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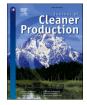
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# A state-of-the-art review of Natural bitumen in pavement: Underlining challenges and the way forward



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#### ABSTRACT

The demand for alternative bitumen which could fully/partially replace Petroleum sourced bitumen for pavement construction is globally increasing. The increase in demand can be associated with several factors: depletion in crude oil resources, advances in crude oil refining processes, increased demand for highway infrastructure, and regional transportation-environmental policies. Since the production of Petroleum bitumen consumes energy and generates emissions, there is an effort to decrease harmful emissions which has inspired researchers to look for so-called "green alternatives". Natural bitumen could be considered a green alternative as it is a mixture of bitumen and mineral matter present naturally on earth, mainly if the Natural bitumen can be transported easily to the construction site. This paper reviews the state-of-the-art information on pavement construction using Natural bitumen from laboratory and field perspectives. The Physico-chemical properties, rheological properties and field behaviour of asphalts pavements containing Natural bitumen were assessed. Many road authorities would hesitate to utilise Natural bitumen for pavement applications due to a lack of available data, knowledge and a systematic research study. To the best of the authors' knowledge, there is no comprehensive literature review article on Natural bitumen. Thus, the presented article aims to comprehensively review Natural bitumen resources and their types, Physico-chemical properties, application in pavement constructions, and reported field performances. At the end of the paper, future research challenges, future recommendations and a methodological framework is proposed.

#### 1. Introduction

The global increase in population, industrialisation, and transportation sector is commensurate to fuel consumption and greenhouse gas emissions (Rahman et al., 2017). These issues are a modern-day concern for most policymakers. To address some of the environmental problems, most countries are committed to the COP 21 Paris agreement. The COP 21 Paris agreement is a global referendum in which countries jointly agreed to cut down on the emissions of  $CO_2$  to reduce the current average global temperature to well below 2 °C as compared to the pre-industrial level (Schreurs, 2016). For example, the Netherlands has agreed to reduce emissions by 49% compared to 1990 (Beer, 2019).

Keeping harmful impacts of excessive greenhouse gas emissions in mind, a 95% reduction in emissions globally by 2050 compared to 1990 levels is expected in the Netherlands. This vision is driven through a modal shift from car to bicycle, private car to public transport, fueldriven vehicles to electric vehicles, etc. (Beer, 2019). Similar trends are also being adopted in other parts of the world. Eventually, such policies and vision would lead to low demand and production of Petroleum products, affecting the quantity and quality of available bitumen. Such trends could be of significant concern for industries that are dependent on bitumen. One of the industries that could be directly affected by such development is the road industry (Sorrell et al., 2010).

As highlighted in the above paragraph, the continual depletion of crude oil resources is also a concern for relevant industries apart from environmental issues. Researchers (Sorrell et al., 2010) have predicted that by 2030, Petroleum resource extractions will gradually start to decline. Consequently, less bitumen would be available for its engineering usages. Bitumen is an important material used in various civil engineering applications, such as adhesives, sealants, preservatives,

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waterproofing agents and pavement constructions (Polacco et al., 2006), with an annual global demand of about 100 million metric tons (García et al., 2010). In case of a reduction in bitumen productivity, various industries would have to look for alternatives. In addition to the depletion of crude oil reserves, the recent advancement within the oil sector has also changed the overall refining crude oil process.

Researchers (Speight, 2004) have reported that such advances in crude oil refining processes aim to make the whole refining process more efficient and, overall, financially beneficial. These focus on obtaining more beneficial products through the middle portion of the crude oil's fractional distillation processes. In turn, this enables Petroleum industries to break down higher chain hydrocarbons to so-called "premium valued hydrocarbons" rather than less financially beneficial product "bitumen". The key process includes catalytic cracking, hydrocracking and propane de-asphalting (Sahu et al., 2015). Moreover, these advances would further affect bitumen's quantity, price, and quality (Zhu et al., 2014).

The changes in the oil sector (as highlighted above) would directly impact the composition of the available residue to produce paving-grade bitumen and affect the construction of Bituminous pavements. It is noted that more than 90% of the global road network is reported to be made up of bituminous pavements (Gautam et al., 2018). Bituminous pavements are constructed by placing a Bituminous mixture on a prepared subbase or subgrade. These mixtures are typically prepared by mixing bitumen, fine and coarse aggregates at a high temperature in a pre-designed proportion. In bituminous pavement design, bitumen plays a significant role that binds other constituents together and resists the ingress of water (McNally, 2011). Therefore, Bituminous pavement's overall performance would depend on the quantity and quality of added bitumen (White, 2016). Bituminous pavements are also known as Asphalt pavements. However, in this paper, Bituminous pavements will be consistently used.

The pavement community has already started to look for alternative bitumen that could replace Petroleum sourced bitumen. At the same time, several research studies are focusing on studying the suitability of such bitumen in bituminous pavement constructions in terms of performance and financial viability. There is an effort to decrease greenhouse gas emissions which has inspired researchers to look for so-called "green alternatives", either naturally present materials or waste materials. These alternatives include bio-bitumen produced from animal waste (Fini et al., 2011), agricultural waste (Raouf and Williams, 2010), wood waste (Yang et al., 2014), Epoxy (Herrington, 2009; Apostolidis et al., 2018), Lignin (Zhang et al., 2019) and cedar wood (Yang and Suciptan, 2016). Other bitumen sources include crude oil shale, biomass, microalgae, and Natural bitumen (Al-Otoom et al., 2013; Chailleux et al., 2012; Halim et al., 2012; Subramanian and Hanson, 1998). However, researchers (Luo et al., 2015) have concluded that some of the above-proposed alternatives have merits and demerits in terms of cost, availability, and impacts on pavement performance. Hence, dedicated research investigations must be carried out before their utilisation in practice.

A Natural bitumen could be considered as a green alternative as it is a mixture of bitumen and mineral matter present naturally on earth. Researchers (Meyer and De Witt, 1990) reported that Natural bitumen resources are available in various parts of the globe. It is noted that the terms Natural bitumen and Natural bitumen are interchangeably used in different literatures. Hence, a consistent definition in the paper would be followed for clarity. Bitumen is usually referred to as a black cementitious in a solid, semi-solid, or viscous state derived naturally or industrially manufactured, for example, Lake bitumen, tars, pitches and Asphaltites according to the American Society for Testing and Materials (ASTM) (D8-13b, 2013). According to ASTM, asphalt is defined as black cementitious material whose main constituents are bitumen occurring naturally or produced industrially (D8-13b, 2013). In Europe, the term "bitumen" is the binding material in the bituminous mixtures, referred to as a sphalt cement in North America. As per the European Norm

(Speight, 2000), Natural bitumen would be consistently referred to as the binding agent otherwise mentioned in this article. Bitumen is the main component that provides the engineering adhesives in binding mineral aggregates in bituminous pavements construction. Similarly, in the paper bituminous mixture will consistently be used for Asphalt mixture.

The utilisation of Natural bitumen in pavement related activities is not new. Prior to the oil boom era, Natural bitumen was the main source of bitumen for pavement construction activities (Speight, 2000). However, as vehicular traffic density and loading increased, durable pavements and alternative paving materials were required. At the same time, gasoline production was ever-increasing to meet the demand for the increasing number of vehicles. Subsequently, more Petroleum bitumen was available. During that time, pavement researchers found Petroleum bitumen as an excellent material to bind different constituents of paving materials. Consequently, Petroleum sourced bitumen replaced Natural bitumen. Obviously, in follow-up years, less research studies were carried out to evaluate & develop this naturally present resource.

As mentioned earlier, the primary source of bitumen and bituminous materials were only from the natural source before 1907 and pavement construction was done using Natural bitumen (Lavin, 2003). At Present, Natural bitumen could be seen as a bitumen alternative that helps pavement researchers reduce harmful greenhouse gases during the construction and maintenance of bituminous pavements (Speight, 2000). These harmful gases are generated during the construction and maintenance of Bituminous pavements (Pratico et al., 2020). Therefore, environmental and pavement researchers have started to pay close attention to reducing such emissions. Consequently, bituminous pavement construction is gradually geared towards green sustainable approaches to limit the environmental impact of greenhouse gases and lower down energy consumption (Thives and Ghisi, 2017; Santos et al., 2017) in various construction processes. These approaches include a) using a bitumen that would use low energy during its sourcing (as already described in previous paragraphs) and b) reducing the amount of energy used during production and laying of Bituminous mixtures. Researchers (Speight, 2000) reported that Natural bitumen could be used directly in its natural form for paving operations, lowering overall energy consumption during paving operations.

In summary, Natural bitumen could be considered a green and sustainable alternative to Petroleum bitumen, particularly if the bitumen from the source is financially suitable for pavement construction. Many road authorities would hesitate to utilise it in pavement activities if comprehensive data is unavailable. To the best of the author's knowledge, such articles containing a comprehensive overview of Natural bitumen are unavailable. Hence, this paper aims to provide a review of the various Natural bitumen that are commonly available around the world. It would also focus on the various technologies and methodologies used in their utilisation. The review would further focus on the physical, thermal, mechanical, and chemical behaviour of Natural bitumen in view of their various road construction applications, such as additives, modifiers, base bitumen, and rejuvenators. In addition, the reported laboratory and field performance of various Natural bitumen will be highlighted.

Considering the potential benefits highlighted in the previous paragraphs, many researchers have started to relook at different Natural bitumen for their potential pavement construction-related applications. At the end of the paper, a simple framework is also proposed, allowing pavement authorities, researchers and organisations to assess the suitability of givenNatural bitumenfor pavement applications by a methodological framework.

#### 1.1. Paper organisation

To make this paper easier to understand, the article has been arranged in different sections. The first and second sections introduce and provide background information on naturally present resources of Natural bitumen. These sections examine and analyse the performances of natural bituminous mixtures based on available literature. The third section discusses the possible functions of Natural bitumen in bituminous pavement construction. The fourth section presents current challenges in using Natural bitumen for paving operations and a framework to check the suitability of available Natural bitumen for road construction applications.

#### 2. Reserves of Natural bitumen around the world

The volume of Natural bitumen reserve is around 1,856,853 billion barrels, as demonstrated and 864,841 billion barrels as inferred (Meyer and De Witt, 1990). Various kinds of Natural bitumen are found in Canada, Venezuela, Malagasy, Utah, Californian Mexico, Kentucky in the United States, Buton Island in Indonesia; Albania; Romania; Kazahstan; France (Gard &Tham); Switzerland (Traver valley); Italy (Ragusa), Greece (Paxos/Antipaxos and Marathoupoli) (Wen et al., 1978), Nigeria(Ondo, Ogun & Lagos) (Adegoke et al., 1980; Aladekoyi et al., 2016). These Natural bitumen are usually found in the form of rock enriched with bitumen (Adedimila, 2004), Gilsonite (Quintana et al., 2016), Trinidad lake bitumen and Bituminous sand. The estimated volume of Natural bitumen deposits is presented in Table 1. These different kinds of Natural bitumen have different physico-chemical properties.

The following sub-sections present an overview of different Natural bitumen that has the potential to fully or partially replace Petroleum bitumen.

#### 2.1. Lake bitumen

Lake bitumen is one of the widely known Natural bitumen found in a semi-solid or viscous state. Trinidad lake bitumen is a lake bitumen which is one of the most commonly known and utilised lake bitumen (Charles and Grimaidi, 1995). Apart from Trinidad, lake bitumen are also found near Venezuela's northeast coast region, constituting the world's largest known deposit (Phizackerley and Scott, 1978). The deposit of Natural bitumen in Trinidad is estimated to be approximately 15million tonnes spread over a surface area of 50Ha (Wen et al., 1978). Lake bitumen found in North-Eastern Venezuela and near La Brea's town island in Trinidad are often referred to as Bermudez lake bitumen, with approximately 5 million tonnes of reserves (Chilingarian, 2011). The Bermudez lake bitumen usually consists of limestone or sandstone with bitumen content ranging between 5 and 30% (Speight, 2000). As reported by researchers (Speight, 2000), in the past, bitumen from Bermudez Lake was exported to the United States and used for different industrial purposes. In addition, it was also used for pavement construction in the early 1900s (Holley, 2003). However, since 1940 Bermudez bitumen has not been used for road construction activities (Speight, 2000).

The popularity of Bermudez lake bitumen decreased since 1940 because of the following reasons: (a) the Petroleum source bitumen was easily available, (b) the penetration values were not consistent (Speight, 2000; EN, 2007; ASTM, 2013), and (c) the market was monopolised

#### Table 1

World Natural bitumen resources (in a million barrels) (Meyer and De Witt, 1990).

Area	Demonstrated	Inferred	
North America	1,708,548	864,809	
South America	50,460	N.A.	
Europe	20,493	N.A.	
Asia	76,263	N.A.	
Africa	1065	31	
Middle East	13	N.A.	
Southeast Asia	11	1	
World Total	1,856,853	864,841	

(Welborn, 1984). Lake bitumen is also reported to be available in south-western Nigeria (Adedimila, 2004; Nciri and Cho, 2018). Previous studies reported that lake bitumen has a high penetration value (ASTM, 2013) and low softening point (ASTM, 2014) (at room temperature in its natural state). Farmers within this region used the bitumen to repair their mud houses and paint their houses (Adedimila, 1987), and it was never used for road construction. An attempt was made to study the bitumen for further applications; unfortunately, the first world war stopped the process (Adelu and Fayose, 1991). The following sub-section presents the physical and chemical properties of Trinidad Lake bitumen from Trinidad and their application in pavement constructions.

#### 2.1.1. Physico-chemical properties of lake bitumen

In paving operations, engineers rely on physical composition to determine the suitability of bitumen for pavement constructions (Murali Krishnan and and Rajagopal, 2003). The physical composition of Natural bitumen can be determined through tests such as softening point (ASTM, 2014; EN, 2015), penetration test (EN, 2007; ASTM, 2013), specific gravity (Testing and Materials, 2014) and ductility (D113-17, 2017). Overall, such properties are governed by the constituting components. The composition of Trinidad lake bitumen includes water, topsoil, and stones, apart from soluble bitumen (Widyatmoko et al., 2005; Fengler et al., 2019). Table 2 provides the composition of soluble bitumen, mineral matter and organic matter as reported by other researchers (Widyatmoko et al., 2005; Fengler et al., 2019).

As presented elsewhere (Liao et al., 2014) and reported in (He et al., 2019), the properties of fresh bitumen modified using Trinidad lake bitumen results in the reduction of penetration values and an increase in the softening point value regardless of the percentage of inclusion, as shown in Table 3, Figs. 1 and 2 (He et al., 2019).

Previous study (Read et al., 2003) reported that Trinidad Lake bitumen blended with 160/220-grade Petroleum bitumen in a 50:50 ratio resulted in a modified bitumen with 50 penetration grade which was successfully latter used in the pavement surface course construction. It is noted that in reality, the penetration value of 50 is a hard bitumen, and most times, it is used when stiffer pavement wearing courses are required. However, FTIR is the most commonly used techniques among pavement engineers to analyse the chemical composition of bitumen (Zhang et al., 2020). It is highlighted that these techniques provide adequate information on the average chemical composition of bitumen.

The FTIR identifies each functional group of bitumen using a lightabsorbing technique (Hofko et al., 2018). The wave number between 1800 and 600 cm is the most important fingerprint region that describes the bitumen's chemical composition. The wave number, as discussed in the previous paragraph, provides useful information about the functional chemical groups, such as carbonyls(1753-1600 cm<sup>-1</sup>) and sulfoxides (1047-995 cm<sup>-1</sup>) (Jing et al., 2019). A detailed description of FTIR can be found elsewhere (Jing et al., 2019). Researchers (Xu et al., 2019) used FTIR to study the performance of Trinidad Lake bitumen. The researchers compared the performance of Trinidad bitumen against a) Petroleum bitumen, b) Trinidad Lake bitumen modified with Petroleum bitumen, C) Petroleum bitumen modified Petroleum bitumen (Xu et al., 2019). The study reported a spectrum of Trinidad Lake bitumen

Table 2
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Composition of Trinidad lake after source (Widyatmoko et al., 2005; Fengler et al., 2019).

Composition	Percentage (Widyatmoko et al., 2005)	Percentage (Fengler et al., 2019)
Soluble bitumen	53–55	52–62
Mineral matter	36–37	33–38
Organic matter	9–10	4

Properties of Trinidad Lake bitumen (Liao et al., 2014).

Test	TLA.
Penetration@25 °C dmm	1
Softening Point °C	99
Ductility@25°C, cm	NA.
Solubility (TCE) %	54
Flash point (C.O.C) °C	>240
specific gravity @25°C	1.37
Ash, %	36

with a peak around  $1700 \text{ cm}^{-1}$  stretching the vibration of carbonyl oxides, as shown in Fig. 3. In addition, they reported that the wavenumber of 1030 cm<sup>-1</sup> was present due to the stretching of the sulfoxide group, which is similar to the Petroleum sourced bitumen (Xu et al., 2019).

Researchers (Peng et al., 2003) reported that the spectra of Trinidad Lake bitumen had a band that appeared around 3800-3500 cm<sup>-1</sup>which is assigned to the vibration of free water(OH). They concluded that the presence of free water(OH) changes the chemical composition of the Trinidad Lake bitumen when modified using Petroleum bitumen (Xu et al., 2019).Furthermore, a recent study showed that the chemical composition of Trinidad Lake bitumen and other Natural bitumen has similar composition to the Petroleum bitumen, as presented in Table 4 (Wen et al., 1978).

#### 2.2. Rock bitumen

Rock bitumen is another form of Natural bitumen confined in a coarse-grained sandstone filled with bitumen (Sun, 2012). Since the early 17th century, Rock bitumen has been used for waterproofing in the

ship industry and the wood industry in the European region and they were commonly obtained from Switzerland, France, and Italy (Read et al., 2003). In Utah and Kentucky, Rock bitumen is available in Bituminous sandstone and schist deposit, which were mainly used until the early twentieth century (Cashion, 1973). The Utah deposit is estimated to have 800 million tons of Rock bitumen (Oblad et al., 1987), which is enough to construct 22 m wide road network three times around the earth (Read et al., 2003). Recently, Button Rock bitumen has been found in Indonesia (Rusbintardjo, 2013; Li et al., 2018; Zou and Wu, 2015). Buton Rock bitumen is reported to have approximately 100million tons of bitumen, which is an attractive alternative to the local pavement authourity (Ma et al., 2020). In general, Rock bitumen is extracted from

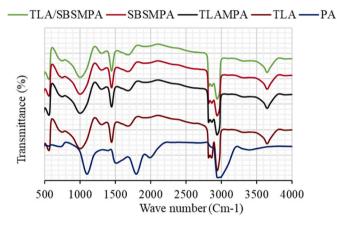


Fig. 3. FTIR results of Trinidad Lake Bitumen (Xu et al., 2019).

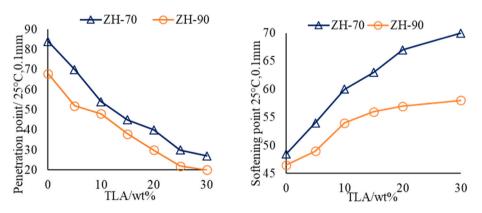


Fig. 1. Effect of Trinidad Lake bitumen on softening point and penetration (He et al., 2019).

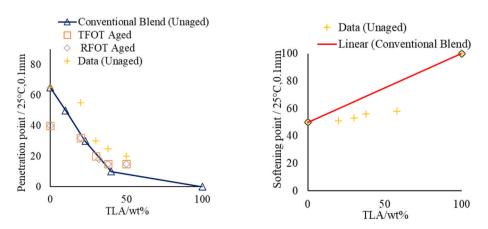


Fig. 2. Effect of Trinidad Lake bitumen on penetration and softening point values (He et al., 2019).

Typical chemical compositions of Natural bitumen of different resources (Wen et al., 1978).

Location	Bitumen content %	Chemical Compos	Chemical Compositions (Average values)				
		Saturated%	Aromatic %	Resins %	Asphaltenes %	Sulfur%	
Trinidad Island	54	5.7	24.8	38.5	31.0	6–8	
Venezuela Alberta	64	NA	NA	NA	NA	5.9	
(Athabasca, Canada)	2-8	17.2	38.2	29.3	15.3	3.54	
		13.6	39.8	40.6	6.0	5.3	
		14.1	20.3	53.3	12.3		
		14.2	20.6	51.6	13.7		
Tham region, France		14.6	30.6	47.9	6.9		
		14.0	25.1	52.0	8.9		
Switzerland		10.8	31.8	30.6	26.8		
Albania		20.5	12.4	53.6	13.5	0.77	
Romania		22.6	27.9	42.3	7.2	1.10	
SW of France	8–14	NA	NA	NA	NA	6.1	
	15–22					0.7	
		1.2-4.6	14.4-35.3	34.6-48.7	19.4-46.8	7.9–10.6	

mines and the percentage of bitumen content depends on their deposit types, sources and origins (Read et al., 2003).

#### 2.2.1. Physico-chemical composition of rock bitumen

The well-known Rock bitumen that has been in use for road construction are from Kentucky, Texas, Oklahoma, Colorado, and California. They are commonly found with varying bitumen content percentages, as shown in Table 5 (Speight, 2000). Researchers also reported that the bitumen grade of Rock bitumen usually depends on the geological location. The physical composition of Rock bitumen is presented Table 6.For decades, pavement engineers have been attracted to the use of Rock bitumen as a natural source of bitumen due to its ability to enhance high-temperature properties (Florence, 1968). In addition, the rutting and the fatigue resistance of the Rock bituminous pavements were found to improve (Jahanian et al., 2017; Ameri et al., 2018; Ma and Zhang, 2013; You et al., 2018a).Other researchers reached similar conclusions for different types of Rock bitumen (Subagio et al., 2003; Huang and Xu, 2013; Cheng et al., 2020; Zhong et al., 2017).

Buton Rock bitumen has an Asphaltene content of about 20% (Lv et al., 2019)in its granular form, and it has been mostly used for Petroleum bitumen modification. The Four-point bending test was conducted on modified bituminous mixtures to evaluate the fatigue performance. It was observed that the Bituminous mixtures modified with Buton Rock bitumen had better resistance to fatigue failure (Karami, 2020). Researchers (Rusbintardjo, 2013) found that the addition of Buton Rock bitumen into Petroleum bitumen reduces penetration value and increases the softening point as the percentage of Buton Rock bitumen to Petroleum bitumen into the Petroleum bitumen enhances the temperature susceptibility of Petroleum bitumen. On the other hand, it was reported that the modified bitumen could be prone to brittleness and could be susceptible to cracking. Furthermore, the authors concluded that the Petroleum bitumen modified with 50% of

Table 6		
Droportion	of Pock	hitumon

Properties of Rock bitumen (Speight, 2000).

Property	Types of Rock bitumen				
	XRA (Zhong et al., 2017)	Buton (Li et al., 2015)	QC (Li et al., 2015)	UM (Li et al., 2015)	
Bitumen content (%)	99.4	28	83	82	
Mineral filler content (%)	-	72	17	18	
Specific gravity	1.06	1.62	1.10	1.16	
Softening Point	-	144	99	N.A.	
Flashpoint. °C	>230	N.A.	N.A.	N.A.	
Ash content	0.54	N.A.	N.A.	N.A.	
Loss of mass	0.0632	N.A.	N.A.	N.A.	
Moisture content	0.1	N.A.	N.A.	N.A.	

NA means Not available.

Buton Rock results in high Marshall stability values (Rusbintardjo, 2013).

A recent study (Limin et al., 2016)shows that the Petroleum bitumen modified with 50% of Buton Rock bitumen has higher rut and fatigue resistance than Petroleum bitumen, as shown in Fig. 4. Another study (Zhong et al., 2017) indicated that Rock bitumen enhances high-temperature performance. However, the addition of Rock bitumen to Petroleum bitumen affects the low-temperature performance of the bitumen and mixtures (Zhong et al., 2017). Hence, engineers and pavement scientists must be careful when using Rock bitumen for paving operations in a negative way.

Researchers (Widyatmoko and Elliott, 2008)have studied the chemical composition of Rock bitumen and reported that different types of Rock bitumen vary in terms of hydrocarbon composition. Another study (Mastalerz et al., 2018) showed that Rock bitumen contains Carbon with varying content from 72 to 93%, Hydrogen from 2 to 7%, and

#### Table 5

Typical composition and properties of Rock bitumen (Speight, 2000).

Property	Kentucky <sup>a</sup>	Oklahoma <sup>b</sup>	Alabama <sup>c</sup>	Texas <sup>d</sup>	Sicily <sup>e</sup>	Germany <sup>f</sup>
Bitumen content, (%) Softening point	7–9	12.5 28	4.5	10–20	8–9 38	4.7–9.5 30–36
Penetration Type of mineral	Too soft Sand sandstone	>360 Sand	15–25 Oolitic Limestone	4 Limestone	Too soft Marlylimestone	soft Limestone

<sup>a</sup> Crushed to fine size, less than 6.4 mm. Spread and rolled at temperatures above 15.6 °C for pavements.

<sup>b</sup> Product blended from various deposits or sand added.

 $^{\rm c}$  Quarried, crushed, sized from 0.95 cm to dust and fluxed with petroleum bitumen for use as cold-applied pavement surfacing.

<sup>d</sup> Mixed with trap rock and petroleum bitumen to obtain mix for cold rolling as pavement surfacing.

<sup>e</sup> Selected grades ground and used for paving purposes.

<sup>f</sup> Enriched by adding petroleum bitumen and marketed for use as pavements or mastic.

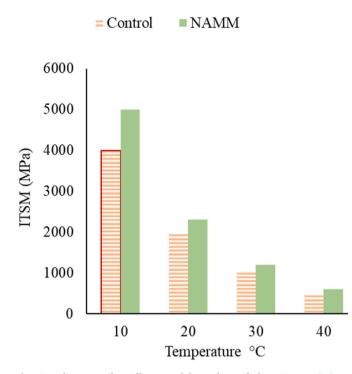


Fig. 4. Indirect tensile stiffness modulus values of the mixtures (Yılmaz et al., 2012).

Nitrogen from <0.1% to 5%, Oxygen content is typically below 1%, whereas Sulfur content ranges from <0.1% to 8%.They reported that Rock bitumen contains Aromatics (54.11%), Resins (11.01%), Saturates (2.43%), and Asphaltenes (32.45%).Table 7 presents the chemical composition of various Rock bitumen with their bitumen content and SARA values (Hosterman, 1990).

Previous researchers determined the chemical composition and the percentage of minerals in Rock bitumen using X-ray fluorescence (Darwis and Massinai, 2018). Researchers reported that the Rock bitumen

Chemical composition of Rock bitumen location (Hosterman, 1990).

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contains  $Fe_2O_3$ , MgO, CaO, and SiO<sub>2</sub>, as main constituents (See Table 8).

Asbuton is another form of Rock bitumen which are obtained from Buton island contains bitumen and mineral with high lime content (CaCO3).The examined sample of Asbuton contained 37.90% Asphalt, 43.28% carbonate, and 18.82% other minerals (Darwis and Massinai, 2018).The following sub-section would discuss about Gilsonite.

#### 2.3. Gilsonite

Gilsonite is a Natural bitumen also known as Asphaltite, Uintaite, or Asphaltum, which are generally found in the United States, Russia, Iran, and China (Sun, 2012). Gilsonite is a by-product of crude oil separated from its reservoir beneath the earth and transported towards the earth's surface through fractures of ground layers (Ameri et al., 2011). It is a soft material that could be easily crushed into powder (Ameri et al., 2011). Since Gilsonite is a brittle and hard material, it has been mainly used for waterproofing engineering structures (Read et al., 2003).

#### 2.3.1. Physico-chemical properties of Gilsonite

Gilsonite is a resinous hydrocarbon, which can be classified as a hydro carbonic modifier. Gilsonite has been used to either blend bitumen before the addition of aggregates or it has been blended with aggregates directly before the addition of bitumen (Bahia et al., 2001).In its natural state, Gilsonite has higher stiffness (50 times) than Petroleum

#### Table 8

XRF data of Button Rock bitumen (Darwis and Massinai, 2018).

Sample	Compound metal oxide(m/m%)				
	Fe <sub>2</sub> O <sub>3</sub>	Fe <sub>2</sub> O <sub>3</sub> MgO SiO <sub>2</sub>			
1	39.07	27.18	22.80	5.78	
2	39.96	25.74	25.50	3.74	
3	5.56	0	0	89.46	
4	44.39	0	0	5.46	
5	12.79	12.79	0	76.78	
6	36.37	28.03	29.37	0.644	
7	6.99	0	4.01	71.86	

Sample id	Number	Available hydrocarbon	Asphaltene	Saturated hydrocarbon	Aromatic hydrocarbon	Polar
CA-1	US06083-1	14.37	18.9	16.9	38.4	25.8
CA-2	US06083-2	2.80	24.7	30.9	16	28.4
CA-3	US06083-3	3.87	22.2	9.7	13.2	54.9
CA-4	US06079-1	12.34	25.6	21.4	30.3	22.6
KY-1	U621141-1	8.24	24.6	20.4	35.1	19.9
KY-2	US21061-1	7.02	40.3	13	24.5	22.2
NM-1	US35019-1	6.95	46.9	20.2	22.3	10.6
OK-1	US40099-1	4.52	25.7	8.6	30.6	35.2
OK-2	US40019-1	17.56	80.9	5.8	5.2	8.1
OK-3	US40099-2	5.69	31.6	15.2	26.2	27.0
OK-4	US40019-2	10.88	81.7	4.4	4.3	9.6
OK-5	US40019-3	26.62	60.8	7.9	8.0	15.4
TX-1	US48323-1	11.42	40.1	5.3	37.7	17
TX-2	US48463-1	6.63	-	_	_	-
UT-1	US49003-1	100	29.9	2.7	52.5	15
UT-2	US49007-1	10.56	80.2	2.9	5.7	11.2
UT-3	US49007-2	15.12	15.12	21.8	31.5	16.5
UT-4	US49047-1	11.38	11.38	17.3	17.2	32.1
UT-5	US49047-2	14.27	14.27	_	_	-
UT-6	US49047-3	100	100	49.5	19.1	26.5
WY-1	US56003-1	15.35	15.35	15.5	41.5	35.5
AB-8	CA01-8	14.73	14.73	_	-	-
TT-1	TD09-1	11.96	11.96	19.7	41.2	27.2
VE-1	VE02-1	100	100	22	44.5	13.9
VE-2	VE16-1	100	100	22.8	46.4	10.8
VE-3	VE16-2	100	100	20	46.3	11.6
VE-4	VE00-1	17.29	17.29	_	_	_

CA California, KY Kentucky, NM New Mexico, OK Oklahoma, TX Texas, UT Utah WY Wyoming AB Alberta TT Trinidad and Tobago VE Venezuela.

bitumen due to its higher softening point value, as shown in Table 9. Moreover, Gilsonite has similar elemental composition as Petroleum bitumen, as shown in Table 10. The addition of Gilsonite in Petroleum bitumen increases the bitumen viscosity and reduces its penetration value.

A study (Quintana et al., 2016) reported that wet process modification enhances the bituminous mixture's mechanical properties. Furthermore, the compatibility of Gilsonite with Petroleum bitumen makes it a good Natural bitumen modifier (Aflaki and Tabatabaee, 2009). The chemical composition of Gilsonite depends on the molecular types and their internal structural arrangements.

Table 10 shows the elemental composition of Gilsonite, which is predominantly composed of Carbon and Hydrogen (Nasrekani et al., 2018). Researchers (Nciri et al., 2014) used FTIR results to show that the broad absorption bands of Gilsonite are between 3200 and 3600 cm<sup>-1</sup>. Also, it was reported that the major compositions of Gilsonite are clay, Alkane, Aromatic rings, Phenyl rings, Alcohols, Carbonyl groups, Organic sulfoxides, and Sulfate salts (Nciri et al., 2014).

The SARA results (Nciri et al., 2014) showed that Gilsonite contains more Asphaltenes and has no Aromatics, while Saturates and Resins yield only for 1.6 wt% and 18.7 wt%, respectively, as presented in Table 11.

#### 2.4. Bituminous sand mixtures

Natural bitumen is also found in the form of bituminous material mixed with fine-grain sands, which is commonly known as Natural Sand Asphalt, Bituminous Sand Mixtures, or Oil Sands (Caro et al., 2015). These are deposits of loose sands or mainly consolidated sandstones saturated with highly viscous bitumen, which contain sands and moisture in varying percentages (Adedimila, 1987). Bituminous sand mixtures are found in various countries such as Canada, Nigeria, Mexico, the United Kingdom, United States, Ecuador, Kuwait, Iraq, Malagasy Republic, and Venezuela (Adebiyi and Omode, 2007).

Bituminous sand in Athabasca Canada, is approximately 900 billion barrels, while Nigeria has a reserve of about a 42.47billion tons of Bituminous sand mixtures, a quantity estimated to be the second-largest in the world (Adegoke et al., 1980; Olabemiwo et al., 2016; Mossop, 1980).

#### 2.4.1. Physicio-chemical composition of Bituminous sand

The average bitumen content of the Bituminous sand sample from Athabasca, Canada is 16%. On the other hand, researchers (Button et al., 1980) reported that the bitumen content varies from 8% to 16% for the sample from Ilubirin, Nigeria. Also, similar bitumen contents varying from 13% to 16% was reported by other researchers (Adedimila, 1987), while an average value of 10.5% bitumen was reported by researchers (Caro et al., 2015). In another study (Enu, 1985), researchers concluded that the average bitumen content of Nigerian Bituminous sand is between 5 and 20% content by weight.

The clay content for samples from Nigeria was reported to be

Га	ы	e	9	

#### Properties of gilsonite (Nasrekani et al., 2018).

Measured parameter	Results	Test Method
Ash Content	9–14	ASTM D3174
Moisture content,	$\leq 3$	ASTM D3173
Volatile matter	63	ASTM D3175
Fixed carbon	29	ASTM D3172
Solubility in CS2	81	ASTM D4
Specific gravity @ 25°C	1.11	ASTM D3289
Normal heptane insoluble, %wt.	86	ASTM D3279
Colour in mass	Black	-
Colour in streak or powder	Brown	-
Softening Point, °C	$225\pm5$	ASTM D36
Penetration @ 25 °C, 0.1 mm	0	ASTM D5
Solubility in trichloroethylene	56	ASTM D2042

Table 10

Constitution of elements in Gilsonite (Nasrekani et al., 20	18	J)	١.
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Element analysis	Result (%)	Test method		
Carbon	74	ASTM D5291		
Hydrogen	7.1	ASTM D5291		
Nitrogen	0.67	ASTM D5291		
Oxygen	3.1	Leco(s) Analyse		
Sulfur	4	Leco(s) Analyse		

Table 1	11
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SARA composition of Gilsonite (Nciri et al., 2014).

Components	Gilsonite	TLA
Saturates	1.6	4.1
Aromatic	0	22.9
Resins	18.7	37.4
Asphaltenes, wt.%	79.7	35.6
Colloidal instability, CII	4.34	0.65
Stability	Very unstable	Stable

between 2 and 7%, while the samples from Athabasca, Canada was between 10 and 25% (Enu, 1985). The void percentages in Bituminous sand from Nigeria ranges between 24 and 35%, which is lower than samples obtained from Athabasca, Canada (35%) (Adedimila, 2004). Table 12 and Table 13 gives the physical properties of Bituminous sand. It was reported (Button et al., 1980) that the chemical components of Bituminous sands vary depending on the resource locations.

Similar to Petroleum bitumen, the bitumen extracted from Bituminous sand can be separated into four broad chemical types: Saturates, Naphthene Aromatics, Polar Aromatics and Asphaltenes (Sörensen and Wichert, 2009).Table 14 shows the composition of bitumen extracted from Bituminous sand, which was reported to be similar as Petroleum bitumen (Al-Otoom et al., 2013; Adebiyi and Omode, 2007; Tileuberdi et al., 2014; Shuyuan et al., 1995; Victor-Oji et al., 2017; Ogiriki et al., 2018; Oluwole et al., 1987; Thomas et al., 1986; Bunger, 1976; Jones, 1993). It is noted that the hydrocarbon composition reported (see Table 15) is also similar to Petroleum bitumen.

The FTIR results on extracted bitumen from Nigerian Bituminous sand are presented in Table 16, Table 17 and Table 18. In general, it can be concluded that Bituminous sand can be used for paving operations if adequately designed.

#### 2.5. Performance of lake bitumen in road construction

Pre-and post-construction performance assessment of paving materials is important for concrete conclusions, further developments and future evaluations. In this section, a brief discussion about the reported performance of Lake bitumen is presented. Researchers (Charles and Grimaidi, 1995) reported that Trinidad Lake bitumen cannot be directly used for Bituminous pavement constructions particularly due to its hardness. Hence, the lake bitumen is usually blended with a softer bitumen to produce the desired blend. For instance, top layer of a steel bridge in Taiwan (Chen et al., 2011) was successfully constructed by mixing Trinidad Lake bitumen with a penetration grade of 20/40 in a

# Table 12 Physical properties of un-modified Bituminous sand (Olabemiwo et al., 2016).

Property	Value
Penetration at 25°C(dmm)	82
Softening point <sup>o</sup> C	47
Specific gravity	1.047
Flash point <sup>o</sup> C	265
Fire point °C	275
Kinematic viscosity(cst)	350
Penetration index	-0.776

Physical properties of Bituminous sand (Ola, 1991).

Test	Result
Liquid Limit (%)	20
Plastic Limit (%)	11
Plasticity Index (%)	9
Linear Shrinkage (%)	5
Specific Gravity	2.58
Bitumen Content of in situ sample (%)	3–5
Moisture content of in situ sample (%)	1.5
Maximum dry density, standard proctor $(kg/m^3)$	1408(88 pcf)
Optimum moisture content, standard proctor (%)	7.1
Maximum dry density	1459(91 pcf)
Optimum moisture content	6.3
Maximum dry density	1493(94 pcf)
Optimum moisture content	5.2

Pcf = pounds per cubic foot.

#### Table 14

Composition of Bituminous sand (Tileuberdi et al., 2014).

Elemental Composition	SB	NBE	KA	E1	E2	ATH
Carbon	84.91	84.36	86.1	84.5	80.8	83.1
Hydrogen	11.01	12.16	11.2	10.1	9.7	10.1
Sulfur	3.050	1.294	0.5	2.6	4.2	4.8
Nitrogen	0.52	0.29	0.3	0.3	0.3	0.4
Oxygen	0.343	1.814	2.0	2.3	4.9	1.1

ratio of 25:75. In another study, SBS polymer-modified bitumen containing 20% Trinidad Lake bitumen was used as waterproof materials for bridge deck pavements (Cao et al., 2009). Researchers concluded that the produced mixture helps in reducing the effect of moisture and increases the life span and durability of the bridge deck.

Researchers (Fengler et al., 2019; Zhang et al., 2009) noted that Petroleum bitumen modified using Trinidad Lake bitumen results in increased stiffness, improved adhesion properties, improved thermal effect and improved permanent deformation. Researchers (LaForce, 2006) used Trinidad Lake bitumen to improve aggregate quality for a Bituminous mixtures overlay for a modified PG76-28 bitumen (LaForce, 2006).They concluded that the increased stiffness may also affect pavement performance and resistance to cracking. On the other hand, the researchers concluded that such mixtures may suffer from poor workability, appropriate compaction, and premature failure.

The National Centre for Asphalt Technology (Timm et al., 2014) utilised an accelerated testing facility to compare the performance of Trinidad Lake bitumen modified mixture against conventional Hot Mix Asphalt. It was reported that the section modified using Trinidad Lake bitumen performed better than the sections without Trinidad Lake bitumen after 2-year of traffic cycles. In addition, it was reported that the Trinidad Lake bitumen modified HMA has more resistance to permanent deformation (Timm et al., 2014).Furthermore, the Trinidad Lake bitumen modified mixture showed an increase in fatigue resistance with an increase in number of reduced traffic cycles. The fatigue life of modified mix was estimated to be three times higher than the reference section with petroleum bitumen (Timm et al., 2014). In another research study (Russell et al., 2008), the Washington state department of

transportation used Trinidad Lake bitumen-modified hot mixtures to overlay a steel bridge deck. The bitumen modified using Trinidad Lake bitumen improved the ability of the bitumen to withstand stress induced due to traffic (Russell et al., 2008). It was reported that the overall performance was below expectation. The reason were cited mainly as unavailable specific design procedure and specification for Trinidad Lake bitumen (Russell et al., 2008).

The Department of Transportation Nevada (Sebaaly et al., 2003) constructed a road with modified mixtures containing 75% AC20 and 25% of Trinidad Lake bitumen on a high traffic route. The Department of Transportation Nevada problems of mineral matters settling in the bitumen storage tank during construction. This resulted in pumping and mixing problems in the hot mix plant, which was later identified as the cause of developed block cracking. It was highlighted that the use of Trinidad Lake bitumen did not meet the desired performance. Based on the description provided above, in general, it could be concluded that an appropriate mix design and specifications should be developed for sustainable utilisation of Trinidad Lake bitumen in paving operations.

#### 2.6. Performance of rock bitumen in road construction

The performance of Rock bitumen have been evaluated and reported by various researchers (Wang and Xing, 2021; Liu et al., 2021a, 2021b). Although reports on the use of Rock bitumen for road construction projects are limited, Kentucky highway research (Rose, 1992) summarised the field performance of Rock bituminous pavements (Rose, 1992) in detail. It was reported that Kentucky Rock bitumen is suitable for paving in its natural state, especially when the selected Rock bitumen has bitumen content within 7–9% (Speight, 2000). Kentucky Rock bitumen was mostly used as traffic bound base and surface course for rural roads (Rose, 1992). It was also used to produce HMA, resulting in a highly resistant surface course against a high traffic road network (Florence, 1968). In addition, it was reported that using seal coats on the

#### Table 16

Functional groups in the paraffinic hydrocarbon fraction of Bituminous sand (Adebiyi and Akhigbe, 2015).

Sample and v	wave number,	cm <sup>1</sup>			Possible
Orisunbare	Agbabu	Mile 2	Illubirin	Olowo- Irele	Functional groups (Bond)
2924.15(s) Stretch	2923.50 (s) Stretch	2924.76 (s) Stretch	2923.58 (s) Stretch	2924.97 (s) Stretch	CAH (CH <sub>3</sub> )
2866.86(s) Stretch	2867.99 (s) Stretch	2866.98 (s) Stretch	2867.76 (s) Stretch	2867.36 (s) Stretch	CAH (CH <sub>2</sub> )
2726.20 (w) Stretch	2726.51 (w) Stretch	2726.92 (w) Stretch	2725.76 (w) Stretch	2726.33 (s) Stretch	СН
1456.30 (m) Bend	1457.30 (m) Bend	1457.18 (m)Bend	1456.31 (m)Bend	1458.00 (m)Bend	CH <sub>2</sub>
1376.54 (m) Bend	1376.55 (m) Bend 724.12	1376.48 (m)Bend 727.87	1376.57 (m)Bend 723.87	1376.43 (m)Bend	CH <sub>3</sub>
724.41 (w) Bend	724.12 (w) Bend	/2/.8/ (w)Bend	723.87 (w)Bend	-	CH <sub>2</sub>

Table 15

SARA data of the Bituminous sand at different sources (Shuyuan et al., 1995; Victor-Oji et al., 2017).

Composition	K	E1	E2	ATH	LU	AB	IL	ID	AY	LA
Saturates	42	22.5	13.9	22	56.64	48.39	52.41	44.14	64.79	69.44
Aromatics	14.7	17.1	7.8	21	35.85	27.35	33.64	21.78	42.63	44.95
Resins	37.2	49.6	54.4	39	19.17	15.78	21.9	11.07	24.61	28.37
Asphaltenes	6.1	10.8	23.9	18	16.37	11.17	14.89	8.63	19.72	29.28

Ath = Athabasca(Canada), Lu = Ludasa(Nigeria), Ab = Agbabu(Nigeria), Il = Ilubirin(Nigeria), Id = Idiobilayo(Nigeria), Ay = Aye-Railhead(Nigeria), La = Ladawo (Nigerian), NBE = Natural bitumen extracted from oil sands in Kazakhstan, SB = Standard paving bitumen60/90, SB = Standard Bitumen, K = Karamay(China), E1 = Erlian1(China), E2 = Erlian2(China).

Functional groups in the aromatic fraction of Bituminous sand from Nigeria (Adebiyi and Akhigbe, 2015).

Sample and wave	number (cm <sup>1</sup> )						
B1 fraction	B2 fraction	B3 fraction	B4 fraction	B5 fraction	B6 fraction	Plausible functional groups (bond)	Compound type
2921.31(s)	2921.49(s)	2921.93(s)	2921.88(s)	2921.49(s)	2921.04(s)	C–H (CH3)	Alkanes
Stretch	Stretch	Stretch	Stretch	Stretch	Stretch		
2854.29(s)	2729.83(s)	2854.64(s)	2854.21(s)	2729.83(s)	2854.85(s)	C-H (CH <sub>2)</sub>	Aromatic ring
Stretch	Stretch	Stretch	Stretch	Stretch	Stretch		
3024.71(m)	3025.11(m)	3028.81(m)	3024.93(m)	3025.01(m)	3028.01(m)	CH	
Stretch	Stretch	Stretch	Stretch	Stretch	Stretch		
728.72; 694.66	728.58; 694.60	728.66(s);	728.66; 694.63	728.58; 694.60	728.74(s);	CH	Phenyl ring
(s) Bend	(s) Bend	694.59 Bend	(s) Bend	(s) Bend	694.67 Bend		substitution bands
1463.50(m)	1463.58(m)	1463.42(m)	1463.43(m)	1465.00(m)	1463.51(m)	C=C	Aromatic ring
1604.64(w)	1604.74(w)	1604.56(w)	1604.80(w)	1604.74(w)	1604.77(w)	C=C	Alkenes
Stretch	Stretch	Stretch	Stretch	Stretch	Stretch		
3584.01(s)	3584.29(s)	3583.77(s)	3543.85(s)	3584.29(s)	3583.98(s)	N–H	Amine
stretch	Stretch	Stretch	Stretch	Stretch	Stretch		
3435.79 stretch	3436.57 Stretch	3436.00 Stretch	3436.11 Stretch	3436.57 Stretch	3382.99 Stretch	OH	Alcohol Organic
1030.75(s)	1030.23(s)	1030.18(s)	1030.69(s)	1030.23(s)	1030.76(s)	S=O	sulfoxides
stretch	Stretch	Stretch	Stretch	Stretch	Stretch		

Where B1 = Olowo-Irele, B2 = Ilubirin, B3 = Mile 2, B4 = Agbabu, B5 = Loda and B6 = Orisunbare bitumen samples.

#### Table 18

Functional groups in the NSO compound fraction of Bituminous sand (Adebiyi and Akhigbe, 2015).

Sample and wave number (cm<sup>1</sup>)

B1 fraction	B2 fraction	B3 fraction	B4 fraction	B5 fraction	B6 fraction	Plausible functional groups (bond)	Compound type
2920.85(s)	2921.54(s)	2921.02(s)	2921.39(s)	2921.74(s) Stretch	2920.74(s)	CH (CH3)	Alkanes Aromatic
Stretch	Stretch	Stretch	Stretch		Stretch		ring
2871.91(s)	2869.65(s) stretch	2870.99(s)	2869.72(s)	2869.27(s) Stretch	2870.75(s)	C-H (CH2)	
Stretch		Stretch	Stretch		Stretch		
3086.55(m)	3086.47(m)	3086.47(m)	3086.45(m)	3086.41(m)	3086.50(m)	CH	Phenyl ring
Stretch	stretch	Stretch	Stretch	Stretch	Stretch		substitution bands
728.86; 694.45(s)	728.96; 694.45(s)	729.00(s) Bend	729.06; 694.46(s)	728.93; 694.45(s)	728.89(s) Bend	CH	Aromatic ring
Bend	Bend		Bend	bend			
1604.65(s)	1604.60(s)	1604.59(s)	1604.58(w)	1604.57(w)	1604.62(w)	C=C	Alcohols, ethers,
Stretch	Stretch	Stretch	Stretch	Stretch	Stretch		carboxylic
1178.72;	1178.68; 1106.62;	1178.68;	1176.64; 1106.65;	1178.63; 1106.52;	1178.70;	C-O	acid, esters
1106.63;	1030.17;	1106.57;	1030.17;	1030.15;	1106.61;		
1030.17;		1030.17;			1030.18;		
1081.42(s)	1081.39(s)	1081.39 (s)	1081.39(s)	1081.36(s) Stretch	1081.41 (s)		Aldehydes, ketones
Stretch	Stretch	Stretch	Stretch		Stretch		carboxylic
1735.58(s)	1735.58(s)	1735.50(s)	1735.00(s) stretch	1735.03(s) Stretch	1735.59(s)	C=O	acids, esters
Stretch	Stretch	Stretch			Stretch		
3584.00(w)	3584.30(w)	3584.02(w)	3583.97(s)	3584.30(s) Stretch	3584.05(s)	N–H	Amide
Stretch	Stretch	Stretch	Stretch		Stretch		
2871.91; 2920.85	2869.65; 2929.54	2870.99; 2921.02	2869.72; 2921.39	2869.27; 2921.74	2870.75; 2920.74	OH	Carboxylic acid
(s) stretch	(s) stretch	(s) Stretch	(b) Stretch	(s) Stretch	(s) Stretch		
1178.72(v)	1178.68(v)	1178.68(v)	1178.64(v)	1178.63(v)	1178.70(v)	C-O-C	Anhydrides
1030.17(s)	1030.17(s)	1030.17(s)	1030.17(s)	1030.15(s) Stretch	1030.60(s)	S=O	Organic sulfoxides
Stretch	Stretch	Stretch	Stretch		Stretch		
1156.44(s)	1156.68(s)	1156.62(s)	1156.69(s)	1156.57(s) Stretch	1156.62(s)		Sulfate salt
Stretch	Stretch	Stretch	Stretch		Stretch		

Where B1 = Olowo-irele, B2 = Ilubirin, B3 = Mile 2, B4 = Agbabu, B5 = Loda and B6 = Orisunbare bitumen samples.

lean Rock bitumen bases improved the resistance against deterioration. However, it was later highlighted that the sealed bases deteriorated rather quickly (Havens and Williams, 1956). In another study, it was highlighted that the bitumen content in the Rock bitumen was too low to provide sufficient bonds to withstand traffic and adverse environmental conditions (Havens et al., 1974). Researchers also reported that Rock bitumen mixture confined with sandstone particleshave goodskid resistance properties (Havens et al., 1974).

HMA produced using Rock bitumen was placed on the Nolin Dam Road and was reported to have good stability and high wet skid resistance due to high air void content (13–15%) (Florence, 1966). As a caution it was advised that unless the base was adequately primed and sealed before surfacing, rainwater could damage the base and underlying soil. It is noted that researchers used conventional dryer pugmill to produce Rock bitumen-modified HMA. The addition of partial Rock bitumen was found to increase the stiffness of the Petroleum bitumen resulting in an improved anti-rutting performance as reported by another study (Li et al., 2015). The Rock bitumen in paving operation could be used in conditions where ageing resistance characteristics are essential and/or waterproofing is necessary. However, there are concerns associated with modifying Petroleum bitumen using Rock bitumen, such as natural segregation, poor workability, and adverse changes in low temperature rheological characteristics (Ameri et al., 2018; Li et al., 2015; You et al., 2018b).

Another studies (Havens and Williams, 1956) reported that the field performance of Rock bituminous cold mixture was not up to expectations. As reported by researchers (Widyatmoko and Elliott, 2008), commercialising the production of Rock bitumen through chemical or solvent extraction process comes with a huge financial burden hence road assets managers should carry proper benefits cost analysis. On a positive note, it must be remembered that Rock bitumen was used to construct wearing course in Kentucky from 1900 to 1950 and was regarded as the choice of highway engineers (Rose, 1992). It provided a good riding quality and enhanced skid resistance. With proper scientific investigations, its ability to withstand modern traffic conditions should be further examined.

#### 2.7. Performance of Gilsonite in paving operation

Researchers (Akbari Nasrekani et al., 2016) added Gilsonite in 5% and 10% by weight of Petroleum bitumen and reported an increase in complex modulus while the phase angle reduced at high temperature, indicating an improvement in rut resistance. Similar study (Yilmaz and. Celoğlu, 2013) reported that the addition of Gilsonite produces mixtures with improved mechanical performance such as water sensitivity, resistance to fatigue, rutting, direct tensile strength. Researchers (Ameri et al., 2011) highlighted that the high temperature performance of mixtures improved when 4%, 8% and 12% of Gilsonite bitumen is added into Petroleum bitumen. Research studies (Polacco et al., 2006; Bitumen, 1995) are relatively inconclusive about the influence of Gilsonite at low temperatures, as contradicting conclusions were reported in different studies. In addition, as shown in Fig. 4, the average stiffness of Gilsonite modified mixtures regardless of the temperature was higher than the Petroleum bitumen (Yılmaz et al., 2012). In another study, researchers (Themeli et al., 2016)showed that the aging resistance of soft bitumen improved when 10% of Gilsonite was added in Petroleum bitumen. As reported by researchers (Liu and Li, 2008), Gilsonite has a high potential to improve the mechanical properties of bituminous mixtures. It has been reported that Gilsonite-modified HMA mixtures extended the expected life of the pavement, improved the stripping resistance, and significantly reduced shoving and rutting distress (Liu and Li, 2008).

Gilsonite-modified bitumen have been successfully used in hightraffic stressed networks in Norway, on toll booth sections in New Jersey, on major city streets and highways in Australia, Singapore, Indonesia, Japan, France and Germany (Jahanian et al., 2017; Yang et al., 2019). A comparative field performance test was conducted in Germany using Trinidad Lake bitumen and Gilsonite modified HMA. In the study, mastic produced using Petroleum bitumen was modified with 24.4% Trinidad Lake bitumen and other samples were prepared to modify petroleum bitumen using 5% and 6% of Gilsonite bitumen. The results show that Gilsonite modified bitumen had an increased softening point, viscosity, and elasticity than the Trinidad Lake bitumen, indicating high resistance to rutting (<AGC-Asphalt-Germany-Oct2020\_CH. pdf>., 2020). In addition, researchers (<AGC-Asphalt-Germany-Oct2020\_CH.pdf>., 2020) reported that Gilsonite modified HMA performances are better as they have shown more resistance against cracking.

#### 2.8. Performance of Bituminous sand in paving operations

It is noted that the reports on the utilisation of Bituminous sand in road construction are limited (Adedimila, 2004), hence the following conclusions are reported on the basis of a handful research studies. As reported in previous studies (Yılmaz et al., 2012), Young's modulus of the Bituminous sand-modified mixtures with 8.33% modifier compared to a Petroleum bitumen mixture were higher. Other studies reported that Bituminous sand could be used as a base course or wearing course with a proper mix design (Adedimila, 1987; Zhou, 2019). The utilisation of Bituminous sand in road construction has advantages as it allows for a lower temperature(104 °C) mixing than HMA(177 °C). Researchers also concluded that consumption using the Bituminous sand mixtures could significantly reduce overall emissions (Vrtis and Romero, 2015). This is associated with lower mixing temperature being more environmentally

friendly, which lowers the generated emissions and helps maintain air quality (Vrtis and Romero, 2015).

The performance of roads constructed using 33% Bituminous sand and 67% coarse aggregates was reported to be an optimal ratio for good bonding based on the studies conducted by researchers (Vrtis, 2013). Hamburg wheel tracking test was conducted on bituminous pavement slabs produced with Bituminous sand. The rut depth was less than 10 mm after 10,000 passes. However, the mixtures were more prone to moisture-induced damage at 50 °C (Vrtis, 2013). In addition, the bending beam rheometer test result produced high stiffness at low temperatures from laboratory and field samples compared to Petroleum bitumen (Vrtis, 2013). Previous research studies (Pei et al., 2020) showed that adding Bituminous sand into bituminous mixtures enhances the mechanical performance of the mixture. Researchers (Nawarathna, 2021) studied the effect of Recycled asphalt pavement (RAP) and Recycled asphalt Shingles (RAS) when added in conjunction with Bituminous sand. They reported that the addition of Bituminous sand improves the high-temperature performance.

In general, the authors feel that more laboratory tests are required to check the suitability of Bituminous sand (with/without RAP) for major road applications.

#### 3. Application of Natural bitumen: current perspective

Natural bitumen has been used in pavement applications for a long time. However, the demand decreased over the years due to the availability of more economic resources. As highlighted in the introduction section, the application of Natural bitumen in the pavement field is regaining popularity due to several reasons. Natural bitumen as directly obtained from their source may not be suitable for different pavement applications. This section aims to provide a summary of potential applications of different Natural bitumen under current perspective. The use of Natural bitumen in pavement construction could be in the form of full replacement (Xu et al., 2019); or partial replacement of Petroleum sourced bitumen depending on different factors: a) Quality of Natural bitumen; b) Type of pavement (Low volume/high volume); c) Economic feasibility etc. Based on the various use of different bitumen sources in the previous sections, a summary of potential applications is provided in Table 19.

As highlighted by many researchers, Natural bitumen has the potential to be used in the field for various applications such as patch work, major maintenance or even road construction. It is also evident from the discussion of the previous section (see Section 2) that there are still many aspects of Natural bitumen which are not well examined. Hence, their large-scale applications in pavement engineering are challenging.

Table 19
Possible application of various Natural bitumen.

Natural bitumen source	Potential application
Lake bitumen	a) Can be used as a modifier in the bitumen of high penetration value (Liu et al., 2019; Yilmaz et al., 2011).
	<li>b) Can be used as a full replacement with soft non-Petroleum bitumen (waste engine oil, bio-oil etc.).</li>
	c) Can be used in all layers of pavements.
Gilsonite	a) Can be used as a bitumen course directly.
	b) Can be used for potholes maintenance.
	c) Can be optimised for wearing course application directly.
Rock bitumen	a) Can be used in bitumen course directly
	b) Can be used for bitumen modification
	c) Can be optimised for wearing course application directly
Bituminous sand	a) Can be used as a bitumen course directly.
	b) Can be used for potholes maintenance.
	c) Can be optimised for wearing course application directly.

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#### 3.1. Difference between Natural bitumen and Petroleum bitumen

Natural bitumen, as earlier defined in the previous section, are derived naturally, while the Petroleum bitumens or Petroleum bitumen are derived from the distillation of crude oils (Read et al., 2003). The major difference between Natural bitumen and Petroleum bitumen is based on the volatile nature of the hydrocarbons fractions such as paraffinic, naphthenic and aromatic (Meyer et al., 2007). Natural bitumenhas been reported to have high asphaltene content and higher molecular weight compared to refinery/Petroleum bitumen (Nciri et al., 2014).

The asphaltene content has a major effect on the rheological characteristics of a bitumen. Increasing the asphaltene content produces a harder, more viscous bitumen with a lower penetration, higher softening point (Bitumen, 1990). It has also been reported that the rheological properties of bitumen are dependent on the asphaltene content (Lin et al., 1996). In addition, the high asphaltene content improves the properties of asphalt mixtures such as stiffness and deformation resistance (Speight, 2009). Furthermore, as reported in (Meyer et al., 2007) the removal of asphaltene or preasphaltene during the refinery process would affect the rheological properties of the Petroleum bitumen. While a higher molecular weight would increase the softening point, reduces the penetration value of bitumen, improving the lower temperature susceptibility properties (Chen et al., 2003). In addition to the previous characteristic and differences highlighted above, the large asphaltic molecules in Natural bitumen as compared to Petroleum bitumen, result in changes (increase or decrease in)the density and viscosity of the oil (Meyer et al., 2007).

Past studies claim that Petroleum bitumen possesses adequate performance characteristics when high temperature-dependent properties are required (Bitumen, 1990). However, low temperature-dependent properties are compromised in such cases (Bitumen, 1990). Researchers have found that blending petroleum bitumen with Natural bitumen enhances the low temperature-dependent properties (Bitumen, 1990). Some of the average chemical and physical properties of the conventional and Natural bitumen are given in Table 20.The corresponding table shows their relative characteristics. In addition, Natural bitumen is relatively hydrogen-deficient when compared to heavy oil and conventional crude oil, and often the addition of substantial hydrogen is added (Bailey, 1991).

Natural bitumen and bitumen from conventional crude oil differ from light oils by their high viscosity at reservoir temperatures, high density (low API gravity), and significant contents of Nitrogen, Oxygen, Sulfur compounds and heavy-metal contaminants, which are similar to the residuum from the refining of light oil (Meyer and Attanasi, 2003). Natural bitumen and bitumen from the refinery are used to produce paving-grade bitumen, and most grades of bitumen are produced by

#### Table 20

The properties of conventional crude oil and Natural bitumen (S	Speight, 2005).
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Property	Natural bitumen	Conventional crude oil
Gravity, <sup>o</sup> API	8	35
Viscosity centipoise@38 °C	500,000	10
Pour point F	50	0
Asphaltenes % by weight	19	5
Resins	32	10
Aromatics	30	25
Saturates	19	60
Metals per million Vanadium	250	10
Nickel	100	5
Carbon residue % by weight	14	5
Heating Value(btu/Ib)	17,500	19,500
Carbon %by weight	83	86
Hydrogen% by weight	10.6	13.5
Sulfur % by weight	4.8	0.1
Nitrogen % by weight	0.4	0.2
Oxygen % by weight	1	0.2

blending with the addition of polymers (Aflaki and Tabatabaee, 2009).

# 4. Current challenges, opportunities and proposed methodological solution

As highlighted in the previous section, there are several challenges in using Natural bitumen for pavement related applications. Hence, it is necessary to develop a framework to evaluate Natural bitumen through scales, i.e., from bitumen scale to mixture scale, for its potential utilisation in bituminous pavement constructions. Some of these challenges and methodological approach to examining Natural bitumen are highlighted in the following sub-sections.

The literature review on using natural bitumen for pavement construction has been discussed extensively. In addition, the review gave an insight into the opportunities and challenges for further research for the utilisation of natural bitumen in the pavement construction industry.

#### 4.1. Challenges and opportunities

Challenges of utilising Natural bitumen as a partial replacement or full replacement of Petroleum bitumen could be linked to life cycle cost, their implication to the environment, best construction practices &tools. To the best of the author's knowledge, standard specifications are not available to practitioners that cover different aspects of Natural bitumen. Hence, a thorough study needs to be carried out to link the physico-chemical properties of Natural bitumen with mixture design. The cost-benefit analysis must be carried out because economic viability pertaining to its technical characteristics must be estimated. In such analyses, factors such as proximity of natural resources, implications on air emissions, cost of importing petroleum bitumen, expected life time etc., should be considered.

#### 4.1.1. Challenges

The use of natural bitumen in the road sector has challenges ranging from its exploration process, available scientific data, sourcing design to fit into current practice, methods of evaluating their use in the road sector etc. The following subsection would highlight some of these challenges elaborately.

4.1.1.1. Inadequate and sufficient scientific data for its utilisation. The unavailability of sufficient scientific data on the performance of natural bitumen among pavement engineers and practitioners is challenging. In addition, policymakers and decision-makers, especially in developing countries where these vast natural resources are available, not be technically equipped to use these resources for sustainable paving operations. Hence there is a need for more studies to cover a lot of design aspects of pavement mixtures such as long term performance. Since only a few publications considered the long-term performance aspect. Hence, more resources are needed to conduct scientific research on sustainable use of natural bitumen for pavement construction (Speight, 2005).

4.1.1.2. Unavailability of advancement and technologies. The availability of technologies and advanced equipment for characterising and exploring natural bitumen for sustainable use has hindered its usage in pavement construction. The area where most of this natural bitumen is located is inaccessible due to farmlands, unplanned settlements, and lack of access roads which affect and dictate the type of machinery used for exploration. The use of equipment that would not affect the ecosystem while exploration would be highly encouraged. This is in addition to processing and transporting natural bitumen to asphalt plants where it would be deployed for paving operations (Caro et al., 2015).

4.1.1.3. The current design of plant production technologies and system. The current plant designs for asphalt production were not designed to incorporate natural bitumen in terms of full deployment and modification of petroleum bitumen because the natural bitumen with some particles can block the bitumen storage tank. Hence there would be a need to design an asphalt plant that could accommodate natural bitumen during the production of asphalt. However, the additional costs of plant upgrading and/or modification should be assessed (Sebaaly et al., 2003).

4.1.1.4. Quality and homogeneity of natural bitumen. Considering the use of natural bitumen in asphalt mix design, the quality and homogeneity of natural bitumen are needed to be evaluated to avoid changes in the performance of asphalt. In addition, utilising natural bitumen care must be taken to ensure that there are no fine particles or impurities. The amount of fine particles may limit the maximum percentage of natural bitumen content because the gradation requirements of the final mix might not be achieved. All these concerns must be considered to achieve proper mix design and production of asphalt concretes containing Natural Bitumen (Tarsi et al., 2020).

However, the quality of the natural bitumen sourced might also depend on the sourced location, milling machine, and speed. It is always difficult and impractical for companies to store natural bitumen from different locations in different spaces before and after processing it. This is due to the limited area of the production plant. In these cases, additional operations should be performed to increase the homogeneity of the material before its characterisation and use in new asphalt mixtures. Adopting a quality control system during the extraction process that includes mixing the material extracted from different zones in the mine to reduce the heterogeneity of the material, as reported in (Caro et al., 2015).

Further elaboration of benefit-cost analysis is beyond the scope of this paper, and it opens the need for future research. It is highlighted that the life cycle cost assessment of Natural bitumen should be conducted keeping the additional cost in mind that the road authorities will be required to overcome the forcible challenges. In the following sub sections, some of the specific challenges pertaining to commonly available Natural bitumen (see Section IV) will be discussed.

4.1.2. Major challenges reported in utilisation of Trinidad Lake Bitumen Researchers (Quintana et al., 2016; Charles and Grimaidi, 1995; Russell et al., 2008; Yang et al., 2019) have highlighted various challenges in using Trinidad Lake bitumen for paving operations. These studies highlighted that fully/partially replaced Trinidad Lake bitumen reduces the bituminous pavement resistance to cracks. Moreover, the researchers also highlighted that if the incorporation of Trinidad Lake bitumen in Bituminous pavement is not properly engineered, it will result in poor mixture workability, segregation, ultimately leading to premature failure.

#### 4.1.3. Major challenges reported in utilisation of rock bitumen

Rock bitumen when used as partial or full replacement of petroleum bitumen may result in mixture segregation and poor workability. In addition, the rheological properties of the bitumen at low temperatures are also adversely affected (making it more susceptible to cracking) (Ameri et al., 2018; Li et al., 2015; You et al., 2018b). Another study reported (Widyatmoko and Elliott, 2008) thatthe usages of chemical or solvent during extraction process from Rock bitumen should be seen as an additional cost. Due to the low bitumen content in the Rock bitumen, the prepared mixtures provide an insufficient bond to withstand traffic and environmental conditions if used directly without any additives, as reported by researchers (Li et al., 2015; Havens et al., 1974; Florence, 1966). Researchers (Rose, 1992) reported that when Rock bitumen was used as a cold mix, performance was not encouraging.

#### 4.1.4. Major challenges reported in the utilisation of Gilsonite

The challenges associated with using Gilsonite for pavement construction include its high cost, mainly associated with the mining process and its labour-intensive extraction/construction processes (Nasrekani et al., 2018). The excessive costs involved with Gilsonite have made it an unattractive alternative for Petroleum bitumen.

#### 4.1.5. Major challenges reported in the utilisation of Bituminous sand

Researchers (Caro et al., 2015) reported that Bituminous sand has varying bitumen content in samples obtained from different regions. Moreover, the sand fractions are reported to be heterogeneous (Caro et al., 2015). The variability in bitumen content and minerals composition is a concern when designing a mixture.

Bituminous sand when used without the addition of additives, usually creates additional friction when being dumped into a hopper. In the process, it hinders the hopper's mixing chain, resulting in selfcompaction, which may ultimately result in the hoper malfunctioning.

#### 4.1.5.1. Opportunities.

- Natural Bitumen may be economical when the price of petroleum sourced-bitumen is higher as projected or when it is scarce due to advancements in the oil sector. In addition, there is a tendency to use natural bitumen to reduce carbon emissions. Ultimately, evaluating the environmental impacts and economic feasibility of asphalt mixtures containing natural bitumen indicates that combining it with other selective additives provides more economical and environmental advantages. However, more research is needed to consider the economic and environmental perspectives fully. The costeffectiveness and carbon footprint of using natural bitumen as an asphalt modifier needs further investigation since only a few publications have considered this aspect.
- The environmental impact and life cycle cost of utilising natural bitumen in pavement construction is an aspect of concerns that researchers need to address. According to a study conducted on a life cycle impact assessment and Life cycle inventory study on the various stages of natural bitumen production. The study reported that comparing natural bitumen and bitumen produced from crude oil refineries, the CO<sub>2</sub> emission released during the production of Natural bitumen is reduced by approximately 44% compared to that of crude oil bitumen.
- The current refining and process technologies for crude oil where bitumen is derived might not be suitable for the refining process of natural bitumen in terms of collection from the site, transportation removal of minerals if present, storage system etc. It is optimistic that many new technological opportunities could arise to provide solutions that would enhance the sustainable use of natural bitumen.
- Natural Bitumen can be designed as a pellet or in a capsule form which could be used for new asphalt mixtures, rejuvenating old asphalts and improving the performance of asphalt. The industry that would be making available natural bitumen in pellets or capsules for the pavement construction industry would take this as an opportunity.
- The world is going into an era predicted that crude oil would begin to deplete. If that happens, the implication will result in more modernisation and advancement, making the production process more innovative and economical. These involve breaking down bitumen, a residue, into more financially viable products, which could significantly result in scarce bitumen for the paving community. Hence natural bitumen sources could bridge this gap in the road sector.

#### 4.2. Approach for assessing Natural bitumen in road construction

As discussed in previous sections, it is of prime importance that the available Natural bitumen is properly examined before real field applications. An experimental strategy for assessing the quality of utilising Natural bitumen is proposed in this section. These include appropriate sampling procedures, characterising the physical properties, rheological properties and chemical properties. It is noted that the proposed approach is to provide road authorities and pavement researchers a simplistic guideline to evaluate Natural bitumen and an in-depth analysis or detailed description of the proposed approach is outside the scope of this paper.

In the following sections, the authors will discuss some commonly available tools and techniques that could be used for field and laboratory performance assessment. These include computational tools, smart sensors, and other non-destructive methods. A proposed methodological approach is shown in Fig. 5 that could be adopted or modified to suit the prevailing conditions in carrying out a small/large scale study.

#### 4.2.1. Laboratory-based approaches

4.2.1.1. Material sampling. Material sampling is an important first step in the whole process and, if not done properly, may affect the research conclusions significantly. It is obvious that during the sampling of Natural bitumen, heterogeneity of material is expected. To reduce the influence on the performance of the location of the bituminous material, the location where it is sourced can be stratified into different zones. During sampling, tools such as hand-held diggers, sample bags, permanent makers, field notebooks and clipboards, and hand-held GPS should be carried. As a good sample collection practice, the following procedure can be adopted:

- i. The number and approximate location of Natural bitumen samples should be marked before going to the field.
- ii. The sample collection location can be marked using GPS or any smartphone having such an application. This information could be helpful when tracking locations where samples were collected in subsequent sampling events for further verification.

- iii. Unnecessary residue from the top soil should be removed before sample collection.
- iv. The sample should be dug out mechanically by inserting the soil probe to the desired depth. However, care must be taken to prevent the probe from tilting.

Once sampling of Natural bitumen is completed, it is important to ensure that samples are stored properly to avoid environmental contamination. After sampling collection, it is advisable to keep the samples in a refrigerator to minimise the chance of the Natural bitumen forming mould growth. Furthermore, excess samples can be air-dried slightly by spreading samples on a flat surface. Oven drying of samples excessively should be carefully done to avoid unnecessary aging damage to the samples.

4.2.1.2. Laboratory testing. Conducting relevant laboratory tests that could be used to scientifically characterise and classify a Natural bitumen (in terms of its suitability) is practically "a must" before further investigations. It is advised that the obtained samples should be tested through different scales. Moreover, Natural bitumen will age over time and the ageing process might be completely different than Petroleum bitumen (Jing et al., 2019). Over a period of time, this will change the Natural bitumen's chemical and mechanical responses, possibly making the Natural bitumen stiffer and more elastic, which in extreme cases may result in an early brittle failure. It is highlighted that ageing of Natural bitumen could also occur during mixing, production, and transportation at a faster rate hence, ageing related laboratory investigations should be properly carried out.

The following paragraphs describe commonly available laboratory equipment which could be use to test and compare aged/unaged Natural bitumen/Petroleum bitumen.

4.2.1.3. *Physical test.* It is proposed that as the first stage of filtering, common physical tests should be performed on Natural bitumen under standard conditions with a reference Petroleum bitumen. These physical tests include: penetration tests, softening points, viscosity, etc. Which would help in determining the consistency of the Natural bitumen as compared to Petroleum bitumen.

As per the authors' views, in case slight variation from the standard

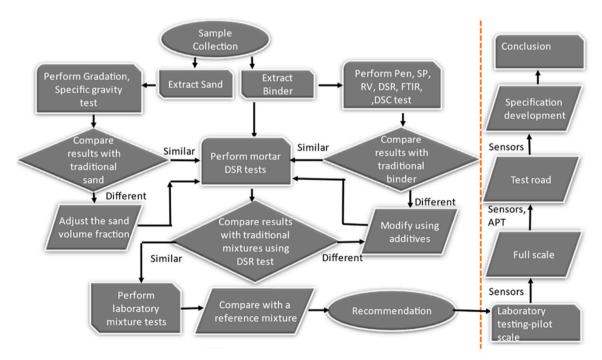


Fig. 5. Schematic of the experimental design.

practices is realised, these samples should not be disqualified immediately, but further investigations should be carried out. Short-term and long-term ageing effects on the Natural bitumen by using a Rolling thin film oven (RTFOT) (EN, 2014) and Pressure ageing vessel (PAV) (Jing et al., 2019; EN, 2012) should be done. Once the standard tests are performed, further advanced tests such as (FTIR) should be conducted to analyse the chemical composition of Natural bitumen through functional groups identifications. Such tests could also assess the compatibility with petroleum bitumen and Natural bitumen (Hofko et al., 2018) in case the Natural bitumen is used as a partial replacement.

The rotational viscosity test should be used to investigate the viscous nature on both aged and unaged samples (ASTM, 2015). The material's sensitivity towards temperature during mixing and compaction could be carried out using Brookfield rotational viscometer (ASTM, 2015). This test will help develop appropriate mixing and compaction temperature for Natural bitumen under field conditions.

The rheological properties (Complex shear modulus ( $G^*$ ) and Phase angle ( $\delta$ ).) of Natural bitumen should be characterised using a dynamic shear rheometer (E. and Bitumen and bituminous binders, 2012). Dynamic shear rheometer can also evaluate the fatigue and permanent deformation of the Natural bitumen (E. and Bitumen and bituminous binders, 2012). It is noted that the identification of thermal effects on the DSC curves depends on the accuracy of quantitative evaluation of the processes (Frolov et al., 2019).

The Atomic Force Microscope (AFM) can help in the evaluation of Natural bitumen's microstructure surface analysis and material property analysis using a sharp tip probing sample at the micro and Nano scale (Pauli et al., 2011). The AFM results can help improve the performance of Natural bitumen in case of polymer, waste rubber, etc., is added. It is noted that the AFM is not readily available to pavement engineers, and in such cases this step can be skipped.

Mixture scale tests should be done according to regional specifications using devices such as four-point bending beam, Indirect Tensile Test, etc., (Rowe et al., 2012). Fatigue test could be performed on Natural bitumen by using the above-mentioned devices. However, all these tests are calibrated for petroleum bitumen and may not be valid for Natural bitumen hence there is a need to design appropriate protocols.

Permanent deformation tests such as the repeated load Triaxial test to determine the sensitivity of Bituminous mixes should be done (Garba, 2002). Permanent deformation can cause rutting which is very dangerous for the road user, especially in wet weather conditions. After coming up with an appropriate mixture design in the laboratory the next phase would be to carry out a pilot scale Laboratory/Field testing.

#### 4.2.2. Computational approaches

In modern days, computational tools in pavement engineering are gaining wide popularity. These tools can numerically simulate the behaviour of bitumen through scales (Zheng et al., 2021). Upon proper validations, such tools cost less time and human effort resulting in minimising laboratory and in-field tests. In this section, some useful computational tools are highlighted briefly. Detailed descriptions and covering all the available tools are beyond the scope of this paper.

4.2.2.1. Molecular dynamics simulation tools. Molecular dynamics simulation has been widely used by pavement engineers to investigate the mechanism and microscopic development of bitumen composition in relation to their higher scale performance (Guo et al., 2020; Ding et al., 2021). Hence, the authors expect that the tools can be utilised to further examine the behaviour of Natural bitumen at the Nano-scale compared to the petroleum bitumen.

*4.2.2.2. X-ray CT scans.* Xray CT scan is a commonly used technique for characterising the internal microstructure of heterogeneous, opaque, composite materials. Past researchers have successfully used Xray CTscan techniques to study different physical processes (in relation to

stress development) of bituminous mixtures (Spinel, 2011). It is recommended that the Natural bituminous mixtures sample should also be deeply studied using these techniques. This would help pavement engineers evaluate various distress propensity for these mixtures(clogging, Ageing, moisture damage, etc) (Caro et al., 2010). In addition to reducing the structural load-carrying capacity of the material, Pavement distresses development such as ravelling are induced due to the presence of moisture in pavements.

4.2.2.3. Finite elements tools. Caro (Spinel, 2011) formulated a 2D FE-based micromechanical model for predicting moisture-induced damage in AC. The developed computational model coupled the effect of moisture transport with the mechanical response of the microstructure. The impact of moisture presence was examined in terms of cohesive and adhesive degradation. Moisture-induced effects on Natural bitumen could be studied using these techniques.

4.2.2.4. Discrete element and finite element to understand behaviour at mixture scale. In the last couple of decades, many micromechanical models based on computational approaches have emerged. Such numerical micromechanical models were mostly based on the Finite Element (FE) or the Discrete Element (DE) method. The viscoelastic behaviour of the Natural bitumen can be modelled with the Burgers/Generalised Maxwell model from the obtained laboratory tests. This technique could be used to study the compaction behaviour of Natural bitumen (Liu et al., 2009; Al Khateeb et al., 2021).

# 4.2.3. Economic and environmental benefits of Natural bitumen application in paving operations

Economic analysis and environmental impact of Natural bitumen is an important aspect of modern days requirements. As mentioned in prior sections, the major application of Natural bitumen in the asphalt paving operation was only done before the availability of petroleum Bitumen. In general, the different phases of pavement construction is known to have a negative impact on the environment (Cui et al., 2020; Chauhan et al., 2010). The bitumen derived from crude oil processing at ambient temperature is stable and poses no health hazard. However, the hydrocarbon emissions produced during production, storage, and transportation may pose a risk to construction workers (Cui et al., 2020; Chauhan et al., 2010). Moreover, the net energy consumption in petroleum and Natural bitumen may vary (Wahjuningsih et al., 2018). Although benefit-cost analysis considering issues such as modern economics, environmental impacts, circularity etc., are complex and must be examined thoroughly because of the limitation of the word limit, this is beyond the scope of this paper.

#### 5. Conclusions

The paper presented a general overview of the use and application of Natural bitumen for pavement construction. Studies have been conducted to evaluate the applicability of Natural bitumen for paving operations. On the basis of the literature review, the following conclusions can be drawn:

- Natural bitumen has been gaining popularity in recent years due to depletion in crude oil resources, advances in crude oil refining processes, increased demand for highway infrastructure, and regional transportation-environmental policies.
- The available reserve of Natural bitumen can be found at several places across the globe 1,856,853 billion barrels as demonstrated and 864,841 billion barrels as inferred. Hence Natural bitumen has sufficient reserves to serve as an alternative to Petroleum bitumen.
- In this article, Natural bitumen was broadly categorised into four major types, i.e., Lake bitumen, Gilsonite, Rock bitumen, and

Bituminous sand. Their respective merits and demerits pertaining to pavement construction were also highlighted.

- Lake bitumen has been found to improve the ageing resistance, permanent deformation resistance, thermal stability, and fatigue crack resistance of bituminous pavements. The addition of Lake bitumen into the petroleum bitumen increases the softening point of the bitumen and reduces the penetration value. However, modified bituminous mixtures may not be easily workable and compatible, making them prone to premature failure.
- Bituminous sand has been found to have variable compositions. Previous studies suggested to divide Natural bitumen resources into different imaginary zones. This way, Natural bitumen would have similar physico-chemical characteristics within a particular zone.
- When modified with Rock bitumen, Petroleum bitumen showed improved fatigue and rutting performance. In addition, it was found to have good anti-skid resistance properties. However, low bitumen content in Rock bitumen results in poor workability, poor rheological properties and aggregates segregation during construction.
- Gilsonite has zero penetration value, and when added to bituminous mixtures, the resistance against permanent deformation was found to be improved. Also, it has been reported that the cost associated with the mining process have made Gilsonite unattractive for road practitioners.
- A simplistic guideline and framework to evaluate Natural bitumen in in-depth analysis or in detail was proposed in the article.
- Modifying bitumen using Natural bitumen may help promote sustainability practice in pavement construction, resulting in reduced energy consumption and CO<sub>2</sub> production that is usually associated with the production of Petroleum bitumen.

In summary, using Natural bitumen for pavement construction can reduce the dependency on petroleum-sourced bitumen. Secondly, adding Natural bitumen improves the Physico-chemical properties and the performance of pavements. However, Natural bitumens had been less utilised for decades due to limited practical and field studies. Thus, to bridge this gap, this review gives future recommendations to increase the utilisation of Natural bitumen as a new binder.

#### 6. Future recommendation

This review paper has shown that Natural bitumen may be a good alternative to Petroleum bitumen for paving operations which can help reduce carbon emissions. However, the Natural bitumen cannot be fully utilised due to lack of scientific evidences. Keeping this in mind, future studies will be carried out on following aspects:

- Optimization of Natural bitumen content in terms of enhancing physico-chemical performances.
- Sufficient data should be provided to predict the long-term performance of Natural bituminous pavements.
- The recyclability of Natural bitumen should also be evaluated to help predict more practically replacement of aged Natural bitumen with new Petroleum bitumen should be studied.
- The environmental, economic, and life cycle cost of using Natural bitumen for pavement construction should be considered to have field-based experimental data.
- Dedicated specifications and codes for Natural bitumen should be developed.
- The actual cost-effectiveness of Natural bitumen for road construction is encouraged to be conducted on an industrial scale to aid commercialization.
- The cohesion and adhesion properties of Natural bitumen on aggregates should be evaluated for better performance.
- Life cycle assessment should be conducted on using Natural bitumens for pavement construction.

#### CRediT authorship contribution statement

Kumar Anupam: Conceptualisation, Supervision, Resources, Writing – review & editing. Daniel Akinmade: Data curation, Methodology, writing – original draft, Investigation, Writing – review & editing Cor Kasbergen: Writing – review & editing, Resources. Sandra Erkens: Supervision, Writing – review & editing, Data management. F. M. Adebiyi Writing – review & editing, Resources. All authors have read and agree to the published version of the manuscript.

#### Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

#### Data availability

No data was used for the research described in the article.

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