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Review article

Durability of bitumen binder reinforced with polymer additives: Towards upgrading Nigerian local bitumen

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ABSTRACT

One of the major obstacles to Nigeria's techno-economic development has been a lack of good road infrastructure. Despite a large deposit of natural bitumen in the form of semi-liquid and bitumen sand, the reliance on imported bitumen/asphalt for road construction and maintenance in Nigeria has reduced road coverage and quality. To use local bitumen as a binder in pavement construction, an efficient upgrading process is required using polymers, nanomaterials, and other chemical additives. However, the selection of an appropriate modifier depends on many factors including the origin, elemental and chemical composition of bitumen. This review presents vital properties of Nigerian bitumen extracted from oil wells and tar sands with the view to identifying potential additives as solutions for its upgrading. Based on predefined selection criteria, we conducted a systematic review of the literature. We gathered information on the current state of knowledge about the major issues encountered during the polymeric modification of bitumen. In addition, data on existing practices used by various road researchers to address such issues was gathered. Effort was made to review waste packaging polymers and plastics for possible utilization to ensure sustainable pavement infrastructure in Nigeria. The results of this review showed relatively little information on Nigerian bitumen upgrading. Many authors have investigated different polymer additives on asphaltic bitumen sourced from different countries and the results has pointed to the capability of polymeric modification to improve some of the properties of bitumen. A knowledge gap however, exists in the optimization of polymer dosage, and characterization of bitumen at the SARA level to aid the understanding of the effects of polymeric modification and mechanisms involved during the pavement degradation. Additionally, it has been challenging to generalize the effects of different polymers due to the variation of bitumen properties from different sources. This review identifies the potential for upgrading Nigerian bitumen using polymer additives, the potential of waste plastics, crumb rubbers, and packaging waste materials as alternative and sustainable additives also highlighted.

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1. Introduction

Bitumen pavements are common in many developing countries due to the improved riding surface and speedy pace of building [1]. Bitumen pavement is made from a bituminous mixture consisting of three phases: aggregate, bitumen, and an interface phase between the two. Generally, road pavement mixtures consist of about 95 % aggregates (stone, sand, and gravel) and 5 % bitumen. This small amount of bitumen, however, has a tremendous impact on pavement efficacy [2]. The degree of adhesion between bitumen and aggregate is principally influenced by the interfacial property which determines the strength of the mixture [3]. Furthermore, bitumen-aggregate adhesion is considered the critical factor influencing pavement performance under traffic loading and environmental conditions. When the interface between bitumen and aggregate is destroyed, looseness, peeling, water damage, and other faults emerge, resulting in performance degradation and a shorter service life for bitumen pavement. Previous research has highlighted age hardening and moisture as primary factors influencing the durability of pavement [4]. Age hardening occurs when pavements are exposed to atmospheric oxygen, causing oxidation and the loss of ductility of the bitumen binder. When bitumen oxidizes, its chemical composition changes due to the volatilization of lighter components, causing pavement embrittlement, increased stress concentration under traffic, and premature pavement failure.

Since bitumen alone cannot fully meet the standards of a high-grade roadway, a growing number of modified bitumen materials are being utilized in the construction of new roads [5]. The rationale for bitumen modification is to improve physical, mechanical and rheological properties when compared with virgin bitumen. The physicochemical and rheological properties of bitumen vary depending on its origin. This variation in properties causes them to react differently to different kinds of polymers and other additives. Even though it has been shown that adding different modifiers might lessen bitumen's oxidative ageing, their efficacy is constrained by agglomeration, which results in poor dispersion. To establish a homogenous bitumen-modifier system, significant heat and mechanical energy are necessary to lower bitumen viscosity before adding modifiers. The viscosity of bitumen decreases with an increase in temperature. To cause bitumen flow requiring an activation energy between 50 and 120 kJ/mol. To attain a viscosity in the range between 1 and 10 Pa s requires heating the bitumen a temperature in the range of 120 and 190 °C [6]. Also, to achieve a fairly uniform dispersion of polymer in the bitumen matrix during the synthesis of polymer-modified bitumen (PMB), high temperatures and high shear rates are thus necessary, particularly when the polymer is crystalline and has a high melting point. This method uses a lot of energy, aggravate bitumen ageing, reduces service performance and harms the environment.

Furthermore, the binding characteristics of bitumen have been related directly to its composition [7]. Bitumen has a complex structure that includes hydrocarbons, aromatics, resins, asphaltenes, alkanes, maltenes, and other components linked together by carbon groups. Regarding chemical composition, ageing causes an initial decrease in aromatics followed by a rise in resin content along with asphaltene. Aromatics produce resins, which in turn produce asphaltene. All of these changes result in a higher glass transition temperature that influence the transition from viscoelastic solid to viscoelastic fluid. In addition to asphaltene, resins, and oils, bitumen may also contain small amounts of other metals and polyaromatic semiconductors [8]. The complexity of bitumen constituents usually influence the treatment procedures required before use as a binder [9].

Therefore, the primary goals of this review were to (i) know the state of research regarding the modification of Nigerian natural bitumen that was extracted from tar sands and oil wells and (ii) collect relevant data from pertinent open literature to aid the process of selecting appropriate, effective, and affordable polymer and other additives for upgrading Nigerian bitumen resources for the

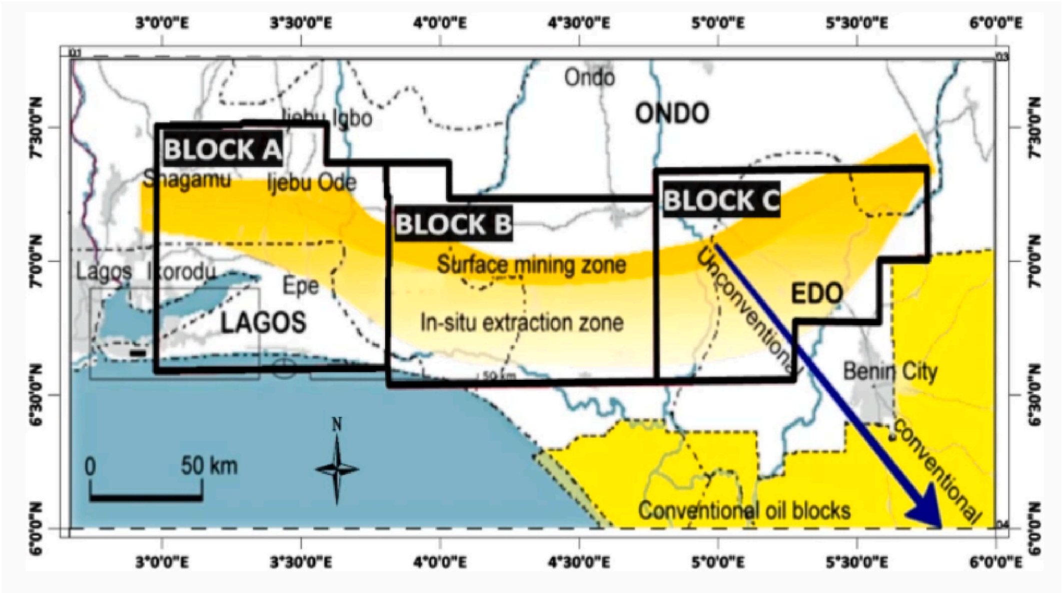


Fig. 1. A map of Nigeria's bitumen belt with highlighted bitumen resource blocks [15].

development of better road infrastructure. This review, therefore, is structures to outline: Nigeria's bitumen reserves, their physico-chemical and rheological characteristics, the earlier studies on the modification of Nigerian natural bitumen concerning pavement construction, factors influencing bitumen properties generally—particularly moisture and ageing—as well as their mechanism and ageing simulation, the effects of reactive and anti-ageing chemical additives, polymer additives and their effects on modified bitumen, and finally, additives for sustainable pavements.

2. Nigerian bitumen and the rest of the world

Previous research has found that the two largest deposits of natural bitumen in the world are located in Canada and Venezuela [10]. The estimated reserve of over 2.2 trillion barrels was confirmed in the Alberta and Saskatchewan provinces of Canada. The Orinoco oil sand deposit in Venezuela is one of the oldest heavy oil deposits (50–60 million years old) with an estimated reserve of about 190 billion metric cubes of oil [11,12]. In Africa, bitumen can be found in Nigeria, Egypt and Algeria [13]. The bitumen deposit in Agbabu, Ondo state, Nigeria has been reported to be the largest with over 16 billion reserves [14]. The sedimentary basin of Nigeria, specifically the Dahomey basin, is where bitumen is typically and primarily found, with Ondo State being the most noted area of bitumen activities in the belt [13]. Other bitumen deposits in the Southern part of Nigeria were located along the Ogun-Lagos axis, as well as at Edo State. Fig. 1 depicts a map of the Nigerian bitumen belt with bitumen resource blocks highlighted [15]. There is a sizable wedge of Cretaceous to recent sediments in the eastern Dahomey basin of the Nigerian sector, which thickens up to 3000 m offshore. Due to the presence of bitumen, limestone, glass sands, and phosphates, the basin has attracted a lot of geological interest [16]. Fig. 2 depicts an overall geological map of the Eastern Dahomey Basin that shows the location and size of the tar sand deposits. Bitumen sands (tar sand) were discovered in a 120 km by 6 km belt in Southwestern Nigeria, stretching from the Okitipupa ridge/western feather edge of the Tertiary Niger Delta to Ijebu-Ode in Ogun State [16]. From topsoil down and from location to location, several distinct bitumen-impregnated hydrocarbon types have been noted within the Nigerian bitumen belt. Outcrop, rich sands, lean sands, shales, and heavy crudes have been observed [13]. The X and Y horizons of two tar sand units, which are located in the Dahomey basin of Nigeria depobelt, are the

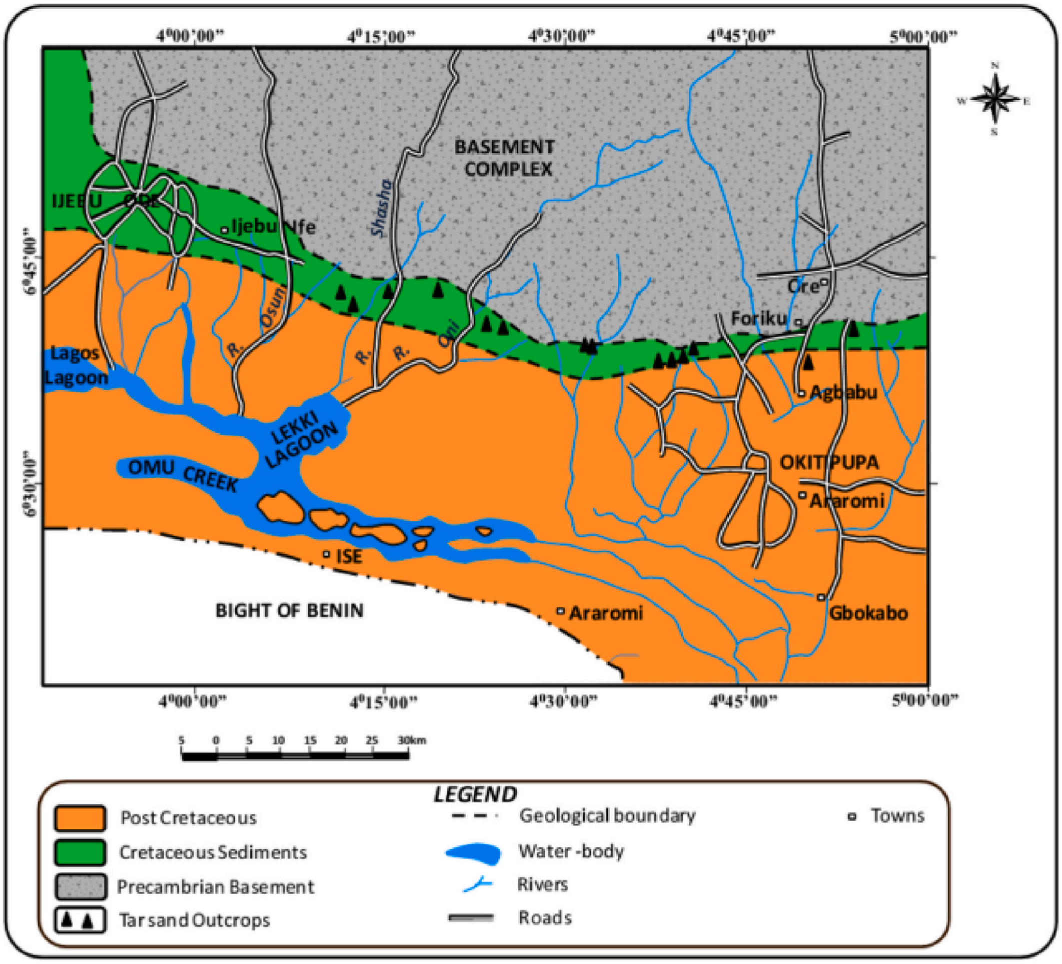


Fig. 2. The area of the tar sand deposits on a geological map of the Eastern Dahomey Basin [16].

source of bitumen seeps, according to earlier authors [17,18]. The observed bitumen sands and tar sands along the coastal region of the same basin flank are thought to be the result of insufficient cap rocks for the bitumen source rock, which allowed it to migrate to the surface as outcrops [16]. In total, a proven bitumen reserve of about 42.5 billion tones has been confirmed in Nigeria which has not been explored for commercial purposes.

3. Review procedure and data sources

This review followed the Preferred Reporting Items for Systematic Reviews and Meta-Analyses (PRISMA) statement 2009 [19]. The review was put together after accessing open-access literature, including but not limited to articles about Nigerian bitumen upgrading for pavement construction. Database article querying was guided using definite keywords in Tables 1 and 2 [20]. The keywords are used as search terms to find articles that cover topics such as natural bitumen, asphalt binder, binder properties, factors affecting pavement binders, types of polymers, modification of natural bitumen and asphalt, a major problem with bitumen modification, previous techniques that have been used to address it, and other alternative additives for long-lasting asphalt pavement construction that mainly spans the last five years. Table 3 displays previous work on Nigerian natural bitumen upgrading for pavement construction obtained via a Google Scholar search link. The improvements recorded on the physical and rheological properties after modification using various additives such as poly-phosphoric acid, EVA, poly-propylene, crumb rubber, sulfur, silver nanoparticles (AgNPs) and Multi-Wall Carbon Nanotubes (MWCNTs) are tabulated as shown in Table 4. Out of the 57 journal publications collected regarding modification of Nigerian bitumen for pavement construction, only 25 (about 44 %) examined bitumen samples that originated in Nigeria. Since the sources of the other publications ($n = 32$) analyzed bitumen samples obtained directly from different construction companies could not be confirmed, they were disregarded. It was also found that Agbabu bitumen was the focus of more than 80 % of earlier research on bitumen modification in Nigeria. Thus, the upgrading of Nigerian natural bitumen has not received enough attention in research articles. Therefore, 165 articles, most of which date back six years, were gathered and examined to provide up-to-date information on bitumen modification that could help improve Nigerian bitumen for pavement construction.

4. Properties of Nigerian bitumen

Like other bitumen from different countries, Nigerian bitumen mixtures contain different types of hydrocarbons such as saturates, aromatics, resins, and asphaltenes, which have different physical and chemical properties. The determination of these fractions is essential to understanding their behaviour and selecting appropriate modifiers for their upgrading. Most researchers and multinational companies have focused on the characterization and geological investigation of Agbabu oil sand and/or natural bitumen [33,34]. Table 5 presents the proximate, chemical and elemental composition of Agbabu natural bitumen extracted from well and tar sand. There were variations in composition and trace metals which are attributed to the method of sample preparation especially the dehydration of the raw bitumen before characterization [35]. Because of a lack of information on bitumen characterization from other locations, samples were extracted from tar sands from the Abigi, Loda and Sunbare deposits and characterized at the SARA level. Table 6 presents the composition of the saturates, aromatics, resins, and asphaltenes fractions while Table 7 compares the maltene and asphaltene obtained on Agbabu bitumen by the previous authors. The results of the SARA fractions can provide some insight into their potential for road pavement applications. Higher percentages of saturates and lower percentages of asphaltenes in the Agbabu samples typically indicate better road pavement potential owing to potential lowest viscosity and better workability. Samples obtained from tar sand are identified with high asphaltene. Although the Agbabu sample has the highest resin content, an indication of better adhesion properties, the resin quantity recorded from the other samples can also provide adhesion to aggregates as there is no significant statistical difference between them and the mean. Therefore, through modification, the percentage resins recorded for these samples can be improved which is essential for ensuring the long-term durability of the road pavement.

Table 1
Popular polymer for bitumen modification [20].

Category	Examples	Advantages	Disadvantages
Plastomers	• Polyethylene • Polypropylene • Ethylene-vinyl acetate (EVA) • Ethylene-butyl acrylate (EBA)	• Excellent high-temperature capabilities • relatively affordable • Good storage stability • high level of rut resistance	• A minimal increase in elasticity • An issue with phase separation • Limited elastic recovery improvement • Slightly low-temperature properties improvement
Thermoplastic elastomers	• Styrene-butadiene-styrene (SBS) • Styrene-isoprene-styrene (SIS) • Styrene-ethylene/butylene-styrene (SEBS)	• Increased rigidity • Reduced sensitivity to temperature • Improved elasticity • High heat, oxidation, and ultraviolet resistance	• Problems with compatibility in some bitumen • Low heat, oxidation, and ultraviolet resistance • Fairly high price • Issues with storage stability • Comparatively lower elasticity • High price

Table 2
Attempted measurement for removing PMB's drawbacks [20].

Attempted measurements	Advantages	Disadvantages
Saturation	• Increased resistance to heat, oxidation and ultraviolet	• issue with phase separation • High price
Sulfur vulcanization	• An increase in storage stability • Excellent high-temperature capabilities	• Applicable only to unsaturated polymer modifiers, such as SBS • Extremely sensitive to dynamic shear and oxidative ageing • Release of hydrogen sulfide • Weak recyclables
Antioxidant	• less oxidation	• High price
Hydrophobic clay minerals	• An increase in storage stability • Good resistance to rutting • Increased resistance to ageing	• Limited improvement in ductility, elastic recovery, and low-temperature properties • Difficult to exfoliate
Functionalization	• Greater compatibility • Other functions are not tried.	• Unpredictability in some situations • High price
Reactive polymers	• Improved compatibility • Improved high-temperature characteristics	• Low-temperature properties have barely improved. • Gelation issues

5. Viscoelastic properties of bitumen

The fundamental purpose of bitumen in pavement construction is to bind the aggregate together. The ability of bitumen to perform this function has been attributed to its viscoelastic properties [38–40]. As the name implies, a material is viscoelastic if it exhibits both viscous and elastic characteristics when undergoing deformation. The knowledge of bitumen viscoelastic behaviour is crucial for obtaining a good design of bitumen-aggregate mixtures that guarantees durability and extended service life of asphalt pavements. Bitumen properties such as stiffness and rheology vary even with a slight change in temperature and/or loading condition [41]. Consequently, the viscoelasticity of bitumen binder is often characterized by low, intermediate and high-temperature [38,42]. To determine the rheology of bitumen, complex modulus (G*) and phase angle (δ) are frequently measured. The complex modulus which depends on the angular frequency (ω) symbolizes the viscoelastic response of bitumen under loading at a given strain level [43]. At a given rate of loading and high temperatures, it is expected that bitumen exhibits a viscous behaviour, whereas when the temperature is low, the response behaviour changes to elastic. The lag distance between the elastic and the viscous responses represents the phase angle (δ). A high value of phase angle indicates the propensity of bitumen towards a viscous behaviour which leads to an increase in ductility, while a low value of phase angle indicates an elastic response with an increase in the bitumen's stiffness and brittleness. The elastic component is otherwise referred to as the storage modulus (G') while the viscous component is the loss modulus (G''). The two components are directly related to G* and to each other through the δ. The ratio of the storage and loss modulus is called the loss tangent (tanδ). Equations (1)–(4) are used to calculate the viscoelastic properties. The consistency of these properties at different temperatures is expressed by the penetration index [44] as expressed in Equation (5) which indicates the variation in the values of penetration point between the value obtained at 25 °C and that obtained at the softening point.

$$G' = G^*(\omega) cos\ \delta \tag{1}$$

$$G'' = G^*(\omega) sin\ \delta \tag{2}$$

$$tan\ \delta = G''(\omega) / G'(\omega) \tag{3}$$

$$G^* = \sqrt{[G'(\omega)]^2 + [G''(\omega)]^2} \tag{4}$$

$$PI = \frac{20(t_{R\&B}) + 500(I_g P) - 1952}{t_{R\&B} - 50(I_g P) + 120} \tag{5}$$

where $t_{R\&B}$ is the softening point (°C), $I_g P$ is the logarithm of the penetration at 25 °C (in 0.1 mm).

The approved limit of physical properties of a virgin bitumen binder, the summary of the standard ASTM test methods, viscosity (kinematic and rotational), and loss on heating are presented as shown in Table 8.

6. Factors affecting bitumen properties

Environmental factors, type, composition, and origin of bitumen [46,47], aggregate type and size, fillers [48], manufacturing process and temperature at which the mix is heated [7], used chemicals [49], are some of the factors that can affect durability and overall performance of bitumen pavements. The effects of these factors have been investigated on bitumen binder and its mixture with aggregates. Investigations on this subject can be grouped into (i). those that primarily focused on asphaltic pavements [38,50–52], (ii). Those that focus on the mixes of bitumen and aggregate [53,54] and (iii). Those that concentrate on the binder in natural bitumen [55,56]. Interestingly, the findings from these researchers have attributed the various damages on pavements to the harm done to the

Table 3
Previous work on Nigerian natural bitumen upgrading regarding pavement construction (n = 12).

	Location & Study type	Objectives	Findings
[21]	AGBABU SARA analysis Column chromatography	Effects of UV radiation	Reduction of aliphatic and polycyclic aromatic hydrocarbon profiles
[22]	AGBABU Polymeric modification	High-Density Polyethylene (HDPE), Polyethylene-co-vinyl Acetate (PEVA) and Polystyrene-co-butadiene (PSCB)	6 % of PEVA and HDPE polymers caused a reduction in penetration value PSCB increased the penetration value
[23]	AGBABU Upgrading of bitumen for road construction	Effects of Sulfur and EVA (ethyl Vinyl Acetate)	14 % EVA, and 5 % Sulfur improved the quality of binders
[24]	AGBABU Comparative study of Fourier Transform Infrared & conventional test	Effect of polyphosphoric acid (PPA)	PPA up to 6 % enhances physical and flow properties
[25]	AGBABU Viscosity prediction models	Investigate viscosity-pressure and temperature relationship and rheological properties of its emulsions	Bitumen and emulsions exhibited shear-thinning behaviour Viscosity sensitivity to temperature decreased with water increment for W/O emulsions Sensitivity increased with increasing water fraction for O/W emulsions.
[26]	AGBABU Study the physicochemical and rheological properties as they are affected by ageing	Poly(styrene-co-butadiene), Poly(ethylene-co-vinyl-acetate), and High-Density Polyethylene	Improved resistance to fatigue cracking and rutting (PEVA at 2 and 6 wt%) Reduction of rate of thermal ageing
[27]	IMERI BITUMEN Upgrading of Tar sand bitumen by polymeric modification	Crumb rubber-modified Tar sand bitumen	Reduction of penetration value Increase of Specific gravity, flash and fire points, viscosity, and softening point Improvement of rutting resistance and reduction of fatigue/reflection cracking
[27]	IJEBU-IMUSHIN BITUMEN Upgrading of Tar sand bitumen by polymeric modification	Effects of Waste Water Sachets (WWS) and shredded Waste PET bottles (SWPB)	It is possible to convert natural bitumen to 60/70 bitumen (1.5 % SWPB and 0.1 % WWS) Softening and flash points are satisfactory at 1.5 % SWPB and 0.1 % WWS addition.
[28]	AGBABU & LODA BITUMEN High and low temperatures susceptibility study	The purpose is to investigate the effects of polyethylene (PE) on mechanical and rheological properties.	Physico-mechanical properties increased with increase in modifiers (10–40 %). Agbabu performed better than Loda, according to reports. For both low and high temperatures, a sheared polyethylene-modified bitumen of 40 per cent is ideal.
[29]	AGBABU Oxidation of bitumen via FTIR analysis	Examining new ways to use silver nanoparticles (AgNPs) as possible anti-oxidative additives	The mechanism of action of silver nanoparticles is the removal of free radicals generated during the oxidation process Reduction of penetration and specific gravity Increase in flash and fire points, kinematic viscosity, penetration index, and softening point temperature
[30]	AGBABU Upgrade Nigerian Bitumen	Effects of powdered rubber tyres, potash and carboxyl methyl cellulose	30 % modifier mixture is optimum for upgrading 200 g of bitumen to international standards.
[31]	AGBABU Ternary modification and optimization study	Effects of Multiwall Carbon Nanotubes (MWCNT) and polypropylene (PP)	1.1 and 1.98 % of MWCNT, and PP caused a large improvement in composite properties The blend is conformable to ASTM and BS standard requirements.

binder's intrinsic properties when exposed to harsh conditions or subjected to large stresses causing damage to the adhesion force. Direct correlation between the durability of the bonds holding the bitumen and aggregates together was established. The correlation between the ultimate performance of pavements under various environmental conditions have also been linked directly to the quality of binder that glued the aggregates together in the mix.

A loss of adhesion force in the bitumen-aggregate mixes resulted in brittle mixes and poor fatigue resistance [52,57]. The grade of bitumen used in pavement also has an impact on its durability. A low-grade bitumen is therefore often upgraded to a higher grade by blending with additives. In the course of doing this, the bitumen will be subjected to high temperature under shearing which changes its properties and ultimately the durability of the binder. For instance, a recent study that investigated the effects of various parameters on the bonding qualities of various bitumen-aggregate mixtures revealed a lower penetration grade bitumen sample exhibited superior bonding quality with aggregates when used in both dry and wet conditions [39]. This perhaps may be a result of the various pre-treatments while upgrading is going on. Also, it was revealed that bitumen alone, as opposed to the type and sizes of aggregate used in the design, had a significant impact on the bonding. The weakening of bitumen properties impacts negatively the bonding capabilities, which decreases the service life of the bituminous pavement. This has been manifested in several types of damages, including ravelling, cracking, and potholes, which are frequently seen on bituminous pavements. Therefore, we can say that the bitumen binding property

Table 4

Some experimental results show the effects of various modifications on the physical and rheological properties of Agbabu bitumen and Oil extracted from tar sand.

	Pen. Point 25 °C (dmm)	Soft. point (°C)	Specific gravity	Flash point (°C)	Fire point (°C)	Kinematic viscosity (cSt)	Penetration index	References
Agbabu Natural Bitumen (ANB)	47–82	47–79	1.047	156–265	159–275	350	−0.776	[22,31]
Bitumen from Tar Sand	45–62.5	50.6–58	0.57–1.03	165	245	22.5	–	[27,32]
ANB+6%PPA	55	60	1.165	165	175	1005	1.271	[24]
ANB+6%PEVA	51	66	1.039	235	250	1460	2.204	[24]
ANB+4.5 wt%AgNPs	75	52	1.046	280	290	395	0.024	[29]
ANB+5%S	18.33	–	–	300				[23]
ANB+14%EVA	18.37			303				[23]
ANB+6%PP	10.5	90		181	224			[31]
ANB+1.98%PP+0.1% MWCNT	10.2	97		224	229			[31]
BTS+15 % Crumb Rubber	45	75.8		250	280	18		[28]
BTS+0.1 % WWS	72.8	50		230		18.2		[27]
BTS+1.5%SWBP	59.7	50		230		15.8		[27]

Table 5

Composition of Agbabu bitumen and oil sand.

Composition of Agbabu bitumen and oil sand	Value	
Bitumen content (%)	83.42	
Moisture content (%)	15.14	
Mineral matter	1.44	
Asphaltene (%)	24.83	
Maltene (%)	21.67–26.8	
Specific gravity	0.6–1.063	
Molecular weight	748	
Elemental composition (%)		
C	81.54–86.62	
H	10.98–12.2	
S	0.92	
N	0.48–1.13	
O	0.83	
Ash	0.17–0.84	
Metal composition		
Mn	28.40	12.41
Fe	3633.10	204.93
Zn	17.50	100.94
Pb	93.78	57.49
Ni	103.65	20.22
V	156.53	7.59

Source: [26,36]; *Metal composition* [22,37]; *Elemental composition:* [21,35].

Table 6

SARA composition of Nigerian bitumen (this study).

Sample Location	Status	Saturate (%) (A)	Aromatics (%) (B)	Resins (%) (C)	Asphaltene (%)
Abigi	Tar sand	14.15	16.72	30.22	35.7
Agbabu	Viscous Oil	17.61	19.14	35.98	23.45
Sunbare	Tar sand	13.1	16.4	33.41	34.5
Loda	Tar sand	16.66	18.82	34.75	27.56

Table 7

Comparison of Maltene and Asphaltene in Agbabu as reported by some authors.

Sample Location	This study	[35]	[37]	[36]
Maltene (%)	76.55	73.05	78.34	74.73
Asphaltene (%)	23.45	26.95	21.66	25.27

Table 8
Physical and rheological properties of virgin bitumen.

Specification	Unit	Test method	Specification limit
Density at 25 °C	g/cm^3	ASTM D 70	–
Penetration degree (100 g, 25 °C, 5 s)	1/10 mm	ASTM D 5	60–70
Softening point	°C	ASTM D 36	49–56
Ductility (25 °C, 5 cm/min)	cm	ASTM D 113	MIN 100
Flash point	°C	ASTM D 92	MIN 232
Solubility	%	ASTM D 2042-76	MIN 99
Penetration index	–	[44,45]	-3-7
Room temperature drop	%	ASTM D 1754	–
Kinematic viscosity (135 °C)	c.St	ASTM 2170	–
Loss on heating	%	ASTM D 6	Max 0.8

has an impact on the pavement's overall performance. This is why serious attention of road engineers has been on modification of bitumen with the view to improving its binding properties.

6.1. Effect of moisture on bitumen properties

When designing and building asphalt pavements, one of the top concerns for pavement experts and other relevant transportation agencies is moisture damage. The separation of the asphalt film from the aggregate surface or a loss of cohesion within the asphalt binder, which lessens the stiffness of the asphalt mix, are the most typical symptoms of moisture damage [58–60]. Damages caused by moisture promote rutting, cracking, and ravelling between pavement layers. Additionally, it hastens the development of potholes, which lowers the effectiveness and serviceability of asphalt pavements [61]. There are two major factors responsible for moisture damage: (i) a reduction in the bond strength between the aggregates and the binder or mastic, including fine aggregates and (ii) a decrease in mastic cohesion brought on by the presence of moisture [61]. In other words, the damage phenomenon can manifest itself in many different ways. It may occur as a result of a breakdown in the cohesive bond within the binder itself, a breakdown in the adhesion bond between the asphalt binder and the aggregate, or a combination of both cohesive and adhesive failures.

Moisture damage is a complex phenomenon. Two aspects have been put forward for understanding the dynamics of moisture in asphalt pavements, the chemical and mechanical characteristics. These characteristics are identified with adhesive failure of the binder or its mixture with aggregates and are summarized as follows:

(i). separation of an asphalt film from an aggregate surface because of the presence of a thin layer of water in the asphalt film. This process is termed detachment. This process resulted in a loss of adhesion causing the peeling off of asphalt films away from the aggregates [62]. Water can reduce the free surface energy at the asphalt-aggregate interface.
(ii). breakage of asphalt film caused by penetration of water to the partially coated aggregates with asphalt film due to the presence of water or rupture of binder films around the aggregates [63]. This process is termed displacement.
(iii). formation of inverted emulsion owing to the combination of water and asphalt which resulted in stripping. The process is termed spontaneous emulsification. The process is facilitated by the presence of emulsifiers such as asphaltene, clay fines or certain additives used to modify the asphalt [64].
(iv). the development of pore pressure in the encapsulated water in the asphalt mix causing the stripping of the asphalt film from the aggregate [63].
(v). reduction of chemical bonds between asphalt and aggregate caused by the shifts in pH of the moisture contacted [65,66].

6.2. Ageing and its effects on bituminous mixture

Binders are said to be durable if they resist deterioration or changes in physical characteristics over time [2]. Vehicle loading, endless exposure to climatic and environmental variables, and other factors make the ageing of bitumen mixtures inevitable. The primary factor affecting the longevity of asphalt pavement has been identified as the ageing behaviour of asphalt in use [67]. Depending on the prevalent activities and related processes, ageing can occur in both the short- and long-term [68]. Short-term ageing is accompanied by the oxidation and vaporization of light components. When the asphalt mixture is produced, mixed, transported, laid, and compacted, there is a chance that it will age quickly in the short term. However, bitumen pavement experiences long-term ageing as a result of environmental factors, which results in significant deformation or deterioration of mixing properties. Both short-term and long-term ageing would harm the durability of bitumen pavements [63,65,69,70].

Road researchers throughout the world have focused their efforts, and industry has invested large resources and time in studying various elements and their interactions as they influence bitumen ageing [41]. Due to bitumen exposure to harsh environmental elements such as precipitation, temperature, air, and ultraviolet (UV) radiation, studies have shown that bitumen ageing has a significant impact on pavement durability [2,5]. For example, at a higher temperature, bitumen softens and loses rigidity, making it more susceptible to rutting. However, at lower temperatures, bitumen's stiffness would rise but its flexibility would decrease, leading to fatigue failure in numerous application areas [71]. The growth of the hardness of asphalt in pavements over time was noted in research conducted four decades ago on asphalt recovered from pavements [72]. Wherein the cracking of pavement and observation of other problems on the sample pavement were attributed to the hardening of asphalt. The hardening of bitumen can occur in two different ways: (i). reversible/physical hardening and (ii). the irreversible type. The former is brought on by the rearrangement of asphalt binder molecules and the process is reversible by raising the temperature of the bitumen mixtures. The irreversible kind, on the other hand, is brought on by chemical oxidation, polymerization, and the evaporation of light components from bitumen or the exudation of oil components from the asphalt binder into a mineral aggregate [69]. The effects of heat, moisture, oxygen, and ultraviolet (UV) radiation on bitumen hardening were studied in the 20th century and have since received a lot of attention [73–75]. There are numerous references to thermal ageing [56,69,76], however, there has been relatively little study effort devoted to understanding ultraviolet (UV) ageing.

6.3. Bitumen ageing mechanism

Bitumen, being an organic substance is highly susceptible to ageing which has significantly limited its applications. The exposure of bitumen pavements to ultraviolet (UV) radiation, air, and temperature during their service life decreased the performance of the pavement [77,78]. Bitumen can undergo three varieties of ageing including; (i). oxidative thermal ageing, (ii). ageing due to photooxidation, and (iii). solution ageing [76]. Regardless of the type of ageing, ageing has a destructive impact on the rheological properties of bitumen by causing an increase in brittleness, triggering cracking and reducing the service life of asphalt pavement [79,80]. The loss of ductility of the asphalt binder, embrittlement of the pavement, and stress concentration under traffic are all effects of bitumen exposure to atmospheric oxygen, which ultimately causes premature pavement failure. Ageing by oxidation results in the hardening of the asphalt.

6.3.1. Thermal and solution oxidation ageing

Thermal-oxidative ageing results from thermal oxidation causing the asphalt binder to lose a light component, especially at high temperatures [76]. Several investigators have simulated thermal-oxidative ageing in the laboratory by using a rolling thin film oven test following the ASTM D2872-04 [67,81]. These investigations have revealed that the ageing mechanism of the asphalt binder is not yet fully understood. The loss in weight of bitumen as a result of the escape of light components would depend largely on the origin, type and the bitumen's composition [82]. Concerning composition, asphalt contains hydrocarbons, non-hydrocarbons and certain heteroatoms each having unique physicochemical properties and displaying a variety of decomposition temperatures during heating [83]. Utilizing the isothermal thermogravimetry/differential scanning calorimetry-Fourier transform infrared spectroscopy (TG/DSC-FTIR) technique, the thermal-oxidative ageing of asphalt binder was examined at the SARA level [76]. The study revealed that the thermal stability, the composition of the volatile components, the amount of SARA fraction released, the chemical compositions and the morphology of aged SARA fractions change during thermal-oxidative reactions. Two chemical reactions were identified the endothermic and exothermic reactions. Endothermic reactions took place in both the saturates and aromatics, and both endothermic and exothermic reactions are equal in resins, and exothermic reactions occur in asphaltenes. There was an increase in the mass of aromatics and asphaltenes but saturate and resin mass losses were measured. Thus, the saturates and resins were most affected by oxidation. This sheds light on how asphalt binder materials age thermally and oxidatively at the SARA fraction level. The authors did clarify, though, that the characterization techniques could not identify the volatile components as the asphalt binder aged.

Currently, there are no established standards for the solution ageing of bitumen, however, several road researchers believe immersion of bitumen in solution such as salt can cause ageing in bitumen. Solution immersion has however reported causing an increase in the asphaltene content, produce oxygen-containing functional groups, increase complex modulus and reduce the phase angle, which are typical characteristics of the oxidation of bitumen [1]. Solution immersion of bitumen was also reported to have degraded the adhesion and cohesion of bitumen [52,84]. These changes indicate that the solution immersion would also age bitumen like thermal-oxidative ageing but with different mechanisms. Ref. [56] investigated and compared both thermal-oxidative ageing and salt solution ageing using four ageing methods. The result revealed the same varying trends under salt solution ageing and thermal-oxidative ageing in terms of oxygen element, physical properties, and low and high-temperature properties, resulting in the ageing of bitumen. However, salt solution ageing results in sharp angles of bitumen pieces, while thermal-oxidative ageing induces the rough surface of bitumen with certain cracks. The study is however limited to neat bitumen and the mechanism of salt solution ageing on bitumen was recommended for further investigation.

6.3.2. Simulation of an ageing system

Bitumen ageing and moisture are now recognized as significant elements influencing the durability of bitumen pavements [57]. Because of this, a proper bitumen mixture design must consider these impacts as well as those of other environmental elements, such as temperature, air quality, and sun radiation, and how they combine to affect the longevity of bituminous pavements. Numerous studies on the ageing of asphalt and bitumen mixtures often take into account the laboratory and simulation ageing techniques. There are two types of laboratory bitumen mixture ageing: short-term oven ageing (STOA) and long-term oven ageing (LTOA). According to the American Association of State Highway and Transportation Officials (AASHTO), asphalt mixtures must be cured for a few hours and days for short-term and long-term ageing, respectively. Rolling thin-film oven test (RTFOT) and pressure ageing vessel (PAV) are the two standardized procedures for carrying out ageing in the laboratory [69]. These processes use high pressure and/or temperature to significantly speed up oxidation and thus shorten the time needed to age the asphalt binder to a level that is comparable to that found in the field [40]. Alternatively, t two thin-film ageing test procedures have been applied, the nitrogen atmosphere oven ageing test (NAAT) which is based on the principle that the inert gas minimizes oxidative ageing and the other, the ambient atmosphere oven ageing test (OAAT) [69]. These methods were employed to assess the effect of thermal and oxidative ageing on virgin asphalt binder properties. In these alternative methods, the virgin bitumen was heated to 163 °C under an inert gas for 4 h.

During short-term ageing, the loose bitumen mixture is aged for 4 h in an oven operated at 135 °C [73]. For long-term ageing, the bitumen mixture is compacted and placed in a convection oven for 5 days at 85 °C [68]. A more realistic technique is simulation, which simulates ageing by mimicking environmental elements such as temperature, UV radiation, and moisture [67]. However, previous studies [85] have shown that creating a realistic model that simulates field circumstances is difficult and imprecise. Typically, an artificial ageing system is developed by simulating a few selected elements, characterizing and evaluating their impact on the ageing of an asphalt mixture based on changes in the mechanical properties of the mixture. The Tecnico Accelerated Ageing (TEAGE) techniques that involved ultraviolet (UV) plus rain-fall simulation were explored in comparative research with the LTOA [57]. The two techniques had similar but not identical effects on the behaviour of bitumen samples. The TEAGE approach, on the other hand, was shown to better replicate the climate impacts on bitumen mixtures. There have also been reports of experiments that incorporated the simulation of volatilization and oxidation as important factors influencing the ageing process [57,86]. The two processes are irreversible and involve chemical reactions that are affected by bitumen grade, and reactive hydrocarbons in bitumen. The main issue with both simulation and laboratory procedures is the difficulty in validating them using field data [73]. This is extremely challenging due to the wide range of variables such as road location, sun exposure, weather, mixture type, and void content. For example, laboratory simulations of ageing for binders are typically performed at temperatures and pressures that differ from the real field conditions under which mixes are aged.

6.3.3. *Effects of anti-ageing chemicals on binders*

The determination of moisture and oxygen content of asphalt binder is crucial for pavement application. Apart from atmospheric oxygen, bituminous mixtures also contain a certain fraction of oxygen that could serve as an oxygen source for oxidative reactions resulting in hardening and embrittlement. The ravelling and cracking of asphalt pavements are fundamentally caused by moisture transport and oxidative ageing [87]. According to Wang et al. [88], bituminous materials with lower oxygen and moisture coefficient of diffusion are capable of reducing the damage rate of pavements and prolonging their service life. Though the determination of the moisture diffusion coefficient is still ongoing research, several studies have been carried out concerning the chemical, physicochemical and mechanical characterization of bitumen [89,90]. To more accurately describe moisture diffusion in bitumen, more work must still be done. To this end, researchers have utilized varieties of antioxidant and ultraviolet absorbers as anti-ageing modifiers in bitumen. Additives such as phosphoric and polyphosphoric acid [91], Zinc dialkyl dithiophosphate additive [92], Microwave activated crumb rubber [5], Intercalated sodium dodecyl sulfate (SDS) and sodium dodecyl sulfonate-containing organic layered double hydroxides (LDHs) [93], nano-ZnO with surface modifications and organically expanded vermiculite [94], diethylene glycol-based polyboron compound [95] etc. had shown better-rutting resistance and improved short and long term ageing performance when used in bitumen. Several other polymers and chemical additives have also been incorporated into bitumen and asphalt binders to improve rheology and mechanical properties. Attempts to modify bitumen with polymers for road construction began after the 1973 and 1979 oil crises because natural bitumen as a binder in the road industry has failed to fully meet the necessities of high-grade highways due to the ageing of bituminous binders which has contributed significantly to the various deteriorations associated with pavements [96,97]. Today, many engineers and managers of the pavement industry are being confronted with serious challenges of poor performance and the short life of asphalt pavements.

However, polymeric modification of asphalt binders has proven to be an effective way to increase pavement ageing resistance, mechanical characteristics and lifespan of binders [98]. Recent studies have indicated that the addition of polymers can improve the rheological, thermal and mechanical properties of modified asphalt binder when compared with the control bitumen samples [99–101].

6.3.4. *Effects of reactive additives on binder properties*

The use of reactive polymer additives to mitigate poor polymer dispersion in binders has also been proposed [102]**.** Reactive polymers have functional groups that can interact with particular bitumen molecules to form various linkages, including anhydrides, isocyanates, and epoxides. Porto et al. [103] attributed the enhancement of properties of bitumen caused by polyolefin to the covalent linkages that were created which subsequently increased polymer compatibility in bitumen. The isocyanate-based polymers have shown a tremendous enhancement of bitumen properties owing to high reactivity. Isocyanate would react with hydrogen-atoms-containing functional groups to form urethane, urea and amide linkages. However, adequate knowledge of types and the correct selection of reactive polymers to use in binder composite production is very critical. Several studies [104,105] have reported the use of MDI (4,4′-diphenylmethane di-isocyanate). MDI significantly improves the properties of binders. The reactivity between the -NCO groups of the oligomer and the most polar groups of asphaltenes and resins in bitumen (primarily -OH and -NH, but also -SH and -COOH) has been credited with the improved properties [105]. The stiffness and storage stability can be increased by the addition of reactive polymers with groups suitable to react with particular bitumen molecules. Between 2 and 2.5 % by weight of binder of a reactive polymer such as ethylene terpolymers functionalized with glycidyl methacrylate (GMA) has been recommended [106]. The good performance obtained from using these additives was attributed to their ability to crosslink and/or chemically bond with the molecules of asphaltenes. However, reactive polymers are highly expensive compared to the other types of polymers. Also, the high number of reactive groups on a single macromolecule often leads to gelation problems.

6.3.5. *Adhesion promoters of bituminous mixture*

The quality of bitumen and bitumen to aggregate adhesion are critical properties of asphalt mixtures that are intimately linked to their durability and resistance to moisture and ageing impacts. It is rampant during construction the use of locally available aggregates to cut construction expenses. However, these aggregates are typically hydrophilic, whereas bitumen binder is hydrophobic, thus the aggregates cannot enable proper adherence to the bitumen binder. Previous studies have indicated that acidic aggregates (granite) would show a greater loss of adhesion than basic aggregates (limestone and marble) under wet conditions [107,108]. The porosity of the aggregates was also found to be significant in this regard, but not as critical as their chemical makeup. To boost the interfacial adhesion in wet conditions, silane, amine, or rubbery polymers may be added to the bitumen. The addition of silane can bridge the interface between the organic bitumen binder and the inorganic mineral aggregate, especially for the silica-rich granites. Amine-based chemicals, anti-stripping agents, are capable of enhancing water resistance especially, for limestone and granite/bitumen mix. In another study, [109] explored a polyamide promoter that is based on fatty acids. It was observed that the adhesive additives positively increase the adhesion of the asphalt binder, even after being supplied to the asphalt mixture in quite a small quantity (0.3 % of the asphalt's binder weight). Therefore, when applied, it should have a positive impact on pavement durability, which will lower life cycle expenses (lower maintenance costs).

Many different adhesion enhancers have been utilized, all of which aim to change the surfaces of aggregates by rendering them hydrophobic through treatment with various chemicals. Bagshaw et al. [110] employed alkyl phosphonate or alkyl phosphate surface modifiers. Following this treatment, the interfacial free energy of the aggregates surface/bitumen interface becomes favoured over that of the aggregates surface/water interface which led to a very strong aggregate/bitumen adhesion that resisted de-bonding by water. Goel and Sachdeva [111] employed hydrated lime and organosilane-based adhesion promoters to increase the resistance of bituminous concrete mixes to moisture damage. The results show that 2 % hydrated lime and 0.05 % organosilane-based adhesion promoter enhances the tensile strength ratio of bituminous concrete mixes which indicates that they are less susceptible to moisture damage. Gunka et al. [112] utilized phenol-cresol-formaldehyde resin produced from a phenolic fraction of coal tar resin and formaldehyde through polycondensation as an adhesion promoter in asphaltic concrete. The efficacy of the chemical was assessed by examining water saturation, coefficient of long-term water-resistance and compressive strength. The result indicates the addition of 1 wt% of the promoter increased the adhesion index, enhanced the bitumen softening point, long-term water resistance, and hermos-stability but reduced the water saturation index, penetration and ductility of bitumen. Valentin et al. [113] compared five different adhesion promoters at multiple scales. The effects on bituminous binders, their interactions with water and aggregates, and the macroscopic characteristics of a particular type of asphalt concrete were evaluated. Phosphate-based and ethylene polyamine-based additives were found to increase the affinity of mineral aggregates but have very limited impacts on the surface free energy of bituminous binders. On the other hand, those that based on silane or vegetable oils were able to improve bituminous binders' adherence, particularly when phonolite aggregates was used.

7. Polymers and polymeric bitumen

Polymers are large molecules containing long and repeated chains or rings of monomers. They vary broadly in properties depending on the types of constituent monomers and the mode of bonding. They can exist naturally or by synthesis. Natural polymers or biopolymers include latex rubber, cellulose, wool, starch, collagen and DNA [114]. Synthetic polymers, on the other hand, are mostly developed via chemical reactions involving various polymerization processes. A recent investigation has shown that not all polymers are suitable for bitumen modification; however, polymer-modified bitumen has been reported to show reduced thermal sensitivity, enhanced resistance to cracking, rutting, fatigue, and moisture [115–117]. Polymeric modification of bitumen has received significant attention and several researchers have studied various polymer modification techniques to address severe rutting and cracking challenges in road pavements [118–120]. The selection of appropriate polymer for bitumen modification has been based on several factors including adhesive and cohesive properties, viscosity, and ductility [121]. Polymers have been classified in various ways because they exhibit different behaviours. However, in the context of asphalt binder modification, they are grouped into elastomer/thermoplastic elastomer and plastomers. This classification was based on the intramolecular forces that hold atoms together within the polymer [64].

7.1. *The plastomers*

The incorporation of plastomers into bitumen has provided the binder with higher strength and modulus properties. However, the elastic recovery of binders will not improve significantly due to weak intermolecular interactions. Plastomers are characterized by a simple structure of carbon atoms connected to two hydrogen atoms. According to Brasileiro et al. [122], they are well-suited for asphalt binders meant for heavily travelled roads and those constructed in areas with warm to hot climates. At low-temperature conditions, plastomers have shown neither detrimental nor significant improvement in the low-temperature performance of the modified bitumen [123]. They are therefore frequently used to cause changes in the phase composition of asphalt and improve engineering properties needed to bear the heavily loaded traffic in slow and rapidly moving traffic conditions. This group of polymers includes ethylene-vinyl-acetate (EVA), polyvinyl chloride (PVC), polypropylene (PP), atactic polypropylene, and polyethene (PE) (EVA) [124].

7.2. *Ethylene vinyl acetate (EVA)*

Prominent among these polymers is Ethylene vinyl acetate (EVA). EVA can be obtained by copolymerization of ethylene and vinyl acetate. The properties of the copolymer vary depending on the percentage of vinyl acetate (VA) which is generally determined through IR, NMR, and TGA [125]. A low content of vinyl acetate yields a copolymer whose behaviour resembles that of Low-density polyethylene (LDPE). Many investigators [126,127] have used various methods to modify bitumen with EVA. The difference in these studies is in mixing temperature, time of blending and rate of shearing the mixture leading to the production of the PMB. In all these applications, the problem of phase separation has been reported owing to bitumen composition, polymer content, characteristics of the polymer and the mixing process. Also, the degree of solvency of polymer and asphaltene for maltene component in the bitumen can affect the stability of the polymer-bitumen system causing poor storage stability. A study [128] has found that the most important factors affecting EVA-modified bitumen are mixing temperature, blending time and shear rate come next to it.

The modification of asphalt by using EVA increases the stiffness which was evident by the decrease in penetration and increases in softening point after adding 2 % EVA to the mix. EVA above 2 % in the mix induces a polymer network and hence, poor asphalt properties. The poor compatibility between EVA and asphalt has been the reason for the poor storage stability of the modified asphalt system [124].

To improve EVA-asphalt compatibility, [129] evaluated the effectiveness of asphalt mixtures made with EVA-modified bitumen and various glass fibre contents. However, 5 % EVA and 0.3 % glass fibre were found to increase the Marshall Stability by about 25 %. The composite additives increased the Marshall Stability more than using either of them separately. This is attributed to the synergistic effect of EVA and fibre additives. Unlike the concentration of polymer and glass fibres, an increase in temperature reduces the resilient modulus of the asphalt mix. Another study revealed that flexible hot mix asphalt made of hot mix asphalt (HMA) modified with 2 % and 4 % EVA could be used in hot, and dry regions, respectively and prevent asphalt cracking [130]. The binder's mechanical property is affected by temperature and EVA composition; it decreases as the temperature increases.

7.3. Polyethylene

Poly Ethylene (PE) is one of the most commonly used plastics produced by the polymerization of ethylene. It is classified based on density into low-density PE (LDPE), high-density PE (HDPE), and linear LDPE (LLDPE). PE was earlier reported with the capability to improve the rigidity and reduce deformation of modified binder under load especially when the concentration of PE was kept in the mix below 1 % by weight of the base bitumen [131]. Subsequent studies had however shown good performance at higher concentrations. For example, Ho et al. [132] reported a 4 % optimum recycled PE modifier. Habib et al. [133] investigated the effect of the mixing process on HDPE and LLDPE-modified bituminous concrete mix properties. The result obtained for LLDPE indicated the

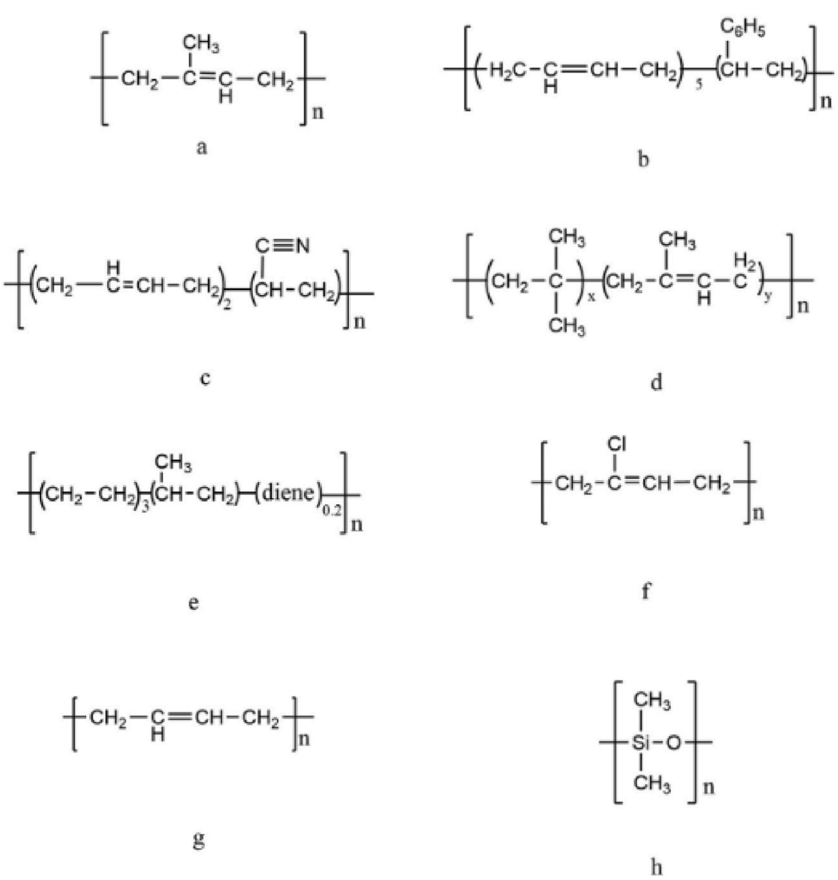


Fig. 3. Chemical structure of (a) Polyisoprene/natural rubber (b) Styrene-butadiene rubber (SBR), (c) Nitrile rubber (NBR), (d) Butyl rubber (IIR), (e) Ethylene-propylene (EPDM) rubber (f) Polychloroprene (CR) rubber, (g) Polybutadiene (BR), (h) Silicone [142,143].

concentration up to 2.5 % yielded a better result in terms of Marshall Stability, resilient modulus, water susceptibility and fatigue life of the modified binder. A more thermodynamically stable structure of LLDPE-modified bitumen was obtained when the concentration of polymer was kept below 4 % which offered resistance in terms of rutting, fatigue and temperature susceptibility [134,135]. The virgin PE used was blended at 1300 rpm for 60 min in a preheated base binder at 160 C. Recycled plastic waste-modified asphalt was investigated in Saudi Arabia and observed that PE between 3 and 5% was optimum [136]. Roman et al. [137] examined the low-density polyethylene (LDPE), high-density polyethylene (HDPE) and polypropylene (PP)-modified bitumen under strain and stress control experiment. From their results, it was observed that PP-modified bitumen showed the lowest fatigue resistance while LDPE, had the highest at 4 wt %. The authors attributed the performance of LDPE to its good dispersion in the bitumen as revealed by the optical microscopy analysis. Desidery & Lanotte [138] investigated the internal structure and performance of PE and PP-modified binders. Both the pellet and powdery forms consisted of 2, 4 and 6 % by weight of bitumen of PP and PE was investigated after 60, 120, 210, and 300 min. Rheological and physical properties improved with concentration irrespective of polymer. PP attained the equilibrium faster than PE and produced extreme stiffness. The powdery forms regardless of polymers accelerated the modification process owing to the surface area and produced different sponge-like polymer networks capable of causing storage issues in the asphalt plant.

7.4. The elastomer modifiers

Elastomers can also be natural or synthetic rubbers that behave elastically when subjected to pressure and tensile stresses. Natural rubber latex is extracted from rubber trees while synthetic rubbers also called "crumb rubber" are produced either by Ref. [1] solution crumb or [2] emulsion crumb methods [114]. Natural rubber has been used to modify bitumen for many years [139] in the form of powder and liquid. Though there is limited research on the use of liquid NR when compared with powder NR, both have shown some beneficial effects on asphalt performance [140]. The NR liquids are said to be significantly more affordable than powder forms and to have many other comparative benefits including accessibility and capability to create viscosity-reducing foams in the modified binder during mixture production [114]. Natural rubber is essentially pure poly-cis-1.4 isoprene and particles vary in size from 0.15 to 3 μm with a chemical formula of C_5H_8 [141]. Fig. 3(a–h) depicts the chemical structure of natural rubber and various types of synthetic rubbers that are readily available. The double bond in the repeated units of isoprene can easily be crosslinked which makes NR to be a stable elastic material. In addition to this, sulfur, peroxide, metal oxides, resins, quinone etc. may be bonded at the ends of its macromolecules to improve the rubber properties. Table 9 displays some typical properties of the NR latex offered by Thai Rubber Latex Corporation.

The characteristics of the latex can be significantly impacted by the presence of non-rubber content. For this reason, the latex is not used directly for bitumen modification, but it is subjected first to either centrifugation or evaporation to achieve highly concentrated solid rubber which prevents bacteria attack [144,145].

Using rubber to modify bitumen is cost-effective and environmentally friendly. Rubbered asphalt binders are characterized by good shear resistance, reduced temperature sensitivity, improved durability, and high resistance to cracks and fatigue [146]. The fundamental idea behind natural rubber-asphalt interaction is that rubber particles can quickly swell when immersed in hot asphalt and absorb components with similar solubility parameters. Additionally, the elastomer's weak cross-links between its chains allow its particles to dissolve more easily in liquid asphalt, allowing the substance to enter the polymer's internal matrix and increase swelling. The other varieties of natural rubber that have been used as a modifier in asphalt binder include skim latex, fresh latex, ribbed smoked sheet (RSS), liquid natural rubber (LNR), and epoxidized natural rubber (ENR) [140].

Recent studies on the use of natural rubber as a modifier in asphalt modification are shown in Table 10. The table contains different base asphalt, the type of rubber used, the percentage latex content investigated, the rubber-binder mixing conditions, and the optimum latex requirement.

7.5. Dispersion of NRL in asphalt binder

Dispersion of natural and synthetic rubber in bitumen to achieve homogeneity remains a serious technical challenge [152]. The poor dispersion has been attributed to the high molecular weight of most elastomers. Another technical challenge is the high

Table 9
The primary characteristics of NR latex [114].

Contents name	Unit	Result
Total solids content	%	≥61.50
Dry rubber content	%	≥60.00
Non-rubber content	%	≤2.00
Ammonia content (on total weight)	%	0.65–0.75
pH value	–	≤11.00
KOH	–	0.85
Volatile fatty acid number	–	≤0.05
MST @ 55 % TS	–	≥650
S.G at 25 °C	–	≥0.94
Mg content (ion solids)	ppm	≤60.00

Table 10
Base binder, mixing conditions and optimum concentration of NRL.

Base asphalt/Rubber type	Percentage	Mixing condition	Optimum content	Author
PG 60/70 NR latex	3, 5, 7 and 9 % NRL	Shear mixer at high speed, 4500 rpm, 40 min, 155 ± 5 °C	7 %	[114]
PEN 80/100 & PG76 binder. Nano polyacrylate and NR latex.	4 % Nano polyacrylate 0 %, 2 %, 4 % and 6 % NR	High-speed shear mixer at 1650 r/min, 60 min, 150 °C	4 %	[147]
PEN 60/70 NR (DRC 60)	0 %, 2 %, 4 % and 6 % NR Latex 5–7% (OBC).	High-speed shear mixer for 20 min,140–170 °C	6 %	[148]
60/70 PEN (AASHTO M20-70) Toluene-treated cup lump rubber	5 %, 10 %, and 15 % cup lump rubber	High-speed shear mixer at 5000 r/min, 120 min, 160 °C	5 %	[140]
60/70 penetration grade Liquid NR latex	3 %, 5 % and 6 %	High-speed shear mixer at 120–130 r/min, 120 min, 160 °C	6 %	[149]
60/70 penetration grade Liquid NRL + Nano-organosilane (ZycoTherm) surfactant additives	3 % and 6 % NR 0.1 % ZycoTherm	High-speed shear mixer at 1000 r/min, 30 min, 160 °C	6 % NR 0.1 % ZycoTherm	[150]
Penetration grade of 60/70 CR and NRL	5 %, 10 % and 15 % CR 5 % and 10 % NRL	High-speed shear mixer at 1000 r/min, 30 min, 160 °C	10%CR and 5%NRL based on the penetration value	[146]
60/70 PEN bitumen	2.5 %, 5.0 %, 7.5 % and 10.0 %, NR Latex, High-speed shear mixer at 1650 rpm for 60 min at 150 °C	700 rpm high-speed shear mixer, 30–45 min, 160 °C	10 %	[151]

temperature and long digestion time requirement during modification [103]. Continuous mixing is required to prevent phase separation of the bitumen-polymer mixture. Moreover, to date, there are no reliable conclusions on the optimum amount of natural rubber required for bitumen modification, even though some authors claim it is significantly dependent on the nature and constituent of the NR [140,153]. Efforts have been made to ensure homogenous dispersion of NRL in bitumen to effect improved physical and mechanical properties. Liquid NR latex has been proposed instead of the powdered form of latex. For example, Usman et al. [149] investigated a liquid NR latex and studied the effects of different shearing rates on elasticity, temperature performance and stability of asphalt binder under high-temperature storage. The liquid NR latex reveals good cohesion due to better NRL dispersion at a mixing speed of 1200 rev/min. The incorporation of LNR latex was observed to relatively stiffen the binder and improve asphalt properties including the low-temperature performance. Also, Sani et al. [150] investigated a liquid NR latex but with the addition of nano-organosilane (ZycoTherm) surfactant as additives. They observed that NRL regardless of ZycoTherm possess the capability to stiffen the asphalt binder the phenomenon which was attributed to its elastic nature. Superpave rutting test conducted on the modified binder indicated an improved rutting resistance of NRL modified asphalt binders. Nevertheless, the addition of 0.1 % ZycoTherm surfactant to the NRL binder showed a better rutting resistance by improving compatibility.

7.6. Styrene butadiene styrene polymers

Styrene butadiene styrene (SBS) polymer is an elastomer of relatively superior efficacy at high- and low-temperature which accounts for its extensive application in high-class bituminous pavements. PMBs are anticipated to be stiffer at high temperatures, softer at low and mid-range temperatures and keep adequate levels of fluidity throughout the creation and application of asphalt mixtures [154]. To this end, macroscopic properties such as moisture susceptibility, ageing resistance and thermal storage stability are commonly assessed. SBS however is characterized by a smooth surface and reduced moisture-induced damage. SBS-modified asphalt binders have exhibited excellent stiffness, temperature susceptibility, elastic recovery, and abrasive resistance [155–157] which has

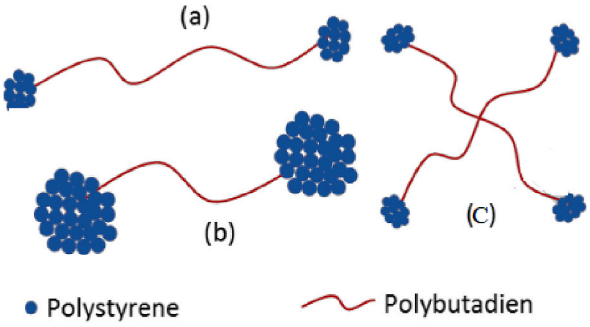


Fig. 4. Structure of (a) linear SBS molecule with high butadiene content (b) linear SBS molecule with high styrene content and (c) radial SBS molecule [53].

been related to its three-dimensional network of structures that formed by physical crosslinking of molecules [53].

The two blocks that made up SBS were the stiff and well-compatible styrene block and the butadiene block, which is highly elastic and compatible with the aromatic fraction of bitumen [158]. The butadiene-styrene ratio (B:S) can be adjusted during the production process (see Fig. 4a–c) with (a) and (b) containing high content of styrene and butadiene, respectively and (c) representing a radial SBS molecule. Both polybutadiene and polystyrene exhibit unique mechanical properties at room temperature, with polybutadiene showing a soft elastic behaviour while polystyrene shows a stiff behaviour at room temperature. Schaur et al. [53] has noted that PS (stiff) or PB (soft elastic) dominates the mechanical properties of SBS below the glass transition of PS, depending on whether PS can establish a continuous network within the SBS polymer. By incorporating SBS into bitumen, the PS end-block cross-links together to form a three-dimensional network. While the PB mid-blocks contribute to the material elasticity. The cross-linking of PS end-blocks may lead to a change from a continuous bitumen-matrix to a continuous polymer-matrix [101].

Numerous researchers have extensively studied the transition to a continuous polymer matrix. The amount of SBS, as well as the butadiene-styrene ratio in the SBS structure, has been found to have a substantial impact on properties such as fatigue, stripping, permanent deformations and the stress relaxation and load-bearing capacity of the modified bitumen. The relaxation mechanism in polymers depends on several factors such as the type and nature of the polymer, fillers and components used, stress limits, straining rates and temperature. Previous studies [158] have reported the extent to which the induced stresses in pavements are been released with the incorporation of polymer modifiers under traffic action. However, stress relaxation in SBS-modified asphalt has been attributed mainly to the interconnected network structures [159]. According to Polacco et al. [160], the interface of SBS with asphalt binder primarily consists of a polymer that absorbs the most compatible fractions of the bitumen while maintaining its structure, causing the polymer to swell in the bitumen. The complication of such mechanisms is generally due to the large compositional variation of bitumen and polymers which can affect their affinity, and ultimately the properties of modified bitumen. The mechanism that governs the interaction of bitumen with SBS has mostly been identified to be physical where the aromatic fractions in asphalt binder diffuse into SBS molecules increasing in volume. Volume increment by approximately 3–5 times the original asphalt volume was earlier reported when SBS was incorporated into the asphalt binder as a polymer modifier [51].

To date, the effects of specific polymers on bitumen cannot be generalized due to variations in their chemical composition and that of base bitumen. The best that we can tell, there have not been sufficient studies that could lead to categorical conclusions on the selected polymer such as SBS in terms of performance in the presence of varieties of base binders collected from different sources or simulated to mimic the properties of many possible binders. However, poor dispersion of different polymers in bitumen after the modification remains a major drawback [124,161,162]. Similarly, several studies have emphasized that the properties of SBS-modified bitumen are primarily dependent on the concentration of the SBS incorporated into the binder [163,164] One of the foremost problems confronting both producers and suppliers of SBS modified bitumen is the property losses during storage and transportation which result in poor quality of roads built using such products. Both Xia et al. and Zhu et al. [165,166], observed that SBS and the bitumen's asphaltenes compete for the absorption of light fraction hydrocarbons present in the bitumen. But if these components are insufficient, phase separation occurs which leads to storage instability. A significant difference in either the glass transition temperature or the molecule size between two components in a mixture system indicates the phase structure of bitumen/SBS blends.

A thorough investigation of how storage temperature, SBS concentration, polymer type, and base bitumen affect the properties of PMB was carried out [51]. Table 11 presents the source and properties of the five SBS-based block copolymers and another reactive elastomeric terpolymer (Elvaloy 4170). The result showed that the base bitumen type has less of an impact. The observed property degradation was largely influenced by temperature, SBS concentration, and type of polymer. The standard SBS-modified binders are normally produced by grafting 2–6% by the weight of the polymer in the asphalt binder [167,168]. The optimum SBS concentration of 3 % was obtained in this study according to the polymer molecules' bitumen network formation threshold. When compared to the D1192 grade (which has a higher vinyl content of >35 per cent), conventional polymer grades showed significantly less property erosion. However, due to its special characteristics, the polymer grade MD0243 maintained perfect stability at 180 °C even after 21 days. The study is constrained by the fact that performance grade (PG) measurements were made at 10 rad/s angular frequency, where bitumen molecules control the rheological response, making it impossible to detect changes in the polymer network.

A report has also shown that the concentration of vinyl in SBS is a major factor in the rate of erosion of property after storage [51]. Compared to SBS polymers with lower vinyl content, high-vinyl SBS polymers slowed the rate of erosion. The use of a high-vinyl content styrene-butadiene copolymer with sulfur as a cross-linker to modify a virgin binder (70/100) was investigated [158]. The authors prepared a high-concentration masterbatch consisting of 7.5 % SBS and then created dilutions to reach various polymer concentrations between 3 and 7 %. After numerous tests, a polymer content of 4.5 % was determined to be stable because it complied with the standards of the current PMB as determined by conventional tests. When SBS concentration exceeded this limit, storage

Table 11
Source and basic properties of SBS polymers [51,169].

Polymer	Properties
D1101	Triblock linear SBS copolymer, Content of styrene: 30–32 %. MFI <1 g
D1192	Triblock linear SBS copolymer, Content of styrene: 28.5–32.5 %. MFI <1 g, Vinyl quantity >35 % Triblock quantity >90 %, Mw: 138–162 kg/mol
D1184	Triblock branched SBS copolymer, Content of styrene: 29–31 %, MFI <1 g
D0243	Triblock linear SBS copolymer, Content of styrene: 31–35 %. MFI: 20 g, Vinyl content >35 %, Diblock content: 75 %
LG411	Triblock branched SBS copolymer, Content of styrene: 29–31 %, MFI <1 g
Elvaloy 4170	Reactive Elastomeric Terpolymer, MFI = 8 g

stability failed and gelation occurred. When compared with a conventional PMB a reduction of stiffness was noticed which was compensated for by a higher elasticity and complex modulus and is more resistant to cracking and rutting.

8. Optimum polymer dosage

Taking the cost of the modifiers into consideration, it is imperative to determine the ideal dosage of polymers and other additives. The common techniques involve the use of instruments to characterize the blended mixture and determine the impact of additive weight percentage on the mechanical, rheological, and physical properties of the bituminous mixture.

8.1. *The use of S-curve*

Determination of the optimum dosage of polymer in asphalt mix is critical for efficient performance characteristics. At optimum concentrations, the modified samples are characterized by a strong 3-dimensional polymer network structure in the binder [51]. Beyond the optimum, the performance of the binder will significantly be affected even by a slight increase in the amount of polymer. The rheological properties of the PMB would change substantially. To assess optimal concentration, the S-curve has been used to determine the optimum concentration of SBS in bitumen [51]. In a related study, Kumar et al. [169] compared the high-temperature properties of 3 distinct SBS-modified binders at different concentration (1–8%). To use S curve the softening point is plotted against the polymer concentration using two different bitumen types [51,169]. It can be observed that the S-curve is more dependent on the polymer type rather than the base bitumen, even among styrene-butadiene based polymers. A typical S-curve can be divided into 3 regions based on the slope as shown in Fig. 5. The lower plateau region corresponds to concentration <3 wt%, the steep middle region corresponds to that between (3–6 wt%) and the upper plateau region falls between (6–8 wt%) concentration. The two curves are similar in many respects. However, a well-developed plateau observed in the case of certain polymers with no significant increment of temperature change as concentration increased between 6 and 8%. The steep rise in SP above 3 % by weight of SBS polymer can bring about a drastic decrease in phase angle (δ) values which points to an increase in the elastic properties of binders [170]. If the concentration is increased from 3 % upwards, the new SBS concentration may take the PMB to the upper plateau region (>6 %) with a higher softening point. This is desirable provided the degradation in the other properties especially after storage would not position the PMB in the steep middle region (3–6%) with a lower softening point [171,172]. Numerous studies have found that the steep middle region of modified binders contains an interconnected maltene-polymer-rich network, which significantly increases the load-carrying capacity of the modified binders [170,173]. In all these studies, it is enough to say that a significant factor in the modified bitumen's property degradation is the SBS concentration.

8.2. *Visual observation of fluorescence images*

Fluorescence microscopy morphology observation has also provided direct information on the distribution of different concentrations of SBS in the modified asphalt mix. Xu et al. [174] analyzed using fluorescence microscopy how the SBS modifier disperses when added to bitumen and how it combines with it. The study seeks to obtain a homogeneous dispersion of SBS in bitumen. The ethyl-vinyl-acetate (MAH-g-EVA) grafted maleic anhydride is used as a compatibilizer to increase the compatibility of polymers. The composite, SBS, MAH-g-EVA, naphthenic oil, and antioxidant, was earlier predicted to encourage the homogenous distribution of SBS modifier in bitumen during the high-speed shear by exploiting the synergy between the two additives. The study also investigated the effects of the SBS concentration on different samples of bitumen. Fig. 6(a–f) shows distinct fluorescent images of polymeric bitumen mixed with 3, 4, 5, 6, 7 and 4.5 wt% SBS, respectively. The images revealed a two-phase system comprised of polymer and bitumen, the polymer phase appears bright while the bitumen phase is the dark portion when viewed under the fluorescent [175]. It is obvious that the image in (a) with a 3 % SBS modifier presents a bitumen-dominated phase with very weak inter-link which can cause weak physical

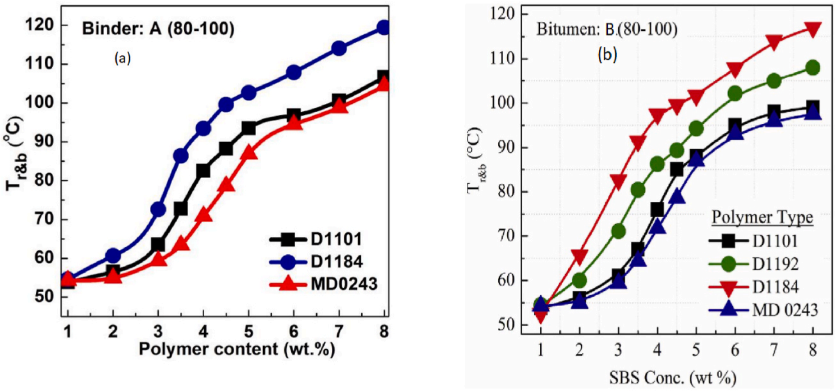
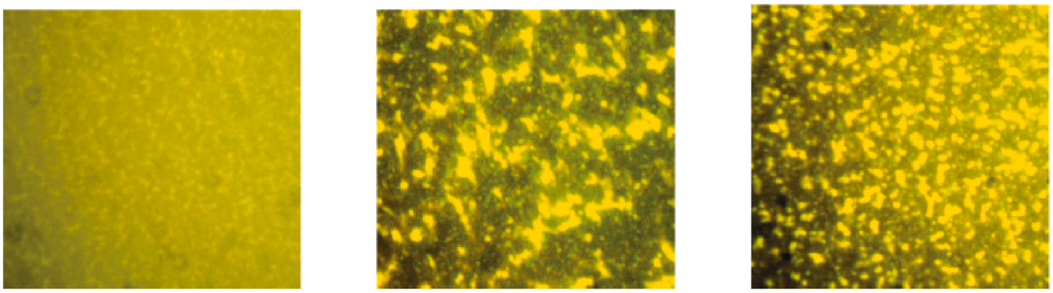
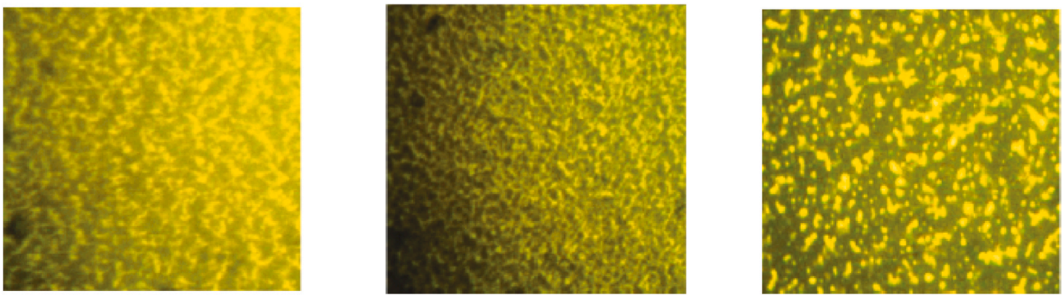


Fig. 5. S-curve of softening point created by adding various styrene-butadiene-based polymers to two different bitumen samples (a) A [80–100] and (b) [80–100] bitumen [51,169]. The S- shape of the curves is preserved regardless of the type of base bitumen used.



(a) SBSC-3 **(b) SBSC-4** **(c) SBSC-5**



(d) SBSC-6 **(e) SBSC-7** **(f) SBS-4.5**

Fig. 6. Fluorescent images of the reformulated bitumen containing (a) 3 wt% SBS (b) 4 wt% SBS (c) 5 wt% SBS (d) 6 wt% SBS (e) 7 wt% SBS and (f) 4.5 wt% SBS [174].

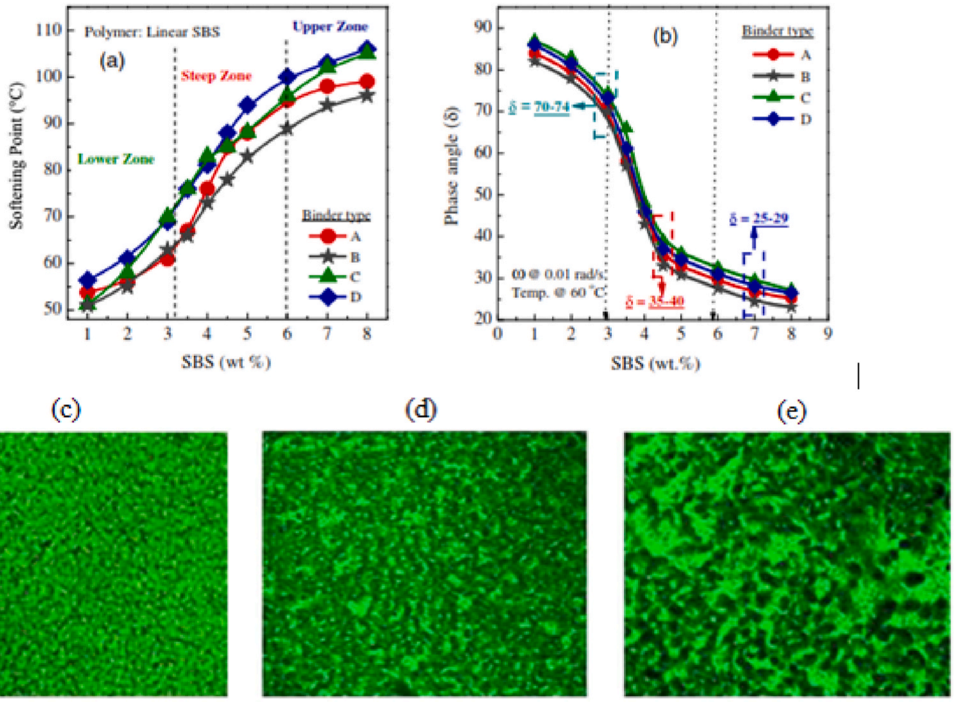


Fig. 7. The response of (a) Softening point and(b) phase separation to type and polymer concentration and corresponding FM images of (c) 3 wt% SBS-MBs (d) 4.5 % SBS-MBs and (e) 7 % SBS-MBs. Dark regions are the asphaltene-rich phase while the fluorescent active light regions represents the maltene polymer-rich phase [177].

and rheological properties.

Bright substances with a dense distribution display a sizable distribution area when the SBS modifier content is increased to between 4 and 5 %, as shown in (c). The dark area and bright area formed the polymer-bitumen continuum co-mingled phase when the SBS content reached 6 %, as can be seen in (d) [176]. The modified bitumen would perform better as a result of this. However, when compared to SBS-6, the polymer in SBS-4.5 had a weaker cross-linking with the bitumen. As can be seen from (f), the SBS modifier was also evenly distributed in the bitumen. This helps to explain why SBS-6 performed better than SBS-4.5 in terms of viscosity, complex modulus, average per cent recovery, and thermal storage stability. Meanwhile, SBS-modified bitumen with a 7 % SBS modifier content had a viscosity that was higher than the limit of 3.0 Pa at 135 °C. To reduce the poor thermal storage stability of SBS-modified bitumen, it was advised that the SBS modifier's content in bitumen should not exceed 7 %.

Similarly, fluorescent images have been used for the investigation of the thermal stability of SBS-modified asphalt. Islam et al. [177] conducted a thorough investigation into the thermal deterioration of the SBS-modified binder. The authors utilized 3, 4.5, and 7 % by weight of bitumen and evaluated the SBS-MB morphology concerning SBS concentration. Fig. 7(a) and (b) illustrate the effects of SBS concentration on softening point and phase separation properties as well as binder classification based on the response to type and polymer concentrations. Fig. 7(c–e) are the FM images showing formation of 2-phase morphology. In Fig. 7(c), the 3 wt% polymer is dispersed more as discrete units with weak interconnections. The polymer phase formed an interconnected network-like morphology when the concentration is increased to 4.5 % and 7 % as can be seen in Fig. 7(d–c). Thus, the enhancement of the softening point (Fig. 7 (a)), and phase angle (Fig. 7(b)), is accompanied by the formation of an interconnected maltene-polymer phase [170]. Thus, in the steep intermediate zone, slight damage to the network can lead to considerable degradation of property.

9. Sustainable pavements

Plastics are ubiquitous in all aspects of daily life. Continuous production of waste plastics will cause major global environmental issues if appropriate waste treatment approaches are not implemented. In addition to the environmental benefits of recycling waste materials, sustainability in pavement construction can be achieved by recycling the waste plastic materials. This method results in a reduction in the consumption of energy required to import new materials. One way to effectively address environmental issues is to turn waste into a resource or move toward a circular economy.

9.1. Waste packaging, plastic scraps and crumb rubber

In recent years, interest from all spheres of society has been drawn towards promoting the recycling of plastics from used packaging materials. Plastic products, such as shopping bags, bottles, and pipes, are widely used. The disposal of these wastes constitutes serious environmental concerns. Waste packaging polymers recycling is one of the ways to achieve sustainable road infrastructural development. Recycled waste packaging polymers can ameliorate environmental pollution and prevent resource wastage. For example, to achieve effective municipal solid waste management and to lessen road potholes brought on by heavy traffic and axle weight, [178] investigate the impact of blending HDPE and PP in conventional AC-20 graded bitumen at different plastic compositions. The blends with 2 % HDPE and 3 % PP polymer loading were found to be the most effective. A homogeneous bitumen-polymer blend was achieved at these concentrations.

Through a variety of experimental lab test methods, [179] use plastic waste materials, such as plastic bottles (PET) and gas pipes (PE), as a bitumen modifier in asphalt materials. The concentrations of plastic by total weight of bitumen used to prepare Marshall samples ranged between 0 and 20 %. The results show that after 30 days, With plastic contents up to 20 %, the stability of asphalt concrete modified with plastic was improved while still meeting standards. [180]The physical, thermal, rheological, and storage characteristics of the MPP-modified binder at different concentrations (2, 4, and 8 %) in asphalt cement were investigated when evaluating the viability of using MPP as an asphalt modifier using a wet method [180]. It was determined that less than 4 % of MPP additives would provide sufficient permanent deformation and higher resistance to fatigue damage in low-temperature climates. However, in hot climates, higher additive concentrations might be preferred to prevent permanent deformation. Waste ethylene-vinyl acetate copolymer (WEVA) has been used to modify asphalt binders to improve flow at high temperatures and reduce cracking at low temperatures. Like other polymers, the dispersion of these wastes in the asphalt remains a major limitation. In the same vein, grafting certain additives with waste polymer materials can improve the dispersion and enhance their properties. Zhang et al. [117,181] investigated the effects of self-prepared graphene oxide (GO) emulsion on the dispersion of WEVA in bitumen. The GO incorporated in 0.08 % amount was able to reduce WEVA particle aggregation in the modified asphalt system. This improvement was ascribed to the adhesion to and accumulation of GO nanoparticles on WEVA particle surfaces. Because a small amount of GO was added, no significant cost was introduced to the overall cost of the asphalt modification using waste polymers.

Certain waste polymers are however often excluded from recycling and they are called plastic scrap. Their rejection is often because of their size, contamination, or physical state. A study [182] has reported pavement service life of approximately 2.81 times higher than that of the virgin binder after incorporating polyethylene terephthalate which is a waste plastic material (PTP). PTP is also harmful to the environment, so reusing it in engineering construction projects like paving roads could enhance the qualities of asphalt binder and asphalt mixtures. Additionally, it will lessen the amount of asphalt binder used in the mix, lowering the overall project cost. The strength of the asphalt mixtures was increased by the addition of PTP. The optimal PTP content for the asphalt mixtures to perform better was 12 %. Movilla-Quesada et al. [183] reported that using plastic scrap by the dry method resulted in reduced moisture damage, increased indirect tensile strength, higher air void content, and a 2 % decrease in the conserved tensile strength ratio. The outcome demonstrates that for any of the sizes examined for improved plastic properties without sacrificing their resistance or stiffness, the ratio of plastic scrap to binder replaced should not exceed 1:1.

Crumb rubber modifier (CRM), is a thermoplastic elastomer used to modify the asphalt binder because of their viscoelastic characteristics. Other frequently used polymers in the same category are Styrene-butadiene rubber (SBR) and Styrene butadiene styrene (SBS) polymers. Applications for CRM have been discovered in the creation of asphalt mixtures for two purposes namely [1] to come up with a different way to dispose of tyres and [2] to enhance the effectiveness of asphalt mixtures. CRM is a rubber by-product made from recycled tyres that, after being crushed and sieved, is used as a feedstock for another manufacturing process. Its use in the construction of road pavement layers offers a ground-breaking platform for significantly reducing residual wastes and valuing waste materials. Because they do not melt in the heat or crack in the cold, they perform better across a wider range of temperatures than bituminous mixtures [184]. Similar to NRL, the use of CRM in bitumen has also been found to have higher resistance to thermal cracking and permanent deformation. The literature claims that depending on the procedures and technology used, its performance varies greatly.

A recent study has queried if CRM can effectively replace polymer-modified bitumen in asphalt mixture [185]. Six plant-produced mixtures—three semi-dense surface courses and three dense binder courses modified with engineered crumb rubber using the dry process—were tested mechanically on a laboratory scale by the authors. At high temperatures, the dense mixtures performed similarly to the control PMB but semi-dense mixtures failed. However, the CRM performed correspondingly or better than the control PMB at low temperatures. From an economic point of view, literature has shown how economical it is to use crumb rubber as an aggregate substitute. Technically however, contrary to the earlier report which highlights that CR-modified mixtures are less susceptible to moisture damage relative to the conventional PMB mixtures [186] another study in the year 2011, has revealed CRM mixes susceptibility to moisture compared to traditional mixes [187]. Rubberized asphalt generally is characterized by low compaction, highly heterogeneous, with obvious density variation between bitumen and rubber. Many investigators agreed they are more susceptible to moisture and are more likely to suffer from storage stability.

Despite these limitations, several studies have reported improved asphalt performance when fortified by crumb rubber. Some of these advantages are enhanced rutting, cracking and fatigue resistance, reduced temperature susceptibility, pollution and pavement maintenance costs, reduced traffic noise, improved durability and, environmental quality. Mohamed and Zumrawi [188] investigated the effect of CR addition to asphalt binders in Sudan. The viscosity and softening point of base asphalt increased by 200 % and 15 %, respectively, after the addition of 15 % CRM. A decrease in penetration and ductility was almost 75 % and 25 %, respectively using the same concentration. Pavements containing CRM binders are characterized by larger amounts of greenhouse gas emissions at the asphalt plant. One viable means to minimize the effects of greenhouse gases is using the exact quantity of components that make up the mix. Bilema et al. [62] used 0–15 % CR by weight of the binder with 1.5 % Sasobit additives, crushed granite aggregate of 9.5 mm nominal maximum size, and determined the ideal bitumen content and assessed the moisture sensitivity of crumb rubber modified asphalt at 125 and 135 °C compacting temperatures. The mixing condition for the CR-asphalt mixture is 177 °C, shearing at 700 rpm for 30 min while Sasobit conducted at 120 °C at 1000 rpm for 10 min. The results demonstrate that moisture damage increased as crumb rubber content increased, but it does not exceed the specification limit of 80 % for the indirect tension strength ratio (ITSR).

Many studies have investigated antistripping agents towards achieving low moisture, good dispersion and improved performance of binders. For instance, the effects of three different Anti Stripping Agents (ASAs) namely EVOTHERM M1 (M1), T9 and AD-here LOF-6500 (LOF-6500) were examined on the rutting, moisture, and low-temperature crack performance of Crumb Rubber Modified Asphalt (CRMA) [98]. The outcome demonstrates that T9 had a less significant impact on low-temperature crack resistance than M1 or LOF-6500. Therefore, they concluded that CRMA mixtures, regardless of ASAs, are appropriate for tropical regions where rutting and moisture problems are major concerns. In many tropical countries like Nigeria, and Malaysia with a high amount of rainfall, distress on roads caused by moisture damage is prevalent. The degree of adhesion at the interface between CR and aggregate decreases in the presence of moisture, causing moisture damage which shortens the service life of asphalt pavement [189]. Ref. [190] used the wet method to investigate the effects of crumb rubber particle size on moisture damage and the strength of hot mix asphalt. The binder was added to the asphalt mixture in three different sizes: 0.075, 0.15, and 0.3 mm, with a crumb rubber content of 5 % by weight. The outcome showed that the addition of small-size CR increased the asphalt mixture's strength while only marginally enhancing its moisture resistance. Wang et al. [60] investigated how moisture affected CR adherence to the asphalt mixture and correlated the interface characteristics at multiple scales. At the macroscale level, a cracking resistance test was carried out on the CR mixture. The surface free energies were calculated at the microscale level, and the effects of moisture on the surface adhesion of asphalt and aggregate were investigated. This study's findings showed weakened interactions between the molecules of asphalt and aggregate, as well as decreased crack resistance and adhesion between the asphalt and the aggregate.

10. Lessons learnt and recommendations

Following reinforcement with various types of polymers, it is clear that bitumen's physical and rheological properties have improved. With the knowledge from this review, it would be possible to create a long-lasting pavement by adding polymer additives to local bitumen from Nigeria. The following can be listed as some of the lessons that were discovered through the use of polymer additives, according to this review:

- There is little research on polymeric modification's potential to improve Nigerian bitumen for paving. The majority of studies in this area have mainly focused on the Agbabu bitumen.

- Research on polymeric studies at the SARA fraction levels is scarce. The macroscopic properties have always been the basis for evaluation. The understanding of the mechanisms involved in the modification and the deterioration of pavement under load may be aided by knowledge of bitumen properties transformation at the SARA level.
- Due to the diversity of bitumen and variations in properties and composition from one location to another, it is impossible to generalize the effects of different polymer additives on bitumen. Research using microscopic analysis can help in this regard.
- In light of the financial implications and sensitivity of properties to additive concentration, determining the optimal polymer concentration is of utmost importance. Only proportionate dosage increments have been used in evaluations, with inconsistent results. It is strongly recommended to use additional optimization techniques in this case.
- Polymer homogenous dispersion is still one of the issues the industry is dealing with. The mixing process has not been thoroughly studied for use in practical highway design applications. Investigating the results of polymer premixing processes is also necessary.
- The cost of energy used during the modification must still be kept to a minimum. The destruction of bitumen and the environment caused by heating bitumen to temperatures as high as 160 °C remains a serious challenge.
- To scale up to the field level, a cost-benefit analysis of polymeric-modified asphalt binder and aggregate mixtures is strongly recommended. There is currently a lack of information to conduct economic analysis in this area. Studies have only looked at the technical advantages.

11. Conclusions and recommendations

The findings of this review indicate that polymers and other chemical additives are superb in enhancing the quality of the original binder if the right polymer is selected and applied in the right dosage. Therefore, depending on the location of the bitumen, Nigerian local bitumen can be upgraded using polymers as additives. Reactive polymers can be used to solve many of the problems inherent in the original bitumen and the functionality of asphaltic pavement can significantly be enhanced. Reactive polymers have also shown promise in assisting bitumen dispersion and resulting in a more homogeneous polymeric mixture with enhanced properties. In specific to the Nigerian bitumen, the following conclusions can be drawn from this study:

- This review shows that there has not been much research done on upgrading Nigerian bitumen to the level needed for building durable roads. The bulk of research that has already been done has mostly concentrated on the Agbabu bitumen, possibly because of its high quality and low preprocessing requirements. Whereas, reports have shown that bituminous sands are widely distributed in various locations in the southwest of the country, including Loda, Abigi, and Sunbare. Investigations on these bitumen deposits are rarely been reported and are therefore recommended for future research.
- Previous attempts to modify Agbabu bitumen have concentrated on the effects of additives such as ethyl Vinyl Acetate (EVA), Polyethylene-co-vinyl Acetate (PEVA) and Polystyrene-co-butadiene (PSCB), Sulfur, Polypropylene (PP), polyphosphoric acid (PPA), High-Density Polyethylene, Crumb rubber, Waste Water Sachets (WWS) and Waste Polyethylene bottles (WPB) on the physicochemical, mechanical and rheological properties. However, for sustainable road infrastructure, plastic items like bottles, pipelines, and shopping bags may be taken into account for future research.
- There are few studies done on the effect of nanomaterials such as Multi-Wall Carbon Nanotubes (MWCNTs) and green synthesized silver nanoparticles (AgNPs) on the physical, rheological and mechanical properties of the bitumen mixtures. Nanotechnology has a prominent role to play in bitumen modification owing to the unique properties of nanomaterials. Therefore, the effects of other nanomaterials such as graphene, nanosilica and nanoclay on performance characteristics of Nigerian bitumen is therefore recommended for future research.
- Research on the modification of bitumen binders with different polymers revealed that several properties of the parent bitumen can be enhanced if the additives are added in the right amount. The effects of the modifications depend on the type and dosage of the polymer material as well as the original bitumen's properties. The modifications with polymer and other chemical additives brought a reduction in penetration, softening, fire and flash points. The effects of these polymers on viscosity, however, vary depending on the dosage and type of polymer. However, the use of performance parameters such as complex modulus (G^*) and phase angle (δ) which are frequently measured before and after modifications are rarely reported and are therefore recommended. Only the effects of property changes on dynamic and kinematic viscosities are commonly documented.
- The studies found in the literature covered the dosage range of 3 %–6 % polymer additives and it was discovered that in all cases, the effects of the modifications increase accordingly with the increase in dosage. However, crumb rubber and other plastic waste can be employed in quantities ranging from 14 % to 20 %. Aside from the environmental benefits, the cost of modification should also be considered.
- The modifications with antistripping and adhesion promoter agents towards achieving low moisture, good dispersion and improved adhesion with different aggregates is not popularly practised with Nigerian bitumen. Also, the optimum dosage of the various polymers seems not determined yet as a situation arose where the performance gains peak and, further increase in dosage worsens the performance. A deliberate investigation to explore better means of obtaining optimal content using an approach such as experimental design and response surface methodology is highly recommended for future research.
- Polymer dispersion in bitumen seems to be a major technical challenge during modification. The review shows a continuous effort regarding the prevention of agglomeration of crystalline polymers in the bitumen mixture which is the reason for heating bitumen to a high temperature during the hot mixing. Liquid additives with low melting points can produce good cohesion due to better dispersion which stiffens the binder and improves binder properties.

- Aside from improved mechanical performance, SBS exhibits an increased ageing resistance when used as a modifier. The inspection of the properties of aged bituminous mixtures revealed the potential of the SBS polymer, which provides a clear indication of the essential life cycle cost evaluation to establish a good balance between construction costs and long-term durability.

Data availability statement

The data associated with this manuscript is included and referenced in the article.

CRediT authorship contribution statement

Salawudeen Taofeeq Olalekan: Conceptualization. **Arinkoola Akeem Olatunde:** Writing – review & editing, Writing – original draft, Methodology. **Salam Kazeem Kolapo:** Visualization. **Jimoh Monsurat Omolola:** Writing – original draft. **Olufayo Augustina Olukemi:** Visualization. **Ayanshola Ayanniyi Mufutau:** Visualization. **Ogunleye Oladipupo Olaosebikan:** Visualization, Supervision. **Abdulkareem Ambali Saka:** Visualization, Supervision.

Declaration of competing interest

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