



Application of Natural and Waste Oils as Rejuvenator in Reclaimed Asphalt Pavement: A Review

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Abstract

Researchers have become interested in using reclaimed asphalt pavement as an economical and environment friendly alternative to flexible pavement. Various natural oils or waste oils are often used in practice as a significant source for modifying and rejuvenating the binder in Researchers have become interested in using reclaimed asphalt pavement (RAP), as they contain light components similar to those found in virgin bitumen. First, basic physio-chemical properties of different oils were introduced, and then a thorough analysis of chemical composition of fresh and waste oils is conducted to offer additional insights into how the chemical contents of fatty acid influence the properties of binders. Further, an assessment was made on how the addition of various oils affects the physical, chemical, and rheological properties of reclaimed binders as well as consequently asphalt mixture properties. This review emphasizes that while both virgin as well as waste oils have vital potential to restore the conventional properties of stiff binders and enhance its fatigue and thermal cracking characteristics, they may compromise the binder's ability to resist rutting. However, increased moisture content and acid value in waste oils due to repeated applications have an adverse impact on physical, chemical, and rheological properties of asphalt binder. Moreover, exceeding the optimal concentration of oils in asphalt mixtures enhances the pavement's thermal cracking resistance but diminishes its resistance to rutting, fatigue, and moisture susceptibility. In summary, this review article presents a comprehensive overview of advantages and disadvantages associated with using oils as an environment friendly rejuvenator.

Keywords Reclaimed asphalt pavement \cdot Rejuvenation \cdot Waste and natural oils \cdot Rutting \cdot Fatigue \cdot Low-temperature cracking

1 Introduction

Asphaltic pavements are well known and widely used due to their excellent performance and easy re-establishment throughout the world. Over time, it becomes non-functional due to its continuous exposure to sun, traffic, environmental conditions, deterioration of materials resulting in modification of its properties such as higher stiffness, viscosity, and lower flexibility which induces several issues such as raveling, rutting, moisture damage, cracking, etc. [1, 2]. To ensure safety and service, maintenance of asphalt pavements becomes necessity which must be cost effective and eco-friendly. During this process, a lot of Researchers have become interested in using reclaimed asphalt pavement

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¹ National Institute of Technology, Kurukshetra, Haryana 136119, India (RAP) is generated and handling of this large volume of RAP is great challenge in pavement industry. Usually, this RAP mixture containing productive asphalt and aggregates is dumped in landfills which causes serious health hazards and environmental issues. On the other hand, re-construction and maintenance of pavement requires a recurring reservoir of the natural resources such as binder and natural aggregate. Even being a non-renewable source derived from crude oil, the annual consumption of virgin asphalt in pavement industry is 1.36 trillion metric tons [3]. Therefore, the recycling and reusing of RAP proves to be an effective measure in sustainable development of pavement industry. Hence, focus is being given to make utilization of RAP. The implementation of RAP in the pavement can not only cut down the demand of natural aggregates but also retain economic benefits.

Asphalt mix can be categorized into three main types, specifically, hot mix asphalt (HMA), warm mix asphalt (WMA), and cold mix asphalt (CMA), based on the temperature at which it is produced. The primary and most commonly utilized mixing technique for pavement construction is HMA, which is prepared at very high temperatures, reaching up to 190 °C [4, 5]. WMA is another type of mixing technique that has been increasing use over the past 2 decades. WMA is produced at temperatures ranging from 100 to 140 °C, making it feasible substitute to HMA [6, 7]. The production temperature for CMA falls within the range of 0-40 °C. Because of this low manufacturing temperature, CMA dose not necessitate heating, leading to substantial energy savings [8]. The primary obstacle in the integration of RAP binder in HMA or WMA is its stiffness due to its oxidation during the aging process of asphalt pavement. In recent years, several laboratory experiments [9, 10], molecular modeling approaches [11] have been employed to study the bitumen aging processes. The variations in the physical, chemical, rheological, and mechanical characteristics of bitumen following oxidative aging [12] allow to evaluates the bitumen aging processes qualitatively as well as quantitatively. The utilization of Density functional theory (DFT) calculations [13, 14] presents a promising approach that can contribute to comprehending the intricate process of asphalt aging. The hard asphalt binder obtained from RAP when mixed with virgin one adversely affects its properties making it more brittle and more prone to cracks and ruts [15] or the same can badly affect workability of mixture so obtained. Specifically, as the asphalt binder ages, its asphaltene-to-maltene ratio increases, inducing poor coherence and adherence characteristics as compared to virgin bitumen [15-17]. The maltenes play a crucial role in asphalt binders to sustain a prolonged service life of pavement road. Nevertheless, to maintain a balance between asphaltene and maltene, some sort of rejuvenators is used in making asphalt mixtures with RAP. Generally, these rejuvenators contain maltene, hence with their addition to aged binder, increase in asphaltene can be compensated. Therefore, to reduce hardness and brittleness of RAP binders, addition of some sort of rejuvenating agent such as polymer, chemical or naturally occurring oil and waste oils is an effective option. Moreover, having RAP content less than 25%, asphalt mixtures prepared with virgin aggregates and binder, exhibit good mechanical properties [18]. While, for high RAP content, there must be an rejuvenating agent in mixture to achieve desirable characteristics [19]. Besides this, rejuvenators also provide an opportunity to maximize RAP content [20], even 100% in HMA design [21]. In fact, rejuvenators can also be added in virgin bitumen or natural hard bitumen to improve their conventional and rheological properties. For the effective and efficient modification of characteristics of RAP binder, there must be appropriate selection of rejuvenator which should consider workability issue, less injurious emissions, and capability to restore the original properties of RAP bitumen. In addition, as properties of both RAP binders and rejuvenators depend upon temperature and loading condition, the consequences of rejuvenating agents are affected by these factors. Rejuvenators with high aromatic constituents could boost colloidal structure, decrease solubility difference between various components of RAP binder, and hence reinstate its original properties [22].

Various types of rejuvenators such as organic, petroleumbased and biomass type have been added to asphalt mixture to diminish their stiffness and enhance their workability and service life. Furthermore, in accordance with National Centre for Asphalt Technology (NCAT), various types of oils which include fatty acids, tall oils, naphthenic oils, and paraffinic oils can improve and modify the properties of RAP mixture [23]. As commercially available rejuvenators may have their financial implications, major thrust is on using waste materials as rejuvenators. Using these waste materials shall also have environmental benefits. During the long-term aged service period of asphalt pavement, it loses its oil components due to repeated wheel loads and environmental effects. To restore its original characteristics, traditional rejuvenators provide unsaturated fatty acids to the binder to compensate its oil loss contents. Further, it has been found that chemical compounds present in oils could better recover the structure and properties of aged asphalt. Numerous researchers have attempted various kinds of oilbased rejuvenators and have observed promising outcomes. This review work primarily focuses on the uses of oils as either a modifier for virgin binder or a rejuvenator for stiff binder, owing to its favorable attributes of lesser viscosity and affinity with asphalt.

Several fresh oils like mustard oil [24], mahua oil [25], tung oil [26], polanga oil [27, 28], cotton seed oil [29], castor and pongamia oil [30], olive oil [31], soybean oil [32, 33], sunflower oil [34, 35], and bio-oils [36, 37] were utilized by researchers as rejuvenating agent to produce fruitful results in areas where these are locally available at reasonable price. Besides, many researchers have investigated the application of waste engine oil (WEO) [38] with RAP due to their similar chemical structure of asphalt. The reuse of WEO with RAP also prevents environmental pollution due to their discharge in water and land sources. Furthermore, a massive amount of kitchen waste, specifically waste cooking oil (WCO) is generated every year due to rising population and its disposal in kitchen sinks, sewerage systems or waste bins [39, 40] is harmful for environment and human health. Keeping this viewpoint, several researchers have studied the impact of WCO [41-44] to rejuvenate the reclaimed binder and improve their physical and rheological properties to the scale of virgin. Recently, Zhang et al. [45] conducted a review on various categories of biomass, as well as the methods of preparing and characterizing bio oils. In addition, they examined the processes involved in producing bioasphalt, along with the conventional, chemical, and rheological characteristics of bitumen that has been modified and rejuvenated using bio-based materials.

This review paper specifically examines the following aspects: (a) physio-chemical properties of fresh as well as waste oils, (b) chemical compositions of fresh and waste oils used as a rejuvenator, (c) impact of oil addition on physical and rheological properties of the binder, (d) the alteration of the chemical properties of the binder resulting from the use of oils, (e) impact of oil addition on performance properties of the asphalt mixes, (f) economic and environmental implications of using oils as rejuvenators.

2 Characteristics of Oils Used as Rejuvenators

2.1 Physio-chemical Characteristics of Oil

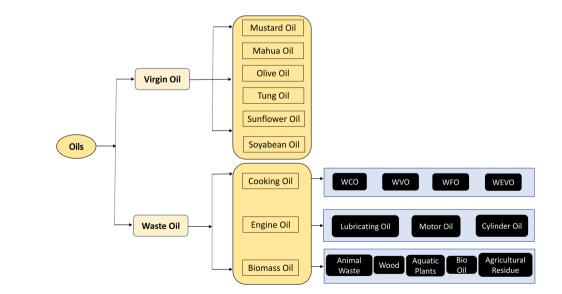
This review elaborates the use of various types of fresh and waste oils as shown in Fig. 1. Natural oils classification includes mustard oil, olive oil, tung oil, mahua oil, soybean oil, sunflower oil, pongamia oil, etc. Waste oils (WO) can be further distinguished into three types: cooking oil, engine oil, and bio-oils. The cooking oil category includes waste cooking oil (WCO), waste vegetable oils (WVO), waste fried oils (WFO), and waste edible vegetable oils (WEVO). Engine oil includes various waste oils obtained from automobile industry, which can be further classified into waste engine oil, lubricating oil, cylinder oil, motor oils, etc. Biooils are obtained from farms and forestland wastes such as crops, animals, wood wastes, other micro-organisms, plants, agricultural residues, etc., which are eco-friendly and renewable. The term 'bio-oil' essentially refers to a type of liquid fuel and a raw material that can be utilized in cooking in place of conventional oils. The physical and chemical properties of oils are shown in Table 1. Although viscosity of various fresh oils depends upon temperatures, it remains

Fig. 1 Waste oil sources

lower than viscosity of a typical asphalt binders at 60 °C. When oil is heated to temperatures above 160 °C during cooking, its density, viscosity, and acidity may increase due to reactions with oxygen and water. Because of variable usage, physio-chemical properties of waste oils do not remain consistent as virgin oils. Long-chain acids present in WO raise the chances of catalytic or thermal cracking, which can result in development of hydrocarbon chains that lead to cracking. Therefore, it can be inferred that the physical and chemical structure of cooking oil cannot be uniform since it varies based on the quality and type of oil used, number of times that oil is being utilized, cooking temperature, food composition, and span of cooking. Moreover, the basic characteristics of oils can be influenced by the oil source, application, and production process, making it important to take these factors into consideration while selecting particular oil as a rejuvenator.

2.2 Chemical Composition of Oils

Generally, gas chromatography–mass spectrometry (GC–MS) has been utilized by several researchers to recognize the chemical structure of representative oils. Table 2 summarizes the possible chemical composition of distinguishing fresh oils as well as waste oils. It can be seen that mustard oil's primary component is erucic acid, which is a monounsaturated omega-9 fatty acid [24]. Due to its non-polar properties, it aids in rebalancing the ratio of asphaltenes to maltenes throughout the restoration process. This is accomplished by supplementing the non-polar saturates within the binder composition since they are typically lacking. In terms of the chemical makeup of tung oil [26], six primary components have been identified and α -eleostearic acid being its major constituent. While mahua oil [25] contains significant amounts of two essential monounsaturated



References	Oil type	Color	Density (g/cm ³)	Viscosity	Flash point (°C)	Acid value (mg KOH/g)	Specific gravity
[24]	Mustard oil	Yellow	_	52.57 @40° C (mPa.s)	320	1.415	0.916
[25]	Mahua oil	Yellow	0.89	-	234	-	0.905
[31]	Olive oil	Golden yellow	-	29.8 @30 °C (mPa.s)	272	-	_
[26]	Tung oil	Yellow	_	62 @60 °C (mPa.s)	234	-	0.938
[34]	Sunflower oil	Yellow	-	61@60 °C (mPa.s)	315	-	_
[32]	Soybean oil	Yellow	-	58.3@40 °C (mPa.s)	-	0.75	_
[46]	WCO	Yellow	0.88	37.52 @60 °C (Pa. s)	_	-	-
[47]	WCO	Yellow	0.785	-	-	5.88	_
[48]	WCO	Yellow	0.91	40.51 @ 60 °C (mPa.s)	222	4.03	_
[49]	WVO	Brownish black	0.97	286.7@60 °C (mPa.s)	-	-	_
[50]	WFO		_	57.5@30 °C (mPa.s)	220.3	-	0.942
[46]	WEO	Dark brown	0.97	60.17 @60 °C (Pa. s)	_	-	-
[51]	Bio-oil	_	0.992	-	200	_	-

Table 1 Physio-chemical properties of natural and waste oils

 Table 2
 Chemical compositions of natural and waste oils

Oil type and its content (%)		·							
Acid type	Mustard oil	Mahua oil	Tung oil	WCO	Waste corn oil	Waste soy- bean oil	Waste sun- flower oil	WFO	Bio-oil
Palmitic	6.26	17.8	2.7	38.35	_	_	9.94	2.51	_
Stearic	-	14	2.6	4.33	-	-	3.44	3.43	-
Lauric	_	-	-	0.34	_	-	-	-	-
Myristic	_	-	-	1.03	-	-		0.98	-
Palmiotetic	_	-	-		-	-	0.14	8.92	-
Cis-11-Eicosenoic	_	-	-	0.16	-	-	-	-	-
9,12-Octadecadienoic	_	-	-	-	38.19	39.21	-	_	-
Heneicosanoic	_	-	-	0.08	_	-	-	_	2.65
9,12-Octadecadienoic (Z, Z)	_	-	-	-	8.08	13.9	-	_	14.56
Linoleic	15.38	17.9	-	11.39	_	-	56.18	11.38	-
Oleic	9.25	46.3	6	43.67	-	_	30.56	54.81	-
Linolenic	10.18	_	8.2	0.29	_	_	-	6.94	-
y- Linolenic	_	_	-	0.37	_	_	-	_	-
Erucic	40.74	_	_	-	-	_	-	_	_
α-Eleostearic	_	_	76.5	-	-	_		_	_
β-Eleostearic	_	_	2.4	-	-	_	-	_	
Octadecanoic	_	_	-	-	3.95	3.71	-	_	6.28
Tetracosanoic	_	_	_	-	0.24	0.32	-	_	
Pentadecanoic	_	_	_	-	2.41	0.95	-	-	
Hexadecanoic	_	_	_	-	10.23	7.97	-	-	3.01
9-Octadecadienoic (Z)	-	_	-	_	34.6	31.42	_	_	_
7-Octadecenoic	-	_	-	_	-	_	_	_	27.46
6-Octadecadienoic	-	_	_	_	1.23	1.46	_	_	_
Docosanoic	-	_	_	_	0.12	0.3	_	_	4.33
(Z)6, (Z)9-Pentadecadien-1-ol	-	_	_	_	0.95	0.76	_	_	_
References	[24]	[25]	[26]	[17]	[54]	[54]	[53]	[<mark>50</mark>]	[52]

fatty acids, namely oleic (46.3%) and linoleic acid (17.9%) and palmitic acid (17.8%) as the major saturated fatty acids. Further, 7-octadecadienoic acid comprises 27.46% of the bio-oil composition [52]. Due to various applications and changes in the lifespan of waste oils, their chemical composition is different from virgin oils as fresh oil contains a high content of unsaturated compounds which are converted into saturated compounds, resulting in increase in acidic value of oils. Consequently, Asli et al. [17] found that the most abundant fatty acids found in WCO are palmitic acid and oleic acid representing 38.35% and 43.67% of the oil's composition, respectively. Similarly, waste sunflower oil [53] and WFO [50] contains linoleic acid and oleic acid in abundance. Nonetheless, waste corn and soybean oils [54] deprived these saturated or unsaturated fatty acids and include methyl esters compounds in profusion. There is significant effect of water content and acid value of oil on rejuvenation mechanism of binder as will be explained in Sect. 5.

Another important factor which affects asphalt rejuvenation significantly is its SARA (saturates, aromatics, resins, and asphaltenes) components. The virgin cooking oil consists of large amount of aromatics and saturates but these contents get reduced by mortification of oils [55]. Finally, the relationship of chemical composition and efficacy of oils as rejuvenator has been scarcely researched. Thus far, research has focused solely on the effects of moisture content and acidic value of oils on binder performance. Further investigation is required to determine how the variability, usage rate, and cooking-related factors influence the effectiveness of oils as an asphalt rejuvenator.

3 Effect of Oils on Physical Properties of Asphalt Binders

The basic properties of rejuvenated asphalt binder include penetration, ductility, softening point, and viscosity evaluated by penetration test, ductility test, softening point test, and Brookfield rotational viscometer, respectively. Penetration test signifies deformation resistance and consistency of bitumen at 25 °C whereas softening point identifies the temperature stability at somewhat high temperatures (i.e., ideally 45-85 °C). Ductility specifies adhesive property of bitumen whereas viscosity helps in determining the viscous behavior of binder. More is the viscosity, more is binder susceptible to thermal cracking although lesser viscosity makes it more prone to fatigue and rutting [21]. The major problems associated with RAP binders are hardness, brittleness, and cracking susceptibility. In the rejuvenation process, oils can overcome these problems and restore their physical properties. Table 3 summarizes the previous research works that have studied the effect of various oils on the basic physical characteristics of RAP binder. From Table 3, it can be deduced that with increasing dosages of oils, ductility and penetration show an increasing trend, although viscosity and softening point show decreasing pattern.

The accumulation of oils also ensures that penetration value, softening point, ductility, and viscosity values of virgin binder can be achieved. Ahmad et al. [24] found that with accumulation of 10% dosage of mustard oil to RAP binder, penetration value and softening temperature of virgin binder (60/70) can be achieved. Moreover, three percentages (5%, 10%, and 15%) of mustard oils are employed and it was found that with 5% and 10% of rejuvenator dosage, penetration value of rejuvenated binder is increased and softening point reduced while 15% of mustard oil increased the softening point and decreased the penetration. Hence, a 10% amount of mustard oil can be regarded as the optimal quantity. Further, Pradhan et al. [25] discussed that concentration of 8% mahua oil is enough to approach the viscosity of target binder (PG70). Bilema et al. [31] investigated that penetration grade of 60/70 can be recovered with virgin cooking oil, natural olive oil, and WCO from aged asphalt binder (30/40) with a fixed oil content of 4%, while a greater amount of virgin as well as WEO is needed to achieve the same. Besides, with only 4% dosage of all types of oils, ductility and penetration value increases, while viscosity and softening point decreases. Further, Yan et al. [26] studied the effects of tung oil on two kinds of asphalts (Pen70 and SBS-modified asphalt) and dictated that 6-8% of tung oil for Pen70 and 4-6% of tung oil for SBS-modified asphalts are sufficient to improve the traditional physical characteristics such as penetration, softening temperature, ductility, and rotational viscosity, hence restoring them up to the level of control binders. By performing the rejuvenation of RAP binders with WFO and waste crumb rubber (WCR) together, Bilema et al. [50] proved that only combination of both the rejuvenators are capable in increasing penetration, decreasing softening point, and improving workability and stability of RAP binder up to the level of control binder 60/70. In analogy, different proportions of WCO (2%, 3%, 3.5%, 4%) and WEO (2%, 3%, 4%, 5%, 5.5%, 6%) were incorporated by Amira et al. [56] in RAP binders to find out the optimum percentages of WO to recover control binder (PG 60/70). Based on their physical testing results, optimum percentages for WCO and WEO are 3.5-4% and 5.5-6% that can increase penetration and decrease softening temperature near to the level of control binder respectively.

Further, Wan et al. [57] studied the modification of PG 60/70 with treated WCO that oil-based modification can improve the physical performances of modified asphalt binders. After determining the acidic value of WCO, they performed chemical treatment of WCO using transesterification and found that this treatment reduced the value of acid

Table 3 Im	Table 3 Impact of natural and fresh oils on conventional properties	n conventional proper	ties						
References Oil type	Oil type	Binder	Aging	Oil content (wt.%)	Pen- etration at 25 °C		Ductility	Softening Ductility Viscosity Findings	Findings
[24]	Mustard oil	60/70	Extracted RAP binder 5, 10, 15	5, 10, 15	S+	S-	P_	S-	The ideal amount for rejuve- nating aged asphalt binder is suggested to be 10% of Mustard oil.
[25]	Mahua oil	PG70	Extracted RAP binder 4, 8, 12	4, 8, 12	<u>ط</u>	4	ď	S-	Mahua oil is effective in concentration of 8% which is quite enough to approach the viscosity of target binder (PG70) and enhancing the workability of mixture.
[31]	Olive/ VCO/ WCO/ VEO/ WEO	60/70	(RTFO+PAV)	4	¢,	Ŷ	\$+	S -	The accumulation of 4% dosage improved the con- ventional properties of the RAP binder, while a greater amount of virgin as well as WEO is needed to achieve the same.
[26]	Tung oil	Pen/ 70SBS	(TFOT+PAV)	2, 4, 6, 8	S+	Ŷ	S+	S-	Adding 6–8% tung oil to Pen70 and 4–6% tung oil to SBS-modified asphalt effectively enhance their physical properties up to the level of control binders
[59]	Castor oil	Pen 50	1	0, 5, 10, 15	\$+	Ŷ	Å	-	Castor-oil-based bioas- phalt effectively modifies bioasphalt binder to the level of PG-50. When 10% bioasphalt is added to base asphalt, penetration increases almost 20-fold in the asphalt binder.
[54]	Soybean oil/Corn `oil	PG-64-22	Extracted RAP binder	2, 4, 6, 8, 10	\$+	Ŷ	\$	Ŷ	The effective impact on aged asphalt can be observed using WVCO (specifically corn oil and soybean oil) at an optimal dosage of 6%.
[63]	WCO/ WEO	VG-40	Short-term aged	1, 2, 3, 4, 5	\$+	Å	S+	S-	The most significant enhance- ment in bitumen modifica- tion occurred when WEO and WCO were in the range of 3%.

Table 3 (c	Table 3 (continued)								
Reference	References Oil type	Binder	Aging	Oil content (wt.%)	Pen- etration at 25 °C	Softening	Ductility	Softening Ductility Viscosity Findings	Findings
[46]	WEO/ WCO/ bio-oil	Pen 70	From RAP	2, 4, 6, 8, 10, 12	S+	Ŷ	S+	Ч-	The ductility cannot be fully rejuvenated by any of the three types of WO, and there is an optimal WO content that achieves the best results.
[61]	WEO/ WCO	Pen 70	(RTFO+PAV)	1, 2, 3, 4, 5	S+	S-	S+	S	WCO is significantly effective in rejuvenating than WEO except for viscosity.
[62]	WEVO	SBS/ 60-80/ 40-60/	(RTFO+PAV)	3, 4, 5, 6, 7	,	Å	* *	S ⁻	Effect of rejuvenation of WVO on softening tempera- ture and viscosity of SBS- modified binder is found to be poor. The optimal content for SBS modified, 60–80, and 40–60 are 4.8, 5.2, and 6.6, respectively.
[17, 64]	WCO	80/100	Short-term aged	1, 2, 3, 4, 5	\$+	γ	\$ *	S_	Optimum dosages of 1%, 3%, and 4.5% of WCO can effi- ciently restore 50/60, 40/50, and 30/40 asphalt binders, respectively, near to 80/100 control binder.
[56]	WCO/ WEO	60/70	Extracted RAP binder	Extracted RAP binder 2, 3, 3.5, 4 2, 3, 4, 5, 5.5, 6	* *	Å	4	S-	Optimum content of WCO and WEO was found to be 3.5-4% and 5.5-6%, respectively, for increasing penetration and decreasing softening point.
[65]	ΟΛΜ	60/70	1	1, 2, 4, 6, 8	S+	Å	\$ *	S-	The optimum dose of WVO improves penetration, duc- tility, and reduce softening temperature and viscosity which can enhance the fluidity.
[50]	CR+WFO	0//09	Extracted RAP binder	I	S+	Å	Ч	S-	The blend of CR and WFO enhances the stability and workability of RAP binder.

Table 3 (c	Table 3 (continued)								
Reference	References Oil type	Binder	Aging	Oil content (wt.%)	Pen- etration at 25 °C		Ductility	Softening Ductility Viscosity Findings	Findings
[66]	WVO/ WEO	40/50	Extracted RAP binder 1, 2, 3	1, 2, 3	S+	S-	S+	S_	The optimum percentages of WVO 1% and WEO 3% effectively regenerate the aged asphalt.
[53]	WVCO	70/100	(RTFO+PAV)	2, 4, 6, 8, 10	\$+	S.	4	S_	After short-term aging, the optimum content of WVCO is 2-4% and, the amount of WVCO is found to be 6-8% after long-term aging.
[58]	олм	Pen 70	(RTFO+PAV)	5, 10, 15, 20	S+	S.	S+	S-	Optimal dosage of 13.4% is significantly effective in enhancing the conventional properties of RAP binder up to control binder.
[57]	WCO	Pen 60/70	Short-term aged	0, 3, 4, 5	s+	Ŷ	4	s -	Treated modified binder with WCO that exhibits improvement in penetration, softening temperature, and viscosity when compared to untreated WCO-modified binder.
[09]	WEO	80/100	(RTFO+PAV)	0, 3, 6, 9	S+	\mathbf{v}_{2}^{L}	4	S	This investigation states that optimum content of WEO 6% has ability to recover the oxidized asphalt to that of fresh bitumen.
[21]	Bio-oil	SBS/ Pen 50	Short-term aged	0.5, 1, 1.5, 2, 2.5, 3	S+	Å	Ŷ	4	The ideal content of bio-oil for Pen 50 and SBS-modi- fied binder is 1.75 and 2%, respectively.
RTFO Rol	<i>RTFO</i> Rolling thin film oven PAV Pressure aging vessel S+increase S-decrease P- not performed	ire aging vessel. S+ inc	rease <i>S</i> -decrease <i>P</i> -n	not nerformed					

RTFO Rolling thin film oven, PAV Pressure aging vessel, S+ increase, S- decrease, P- not performed

content of untreated WCO from 1.66 to 0.54 mL/g in case of treated WCO. Reduction in acidic value results in improvement of penetration, softening, and viscosity of rejuvenated asphalt binder with treated WCO as compared to binder rejuvenated with untreated WCO. Furthermore, using WVO [58] optimal dosage of 13.4% is significantly effective in enhancing the conventional properties of aged asphalt to the level of control binder. In addition, Zeng et al. [59] proved that castor-oil-based bioasphalt can also effectively modify the properties of bioasphalt binder to PG-50. When 10% bioasphalt is introduced into base asphalt, there will be an approximate 20 times increment in the penetration of asphalt binder. In other study [51], it was noticeable that the ideal amount of bio-oil for penetration grade 50 and SBS-modified binder is 1.75 and 2%, respectively, to recover asphalt's ductility and penetration near to virgin level. The PG 80/100 of bitumen was rejuvenated with WCO [17] as well as WEO [60] to determine its optimum percentage for its usage as rejuvenating agent in asphalt binder. Interestingly, it was found that 6% of WEO and 1%, 3%, and 4.5% of WCO have the potential to revive physical properties of 50/60, 40/50, and 30/40 aged binders to 80/100 control binder, respectively. Similarly, other studies [46, 58, 61] with WVO and WEO were conducted to reinstate the physical characteristics of oxidized asphalt (RTFO aging) to the penetration grade 70. Further, Chen et al. [62] performed rejuvenation of three artificially aged (RTFOT + PAV) binders SBSmodified asphalt, 60-80 and 40-60 with WEVO and found optimum proportions of 4.8, 5.2 and 6.6, respectively, for restoring their physical properties to virgin one. Researches have also been conducted with waste soybean and corn oils [54] to show that the optimum dosages of 6% are sufficient to restore penetration, ductility, and softening temperature of aged binder to virgin asphalt PG 64-22. As per the literature mentioned above, by utilizing representative oils in rejuvenation mechanism of RAP, the physical parameters of asphalt binders can be enhanced to cater the diverse requirements of pavement engineering. However, the quantity of oils that is optimal depends on the specific characteristics of aged binder, particularly its stiffness. If too much content of oil is used, it can lead to softening and adversely affect the properties of asphalt pavements. Moreover, the effectiveness and optimum content of distinguished oils in rejuvenating asphalt binder are also impacted by the quality and chemical properties of both the virgin and waste oil.

4 Effect of Oils on Rheological Properties of Asphalt Binders

Rheological properties of asphalt binders comprise mainly rutting, fatigue, and thermal cracking resistances that validate the performance of binders with the accumulation of oils. The rheological characteristics of aged and unaged binders are assessed at different temperatures, loads, and frequencies. Dynamic shear rheometer (DSR) is used to measure high and intermediate-temperature properties such as complex shear modulus (G^*), phase angle (δ), rutting parameter ($G^*/Sin\delta$), and fatigue resistance (G^* . $\sin\delta$) in accordance with Superpave Program [67]. Lowtemperature properties including creep stiffness (s) and *m* value are determined using bending beam rheometer (BBR). Frequency sweep tests are used to study viscoelastic properties at different frequencies and temperatures. ' G^* ' quantifies a material's overall ability to withstand deformation when subjected to shear loading, and δ signifies the delay between the application of shear stress and resulting strain. Table 4 summarizes the rheological properties of aged and RAP binders, utilizing natural and waste oils as rejuvenator, along with virgin binder. The general trend that can be concluded from Table 4 is, with the increase in oil proportion, ' δ ' increases, while 'G*' and G*/Sin δ decrease. As a result of declining in G*/ $\sin \delta$, rutting failure temperature decreases, while reduction in $G^*Sin\delta$ demonstrates increase in fatigue resistance of asphalt binder. Ahmad et al. [24] found that optimum dose of mustard oil to restore aged binder (PG 76-4) to neat binder (PG 64-16) is 10%, while Pradhan et al. [25] observed that only 4% of mahua oil can restore extracted RAP binder (PG-82) to neat binder (PG-70) according to rheological characterization. Likewise, 4% of tung oil [26] had significant positive effect on G^* , δ , and $G^*/Sin\delta$ parameters for stiff binder, though high oil concentration could lead to compromise with rutting parameter. From the other investigations carried out by Navak et al. [30] and Chen et al. [29], it can be deduced that with only 5% dosages of oils, artificially aged (RTFO + PAV) binder and RAP binder can be restored to the control binders (VG 20 and PG 64-22), respectively. Besides, Saboo et al. [68] examined the combined effect of WCO and nano clay (NC) in binder modification and found that optimum content of 2.5% WCO in fusion with 6% NC results in significant improvement in rheological characteristics. An another study [69] made combined use of WCO, WEO and styrene butadiene rubber (SBR) for rejuvenation of extracted RAP binder, recommended 10% and 3% as optimum dosage of waste oils and SBR, respectively. Moreover, Liu et al. [70] recommended an optimum proportion of 4-8%of WEO which gives a good aging behavior for WEOmodified asphalt binders. In another study [33], authors recommended 3-4% of WCO are optimal for reinstating high-temperature rheological parameters of artificially aged (RTFO + PAV) bitumen sample of 80/100 grade. Similar studies revealed an optimum dosage of 10% biooil and 6% of WCO rejuvenated with extracted RAP binder as per rheological testing program [37, 71]. Furthermore,

Table 4 Impact of natural and fresh oils on rheological properties

References	Oil type	Binder	Type of aging	Oil content (wt.%)	$G^* \delta$		G*/Sinô G*Sinô MSCR	nő MSCF	k LAS	(s) m va	<i>m</i> value Findings
[24]	Mustard oil	60/70	Extracted RAP binder	5, 10, 15	S- P	P- S-	∼ L	4 4	d'	P+ P+	The optimum dose of 10% mustard oil is able to revive the PG of aged binder (76-4) to the level of control binder (PG-64-16)
[25]	Mahua oil	PG70	Extracted RAP binder 4, 8, 12 (PG-82)	4, 8, 12	S - S	+ ×	Å	-Ч	4 +	Р-	This investigation shows the effect of mahua oil blended with aged binder and 4% dose of mahua oil was found to be suit- able for rheological characterization
[31]	Olive oil	60/70	(RTFO+PAV)	4	S S	S+ S-	Ч	ď	ď	Р- Ч	The dose (4%) of virgin olive oil had significant effect on G^* , δ , and $G^*/Sin\delta$ parameter
[26]	Tung oil	Pen 70/SBS	(TFOT + PAV)	2, 4, 6, 8	ς Υ	S+ S-	4	4	ď	+4 +4	According to their results, the optimum contents of tung oil can restore $G*/Sin\delta$ of RAP binder but at the same time, it can- not regenerate phase angle values up to the level of target binder
[30]	Pongamia and com- posite castor oil	VG-20	(RTFO+PAV)	5, 10, 15	<u>-</u> Р	- - -	Ŷ	4	ď	십	From their results, it is evident that with 5% dose of both oils, the aged binder was recovered even better than virgin binder
[29]	Cotton seed oil/WCO	PG-64-22	Extracted RAP binder 0, 5, 10	0, 5, 10	S S	S+ S-	P	4	ď	Р- Р-	The 5% dose of WCO is more effective than cotton seed oil in reducing rutting resistance factor and increasing phase angle of stiff binders

Table 4 (Table 4 (continued)										
Reference	References Oil type	Binder	Type of aging	Oil content (wt.%)	$G^* \delta$	$G^*/Sin\delta$	G*/Sinô G*Sinô MSCR LAS	SCR LA	S (s)		m value Findings
[73]	WCO/ WEO	VG-10	Extracted RAP binder (PG-88-XX)	2, 4, 6 5, 7.5, 10	S- S+	Å	S- P+	- + +	4	ď	The optimum dose of 6% and 10% for WCO and WEO, respectively, was substantially effective in restoring oxidized binder
[68]	WCO+NC	VG-30		2.5, 5 2, 4, 6	P- P-	S I	P- P+	+ -	Р-	4	Their investigation reveals that 2.5% dose of WCO produces the suitable results with reference to economic and performance properties
[69]	WO+SBS		Extracted RAP binder 5, 10, 15+1, 2, 3	5, 10, 15+1, 2, 3	S- P-	γ	S- P+	- ⁴	+ 4	+ 4 + 4	In this study, optimum dose of 10% WO (WCO+WEO) was remarkably effective in terms of improving fatigue performance and rutting resistance, respectively, and 3% dose of SBS was noticeable in improv- ing temperature sensitivity and aging resistance
[64]	wco	80/100	Heating stir	1, 2, 3, 4, 5	S- S+	4	Р-	<u>ч</u>	Р	4	Above 65 °C, 8 will resemble to virgin bitumen at 4% dosage of WCO. Similarly, G* reaches to the level of original asphalt at 3% dosage
[47]	WCO	80/100	(RTFO+PAV	0, 2, 4, 6, 8, 10	s-	\$	Р-	+ -	P+	P+	The rejuvenating effect of WCO is more efficient in improv- ing low-temperature properties as compared to high temperature

Table 4 (continued)

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References	Oil type	Binder	Type of aging	Oil content (wt.%)	$G^* \delta$	$G^*/Sin\delta$	G*/Sinô G*Sinô MSCR LAS	MSCR		(s) m value	e Findings
[12]	WCO/ WEO/SP	VG-30	Extracted RAP binder	2, 4, 6	Р- Р-	S.	S_	Р_	P-	P- P-	Optimum dose (6%) of both the rejuvenators (WCO/WEO) has given fruitful results in improving rutting and fatigue parameter
[56]	WCO/ WEO	0//09	Extracted RAP binder	2, 3, 3.5, 4/ 2, 3, 4, 5, 5.5, 6	P- P-	S I	s -	ď	4	P+ P+	WCO and WEO improved the low- temperature cracking properties and 3.5% content of WCO improved the rutting resistance property
[54]	Waste corn oil/ waste soybean oil/	PG-64-22	Extracted RAP binder	2, 4, 6, 8, 10	-ч -ч	Ś	\$ 	ط.	- -	P+ P-	The 6% dose of both waste cooking vegeta- ble oils enhances the <i>G*</i> /Sinð and <i>G*</i> Sinð parameters identi- cal to virgin asphalt and also improves the low-temperature grade
[58]	ovw	Pen 70	(RTFO+PAV)	5, 10, 15, 20	P- P-	ď	4	P+		P+ P+	WVO improves the low-temperature property but rut- ting resistance was compromised with optimal content of 13.4%
[62]	WEVO	40-60/ 60-80/SBS	(RTFO+PAV)	0, 3, 4, 5, 6, 7	-S -	~ 	d.	<u>ط</u>	- -	4	Their results indicate that WEVO is a promising rejuvenator for improving rutting resistance, δ and G^* to the level of virgin binder as compared to SBS-modified asphalt

V. Kumar, P. Aggarwal

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References Oil type	oil type	Binder	Type of aging	Oil content (wt.%)	$G^* \delta$		$G^*Sin\delta$	MSCR	LAS	$G^*/Sin\delta G^*Sin\delta MSCR LAS$ (s) m value Findings	Findings
[02]	WEO	Commercial SBS copolymer	(RTFO+PAV)	0, 4, 8	S- S+	S+ S-	s,	Р_	4	P+ P+	WEO-rejuvenated samples are favorable for improving fatigue behavior and low- temperature proper- ties. At the same con- tent, it is unfavorable for rutting resistance
[72]	Bio-oil	SBS-modified PG 64-22		0, 4, 8, 12, 16	Р- Р- Р-	- Ч	\mathbf{P}_{-}	P_	\mathbf{P}_{-}	P+ P+	Bio-oil gives positive impact on <i>m</i> values but negative effect on <i>s</i> values up to some extent
[37]	Bio-oil	VG-30	Extracted RAP binder 2, 4, 6, 8, 10	2, 4, 6, 8, 10	십	P	4	P+	P+	P+ P+	The optimum dose of 10% of bio-oil gives negative effect on rutting resistance but fatigue and low tem- perature performance may be enhanced using bio-oil
[36]	Bio-oil	40/60		0, 2, 4, 6, 8	S- S+	S+ P-	ď	ď	Ч	P+ P+	Bio-oil addition signifi- cantly enhanced δ and <i>m</i> value, but reduces G^* and <i>s</i> value
S+ increase	S+ increase, S - decrease, P + performed, P - not performed	rmed, P- not performed									

Table 4 (continued)

usage of bio-oils [36, 72] for the modification of binders considerably improved low-temperature properties while compromised high-temperature properties. Other authors [54, 58, 62] also claimed that WVO serve as an excellent rejuvenator to regain the rheological constraints of aged asphalt.

4.1 Rutting Resistance

Moreover, to discuss the effect of oils on fatigue and rutting behavior of the asphalt binder, modern characterization tests like linear amplitude sweep (LAS) and multiple stress creep recovery (MSCR) are being employed [24, 25, 58, 68, 73, 74]. To generate a cumulative strain curve, MSCR test was conducted at two distinct stress levels, i.e., 0.1 and 3.2 kPa. The non-recoverable creep compliance (Jnr) and recovery percentage (R) are two important parameters obtained by MSCR test. These parameters strongly correlated with rutting resistance of modified binders. With increase in load and oil proportion, 'R' decreased while Jnr increased and is maximum for asphalt binder rejuvenated with 15% of mustard oil [24] and 12% of mahua oil [25]. From the outcomes of MSCR test, it is perceived that addition of 15% of mustard oil results into higher Jnr value and lower R as compared to addition of 12% of mahua oil, which indicates addition of 15% mustard oil makes it more prone to rutting or permanent deformation. Ishfaq et al. [73] exhibited that RAP with WEO softens the binder more than RAP with WCO as it has higher Jnr values.

Another study indicated that control binder demonstrated a large amount of cumulative deformation than RAP binder, cumulative deformation of rejuvenated binder increased with increase in WCO content [58]. Thus, it can be inferred that addition of oils improves softness of binder which in turn results in increase of Jnr as well as decrease in rutting resistance. Therefore, it is important to consider the rutting resistance before incorporating various oils into the binder, and caution should be exercised based on the site's climatic conditions.

4.2 Fatigue Resistance

To assess fatigue resistance of binder, (LAS) linear amplitude sweep test is used by researchers [37, 68, 73]. The test is performed in two phases. Initially, a frequency sweep test is carried out to determine ' α ' (an inherent property of the material) at a strain rate of 0.1%, with frequency ranging from 0.2 to 30 Hz. Subsequently, an amplitude sweep test is performed at frequency of 10 Hz, with a gradually increasing load amplitude ranging from 0.1 to 30%. With the increase in amount of mahua oil [25], the fatigue life of rejuvenated binders increases as compared to RAP and control binder, similar results are observed with bio-oils also [37]. The impact of WCO inclusion on fatigue life of RAP binder was also explored by Wang et al. [75] and found that with repeated loading cycles, fatigue resistance improved significantly. Upon reviewing the available literature, it was noticed that very few studies have employed sophisticated characterization methods such as MSCR and LAS tests to investigate the influence of distinguished oils on the rheological characteristics of RAP binders. Based on rheological tests results, it can be concluded that incorporating natural or waste oils into the aged binder indicates an improvement in fatigue resistance.

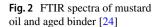
4.3 Thermal Cracking Resistance

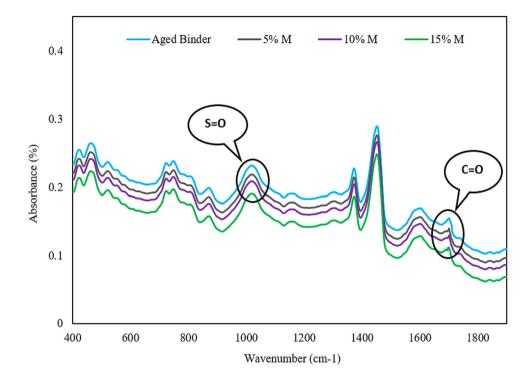
At low temperature, cracking resistance of asphalt pavement is evaluated by two parameters *m* value and *s* value. These properties are determined by BBR, generally performed on PAV-aged binders. The ability of binder to withstand loads over time is reflected by its s value, while the m-value characterizes the changes in asphalt binder's stiffness as loads are applied. Ahmad et al. [24] performed BBR test on aged binder rejuvenated with mustard oil, at different temperatures of -4, -10, -16 and -22 °C. Authors found that at -16 °C temperature, aged binder rejuvenated with 10% and 15% mustard oil, improved low temperature properties, similar to that of virgin binder. Similar improvement in low-temperature properties was observed using tung oil as rejuvenator [15]. Pen 70 and SBS-modified binders could not match the SHRP specifications at -18 °C, however could match the specifications at -12 °C. Other studies with waste oils [56, 58, 70] also showed similar results, where s value decreased and m value increased with increase in temperature and oil proportions, suggesting rejuvenation in improving thermal cracking resistance of aged binders effectively. Although, both parameters m value and s value are influenced by the properties/characteristics of oils used as rejuvenator.

The literature cited above demonstrates that asphalt that has been rejuvenated with natural as well as waste oils exhibits excellent performance at low and medium temperatures. However, its performance in rutting is compromised.

5 Effect of Oils on Chemical Properties of Asphalt Binders

A crucial factor that affects the chemical properties of rejuvenated asphalt binder is acidic value of oil which include proportion of free fatty acids and water content present in oil. As already discussed in Sect. 2.2, natural oils contain higher saturates and as saturates are constituent of maltene, it is anticipated that oils can functionally restore physical and chemical properties of aged binders. However, during the



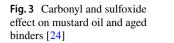


formation of food, oil is continuously heated up to 200 °C due to which the quality of oil degrades and it undergoes various chemical reactions like hydrolysis, oxidation, and polymerization. During these reactions, moisture present in food breaks the oil dual bond and enhances the saturation degree and fatty acid compositions [76]. Azahar et al. [77] compared effect of chemically treated and untreated WCO in aged asphalt binders and found that untreated WCOrejuvenated binders having excessive water content and acidic value exhibited poor results with respect to chemical, physical, and rheological characteristics. In this direction, Zhang et al. [78] outlined that the acidic content of oil has a remarkable impact on the performance characteristics of the rejuvenated binder. As per literature, the properties of rejuvenated asphalt binder are greatly influenced by the acidic value of oil used as rejuvenator. The use of fresh oils or waste oils with lesser acid value in rejuvenating the binder can effectively bring it back to the level of control binder. The limit of 0.4-7 mg KOH/gm is recommended for acidic value of oils to reinstate the properties of aged binder. A high acid value imparts to rise in the viscosity, softening temperature, and colloidal instability of binder. As a consequence, ductility, penetration value, and permanent deformation of the binder are reduced. Therefore, the acid value of selected oil can be considered as a reliable indicator for its effectiveness as rejuvenator. To achieve a higher level of rejuvenation, it is suggested to carry out preliminary chemical treatments aimed at reducing the levels of free fatty acids and water content.

5.1 Functional Groups

To access chemical changes that resulted in the rejuvenation process of asphalt binders with oils, characterization of its functional groups is accomplished by Fourier transform infrared spectroscopy (FTIR). During the aging process of asphalt, its lighter oily components are lost and carbonyl (C=O) and sulfoxide (S=O) groups get modified due to intermolecular interactions between the oxidized components of asphalt binders. Further, the software, which is compatible with FTIR spectroscopy apparatus, can be employed to compute the areas of specific infrared bonds, enabling the determination of the quantity of sulfoxide and carbonyl groups generated throughout the aging process. It can be observed from the analysis of FTIR shown in Fig. 2 that by addition of mustard oil, both sulfoxide (1030 cm^{-1}) and carbonyl peaks (1705 cm^{-1}) get reduced, which implies that mustard oil has capability of decreasing asphaltenes, enhancing non-polar saturates, and restoring colloidal structure of aged binder [24]. Equations (1) and (2) are employed to determine structural indices that depict the level of aging introduced in a bitumen sample in response to an increase in carbonyl and sulfoxide groups.

$$Sulfoxide index = \frac{Area under the peak of sulfoxide groups}{Area under the peaks of ethylene and methyl groups}$$
(1)
$$Carbonyl index = \frac{Area under the peak of carbonyl groups}{Area under the peaks of ethylene and methyl groups}$$
(2)



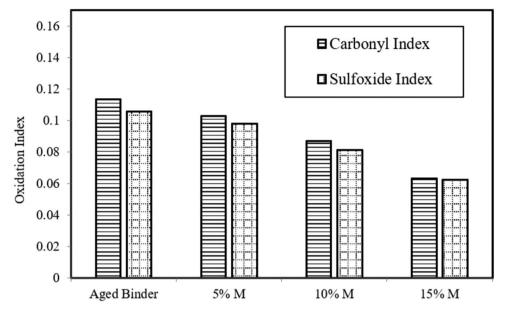


Figure 3 illustrates the sulfoxide index (Is = 0) and carbonyl index $(I_c = 0)$ for both aged binders and binders modified with mustard oil. Similarly, mahua oil (due to the presence of maltene) can also counteract the aging process, as FTIR test performed on rejuvenated RAP binders showed that both $I_{C=O}$ and $I_{S=O}$ indices reduced significantly [25]. Further, the rejuvenation of asphalt binders with sunflower oil induces new peaks at wavenumber 1745 cm⁻¹ and 1163 cm⁻¹ that resides to ester carbonyl and alkyl group, respectively [34]. FTIR results of Pen70 and SBS-modified aged binder with tung oil indicate that sulfoxide and carbonyl indices of the Pen70 could be constructively reduced by adding tung oil; however, such improvement could not be achieved in case of SBS-modified aged asphalt [26]. Besides fresh oils, WO-treated binders also exhibit reduction in carbonyl peak when compared with the RAP binder, which leads to improvement in performance of aged binder [77, 79]. The impact of 4% and 8% WEO on chemical composition and molecular weights of aged binder was examined by Liu et al. [80] who observed that addition of WEO results in reduction in the proportion of large-sized molecules and decrease in occurrence of C=O groups.

Based on the analysis of aforesaid studies, it is evident that an increase in polar compounds (high molecular weight molecules) during asphalt binder aging results in an increase in carbonyl (C=O) peak. However, addition of oil-based ester functional and alkyl groups can decrease the asphaltenes concentration, leading to a decrease in carbonyl (C=O) peak and retarding aging process. However, the carbonyl (C=O) peak cannot be fully restored to the level of virgin binder.

5.2 SARA Components

The complex molecular structure of asphalt binder consists of several hydro-carbons, organic compound, and molecular groups. These molecular groups are characterized by size, polarity, and molecular weight or commonly consists of asphaltenes and maltenes. The maltenes are further divided into aromatics, resins, and saturates. All these molecules collectively are known as SARA in asphalt. Physical, chemical, and rheological properties of binders are function of SARA components in asphalt. As the asphalt binder ages, oxidation and volatization causes decrease in maltenes composition and aromatization increases the asphaltene concentration which leads to disturbance in asphaltene-to-maltene ratio making the binder stiffer. The reason behind this is that the formation of aromatic rings due to aromatization can increase the strength of π - π interactions between them. Consequently, they cause the formation of asphaltene-like layered structures [81]. Hence, it can be said that variation of SARA fractions with aging, causes degradation of physical, chemical, and rheological performance of binder [82]. Zheng et al. discussed the effects of WCO on SARA components of control, artificially aged and rejuvenated bitumen. Treated WCO contains large amount of aromatics and saturates, less concentration of resins, and approximately half amount of asphaltenes in comparison with aged binder [78]. Likewise, in other research works [51, 61, 83, 84], the influence of WCO concentration on SARA components of rejuvenated binders was elaborated; it was deduced that addition of WCO decreases asphaltenes and increases maltenes in aged asphalt that can revive aged binders by considerably infecting the conventional properties. An investigation revealed that rejuvenated binders exhibited carbonyl indices (CI) of 0.54 for fresh cooking oil and 1.13 for WCO. This suggests that the rejuvenation process was more pronounced in fresh cooking oil, as evidenced by its lower C=O bond content, compared to WCO [55]. Alternatively, Li et al. [61] investigated the impact of WEO on the chemical composition of rejuvenated asphalt and found that addition of WEO led to an increase in saturates, resin, and asphaltene contents, while aromatics content decreased. Besides, the relative content of each SARA component determines the colloid structure of asphalt binder. Hence, colloidal index (I_c) is frequently employed to assess alterations in the colloidal structure of reclaimed asphalt. This index can be computed using the following equation:

$$I_{c} = \frac{R+A}{A_{SP}+S}$$
(3)

where *A*, *R*, *S*, and A_{SP} denote the content of aromatics, resins, saturates, and asphaltene in asphalt binder, respectively. Greater improvement is noticed in properties of RAP binder rejuvenated with oils whose colloidal index closely matches to that of original asphalt. Moreover, the introduction of WCO can lead to the restoration of I_c value, attributed to its higher aromatic content in comparison to asphaltenes [46, 85]. Despite using the colloidal index to make predictions, determining the molecular interactions between the binder and rejuvenator's chemical components remains highly complex intricate, and the outcome typically involves only a broad reorganization of various fractions. Moreover, further research is needed in this direction as limited investigation on the impact of fresh oils on SARA constituents in asphalt is available.

5.3 Microscopic Properties

The performance characteristics of asphalt binders are influenced by its microscopic properties. The ongoing methods to estimate microscopic properties of asphalt binders include atomic force microscopy (AFM), scanning electron microscopy (SEM), and transmission electron microscopy (TEM). In the context of existing literature, a considerable number of studies have concentrated on the distinctive microstructural features present on the surface of bitumen. These investigations primarily center around the chemical composition and the mechanisms that lead to the creation of the 'bee-structure.' Furthermore, these studies extensively examine the phase attributes that differentiate between the outer surfaces and internal compositions of bitumen [86-95]. AFM technique is widely used to explore three-dimensional surface structure and morphological nature of asphalt binders. The microstructure image produced by AFM provide information about surface force (adhesion) and surface topography (roughness). In addition, the microstructure image of AFM

consists of three phases catana phase, peri-phase, and para phase [84]. The catana phase is associated with bee-like structure owing to valleys and ridges present in the phase. The rheological properties of asphalt binder are related with bee structure and with the aging process of binder, bee structure increases and rejuvenation process with oil as rejuvenators can reverse it. Lushinga et al. [96] utilized AFM to assess how silicone oil influenced the effectiveness of crumb rubber-modified bitumen. Their findings revealed that with increasing silicone oil content, surface roughness of the modified bitumen decreased. El-Shorbagy et al. [56] using SEM technique compared the configuration of aged and WCO rejuvenated binder and observed that SEM images of rejuvenated WCO binder displayed that even surface as SiO₂ was fully dispersed, which leads to widespread bores of various sizes. WCO has the potential to restore microscopic properties of aged binder to virgin binder. WCO-rejuvenated binder causes depletion in peri and catana phase which enhances the para phase in aged RAP binder, thus redeems the performance of control binder [41]. The microscopic properties of short-term aged binder were restored by rejuvenating binder containing 2% WCO [97]. This restoration occurred through the reduction of binder's bee structure and the filling of intermolecular gaps. Furthermore, while WCO exhibited greater success in revitalizing the asphalt following short-term aging, it was ineffective in rejuvenating the long-term aged binder [98]. In a recent study, Shi et al. [99] found that the micromorphology images obtained through AFM showed a decrease in the quantity of the bee-like structure present in waste soybean-oil-rejuvenated binders when compared to that of aged asphalt. Cavalli et al. [100] implemented various types of bio-based rejuvenators such as a tall-based oil, cashew nut shell-based oil, and vegetable oil into RAP binders. They observed the emergence of a distinct region on the bitumen surface, with the characteristics of this region being influenced by the specific chemical composition of the rejuvenating agent. In addition, Hossain et al. [101] documented that inclusion of a rejuvenator derived from oil brought about changes in both the nanoscale surface structure and the mechanical characteristics of aged bitumen. Despite the existence of some studies utilizing AFM and SEM techniques to examine microscopic characteristics, there is a need for additional assessment of the microscopic behavior of asphalts rejuvenated with fresh oils, specifically.

6 Effect of Oils on Performance Characteristics of Asphalt Mixtures

The mechanical characteristics of asphalt mixtures involves high-, intermediate-, and low-temperature properties. Mostly, high-temperature properties can be assessed by Hamburg wheel track testing (HWWT), dynamic creep test (DCT), and Marshall test. While indirect tensile strength (ITS), resilient modulus $(M_{\rm R})$, and semi-circular bending (SCB) fracture test are used for assessing intermediate-temperature properties. Low-temperature characteristics such as thermal behavior of mixes and moisture susceptibility are evaluated by three-point bending and ITS tests. The rejuvenation or modification of asphalt binders with representative oils make them softer and more susceptible to rutting that induce permanent deformation of road pavement. Recently, Sujit et al. [102] compared the performance of standard asphalt mixtures with mahua-oil-rejuvenated asphalt mixtures having varying RAP content 30-70% in respect of moisture susceptibility, rutting resistance, fatigue resistance, thermal cracking resistance, indirect tensile strength, and resilient modulus. From their study, it can be concluded that asphalt mixture with 60% RAP and 4% of mahua oil can meet the performance properties of asphalt pavement. The wheel tracking test performed to evaluate rutting resistance indicates that asphalt mixtures rejuvenated with mahua oil showed lower rut depth compared to virgin mixes. The rise in both ITS and $M_{\rm R}$ values with an increase in RAP content can be attributed to the presence of a firmer aged binder in the recycled mixes. Thus, it can be inferred that as a rejuvenator, mahua oil has the potential to enable the use of a greater proportion of RAP content in hot in-plant recycling process. Yan et al. [103] compared the feasibility of tung oil and WCO as possible rejuvenators by evaluating the performance of binder and coarse-graded mixtures. It was found that performance properties of asphalt mixtures rejuvenated with tung oil are better than that containing WCO. Overall, it can be inferred that tung oil exhibits superior performance at both high and low temperatures along with better susceptible to moisture. While, another study demonstrates recycled mixtures incorporating WCO demonstrated an extended fatigue lifespan in contrast to recycled mixtures containing tung oil [45]. Bilema et al. [104] found that the incorporation of WFO and crumb rubber (CR) effectively restored the properties of RAP and yielded comparable levels of stiffness, rutting resistance, and moisture damage to that of control mixture. Hence, their combined use can improve the properties of recycled mixtures considerably. From various combinations considered, performance of WEO (7%) with 40% of RAP and WCO (13%) with 50% of RAP was best [105]. When asphalt mixture comprises 50 or 75% RAP, the WEO addition can lead to an augmentation of high-temperature characteristics [106]. According to the literature on dynamic repeated creep load test, it was found asphalt mixtures containing 75% RAP demonstrate improved rutting resistance. However, it is important to be cautious when selecting the optimal dosage of WCO. The study showed that 10% concentration of WCO provides better rutting resistance, which is analogous to control mixture, while 5% and 15% WCO dosages resulted in poorer rutting performance. Moreover, excessive dosage of WCO resulted into decline in fatigue resistance property of bituminous mixes with RAP [107]. Jia et al. [108] observed that addition of RAP, with optimal dosage of WEO in bituminous mix can reduce the number of load cycles in the mixture, while addition of WEO have a rejuvenating effect by minor improvement in fatigue resistance. A comparable investigation confirmed this finding by [109] demonstrating that recycled mixtures comprising 60% RAP exhibited the weakest fatigue resistance, which can be enhanced by the accumulation of WO. Various studies have investigated thermal and fatigue cracking characteristics of asphalt mixtures with different waste oil as rejuvenators but limited studies are available with fresh oils.

6.1 Moisture Susceptibility

Moisture susceptibility of asphalt mixes is commonly evaluated by residual stability (RS) and tensile strength ratio (TSR). Ingress of moisture in bituminous pavement results into loss in holding capacity of binder which in turn separates binder and aggregate. This results into development of potholes as well as other distresses in the pavement structure which hinder traffic flow and compromise safety [110, 111]. In their research, Wen et al. [112] examined impact of incorporating WCO-based bio-oil on moisture susceptibility of bituminous mixtures, and found that all specimens comprising bio-oil met the minimum AASHTO TSR requirement of 80%. The extent to which the addition of WO affects moisture susceptibility is contingent on both the mix design and WO concentration. Contrarily, Majidifard et al. [113] observed that bituminous mixes with control bitumen exhibited highest level of moisture susceptibility, whereas addition of 60% RAP and WCO resulted in a reduction in moisture susceptibility. Similarly, according to Zaumanis et al. [114], WVO-rejuvenated asphalt mixture exhibited poorer resistance to moisture damage compared to base asphalt mixtures. This suggests that using WVO is not effective in restoring moisture damage resistance of aged asphalt mixtures. This could be due to inability to fully rejuvenate the aged asphalt in RAP and presence of fatty acids in WO, which may adversely affect the moisture susceptibility of mixture [45, 115, 116]. However, the mixtures having RAP binders rejuvenated with treated WCO exhibited notably enhanced resistance against moisture-induced deterioration. Treated WCO has lower acidic value while untreated WCO contains greater saturated acids that contribute high acidic components to asphalt binders leading to increased moisture-induced damage in comparison to treated WCO [117]. Majidifard et al. [113] observed that asphalt blends comprising 60% RAP and WCO exhibited greater vulnerability to moisture than the control mixture. It should be noted

that effects of oils as rejuvenating agents on asphalt mixture properties depend on type, dosage of rejuvenating agent, and specific characteristics of asphalt mixtures. Therefore, careful selection and dosage of the rejuvenating agent are essential to achieve desired results.

7 Environmental and Economic Perspective

In addition to its financial advantages, RAP provides various environmental benefits also as utilization of RAP in pavement construction is a beneficial approach for conserving energy and decreasing CO₂ emissions. In their study, Aurangzeb et al. [118] carried out a life cycle assessment on HMA containing high amounts of RAP, and demonstrated that incorporating RAP in HMA mixtures resulted in a 28% reduction in greenhouse gas emissions and energy consumption as compared to conventional HMA. According to Chiu et al. [119], utilization of RAP shall reduce the requirement of fresh bitumen which in turn shall lower burden on environment. Life cycle analyses indicate that using RAP results in a decrease in global warming potential, energy preservation, and dangerous waste production [120]. In addition, many fresh or virgin oils which are unsuitable for consumption (due to its high content of erucic acid, which can cause heart disease and elevate blood cholesterol levels in humans) may be used as potential rejuvenators with RAP mixes. Oils produced in excess of demand/consumption can be explored as rejuvenators. Disposal of waste oils is not only an ecological challenge but also a potential threat to human health [80, 121, 122]. According to a recent study, a single liter of WEO has the potential to contaminate one million liters of freshwater, posing a threat to both plant and animal life [123]. In their study, Veeraragavan et al. [124] discovered that utilizing a reusable asphalt mixes consisting of 50% RAP and WVO can result in cost savings ranging from 34 to 40% as compared to HMA. In another research, Samieadel et al. [125] demonstrated that asphalt binder modified using 10% bio-oil, that originated from swine manure, can reduce CO₂ CH₄ and NO₂ gas emissions, resulting in a lower GWPI (global warming potential index).

Therefore, from the literature, it can be inferred that usage of RAP and rejuvenation of RAP mixes with fresh as well as waste oils have environmental and economic benefits.

8 Conclusion

The focus of this review paper is to examine different facts of utilizing natural and waste oils for rejuvenating aged asphalt binders and mixtures for revival of their original properties. From the literature review conducted in this study, the various conclusions can be withdrawn:

- (1) The basic physical properties of asphalt binder are greatly influenced by addition of fresh as well as waste oils, particularly the lighter oil components. Increasing oil concentration resulted into decrease in viscosity of bitumen, leading to high penetration, greater ductility, and decrease in softening point. Thus, it can be deduced that various oils have potential to restore fundamental physical properties of aged binder to the level of control binder.
- (2) Rheological assessment of aged RAP bitumen rejuvenated with various natural and waste oils indicates a decrease in overall stiffness or hardness. With an increase in oil dosage, less-viscous oil component leads to reduction in creep stiffness (*s*) and *G**, while *m* value and δ exhibit enhancements. Therefore, addition of oils in asphalt binder leads to lowering rutting resistance and enhancing thermal cracking and fatigue resistance of bitumen.
- (3) Effectiveness of rejuvenation is greatly influenced by the quality of oils, including its water content, acidic value, and overall condition. As oil degrades after its usage, water content and acidic value tend to increase, resulting in declination of rejuvenation potential. Certain studies have demonstrated that pre-treated waste oils can mitigate adverse effects of higher water content and acid value, thus improving its chemical and rheological performance as rejuvenator.
- (4) Findings of oxidation analysis using FTIR indicate that aging causes reduction in aromatics component present in asphalt due to crosslinking of polar functional groups within asphalt molecules. In addition, it has been noted that intensity of C=O and S=O bond increases following asphalt aging. As ester group is the distinctive peak of oils, its inclusion will reduce the C=O and S=O of asphalt binders. It suggests that representative oils can be a viable rejuvenator for restoring properties of aged binder.
- (5) Use of oils as rejuvenator can restore the thermal cracking properties of recycled mixes to their original levels; however, moisture susceptibility, high temperature performance, and fatigue resistance could not be restored fully. Moreover, available studies on performance characteristics of bituminous mixtures for natural oils are limited.
- (6) The AFM analysis of rejuvenated asphalt binder shows that use of oil as rejuvenator in asphalt binder improves nanoscale surface topography, adhesion, and reduces nanoscale microcracks, enhancing binder durability.

This review article highlights the potency of using natural and waste oils as rejuvenator to enhance the performance of both asphalt binder and mixtures. Numerous studies have demonstrated encouraging outcomes indicating enhancements in rheological, chemical, physical, and microscopic properties of asphalt binder and mixtures. Nonetheless, present research still has certain shortcomings discussed as follows:

The appropriate amount of rejuvenator and RAP content to be used in field applications is still a topic of interest. Limited study is available for chemical characterization of RAP binders with fresh oils specifically.

There is a scope of research in field performance evaluation of bituminous mixtures that have been rejuvenated with oils.

Life cycle analysis for environmental effects may be carried out as future research.

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Data Availability The authors declare that the data supporting the findings of this study are available within the paper.

Declarations

Conflict of Interest The authors states that they do not possess any conflicting interest.

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