incorporate CLS powder, which was sieved through a No.100 sieve, into base bitumen (Fatemi et al. 2022). The mixing temperature, time, and speed were set to 150°C, 30 minutes and 3000 rpm, respectively.

PROPERTIES AND PERFORMANCE OF MODIFIED BITUMEN

The addition of waste materials as bitumen modifiers or extender generally increases the bitumen stiffness by reduce the penetration and increase the softening point temperature. However, modifier in liquid form found to increase the penetration such as waste cooking oil (Khan et al. 2019) and liquid waste lignin (Pérez et al. 2019 and Pérez et al. 2020). At optimum content of 20%, liquid lignin changes the 50/70 penetration grade bitumen to 80/100 penetration grade bitumen by increase the penetration value from 66 dmm to 97 dmm (Pérez et al. 2019).

The incorporation of lignin (excluding Klason) increases the complex shear modulus (G*) and decrease the phase angle (δ) which the parameter that describe the viscoelastic response of bitumen (Van Vliet et al. 2017). Higher G* indicates a stiffer bitumen which able to resist deformation while lower δ indicates a greater elastic. These two rheological parameters have been established by the Strategic Highway Research Program (SHRP) as the indicators to rutting and fatigue cracking resistance. Using the value of complex modulus (G*) and phase angle (δ) of the sample, the rutting resistance parameter is measured by G*/sin δ and the fatigue cracking resistance is determined by G*sin δ (Ali et al. 2013; Hunter et al. 2000). In the previous studies, waste materials namely lignin, combination of waste cooking oil, tires rubber powder and bagasse ash, and calcium lignosulfonate increase the resistance of bitumen toward rutting deformation. Combination of waste cooking oil, tires rubber powder and bagasse ash also found to increase the fatigue and low temperature cracking resistance.

Table 2 shows the summary of bitumen extenders, mixing methods and the outcomes from the bitumen modifications.

TABLE 2. Extenders, mixing method and effects of the bitumen modification

References	Type of extender and content (%)	Grade of bitumen	Mixing temperature, time and speed	Properties of modified bitumen	Optimum Content (%)
Van Vliet et al. (2017)	Lignin (Organosolv, Klason, Kraft and SuperHeated Steam (SHS)) 25%	70/100	155°C	Viscoelastic: Klason -, organosolv, kraft & SHS ↑ G* ↓δ	25%
Pérez et al. (2019) and Pérez et al. (2020)	Industrial waste: Liquid vegetal lignin 5% 10% 20% 40% 60%	50/70	Manual mixing at 170°C for 10 minutes	- Penetration ↑ - Softening point ↑	20%

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Saeed et al. (2019)	Silicon Waste 10% 20% 30% 40% 50%	60/70	160–170°C	- Penetration ↓ - Softening point ↑ - Ductility ↓	40%
Khan et al. (2019)	Waste cooking oil (WCO), tires rubber powder (TRP) and bagasse ash (BA) First Trial 5% WCO 10% WCO 15% WCO 20% WCO Second Trial 5% WCO + 5% TRP 5% WCO + 8% TRP 5% WCO + 12% TRP 5% WCO + 15% TRP 5% WCO + 18% TRP Third Trial 5 % WCO + 5 % TRP + 5 % BA 5 % WCO + 8 % TRP + 7 % BA 5 % WCO + 12 % TRP + 7.5 % BA 5 % WCO + 15 % TRP + 8 % BA 5 % WCO + 18 % TRP + 10 % BA	60/70	TRP: 150°C for 30 minutes at 900 rpm. WCO & BA: 135°C	- Penetration ↑ - Softening point ↑ -Rotational Viscosity ↑ - Rutting Resistance ↑ - Fatigue Resistance ↑ - Low Temperature Cracking Resistance ↑ - Age Hardening Resistance ↑	5% WCO and 15% TRP (with 8% BA)
Wu et al. (2021)	Klason lignin 25%	70	160 – 168°C for 45 minutes at 1200 rpm	- Penetration ↓ - Softening point ↑ - Ductility ↓	25%
Fatemi et al. (2022)	Calcium Lignosulfonate 5% 10% 15% 20%	60/70	150°C for 30 minutes at 3000 rpm	- Penetration ↓ - Softening point ↑ - Kinematic and Rotational Viscosity ↑ - Rutting Resistance ↑	15%

↑increase, ↓ decrease, - no changes

COST EFFECTIVENESS ANALYSIS

Analysis of cost effective using waste materials as extender has been reviewed in this paper using RM (Ringgit Malaysia) as the currency. Perez et al. (2019) carried out a cost-effectiveness analysis for the control bituminous mixture (0% lignin) and the modified bituminous mixture with the optimum lignin content (20%). The mixtures were evaluated at their optimum bitumen content, which was 4.5% and 4.1% for the control mixture and modified mixture, respectively. As shown in Table 3, the cost of producing the modified mixture containing 20% industrial waste was 16.5% less than the control mixture.

Saeed et al. (2019) estimated a 4.2% cost saving in bitumen with silicon waste. The optimum bitumen content was 7% by weight of bituminous mixtures and the optimum silicon waste content was 40% by total weight of bitumen.

The replacement of bitumen by the calcium lignosulfonate as bitumen extender can be up to 15% (Fatemi et al. 2022). However, the cost of this waste materials if obtained commercially can be up to RM1422/tonne. Another waste material by-products from paper industries are lignin. Lignin costs in China, around RMB 1500 (RM984) per tonne, while bitumen costs around RMB 3500 (RM2296) per tonne (Yao et al. 2022). The replacement of bitumen by up to 25% (Van Vliet et al. 2017; Wu et al. 2021) can reduce the cost of binder in road construction by up to 11.42%.

Asphaltic concrete wearing course (ACW14) premix costs roughly RM184/tonne in Malaysia. Since aggregate costs are around RM30-40/tonne, a large portion of the premix cost comes from bitumen despite accounting for only about 5% of the bituminous mixture. The cost of premix can be reduced by RM25/tonne by partially replacing the bitumen with lignin (RM984/tonne).

TABLE 3. Cost-effective analysis

No	Reference	Materials	Cost (RM/tonne)	Cost of bituminous mixture (RM/tonne)	
				Control	Modified
1	Perez et al. (2019)	Bitumen (B50/70)	1,803.7	81.17	61.64
		Aggregate (hornfels)	38.36	36.64	36.78
		Industrial waste	0.00	0.00	0.00
		Total		117.81	98.42
2	Saeed et al. (2019)	Bitumen (60/70)	2370	165.9	158.79

Reproduced from Perez et al. (2019) & Saeed et al. (2019)

CONCLUSION

The modification of bituminous mixture for the road construction using a waste material can provide a cost-effective and a sustainable pavement besides provide a proper solution for the disposal of waste. This review highlights several waste materials for bitumen extender which efficient, sustainable, eco-friendly, and low in cost. From the study, it has been shown that silicon gives higher bitumen replacement. However, the study on silicon waste as a bitumen extender was in the initial stage and the preparation process of the silicon waste prior to mixing with bitumen can be costly and time exhausting.

In the case of waste cooking oil, as a single modifier, this waste material can be incorporated up to 5% by weight of bitumen. However, waste cooking oil can be mixed with tire rubber to form a composite modifier which can replace 20% of the binder. In spite of this, the trial process to determine the feasible combination can be tiresome. Waste cooking oil might also contain a variety of residues, thus the oil needs to be filtered, and thoroughly inspected so that its effects on the bituminous mix can be controlled and properly maintained. Lignin and calcium lignosulfonate are both in powder form and can be directly mixed with bitumen. Lignin in liquid form also shows a promising result, however, it is not suggested to be used in higher temperature areas since the addition of this liquid waste increases the penetration of bitumen, therefore reducing its hardness.

Based on the findings, it has been suggested that lignin can be feasible as the bitumen extender. With 25% replacement of bitumen, lignin reduces the consumption of non-renewable bitumen and improve the properties of bitumen. With a higher demand, the extraction cost of lignin is expected to be reduced. Thus, this will in turns reduce the materials cost and maintenance costs of the pavement.

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DECLARATION OF COMPETING INTEREST

None

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