

Review

Waste Cooking Oil as a Sustainable Bio Modifier for Asphalt Modification: A Review

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Abstract: The rising demand for non-renewable resources such as asphalt binder is a significant issue in the pavement industry. Flexible pavement consumes a significant amount of asphalt binder, which has become a major issue in terms of environmental sustainability and from an economics viewpoint. Hence, researchers strive to find other alternatives to solve these problems, to enhance the performance and lifespan of flexible pavement. Biomass-based bio-oil, such as waste cooking oil (WCO), as a modifier has illustrated favorable effects for asphalt binder and mixture. However, in the pavement industry, its adoption as a modifier is still in an empirical stage. Hence, this paper aimed to give an overview by analyzing literature in-depth to reveal the potential of WCO as a modifier in the pavement industry. The low- and intermediate-temperature performance of the WCO-modified asphalt binder are superior. However, it compromises physical properties and high-temperature performance. Hence, it can be improved by controlling the quality of WCO or by further modification by additives such as ground tire rubber (GTR) and waste plastic. This paper also attempts to review available and potential physical and chemical technologies to minimize the negative effects of free fatty acid (FFA) and water content of WCO on modified asphalt binder properties. For WCO-modified asphalt mixture, the overall performance depends on the dose, quality of WCO, and type of additive added in the WCO-modified binder. Finally, future recommendations are provided to broaden the scope of WCO as a modifier in the forthcoming sustainable pavement industry.

Keywords: waste cooking oil; transesterification; sustainability; free fatty acid; asphalt binder; rheological properties



Citation: Elahi, Z.; Mohd Jakarni, F.; Muniandy, R.; Hassim, S.; Ab Razak, M.S.; Ansari, A.H.; Ben Zair, M.M. Waste Cooking Oil as a Sustainable Bio Modifier for Asphalt Modification: A Review. *Sustainability* **2021**, *13*, 11506. <https://doi.org/10.3390/su132011506>

Academic Editor: Hamzeh F. Haghshenas

Received: 20 September 2021
Accepted: 15 October 2021
Published: 18 October 2021

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1. Introduction

Amongst many global issues, the rapid growth of cities and the increasing number of vehicles have dramatically enhanced the demand for pavement construction and maintenance [1]. Around 95% of the world's roads are made of flexible pavement, and asphalt binder serves as a conventional material for pavement construction [2]. Asphalt binder is a byproduct of crude oil, which is well known as a non-renewable resource. A dramatic increase in the price of petroleum-based binders, due to the depletion of petroleum reserves, forces road authorities to explore alternative materials. Currently, the global practice is moving towards a sustainable, economical, and green environment. The scientific community is interested in optimizing bio resources for renewability, sustainability, and local production in the pavement industry, due to reduced costs and lower energy consumption.

Biomass is the primary source of bio material, whereas biomass refers to living things that store solar energy, such as plants, animals, and microorganisms [3]. Among

various forms of biomaterial, bio-oil is a widely accessible material and is categorized as waste oil from the forest/agriculture industry, animal, and residual. Hence, modifying the asphalt binder with bio-oil significantly minimizes the issues regarding non-renewability and fluctuation of binder cost [4]. The compatibility and likelihood of bio-oil modified asphalt are somewhat satisfactory, and its quality differs with source and the content [5]. However, the addition of agricultural waste oil (5–10 wt.%) improves the bio asphalt viscosity and low-temperature properties, whereas there is marginal improvement in the high-temperature properties [6,7]. Similarly, the waste wood oil-based asphalt mixture improved the anti-fatigue and crack resistance properties [8]. Conversely, animal waste oil is commonly utilized less than 10 wt.% by asphalt to achieve better performance [9,10]. The addition of animal waste based bio-oil reduces the viscosity and stability of asphalt at higher temperatures and increases low-temperature crack resistance [1]. Yet the agriculture and animal-based oils are subjected to time-consuming and expensive refining procedures, such as pyrolysis and hydrothermal liquefaction [3,11]. In this way, the current implementation of bio-oil to modify asphalt binder is mainly focused on plant-based oil such as waste cooking oil/waste vegetable oil, as these soft oils are a ready substrate with lower production cost [3,12–14].

Waste cooking oil (WCO) is considered a potential bio-based waste material because it can lead to multiple topologies of the product [15]. WCO is generated after frying activities, and the rising population has increased its quantity due to the increased demand for food. WCO is related to the vegetable oil family and mainly arises from the kitchen and food industry. The global consumption of vegetable oil in 2021 has reached 209.14 million metric tons, and each year consumption has increased by 2% (Figure 1). In contrast, a significant problem regarding the proper disposal and collection of WCO has still gone unsolved for a long time. The lack of awareness, facilities, and improper disposal of WCO are leading to environmental, ecological, and municipal problems [16,17]. Hence, by heavily consuming oil and fats, many countries face sewer blockage problems associated with an increased annual cost to remove blockage [18].

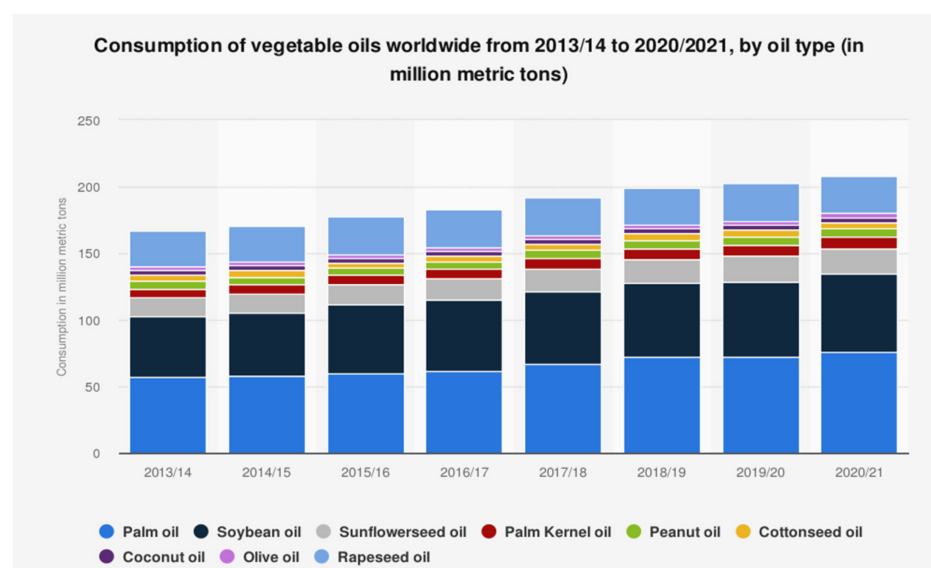


Figure 1. Global vegetable oil consumption worldwide by oil type [19].

One approach to solve this issue is to adopt a circular economic strategy for WCO by its reconversion, which can reduce environmental impacts while increasing economic efficiency [20]. Recycling WCO provides a renewable feedstock for biofuels (i.e., biodiesel) and other bio-based goods (e.g., soap and animal feed) due to its chemical composition [21]. Although, it also helps to minimize pollution by reducing greenhouse gases (GHG) produced by inappropriate handling [22]. A typical application of WCO as a modifier in the

pavement industry dates back to 2013, since many studies were conducted in laboratories to examine WCO-modified binder performance [13,23]. Based on previous research, it can be concluded that WCO is a potential waste material due to its consistent performance, specifically at low and intermediate temperatures [19,24,25]. On the other hand, high-temperature properties of WCO-modified binder are still compromised, as WCO can soften the binder, which causes higher penetration and lower softening. Thus, it is believed that soft binders cannot withstand high-temperature exposure and lead to poor rutting resistance [12,26–28]. The quality of WCO, which includes acid value as a quality assessment parameter, influenced the high-temperature performance of the modified asphalt binder. The high acid value is associated with free fatty acid (FFA) and water content in WCO. However, the undesired characteristic of a high acid value limits the application of WCO as an asphalt binder modifier.

Azahar et al. [25] investigated the effect of acid value and water content on asphalt binder modification and revealed that chemically treated WCO with lower acid values and water content showed better high-temperature performance. Although, the physical performance of treated WCO was still below the control binder and compromised. To overcome the deficiencies encountered by the WCO-modified binder, researchers recently utilized additional modifiers such as waste polymers, palm oil fuel ash (POFA), and nano clay (NC). It was concluded that WCO-modified binder with other modifiers could withstand hot climate regions. Moreover, a higher percentage of WCO can be used along with additional modifiers to enhance the performance while ensuring a more sustainable environment [29–31].

Therefore, this review paper focuses on WCO-modified asphalt and aims to enlighten possible solutions for the cited problems by encouraging sustainability. Thus, an overview of technology development over the last few decades has been summarized and discussed. The production and treatment methods of WCO and the physical, rheological, and mixture performance of untreated, chemically treated WCO, and WCO/additional modifier binder are also reviewed.

2. Production of Waste Cooking Oil

The vegetable oils market is dominated by palm and soybean oils, which account for 36% and 28% of the market. Other vegetable oil fractions include sunflower (9.09%), rape (13.2%), peanut (2.95%), corn (2.1%), coconut (1.75%), olive (1.48%), and sesame (1.1%) [17]. Amongst all continents, Asia is on top of the list for highest production of vegetable oil. Domestically, edible oil is used for frying purposes, whereas food processing industries and fast food companies largely use oil [32]. Frying is a process in which food is fried at a temperature between 150–200 °C in the presence of moisture, anti-oxidants, and pro-oxidants [33]. During frying, various reactions occur, such as hydrolysis, polymerization, isomerization, and oil decomposition. After a frying operation, the leftover oil is considered waste material and named waste cooking oil or used cooking oil [34]. The rapidly growing world's population has dramatically increased the food demand that ultimately affects the production of WCO. Recent studies estimate that the generation of waste cooking oil is almost 20–32% of total edible oil consumption [17,35]. China and India are the most populated countries and produce the most WCO, around 5.6 million tons and 1.135 million tons per year. Canada, Denmark, Spain, Italy, Japan, South Korea, Malaysia, and the United Kingdom have average generations, ranging from 0.1–0.5 million tons per year [36]. The other remaining countries produce no more than 0.1 million tons per year, and as per capita production of WCO, South Korea is on top of the list, followed by EU countries [17].

3. Physical and Chemical Properties of Waste Cooking Oil

WCO is a thick, dark brown or black liquid at room temperature and is considered an undesirable material, as it is composed of complex organic compounds such as acids, ester, and aldehydes [19]. The composition and variation in organic compounds depend on the oil source and degradation reaction during frying activity [37]. The degradation reaction

increases the density, viscosity, specific heat, foaming, and total polar material value of oil. Variation in viscosity and density of WCO varies due to the formation of other complex organic compounds such as glycerides, dimeric, and polymeric acids [38]. Table 1 shows the physical properties of WCO.

Table 1. Physical properties of WCO.

Reference	Viscosity @ 40 °C (m ² /s)	Density (g/cm ³)	Water Content (%)
[39]	4.2×10^{-6}	0.91	1.9
[40]	36.4×10^{-6}	0.924	0.42
[38]	144.5×10^{-6} – 179.9×10^{-6}	0.932–0.936	–
[41]	30.5×10^{-6} – 31.77×10^{-6}	0.920	–
[42]	64.1×10^{-6}	0.933	–
[12]	4.9×10^{-6}	0.9013	0.140

The major components of edible oil are triacylglycerol with fatty acids, with variable degrees of unsaturation, ranging from C16 to C18 carbon chain. Triglycerides are made by linking glycerol to three fatty acids, whereas all fatty acids contain an even number carbon chain [35]. Moreover, natural edible oil is also composed of other minor components such as free fatty acids (FFA), acylglycerols, carotenoids, tocopherols, tocotrienols, sterols, phosphatides, hydrocarbons, and trace metals [43]. For the preparation of food, deep-frying is one of the traditional and widely used methods. While frying at high temperatures such as 150–200 °C, food reacts to the heated oil, air, and moisture present in the food. Thus, hydrolysis, oxidation, and polymerization are common reactions and produce volatile and non-volatile compounds (Table 2) [37]. Therefore, it is concluded that the chemical and physical changes during deep-frying effect the overall composition of waste cooking oil from that of edible oil, as shown in Figure 2.

Table 2. Variation in the chemical composition of oil after the frying process.

Chemical Reaction	Reason of Reaction	Variation in Chemical Composition
Hydrolysis	Water, steam, oxygen	Production of di-mono-acylglycerols, glycerol, and free fatty acids
Oxidation	Surrounding air or oxygen	Production of CO, CO ₂ , and dimeric compounds
Polymerization	Heating oil at high-temperature range 260–340 °C	Formation of dimers including oligomers that increase the molecular weight of oil

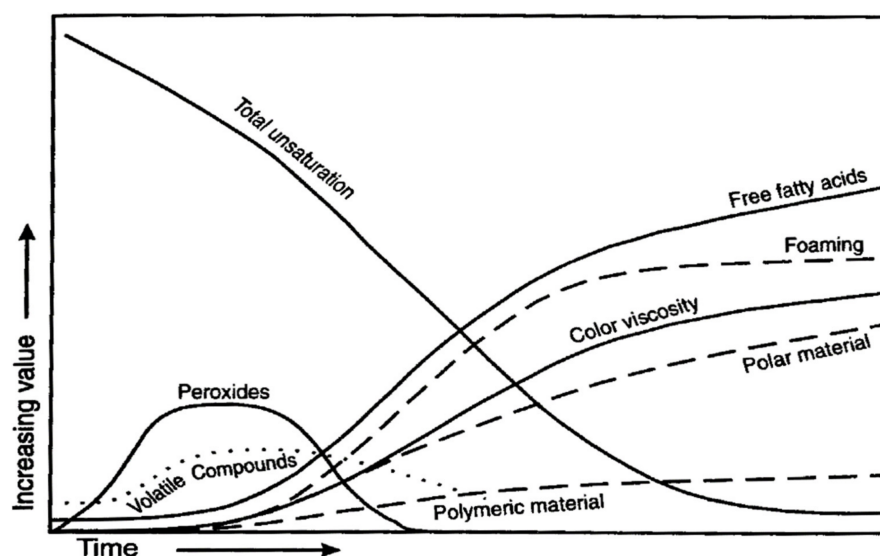


Figure 2. Physical and chemical changes in oil during deep-frying [37].

4. Free Fatty Acid in Waste Cooking Oil

Edible oil undergoes chemical and physical changes during frying activities, and the quality of the oil degrades. The steam generation during frying reacts with the oxygen and water in the food, initiates the hydrolysis process, and produces the FFA. The concentration of the FFA varies and depends on diverse conditions, such as heating temperature, moisture content, and storage time. Thus, during the additional reaction time the triglyceride bonds break down and produce FFA, including glycerol, diglyceride, and monoglycerides, as shown in Figure 3. Furthermore, at a temperature above 150 °C, the glycerol accelerates the hydrolysis process, leading to further FFA production [44]. As the degradation process continues, the production of FFA accelerates, which ultimately decreases the unsaturation level of the cooking oil. Hence, the oil becomes no longer suitable for further frying and needs to be discarded as waste material [45]. The WCO quality can be monitored by the acid value parameter, which identifies the concentration of FFA and the amount of water content [19,46]. However, researchers have utilized the gas chromatography–mass spectrometry (GC–MS) technique to analyze the concentration of FFA in WCO. The acidic compounds of WCO are categorized as saturated, monounsaturated, and polyunsaturated compounds. The chemical composition of WCO studied by Azahar et al. [47] indicated that palmitic acid, stearic acid, and meristic acid are examples of saturated fatty acids, whereas monounsaturated fatty acids contain oleic acid and cis-11-eicosenoic acid. Table 3 indicates that the main components of WCO are oleic acid, palmitic acid, and linoleic acid, which account for 43.67%, 38.35%, and 11.39%, respectively [19,24].

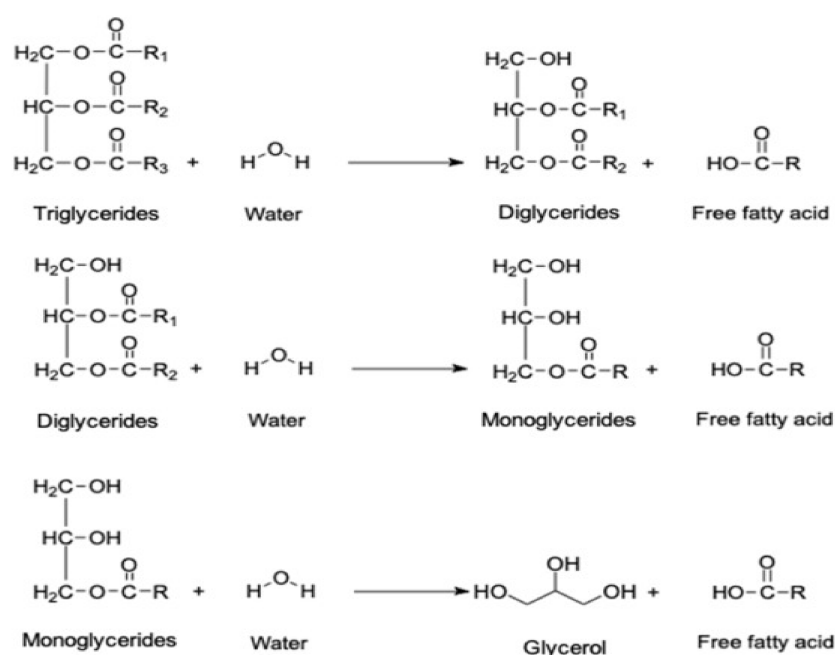


Figure 3. Chemical compositions of WCO [19,24,47].

Table 3. Hydrolysis of triglycerides.

Formulation of Fatty Acids	Type of Free Fatty Acid	Possible % in Waste Cooking Oil (WCO)	Type of Saturation
C18:1 (Cis 9)	Oleic Acid	43.67	Unsaturated
C16:0	Palmitic acid	38.35	Saturated
C18:2 (Cis)	Linoleic acid	11.39	Unsaturated
C18:0	Stearic acid	4.33	Saturated
C14:0	Myristic acid	1.03	Saturated
C18:3 alpha	γ-Linolenic acid	0.37	Unsaturated
C12:0	Lauric acid	0.34	Saturated
C18:2 t	Linolenic acid	0.29	Unsaturated
C20:1	Cis-11-Eicosenoic acid	0.16	Unsaturated
C21:0	Heneicosanoic acid	0.08	Saturated
	TOTAL	100	

5. Effect of Acid Value and Water Content on Asphalt Binder Performance

The FFA composition of various waste oils from different sources is not similar to each other. It depends on food type, oil type, oil quality, duration, temperature while frying, and the number of times the oil was used [19]. The presence of FFA and water content weakens the adhesion bond between asphalt binder and WCO. Furthermore, it decreases the rheological characteristics of the binder, which has a negative impact on the performance of the pavement mixture. Zhang et al. [48] reported that the acid values significantly impact the asphalt binder properties. Similarly, the effect of acid value and water content on asphalt performance was investigated by Azahar et al. [23] and revealed that WCO-modified binders with lower acid values and moisture content enhanced the performance in terms of rheological and chemical properties. However, the higher the acid value interchange, the more the performance improved and behaved otherwise. In another study, Azahar et al. [25] evaluated the effects of untreated and chemically treated WCO-modified binder and concluded there was improvement in physical, rheological, and chemical properties of the treated WCO with a lower acid value and higher water content.

Table 3 indicates the chemical composition of WCO; the data present that palmitic acid, oleic acid, and linoleic acids occupy the highest percentage of acid concentrations compared to other acids. In binder modification by WCO, these chemical contents play a fundamental role. These three acids indicated the water affinity of WCO, as the chemical structure exhibits both polar and non-polar ends [49]. It has been reported that oleic acid and palmitic acid can easily absorb siliceous stones such as quartz and granite in dry conditions, whereas linoleic acid is a polyunsaturated omega-6 fatty acid and is insoluble in water [50]. These acids are quickly replaced by water and thus give rise to moisture susceptibility in WCO-modified binder, leading to premature pavement failure [46].

To conclude, the acid value is an excellent indicator of the modification efficiency of WCO. The lower acid value and water content contribute toward enhancing the properties of WCO-modified binder. Therefore, to achieve better performance, it is recommended to perform a preliminary chemical or physical treatment to reduce FFA and water content from WCO.

6. Available Technologies to Minimize FFA from Waste Cooking Oil

The multiple numbers of FFA production indicate a higher acid value of WCO. Besides the changes in the waste oil's quality caused by FFA, various chemical reactions occur between food and the oil's protein and amino acid. In return, these leached components can increase phosphorus, sulfur, and nitrogen compounds in WCO. The generation of these degrading compounds is mainly responsible for the darkening and odor changes in oil that reduce cooking oil's safety and nutritional value. Moreover, prolonged storage practices in the presence of moisture also play a significant role in further degradation of WCO's chemical composition fostered by hydrolysis [51]. The degradation products, including FFA, have harmful effects on human health and the environment [52]. Despite the vast range of degradation products detected in WCO, they only make up a tiny proportion of triglycerides. Therefore, the removal of FFA must be prioritized to reduce the acid value of WCO. In many industrial processes, WCO can be used as a raw material such as bio-lubricant, in fuel production, or as an asphalt rejuvenator or modifier [15]. The highly complex nature of WCO limits its possible implementation in any field of study. In this case, pretreatment must be ensured if WCO's byproducts are intended to be used for any specified purpose [35]. The following section explains available methods and technologies that need to be implemented before utilizing WCO in the pavement industry. Processes are categorized as physical and chemical treatment of WCO to reduce solid impurities, FFA removal, and moisture elimination.

6.1. Technologies for Treating Waste Cooking Oil Physically

The aim of the physical treatment of WCO is basically to remove undesirable components to obtain an improved quality of regenerated oil.

6.1.1. Filtration and Centrifugation

The removal of fine particulates and suspended particles is the initial stage in WCO's treatment. These are small food residue particles (such as skin, bones) or other paper or plastic materials. Based on frying and collection activities, WCO contains solid impurities of almost 10–20% by volume. Mainly, simple filtration and centrifugation are two methods commonly used to remove solid impurities that enable the reduction in obstruction for further treatment of the oil. The filtration process involves fabric net bags or steel strainers with 150–200 μm openings to filter granular particles. Afterwards, a fine filtration by 70–100 μm steel meshes is generally carried out, followed by 5 μm mesh for further removal of tinny particulates. Regardless of solid impurities separation, the centrifugation method can also be used to eliminate water content from WCO.

6.1.2. Distillation

The simplicity of distillation deserves to be described, as it is the most used physical method. The method aims to reduce volatile compounds and water content by taking advantage of the fact that FFA has higher volatility than triglycerides. This process is carried out by simply heating filtered WCO in boiling kettles under vacuum conditions for about ≤ 90 min [35]. Table 4 summarizes key parameters for acidity and water reduction by means of the distillation process. Thus, this process purifies WCO of volatile compounds and water content up to the maximum level. Besides this, another consequence is eliminating other volatile compounds such as ketones, aldehydes, sulfur, and nitrogen. The removal of these undesirable compounds by distillation further improves the quality of WCO [35].

Table 4. Water content and acidity value reduction by distillation process.

Reference	Temperature ($^{\circ}\text{C}$)	Reduction in Water Content (%)	Reduction in FFA (%)
[53]	220	From 1.15% to 0.062%	From 29% to 2%
[52]	200–280	From 5% to 0.1%	Less than 0.5%

6.1.3. Membrane Separation

Over the last few years, membrane technology in WCO processing has grown in popularity due to it having less than 50% of the energy consumption of the conventional filtration process [54]. This process involves a semipermeable membrane under pressure gradient, which retains undesirable particles while allowing specific components based on a difference in permeability value. The difference could be between sizes, shapes, molecular weight, and physiochemical interaction with membrane materials [55]. Commonly used materials for membrane separation include cellulose polymers, ceramic material, clays, nano/ultra-filtration membranes, and metal screens [56]. The FFA retained up to 35–40% by ultrafilter membrane, whereas maximum triglycerides permeated through the membrane compared to other polar compounds [57]. Despite the high performing capability of conventional filters, synthetic fibers of polyester and nylon have shown about 93–98% removal of FFA [58]. Similar to the distillation process, the initial purpose of membrane separations was to retain the water content of WCO. Thereby, a reduction in FFA might be considered an additional benefit of such a method [54].

6.2. Technologies for Treating Waste Cooking Oil Chemically

The chemical transformation aims to take advantage of chemical functional groups present in WCO to synthesize high-value products. Consequently, alcohol is added to WCO, and a catalyst is employed to run the reaction fast. Based on the catalyst, the transformation reaction can be categorized as base-catalyzed transesterification or acid-catalyzed esterification.

6.2.1. Base-Catalyzed Transesterification

Transesterification is also known as alcoholysis, and it is the best conventional method for reducing the acid value of WCO by converting the triglycerides of an oil into ester by

direct reaction with alcohols. A variety of alcohols can be used, such as methanol, ethanol, butanol, and propanol. Methanol is a more feasible option due to its wide availability, low cost, and quick reaction with triglycerides [59]. Generally, a 3:1 molar ratio of alcohol and oil is needed in the transesterification reaction to complete the reaction (Figure 4). The kinetic analysis of the transesterification process is highly challenging, since the reaction is under an equilibrium process. Alcohol must be present in excess in the reaction mixture to shift the reaction towards the formation of the products [60]. However, to accelerate the transesterification reaction, commonly used catalysts are sodium hydroxide (NaOH) and potassium hydroxide (KOH). These catalysts are highly demanded for several reasons: cheap price, wide availability, and maximum yield at minimal time duration [61]. KOH catalyst reacts faster than NaOH due to strong bonding with alcohol. Tomasevic and Siler-Marinkovic [38] investigated the effect of KOH and NaOH as a catalyst; the result concluded that higher conversion of triglycerides was obtained at a 6:1 molar ratio of methanol to oil, a 90 min reaction time, 1% KOH, and 25 °C temperature. On the other hand, ester covers a wide range of end products and can be utilized in different applications, such as biodiesel production, asphalt industry, detergent, and cosmetics [25,41].

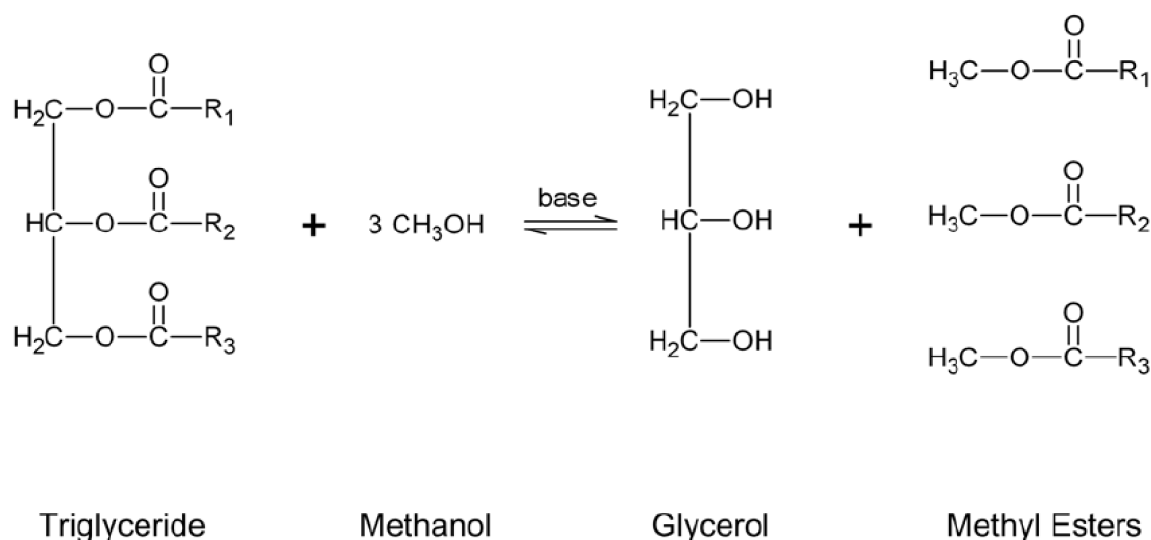


Figure 4. Base-catalyzed transesterification [62].

However, the ultimate maximum ester application of the base catalyst in transesterification has certain limitations, as this process is susceptible to water content and FFA presence in WCO. A higher percentage of FFA reacts with the catalyst, whereas water content partially converts the reaction to saponification, and excessive soap may foam a semi-solid mixture at room temperature, which creates problems during the separation process [60,63]. Thus, the base catalyst cannot tolerate higher FFA content in WCO, and the desirable range is from less than 0.5 to 1%, as shown in Table 5.

Table 5. Desirable ranges of FFA.

Author/Reference	Recommended FFA (%)
[59]	Less than 1
[64]	Less than 0.5
[65]	Less than 0.5
[66]	Less than 2
[67]	Less than 2
[25]	Less than 1

6.2.2. Acid-Catalyzed Esterification

The restriction of base-catalyzed transesterification is its sensitivity to the purity of reactants, particularly with water and FFA. Hence, acid catalysts are proposed to have better results for waste oil with FFA greater than 1% [68]. Sulfuric acid (H_2SO_4) and hydrochloric acid (HCL) are the most frequently used catalysts for acid-catalyzed esterification due to their fast reactivity, low cost, and better stability concerning impurities [35,63]. This process is generally carried out by direct mixing of acidified alcohol with WCO to convert FFA into an ester as a reaction, as shown in Figure 5. In a study of acid-catalyzed transesterification, Wang et al. [69] used WCO with an acid value of 75.92 ± 0.04 mg KOH/g. The satisfactory conversion was carried out when the acid value was reduced to 2.2 mg KOH/g at a 16:1 molar ratio of alcohol and oil and a reaction time of 10 h at 95 °C temperature. However, acid-catalyzation is preferable for a higher content of FFA in waste oil. Although, this process requires a longer reaction time and a higher amount of alcohol and catalyst than base-catalyzed transesterification.

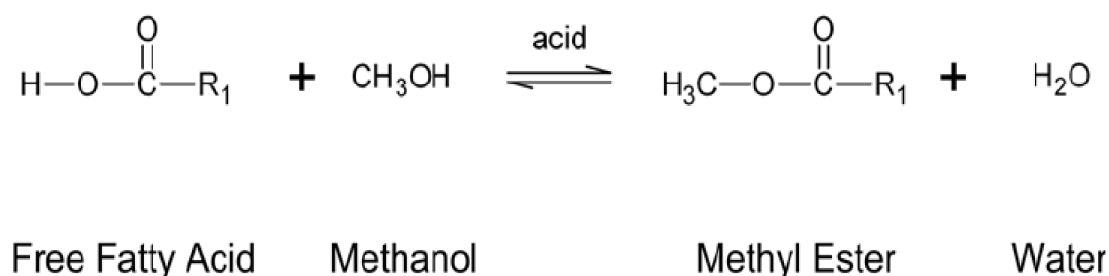


Figure 5. Acid-catalyzed transesterification [62].

6.2.3. Microwave Irradiation Transesterification

In recent years, many researchers have been interested in electromagnetic waves utilization. Hence, microwaves are one example of accelerating chemical reactions, such as transesterification/esterification [70]. Microwaves can effectively transfer heat to the sample, and this energy completes the reaction. However, in conventional processes, heat is not distributed uniformly, resulting in more energy and time-consumption. Thus, the heat transfer mechanism is more effective than traditional heating [71]. Further advantages of microwave irradiation for treating waste oil include a lower alcohol to oil molar ratio, being environmentally friendly, water content removal, and an immediate decrease in FFA [63,72]. Microwave transesterification of WCO with a high FFA content indicated additional benefits in the separation and purification process between treated oil and residues compared to the conventional method [73]. Transesterification of WCO by sodium hydroxide as the catalyst (NaOH) was carried out in a domestic microwave (800 W) under varying parameters, such as molar to ethanol ratio and reaction time. The result indicated that the conversion time of FFA to reduce the acid value of the oil by microwaves is 10 times shorter than the conventional method [71].

7. Available Technologies to Minimize Water Content from Waste Cooking Oil

The degradation in WCO during storage and processing can be minimized by keeping moisture content as low as possible. Moreover, as previously stated, water must be removed to prevent it from effecting subsequent treatment, such as transesterification. For example, in typical base-catalyzed transesterification, the moisture content in WCO should be less than 500 ppm for effective reduction in acid value [74]. Hydrolysis reactions of triglycerides in WCO are caused by water, as shown in Figure 6.

Water from WCO can be removed by different methods, some of which have previously been discussed. Among various methods, water can be extracted by membrane separation, distillation, or acid-catalyzed transesterification. A few of the materials used to retain FFA (such as ceramic powder and clays) can absorb a significant amount of water present in

WCO physically. Moreover, treatment with silica gel, magnesium sulphate, and anhydrous sodium has been evaluated for water removal in WCO [35]. Table 6 lists some of the operating parameter procedures that have been assessed for water removal from WCO.

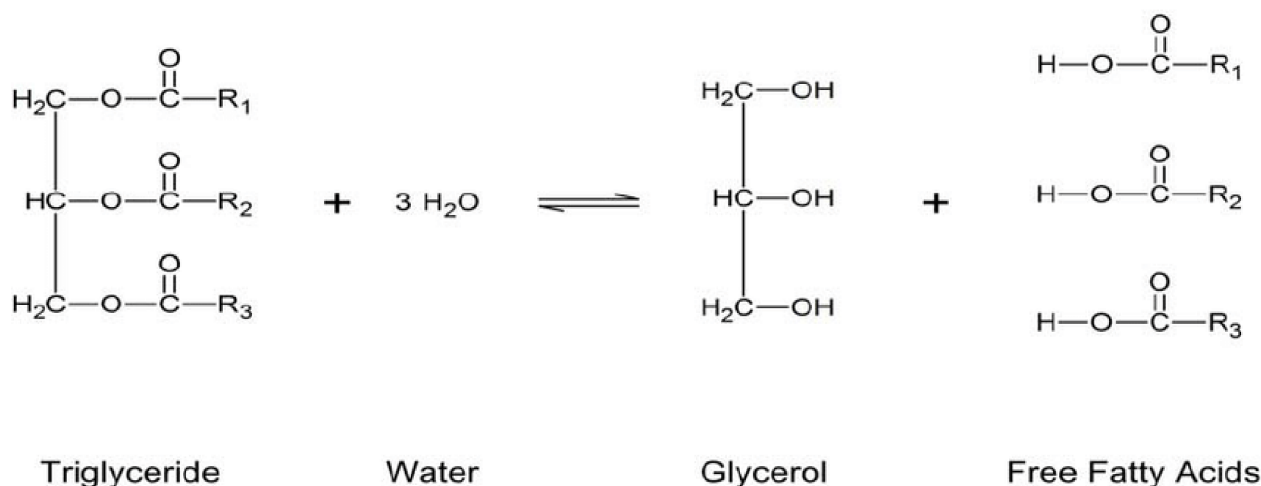


Figure 6. Hydrolysis of triglycerides [62].

Table 6. Operating parameters for water removal from WCO.

Operating Parameters	[75]	[76]	[77]
Solvent	Anhydrous sodium sulphate	Magnesium sulphate	Silica gel
Filtration type	Vacuum	Vacuum	Vacuum
Pre-moisture	—	—	0.7%
Post-moisture	—	0.07–0.13%	0.2 ± 0.1%

Moreover, moisture from WCO can be removed by evaporation technique at high temperatures. Hence, the drawback of this method is more time and energy consumption as water molecules take enough time to liberate from WCO [74]. Alternatively, microwaves can reduce time and energy consumption during the reduction of water from WCO. Many studies have used different separation models for oil and water separation, such as cake separation, deep bed separation, and full and intermediate blocking models. However, their application is limited at a large scale due to their sensitivity to mechanical and thermal stresses [70].

8. Application of Waste Cooking Oil as a Bio-Asphalt Binder in Pavement Industry

A typical approach to prepare a bio-asphalt binder is simply blending biomaterial (such as WCO) with asphalt. Bio-asphalt binder is further classified into three categories based on oil content in the binder (Table 7). However, 100% replacement of conventional binder with a bio-asphalt binder is not suggestible as the saturates, aromatic, resins, and asphaltenes (SARA) fraction of traditional asphalt and bio-oil is not similar. In addition, the volatile matter and water content in bio-asphalt binder could negatively impact the performance and durability of the asphalt mixture [5]. Therefore, bio-asphalt can be used as a modifier or extender.

Table 7. Classifications of bio-asphalts [78].

Category of Application	Content in the Binder	Current State
Modifier	<10%	The application mode is relatively mature
Extender/substitute	25–75%	The application mode is common
Alternative	100%	The application mode cannot be achieved due to the limitation of the performance of bio-asphalt

Initially, the WCO was utilized as a rejuvenator due to its fluidity nature for aged asphalt. It has the potential to restore low molecular weight compounds and viscosity lost due to the aging

of a binder. Similarly, following this attempt, researchers started exploring the opportunity for using WCO as a modifier to improve asphalt binder [13,24,25,49,79]. The modification of the asphalt binder with WCO needs proper blending variables. However, there is no specific standard available for the WCO-modified asphalt. Table 8 lists primary preparation variables in producing the WCO-modified binder. Su et al. [78] reported that a higher mixing temperature and shearing rate could generate a suitable vortex for WCO blending with asphalt binder. On the other hand, Wang et al. [80] stated that mixing variables should not exceed the specified limit, as too high a temperature and shearing rate for a long duration could destroy the quality of WCO-modified binder. Therefore, the literature concluded that the most utilized mixing condition of WCO in asphalt binder uses a shear rate of less than 5000 rpm at 135 °C–160 °C for about 30–60 min.

Table 8. Mixing variables for WCO-modified binder.

Reference	Shearing Time (min)	Shearing Speed (rpm)	Shearing Temperature (°C)
[81]	50	4000	170
[23]	60	1000	160
[82]	30	—	190
[83]	5	1600	145
[47]	60	1000	160
[84]	30	4000	140
[25]	60	1000	160
[85]	60	4000	160
[86]	60	1000	163
[87]	120	2000	150

9. Effect of Untreated and Chemically Treated Waste Cooking Oil on Conventional Properties of Asphalt Binder

The incorporation of WCO at various concentrations affects the physical properties of WCO-modified asphalt binder, as shown in (Table 9). Thus, a higher concentration increases the penetration while effectively reducing modified asphalt consistency [47]. According to the conclusions drawn by Al-Omari et al. [83], the penetration was observed to increase when 1–8 wt.% of filtered WCO was added to 76–145 dmm of asphalt, compared with the penetration of the 65 dmm of neat asphalt. Consequently, the softening point results have followed a similar trend, with a decreased temperature to 44 °C compared to 53 °C for neat asphalt. A similar study was conducted by Eriskin et al. [85], in which asphalt was modified by adding 1–5 wt.% of WCO to asphalt. The experimental outcomes have shown an increase in the penetration resistance to 102 dmm on a high content of the WCO when using a grade 50/70 asphalt. Similarly, the softening was reduced from 52.1 to 43.3 °C on high usage of WCO. Moreover, Somé et al. [82] utilized the WCO and reported the decrement in penetration and softening point results. The dilution of WCO distributes the asphalt particles more uniformly, which lowers the interaction between the saturation level of WCO and binder and can significantly lower the grades of a modified binder. Thus, many researchers have suggested that filtered WCO-modified binder could perform better in cold regions than conventional base asphalt [49].

On the other hand, the quality of WCO has a significant role in asphalt binder modification. The higher acid value of WCO weakens the bond between WCO and asphalt binder, as discussed in previous sections. In this way, Azahar et al. [23] evaluated the effect of acid values on the performance of modified asphalt. The WCO samples from April, August, and December were used with acid values 3.5, 2.8, and 1.6 mL/g, respectively. The 60/70 base asphalt was replaced by 3–5 wt.% of a binder. It was noticed that penetration was minimum for December compared to the other samples due to its lower acid value. Similarly, in another study, Azahar et al. [25] used treated WCO (transesterification) and compared the performance with untreated WCO in asphalt binder. The 28, 46, and 52% reductions in penetration were observed while adding treated WCO by 3, 4, and 5 wt.%, respectively. However, the results of the softening points are directly proportional to the penetration resistance, and improvement of 3, 4, and 5% was noted compared to the untreated WCO-modified bitumen (Figure 7a). The improved penetration value indicates that chemically treated WCO-modified binder is much harder than untreated WCO-modified

binder due to strong interaction bonding between the saturation level of treated WCO and asphalt binder [46]. Therefore, the chemical treatment of WCO might be a valuable alternative for WCO-modified asphalt binder in hot climate regions.

Table 9. Summary of the reviewed literature on the use of WCO and additives at various concentrations.

Reference	Modifier	Content of WCO/Additives (wt.%)	Penetration (dmm)	Viscosity @ 135 °C (Pa.s)	Softening Point (°C)	Ductility (cm)	Rutting Resistance (Pa)	Fatigue Resistance (Pa)	Low Temperature Stiffness (Mpa)
[13]	Simply filtered WCO	10, 30, 60	↑	↓	↓	—	↓	↓	↑
[82]		17.9	↑↑	—	↓	—	↓	↓	↑↑
[83]		1, 2, 4, 6, 8	↑	↓	↓	↑	↓	↑	↑
[88]		10	—	—	—	—	↓	—	—
[23]		3, 4, 5	↑↑	↓	↓	—	↓	—	—
[85]		1, 3, 5	↑↑	↓	↓	—	↓	—	—
[86]		1, 2, 3	↑↑	↓	↓	—	↓	—	—
[89]	Chemically treated WCO	2, 4, 6	↑↑	↓	↓	—	↓	—	—
[25]		—	↓↑	—	—	—	—	—	—
[90]		3, 4, 5	—	—	—	—	↑	—	—
[81]	SBS/PE/WCO	5, 3, 5	↓	↑	↑	↑	↑	—	↓
[31]	HDPE/WCO	2.5, 5, 7.5	↓	—	↑	—	—	—	—
[29]	GTR/WCO	5, 10, 15	↓	↑	↑	↑	↑	↓	↓
[91]	LDPE+PVC/WCO	2.5, 5, 7.5, 10	↓	↑	↑	↑	—	—	—
[92]	WCO/TR/POFA	1, 3, 5	↓	↑	↑	↑	—	—	—
[93]	NC/WCO	1, 2, 3	↑	↑	↓	—	↑	↑	—
[87]	WCO in SBS-modified binder	5, 7, 5, 10	↑	↑	↓	—	↑	↑	—
[94]	SBS/EVA/WCO	2, 4, 6	↔	↓	↓	—	↑	↔	—
[30]	WCO/TR/Bagasse	2.5, 5, 5	↑	↓	↓	↔	—	—	—
[95]	HDPE/WCO	1, 3, 5, 7, 9	↑	↓	↓	↔	—	—	—
[94]	SBS/EVA/WCO	3.5	↓	↑	↑	↑	↓	↑	↑
[30]	WCO/TR/Bagasse	5.5	↓	↑	↑	—	↑	↑	↑
[95]	HDPE/WCO	5, 10, 15, 20	↑	↑	↑	—	↑	↑	↑
[95]	HDPE/WCO	5, 8, 12, 15, 18	↑	↑	↑	—	↑	↑	↑
[95]	HDPE/WCO	7, 7, 7.5, 8, 10	↓	—	↑	—	—	—	—

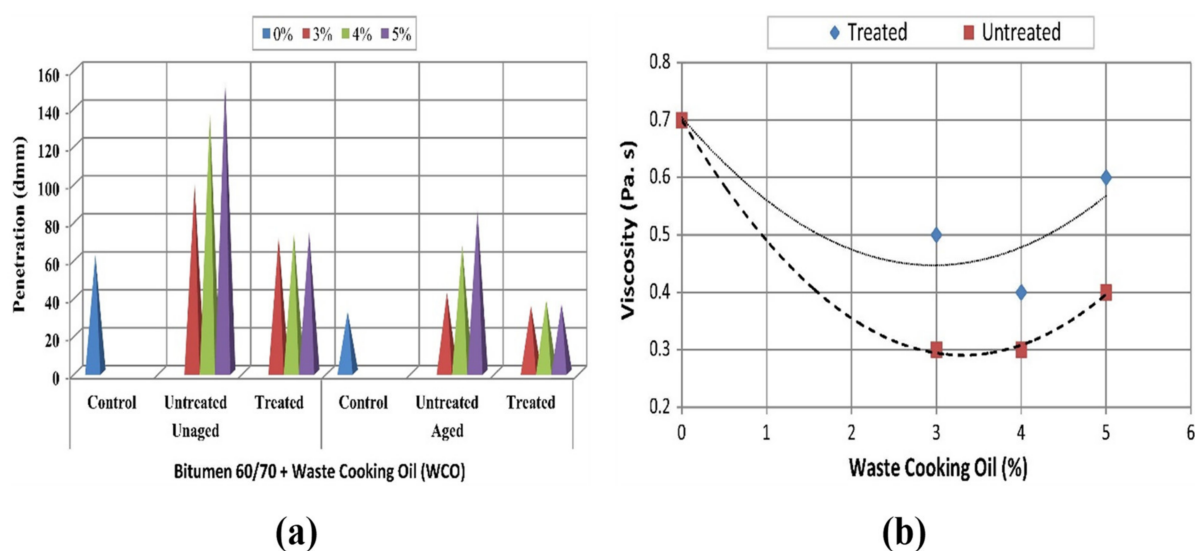


Figure 7. Effect of untreated and treated WCO content on asphalt binder: (a) penetration; (b) rotational viscosity [25].

Further reviewing the WCO-modified asphalt binder, the flow characteristic is worth considering. In a study, Rasman et al. [86] studied the viscosity of the untreated WCO-modified asphalt binder. Hence, the results revealed that the rotational viscosity at 135 °C was decreased linearly with an increase in the dosage of the WCO. Thus, the lowest viscosity was 0.2 Pa s for 3% WCO compared to 0.3 Pa s of the neat asphalt. Similar results

were reported by Bilema et al. [89] while adding the WCO at 2–6 wt.% of the 60/70 asphalt binder. The viscosity was reduced to 0.388 Pa s compared to the viscosity of a neat asphalt binder of 0.628 Pa s. Similarly, in studies by Al-Omari et al. [83] and Mahrez et al. [96], incorporating the untreated WCO softens the binder and reduces the viscosity. Thus, it is perceived that the WCO, without proper treatment to reduce the acid values, caused the asphalt binder to soften and ultimately declined the performance. The reduction in the consistency of the asphalt binder is due to the dilution and less internal friction. Thus, there is an inverse relationship between the viscosity of the asphalt binder and the content of WCO. Consequently, in another study, Azahar et al. [23] used different months of samples of WCO from April, August, and December to modify asphalt binder. The results indicated that the lower acid value sample of December showed some enhanced viscosity at 135 °C compared to other samples of WCO-modified asphalt binder. Azahar et al. [25] followed the transesterification process to reduce the WCO acid value. It is noticed from the outcomes that the viscosity of the treated WCO-modified asphalt binder is 33% improved than the untreated modified binder. However, with WCO at 5% replacement it can also be observed that viscosity has shown an increasing trend, as shown in Figure 7b. The viscosity increment at 5% is attributed to the adhesiveness between the particles of WCO and asphalt binder, with no dilution effect. From an economic standpoint, the marginal reduction in the viscosity is favorable for the ease of mixing and compaction temperature and to save the cost.

10. Effect of Untreated and Chemically Treated Waste Cooking Oil on Rheological Properties of Asphalt Binder

The asphalt binder demonstrates viscoelastic properties and mainly depends on the temperature and the frequency variation. The rheological properties are characterized by the complex modulus (G^*) and the phase angle (δ). The dynamic shear rheometer is widely used to identify the shear resistance of the asphalt binder. The G^* refers to the total resistance of the asphalt binder against deformation generated by shear loading, whereas the δ refers to the time delay between shear strain and shear stress. For a better understanding, the WCO modification and its effect on the viscoelastic behavior of the asphalt binder are worth analyzing. Thus, Bilema et al. [89] studied the G^* and δ of the untreated WCO-modified asphalt binder. They observed a decline in the values of G^* for the WCO content of 2–6 wt.% of the asphalt. The G^* values were more pronounced at a temperature range of 46–58 °C and particularly on the higher dosage of the WCO. This elucidated that the G^* of the 6% WCO-modified asphalt was reduced by 84% compared to without the modification. However, the G^* has not shown any notable difference in the values at a higher temperature. On the contrary, the δ is approaching 90° with the increasing content of WCO, which indicates the shift towards the viscous behavior of asphalt binder. In other comprehensive studies by Somé et al. [82] and Wen et al. [13], the performance of untreated WCO-modified asphalt binder on the higher content has similar trends of the G^* and δ , as concluded by [89].

The rutting ($G^*/\sin\delta$) and fatigue ($G^*\sin\delta$) parameters are the evaluation indicators for determining high and intermediate temperature resistance as per the Superpave specification, respectively. The reflective values to resist against the load for the $G^*/\sin\delta$ and the $G^*\sin\delta$ should be greater than 1.0 KPa and equal or less than 5000 KPa, respectively. In a recent study by Al-Omari et al. [83], the $G^*/\sin\delta$ was evaluated by adding untreated WCO 1–8 wt.% of the asphalt binder. The results indicated that the increasing content of WCO reduces the failure temperature of the modified asphalt. Figure 8a shows a slight variation in the $G^*/\sin\delta$ up to 4% of WCO, satisfying the Superpave criterion. However, on the higher content of untreated WCO, the $G^*/\sin\delta$ of the modified asphalt dropped drastically. The reduction in the $G^*/\sin\delta$ at higher contents of WCO for asphalt binder modification was also reported by Rasman et al. [86]. The results indicated that $G^*/\sin\delta$ is nearly similar to WCO-modified asphalt binder at lower contents. However, there was a sudden reduction in values when incorporating higher contents of WCO, as shown in

Figure 8b. Nordiana et al. [90] reported a reduction in the $G^*/\sin\delta$ when incorporating WCO more than 5 wt.% of the asphalt binder, similarly to [83,86].

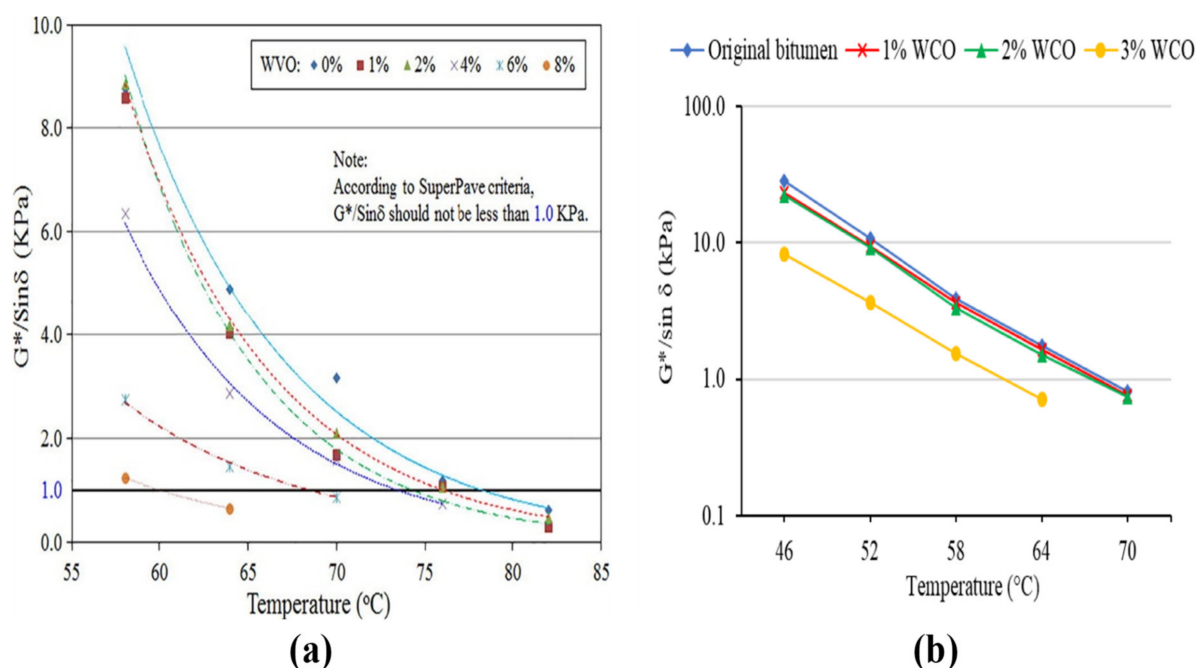


Figure 8. Rutting parameters of asphalt binders at various contents of WCO [83,86]. (a) Study done by Al-Omari et al. [83] and (b) Study done by Rasman et al. [86].

The quality of the WCO greatly influences the rutting performance of the WCO-modified asphalt binder, and acid value is the key parameter. In another study, Azahar et al. [25] evaluated the $G^*/\sin\delta$ by incorporating the three different qualities of WCO with acid values of 3.5, 2.8, and 1.6 mL/g. The results imply less $G^*/\sin\delta$ of 1.6 mL/g acid value WCO-modified asphalt compared to 3.5 mL/g. The failure temperature was 64 °C for the 1.6 mL/g acid value WCO-modified asphalt, which is higher than the 58 °C and 58.2 °C for 3.5 and 2.8 mL/g acid value WCO-modified asphalt, respectively. Azahar et al. (2016c) recently employed the transesterification process to treat WCO and significantly reduce the acid values from 1.66 to 0.54 mL/g. The treated and untreated WCO-modified asphalt binder failed at 70 °C and 64 °C, respectively (Figure 9). It was observed that the chemical modification of WCO improves the quality of WCO, which ultimately enhances rutting resistance. Acid values improvement provides better chemical bonding between WCO and base binder, producing a stiffer WCO-modified asphalt binder [3,48,97].

Apart from the higher temperature (rutting resistance), the intermediate temperature (fatigue resistance) also significantly impacts the performance of the WCO-modified asphalt binder. Usually, fatigue damage occurs in pavements due to repeated load and propagates within the bitumen mastic, resulting in microdamage. WCO can modify the asphalt binder and disperse energy by rebounding without cracking the pavement, due to improved adhesion. Al-Omari et al. [83] reported on the $G^* \cdot \sin\delta$ and exhibited that as the WCO content increases, fatigue resistance decreases at the intermediate temperatures, compared with the neat asphalt binder. The reduction in the $G^* \cdot \sin\delta$ for the modified asphalt binder with WCO 1–8 wt.% at 28 °C was 9–33%, respectively. Hence, the adding of WCO can enhance the fatigue resistance. Similarly, in other studies by [88,98], WCO can increase fatigue resistance at intermediate temperatures, and the WCO-modified binder has more improved performance than a base binder.

Al-Omari et al. [83] further investigated WCO-modified asphalt binder and revealed that WCO enhances the binder elasticity at low temperatures. The S-values of the modified asphalt with WCO decreased and met the Superpave criterion at −6 and −12 by 45%

and 54%, respectively. Yet, there are pronounced, changed m-values when more than 4% WCO was incorporated into the asphalt binder. Similarly, other comprehensive studies by Sun et al. [97], Gong et al. [99], Zhu et al. [100], and Zhang et al. [48] supported the same result of WCO-modified binder in low temperatures. It has been shown that adding more WCO improves the creep stiffness, whereas the m-value decreases. The lower temperature makes the binder stiffer, light components in WCO can soften the binder, and the m-value increases. On the other hand, due to the hydrolysis process, the presence of water content has a negative effect on the quality of WCO and the compatibility of asphalt binder with WCO, which consequently affects the low-temperature properties of WCO-modified asphalt binder. Thus, the WCO refinery must control the moisture content, using any water removal method to enhance the properties of the WCO-modified asphalt binder [101].

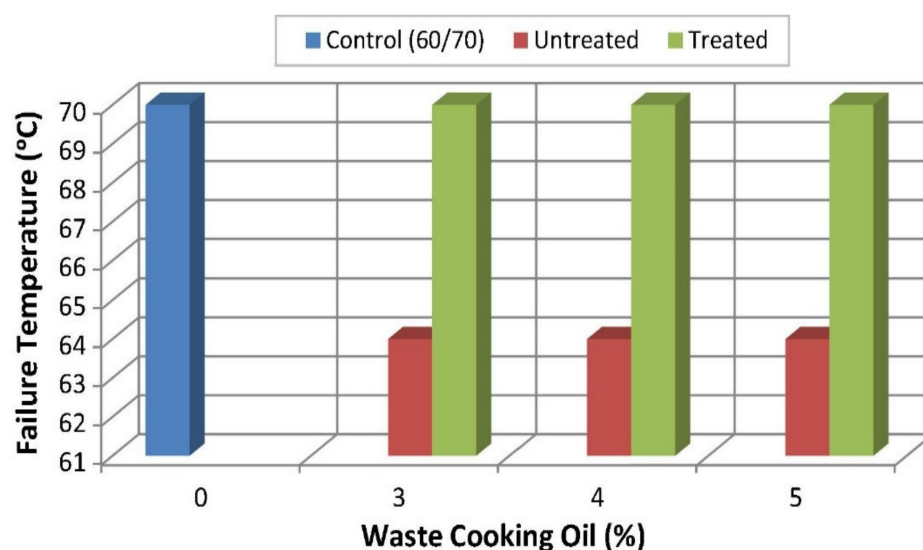


Figure 9. Failure temperatures of untreated and treated modified asphalt binders [25].

Generally, these studies constantly revealed that WCO changes the rheological characteristics of asphalt binder. The incorporation of WCO lowered the shear strength and decreased the resistance to flow. Therefore, based on the results of the rheological tests, it can be inferred that using WCO as a modifier offers superior fatigue resistance and low-temperature performance. However, reducing rutting resistance may cause concerns, which may be addressed by modifying WCO chemically or physically by adding other polymers as modifiers, such as SBS, LDPE, and GTR, which could be a better alternative for ensuring practical application.

11. Effect of Waste Cooking Oil with Additional Additives on Conventional Properties of Asphalt Binder

The incorporation of the untreated and treated WCO in asphalt binder has been investigated by several researchers, who observed an insignificant improvement in the physical and rheological performance [25,83,89,90]. For instance, the chemically treated WCO performs better in comparison to the untreated WCO. However, further enhancement had been made when researchers utilized other additional modifiers in conjunction with the WCO, such as high-density polyethylene (HDPE), low-density polyethylene (LDPE), ground tire rubber (GTR), and nano clay (NC). Adesina and Dahunsi (2021) studied the effect of WCO and HDPE as a modifier on the physical properties of asphalt binder. The penetration resistance increased by 20.7% by adding WCO 15 wt.% of asphalt. Incorporation of the WCO softened the asphalt, thus causing the decrement in softening properties by 41%. However, the reduction in physical properties was counterbalanced by adding the HDPE (Figure 10). A 11.6% reduction in penetration resistance and 37% increase in softening point were observed when adding HDPE 7.5 wt.% of asphalt.

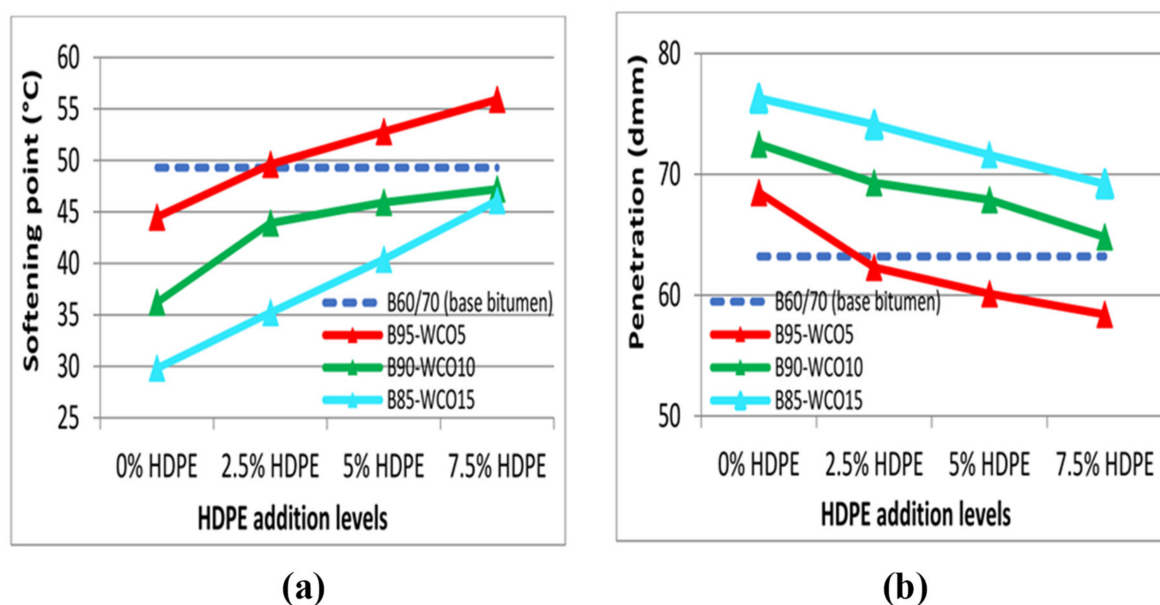


Figure 10. Effect of WCO and HDPE contents on asphalt binder: (a) softening; (b) penetration resistance [31].

Similarly, Joni et al. [91] studied the influence of LDPE and PVC on the WCO (1 wt.%) modified asphalt binder. The results indicated that adding PVC and HDPE 5 wt.% of WCO-modified asphalt decreased the penetration by 31% and 33%, respectively. The softening point trends were in a similar manner, with an enhancement of 7.2% and 7.4% compared to WCO 1 wt.% modified asphalt binder results. Another similar study by Dahunsi and Adesina [95] investigated the physical properties of WCO-modified asphalt binder in conjunction with the HDPE. The results were similar to those by [31,91]. The HDPE's significant tolerance to high temperatures made a gel type structure within the binder, thereby enhancing the modified binder's elastic properties.

In a recent study by Niu et al. [29], the effect of adding WCO (2.5–10 wt.%) to asphalt in conjunction with GTR (10–20 wt.%) to asphalt on a rotational viscosity at 135 °C of binder was analyzed. The results indicated that the viscosity of WCO/GTR-modified asphalt increased with increasing the dosage of GTR. The asphalt with 20% GTR has shown significant improvement with different contents of WCO and temperature. Moreover, the increasing content of WCO in the GTR-modified asphalt lowers the viscosity from 8 to 2.5 Pa s, which signifies that the WCO causes reduction in the compaction and mixing temperature of the mixture (Figure 11). A similar enhancement in the binder's viscosity by incorporating WCO, GTR, and POFA was reported by [92]. The results implied that the 15% replacement of composite material to asphalt resulted in the enhanced rotational viscosity. The 5% WCO, 10% GTR, and 7.5% POFA were the optimal combinations, which increased the 60% viscosity of asphalt compared to neat asphalt. High POFA and tire rubber powder make the binder viscous, since they remain solid even as the temperature rises. Similarly, Khan et al. [30] also reported a similar result considering improving the viscosity of WCO/tire rubber(TR)/bagasse fiber-modified binder. Bagasse fiber was added as an additive. It was observed that, due to the presence of tire rubber and bagasse fiber, almost 20% of the base binder could easily be replaced by composite materials, which will minimize environmental degradation caused by improper waste disposal.

The addition of nano clay (NC) in the WCO-modified asphalt binder also positively impacted the softening point. The WCO 2.5% wt. and 6% dosage of NC in the high-temperature base binder PG 70 improved it to PG 76. This demonstrates that NC has a higher specific surface area, leading to an increase in stiffness of the WCO-modified binder [93]. Consistently, styrene–butadiene–styrene (SBS) and ethylene–vinyl acetate (EVA) can improve low-temperature deformation by increasing the ductility value of asphalt binder. A study conducted by Liu et al. [94] investigated the change in the physical

behavior of SBS/EVA-modified asphalt and it was concluded that by the addition of 10% WCO in the SBS/EVA-modified binder improved the ductility value. This explains that SBS and EVA make a crosslinking translation in asphalt, which allows it to utilize more quantities of WCO and collectively leads to a pronounced increase in ductility value.

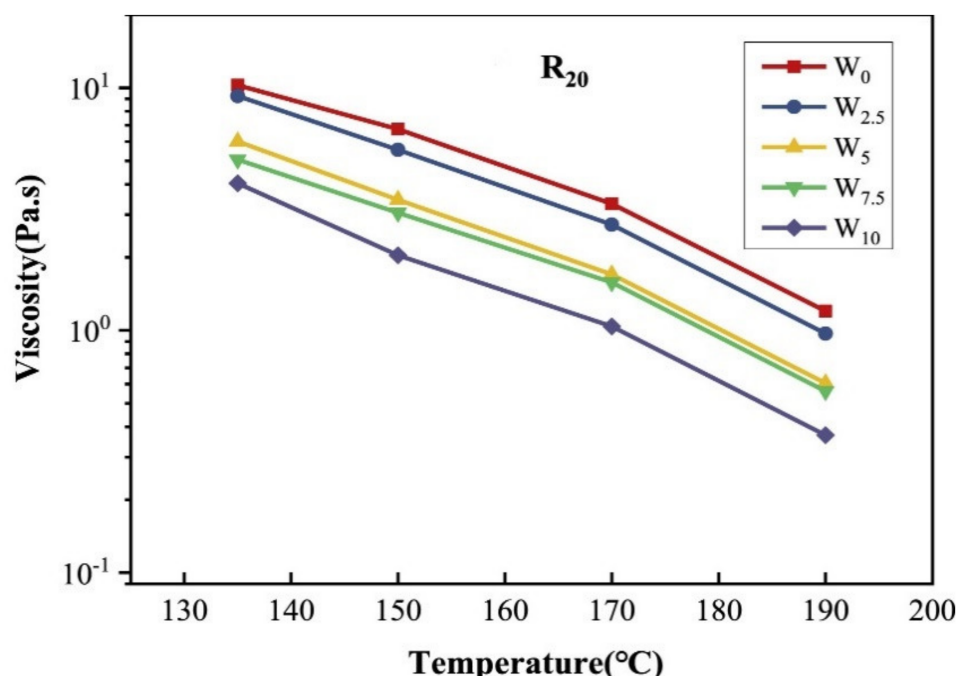


Figure 11. Effect of WCO and GTR content on the rotational viscosity at 135 °C [29].

Other comprehensive studies, by Luo et al. [81] and Oruc [87], reported experimental findings for improving the ductility of SBS/LDPE-modified asphalt with the incorporation of WCO, which demonstrated that adding WCO with polymers can increase flexibility and low-temperature fracture resistance. However, it is evident that the viscosity of WCO is lower. Thus, the addition of WCO in the binder makes it more prone to rutting at a higher temperature. To fill this gap, researchers combined different additional modifiers, such as waste plastic or other polymers, with WCO to make a composite material and replaced different percentages of a base binder with the composite material. This new study not only solved the issue regarding adhesive performance of asphalt binder, but it also worked well as an eco-friendly renewable binder in the pavement industry [30,92,102].

12. Effect of Waste Cooking Oil with Additional Additives on Rheological Properties of Asphalt Binder

The impact of composite materials on the high, low, and intermediate temperature properties of WCO-modified binder is shown in Table 9. Multiple stress creep recovery (MSCR) and linear amplitude sweep (LAS) have gained popularity to test the rutting resistance and fatigue behavior of the asphalt binder using a dynamic shear rheometer [13,93,103], whereas a few studies evaluated the rutting and fatigue properties of WCO-modified binder through MCSR and LAS tests. MSCR provides two essential characteristics to describe the reaction of a modified asphalt binder under stress: non-recoverable creep compliance (J_{nr}) and percentage recovery (%) [104]. The increased J_{nr} indicates weak thermal resistance to stresses and low bitumen rutting resistance [13].

The Saboo et al. binder [93] revealed that J_{nr} varies in response to changes in the percentages of WCO and NC. A higher concentration of WCO responded to the increased J_{nr} value due to the binder softening, resulting from the addition of WCO. On the other hand, the value of J_{nr} dropped as the proportion of NC increased, which simultaneously improved the rutting resistance of the modified binder. Yet at the higher temperature, NC

can retain elasticity, thus improving the rutting resistance of NC/WCO-modified binder. Similarly, Niu et al. [29] reported the J_{nr} value of the GTR/WCO-modified binders under a single creep stress level (3.2 KPa); results showed that WCO and GTR exhibit opposing impacts on the J_{nr} (3.2) value of the binder, suggesting that the negative effect of WCO on the binders' rutting resistance can be balanced by properly increasing GTR content.

The LAS test is conducted to measure the potential of fatigue cracking at intermediate temperatures caused by a repeated load with a linear amplification of strain at a constant frequency. The test consists of two stages: (1) a frequency sweep test to determine the alpha " α " parameter for damage analysis, and (2) an amplitude sweep test to assess damage buildup in the samples versus loading cycles. Niu et al. [29] demonstrated the fatigue life of GTR/WCO-modified binder at 2.5% and 5% strain levels. It was found that at a strain level of 2.5%, modified asphalt binder with 5 wt.% WCO and 20 wt.% GTR enhances the fatigue life 9.6 and 4.6 times longer than 10 and 5 wt.% GTR, as shown in Figure 12. Similarly, Saboo et al. [93] quantified the fatigue behavior of NC/WCO-modified asphalt binder. Based on the evaluation of stress versus strain, it was observed that the addition of NC in WCO-modified binder has no substantial effects on the fatigue resistance of asphalt binder at intermediate temperatures. However, an increase in the percentage of NC leads to an increase in the small stiffness of the binder, thus increasing strain susceptibility.

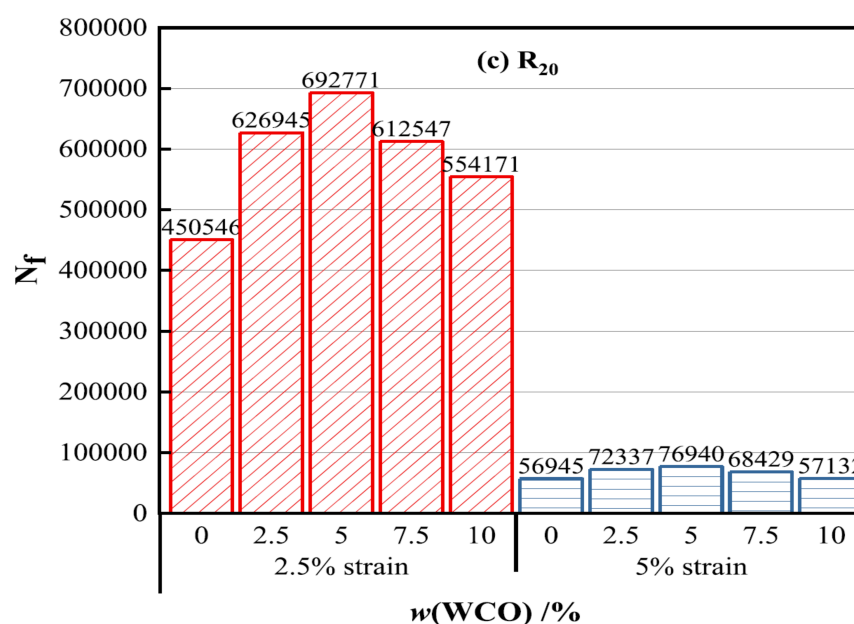


Figure 12. Predicted fatigue life of WCO and GTR modified asphalt binder [29].

Consequently, Rahman et al. [92] reported the $G^*/\sin \delta$ and $G^*\sin \delta$ performance of 50 wt.% and 30 wt.% replaced the base binder with tire rubber (TR) and WCO (under different percentage combinations). At the same time, POFA was added as a stabilizer in a modified binder. The presence of WCO enforced the modified binder particularly close to an 80/100 binder. The result concluded that all modified samples exhibit poor resistance against rutting and higher fatigue resistance compared to base binder 60/70. Furthermore, few other studies observed a mixed trend of rutting and fatigue parameters of the modified binder incorporation of WCO with additional modifiers [30,81,94].

In conclusion, limited studies are available in terms of MSCR and LAS testing on WCO-modified binders. Adding WCO to the binder softens it, thus offering fatigue enhancement, whereas non-recoverable creep compliance lowers permanent deformation resistance. As a result, to improve rutting resistance, additional additives or composite materials should be considered before including WCO into the binder, with cautions based on the site's climatic circumstances.

Furthermore, Liu et al. [94] reported the SBS- and EVA-modified asphalt outcomes in conjunction with the WCO. The low-temperature property of the modified binder was compromised and does not appear to enhance the performance. However, SBS/EVA can increase rutting resistance at high temperatures. Thus, when 5 wt.% and 10 wt.% WCO were mixed with SBS/EVA-modified asphalt, the asphalt met the Superpave requirements at -18°C and -12°C test temperature, respectively (Figure 13). These findings show that WCO increases low-temperature fracture resistance significantly. This is because WCO-modified binder is softer than polymer-modified asphalt binder. Furthermore, similar results have been observed by [29,30]. For instance, Niu et al. [29] compared 10 wt.% GTR-modified binder with and without the addition of WCO. Thus, it was deduced that the creep with higher WCO and GTR had stiffness decrease, whereas m-value increased. The addition of WCO accounted for the soft asphaltene absorbed by GTR, allowing GTR to play a more elastic supplemental function while improving low-temperature relaxation properties. As a result, the WCO/GTR-modified asphalt demonstrated satisfactory deformation ability at low temperatures. It can be inferred that adding WCO to the asphalt binder enhances thermal cracking resistance. Although, blending and concentration of WCO with other types of composite material effects both creep stiffness and m-value.

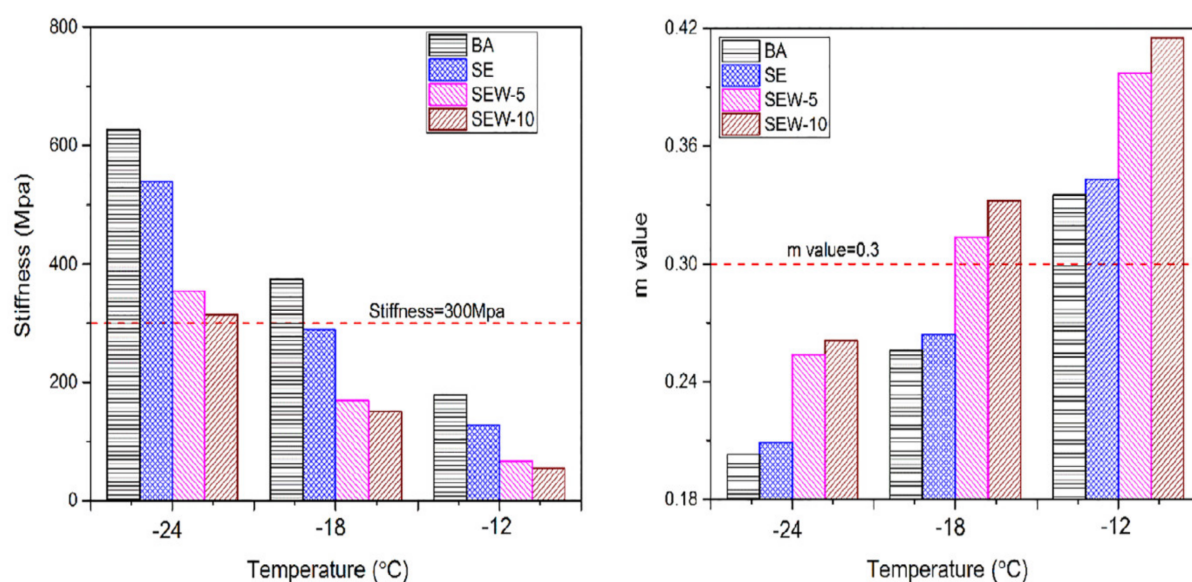


Figure 13. Creep stiffness and m-value of WCO and SBS/EVA modified asphalt binders [94].

Researchers have conducted studies on the performance analysis of the WCO-modified asphalt binder and in conjunction with other additives. The enhancement was demonstrated in the rutting and fatigue performance of the treated and composite asphalt binder. A study was conducted by Wen et al. [13] to evaluate the influence of WCO on the rutting performance of the asphalt mixture. It was evaluated that the rutting potential of asphalt mixes reduces with a high dose of WCO using a dynamic repeated creep load. These findings support what was previously stated for the simply filtered WCO-modified binder rutting tests, based on complex shear modulus, phase angle, and J_{nr} [82]. On the other hand, Azahar et al. [47] reported that the permanent deformation of the WCO-modified mixture could be improved by chemically treating WCO.

13. The Impact of Waste Cooking Oil on the Performance of Asphalt Mixtures

Hence, the result indicated that at 40°C after chemical modification of WCO, the strain rate reduced by 17.5% relative to the control mixture. The non-polar group in the untreated WCO converts into a polar group, and polar attraction between treated WCO molecules and polar aggregate surface improves adhesion bonding, thus resulting in improved rutting resistance performance in the asphalt mixture [105]. Furthermore, Niu et al. [29]

performed a permanent deformation test by dynamic stability (DS) to demonstrate the dynamic stability of three mixes at the high temperature of 60 °C, and the results are shown in Figure 14. In comparison to the original asphalt mixture, the WCO-modified asphalt mixture showed less dynamic stability. Hence, the WCO/GTR-modified asphalt mixture has a much higher average dynamic stability than the other two asphalt mixes.

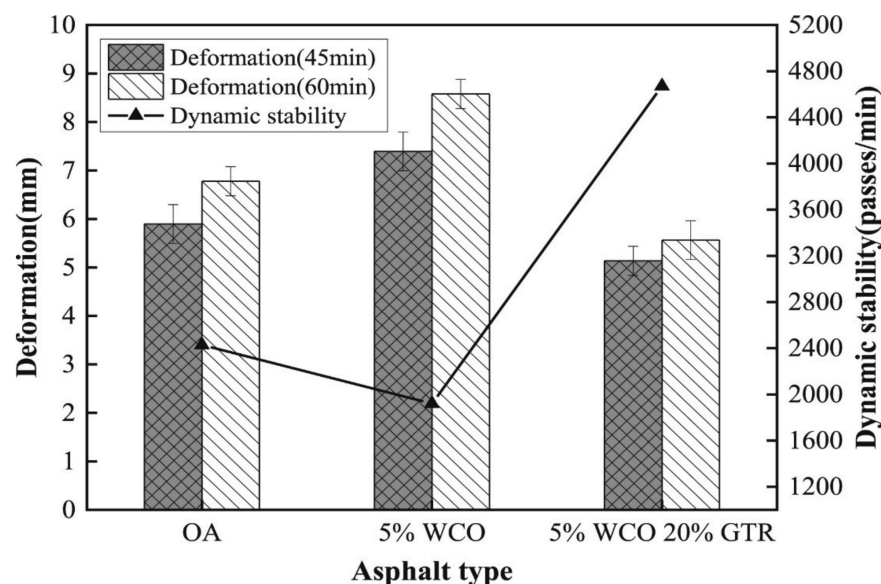


Figure 14. Stability test result [29].

The presence of WCO reduces the high-temperature performance of the asphalt mixture, whereas the addition of GTR counteracts the negative effects. The ability of GTR to modify the viscoelasticity of asphalt through physical and chemical interactions results in a mixture with high stiffness and modulus that effectively reduces deformation under load. A similar result was identified by [10] for optimized bio-asphalt (OBA), Pen 70 and SBS-modified mixture. However, it was observed from Figure 15 that all mixtures satisfy the critical high-temperature dynamic stability value 2400 times/mm for the OBA and 800 times/mm for the SBS-modified asphalt mixture. OBA contains different polymer concentrations along with WCO, which stiffens the binder and improves rutting resistance compared to the WCO-modified mixture. On the other hand, the addition of WCO into the asphalt mixture has a positive impact on fatigue resistance, whereas excessive dose may reduce the fatigue resistance of asphalt mixtures [106].

Wen et al. [13] revealed fatigue test results and indicated that as the amount of WCO in the mixture increases, asphalt fatigue decreases. On the other hand, an incremental dose of WCO enhances the resistance to thermal cracking, indicated by a strength test at low temperatures. Similarly, three-point bending tests revealed that WCO-based asphalt mixes had a high flexural failure strain (5200 $\mu\epsilon$) when compared to control asphalt mixtures (2300 $\mu\epsilon$) and WCO/GTR-modified asphalt mixtures (5000 $\mu\epsilon$). This suggests that WCO can increase the deformability of asphalt at low temperatures and that adding GTR to the mix will reduce the effect of WCO. This demonstrates that a WCO-based asphalt mixture is resistant to low-temperature cracking [10]. It is concluded that the high concentration of WCO reduces the cohesiveness of asphalt binder, which causes specimens to fail at lower deformations, resulting in less fatigue resistance. At the same time, it improves the asphalt mixture thermal cracking resistance. However, very limited studies are available in fatigue and thermal resistance of WCO-modified asphalt mixtures. Besides the mechanical performance of the WCO-modified mixture, the asphalt pavement with weak moisture resistance may experience moisture damage, which ultimately affects the traffic flow, safety, and limited service life in the future [3].

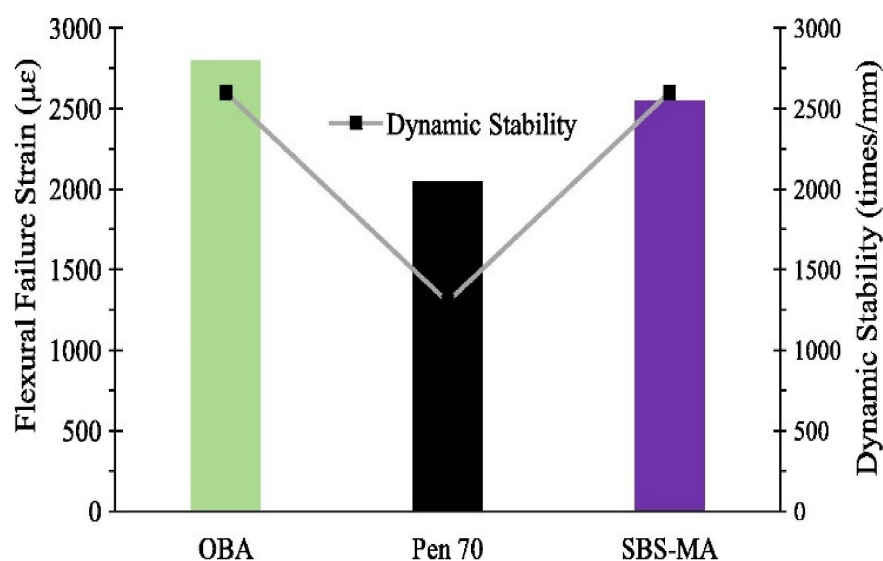


Figure 15. Dynamic stability of different HMA [10].

Wen et al. [13] investigated the effect of simply filtered WCO addition on the moisture susceptibility of asphalt mixes and concluded that all samples containing WCO met the AASTHO tensile strength ratio (TSR) minimum requirements (i.e., >80%). However, similar to results reported by [85], TSR values for each sample prepared with WCO were in the acceptable range shown in Figure 16. It is worth noting that moisture susceptibility depends on adhesion between asphalt and the aggregate surface. WCO-modified asphalt mixture showed better TSR value than WCO/GTR-modified mixture, which identifies an adhesion bond between asphalt and aggregate. The adhesion is correlated with the quality of the WCO. The presence of FFA in WCO may enhance the water affinity for the binder, resulting in increased moisture susceptibility. It has been noticed that linoleic acid and hexadecenoic acid are water-insoluble fatty acids and can easily be absorbed effectively in dry conditions. Thus, in a wet environment, these fatty acids are immediately replaced by water molecules, which in turn raises moisture susceptibility [107,108]. It can be inferred from the summarized studies that WCO-modified asphalt mixtures may fulfil the specification criteria. However, the impact of WCO on moisture susceptibility varies depending on the individual mix and quality of the WCO. Thus, it is needed to have an in-depth analysis of WCO-modified mixture performance prediction.

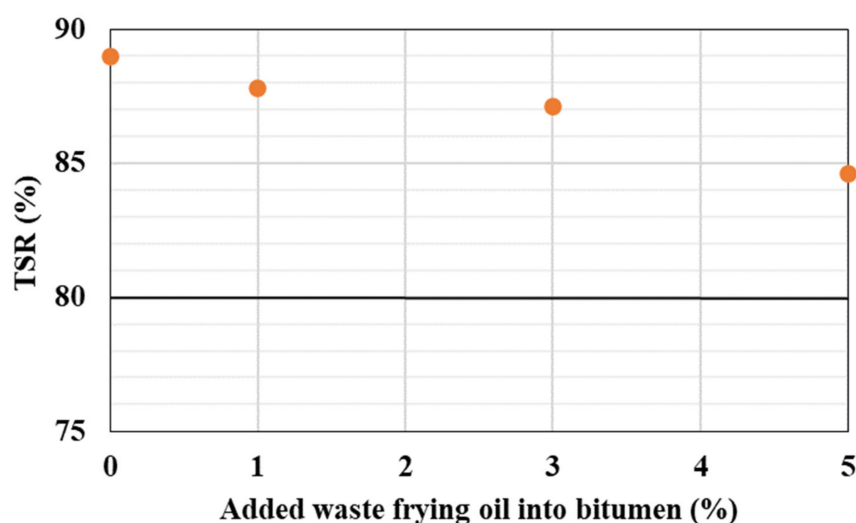


Figure 16. Tensile strength ratio for WCO-modified asphalt mixture [85].

14. Fourier Transform Infrared Spectroscopy of WCO Modified Asphalt Binders

Fourier transform infrared spectroscopy (FTIR) is used to identify the characterization and identification of functional groups in the various samples. The only analytical approach operates at room temperature while simultaneously monitoring the vibrations of the functional groups that describe the molecular structure [109]. According to a previous experimental study, the area between 2.5 mm and 25 mm (4000 to 400 cm^{-1}) is the most acceptable for chemical investigation. Oldham et al. [50] showed FTIR of untreated and chemically treated WCO. The absorbance peaks at 1446 cm^{-1} and 1196 cm^{-1} are attributed to $-\text{CH}_3$ and $\text{O}-\text{CH}_3$ stretching, which are noticed only in the case of treated WCO. However, the absence of a glycerol peak at 1377 cm^{-1} detected esters in large abundance by area in treated WCO (Figure 17). According to Wang et al. [4], the presence of many ester substances in WCO can effectively improve adhesion between WCO-modified asphalt and the aggregate. Besides this, Azahar et al. [110] observed one peak in untreated WCO-modified binder at 1743 cm^{-1} , which shows the presence of a $\text{C}=\text{O}$ bond, which is a type of aldehyde and ketone bond. This kind of bond forms due to the oxidation process that occurs in the WCO sample during the frying process. This peak softens the binder, making it more prone to rutting. In another study by Azahar et al. [25], it was revealed that with the chemical modification of WCO, the $\text{C}-\text{H}$ bond and the $\text{C}=\text{C}$ stretch aromatic bond increased for all percentages of treated WCO, and the $\text{C}=\text{O}$ bond at 1743 cm^{-1} was noticed to be negligible. Thus, from the analysis of the above studies, it can be observed that by chemical modification of WCO, the peak of $\text{C}=\text{O}$ can reduce drastically, resulting in the enhancement of physical and rheological properties of WCO-modified asphalt binder, which ultimately enhance the performance of the asphalt mixture.

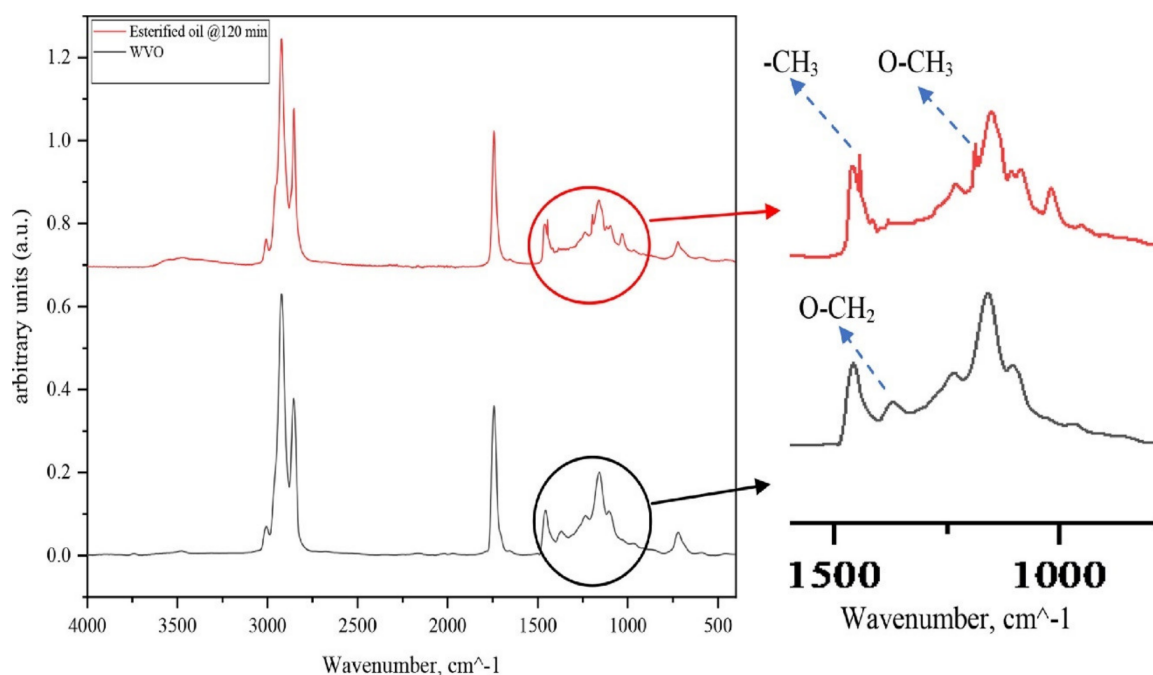


Figure 17. FTIR spectra of untreated and chemically treated WCO [50].

15. Conclusions and Recommendations

In the production of flexible pavement, asphalt binder is the most important component. Therefore, new alternatives must be found to reduce dependency on non-renewable petroleum-based asphalt, to create environmentally friendly, cost-effective, broadly available, and technically acceptable pavement. Much research has been conducted or is being conducted in recent years to see if WCO can be utilized as modifying materials in the pavement industry. This paper reviewed the basic background of WCO, available technologies

to mitigate the effect of FFA and water content, the adverse impact of the quality of WCO on asphalt binder, and HMA performance with and without additional modifiers. Based on the present review, the following conclusions may be drawn:

- The acid value, water content, and overall quality of WCO have a major impact on the efficiency of the modification process. The limited available technologies have been utilized to minimize the effect of FFA and water content on asphalt properties.
- The concentration and quality of the WCO have a major impact on the properties of filtered WCO-modified asphalt binder. As the incremental dose of WCO introduces additional lighter oil components, the physical, rheological, and mixture performance are greatly influenced. However, at lower temperature, addition of WCO enhance the binder flexibility, thus leading to improved thermal and fatigue resistance.
- Few additives, such as GTR, NC, waste plastic, and POFA, have been added in WCO-modified binder to analyze their effect. Improved physical properties with the mixed trend of rutting and fatigue parameters were observed based on types and concentrations of additive along with WCO %.
- The oxidative study using FTIR revealed that untreated WCO has undesired components, such as C=O stretch, responsible for the poor rutting performance of WCO-modified asphalt binder. The treated WCO-based binder contains an increased peak of C–H bond and C=C aromatic bond, with a negligible amount of unwanted chemicals, such as C=O stretch.
- The simply filtered WCO does not work well on mixture performance at high temperatures, whereas treated WCO-modified or additive/WCO-modified asphalt mixture favors improved resistance to permanent deformation.
- All the WCO-modified asphalt fulfilled the TSR requirement. However, in few studies, adding additives in WCO-modified mixture showed comparatively less TSR value than WCO-modified mixture.

As a novel modified asphalt material, WCO has caught the attention of researchers. The consumption of WCO has been proven to be beneficial in many ways for the construction of highway pavements. However, WCO is still used in a very limited way, perhaps because of a lack of research and mixed results and recommendations. Thus, this study can help overcome these limitations and recommend future research to enhance the use of WCO for binder modification:

- Available technologies can be utilized for the physical and chemical treatment of WCO prior to incorporating it into the asphalt binder.
- The physical and chemical interaction of WCO with asphalt binder should be investigated further.
- The analysis of the bond between WCO and different modifiers should be defined at a micro-level, as the focus on this area of study needs to be in-depth.
- Further in-depth evaluation of the performance of asphalt mixture with different modifiers and gradations is required to fully analyze the behaviors of mixtures.
- More studies should be conducted to evaluate the mixing variables for proper blending of WCO with asphalt binder, also in conjunction with other additional additives.

Author Contributions: This article is the result of joint work by all authors. Designing, carrying out and writing the original draft, Z.E.; review and editing, F.M.J., A.H.A. and M.M.B.Z.; conceptualization F.M.J., R.M., M.S.A.R. and S.H. All authors have read and agreed to the published version of the manuscript.

Funding: This research receives no external funding.

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Conflicts of Interest: The authors declare no conflict of interest.

References

1. Fini, E.H.; Al-Qadi, I.; You, Z.; Zada, B.; Mills-Beale, J. Partial replacement of asphalt binder with bio-binder: Characterisation and modification. *Int. J. Pavement Eng.* **2012**, *13*, 515–522. [\[CrossRef\]](#)
2. Ansari, A.H.; Jakarni, F.M.; Muniandy, R.; Hassim, S.; Elahi, Z. Natural rubber as a renewable and sustainable bio-modifier for pavement applications: A review. *J. Clean. Prod.* **2021**, *289*, 125727. [\[CrossRef\]](#)
3. Zhang, X.; Zhang, K.; Wu, C.; Liu, K.; Jiang, K. Preparation of bio-oil and its application in asphalt modification and rejuvenation: A review of the properties, practical application and life cycle assessment. *Constr. Build. Mater.* **2020**, *262*, 120528. [\[CrossRef\]](#)
4. Wang, H.; Ma, Z.; Chen, X.; Hasan, M.R.M. Preparation process of bio-oil and bio-asphalt, their performance, and the application of bio-asphalt: A comprehensive review. *J. Traffic Transp. Eng.* **2020**, *7*, 137–151. [\[CrossRef\]](#)
5. Aziz, M.A.; Rahman, T.; Hainin, M.R.; Abu Bakar, W.A.W. An overview on alternative binders for flexible pavement. *Constr. Build. Mater.* **2015**, *84*, 315–319. [\[CrossRef\]](#)
6. Yang, X.; You, Z.; Dai, Q. Performance evaluation of asphalt binder modified by bio-oil generated from waste wood resources. *Int. J. Pavement Res. Technol.* **2013**, *6*, 431–439. [\[CrossRef\]](#)
7. Yang, X. The Laboratory Evaluation of Bio Oil Derived from Waste Resources as Extender for Asphalt Binder. Ph.D. Dissertation, Michigan Technological University, Houghton, MI, USA, 2013.
8. Mohammad, L.N.; Elseifi, M.A.; Cooper, S.B.; Challa, H.; Naidoo, P. Laboratory Evaluation of Asphalt Mixtures that Contain Biobinder Technologies. *Transp. Res. Rec. J. Transp. Res. Board* **2013**, *2371*, 58–65. [\[CrossRef\]](#)
9. Fini, E.H.; Kalberer, E.W.; Shahbazi, A.; Basti, M.; You, Z.; Ozer, H.; Aurangzeb, Q. Chemical Characterization of Biobinder from Swine Manure: Sustainable Modifier for Asphalt Binder. *J. Mater. Civ. Eng.* **2011**, *23*, 1506–1513. [\[CrossRef\]](#)
10. Sun, D.; Sun, G.; Du, Y.; Zhu, X.; Lu, T.; Pang, Q.; Shi, S.; Dai, Z. Evaluation of optimized bio-asphalt containing high content waste cooking oil residues. *Fuel* **2017**, *202*, 529–540. [\[CrossRef\]](#)
11. Rogers, J.; Brammer, J. Estimation of the production cost of fast pyrolysis bio-oil. *Biomass-Bioenergy* **2012**, *36*, 208–217. [\[CrossRef\]](#)
12. Ramadhansyah, P.J.; Masri, K.A.; Azahar, W.N.A.W.; Mashros, N.; Norhidayah, A.H.; Warid, M.N.M.; Satar, M.K.I.M.; Yaacob, H. Waste Cooking Oil as Bio Asphalt Binder: A Critical Review. *IOP Conf. Ser. Mater. Sci. Eng.* **2020**, *712*, 012040. [\[CrossRef\]](#)
13. Wen, H.; Bhusal, S.; Wen, B. Laboratory Evaluation of Waste Cooking Oil-Based Bioasphalt as an Alternative Binder for Hot Mix Asphalt. *J. Mater. Civ. Eng.* **2013**, *25*, 1432–1437. [\[CrossRef\]](#)
14. Al-Sabaei, A.M.; Napiah, M.B.; Sutanto, M.H.; Alaloul, W.S.; Usman, A. A systematic review of bio-asphalt for flexible pavement applications: Coherent taxonomy, motivations, challenges and future directions. *J. Clean. Prod.* **2020**, *249*, 119357. [\[CrossRef\]](#)
15. Mannu, A.; Garroni, S.; Porras, J.I.; Mele, A. Available Technologies and Materials for Waste Cooking Oil Recycling. *Processes* **2020**, *8*, 366. [\[CrossRef\]](#)
16. De Feo, G.; Di Domenico, A.; Ferrara, C.; Abate, S.; Osseo, L.S. Evolution of Waste Cooking Oil Collection in an Area with Long-Standing Waste Management Problems. *Sustainability* **2020**, *12*, 8578. [\[CrossRef\]](#)
17. Teixeira, M.R.; Nogueira, R.; Nunes, L.M. Quantitative assessment of the valorisation of used cooking oils in 23 countries. *Waste Manag.* **2018**, *78*, 611–620. [\[CrossRef\]](#)
18. Zhao, Y.; Wang, C.; Zhang, L.; Chang, Y.; Hao, Y. Converting waste cooking oil to biodiesel in China: Environmental impacts and economic feasibility. *Renew. Sustain. Energy Rev.* **2021**, *140*, 110661. [\[CrossRef\]](#)
19. Zahoor, M.; Nizamuddin, S.; Madapusi, S.; Giustozzi, F. Sustainable asphalt rejuvenation using waste cooking oil: A comprehensive review. *J. Clean. Prod.* **2021**, *278*, 123304. [\[CrossRef\]](#)
20. Ramos, T.R.P.; Gomes, M.; Barbosa-Povoa, A. Planning waste cooking oil collection systems. *Waste Manag.* **2013**, *33*, 1691–1703. [\[CrossRef\]](#)
21. Li, L.; Xin, C.; Guan, M.; Guo, M. Using Molecular Dynamics Simulation to Analyze the Feasibility of Using Waste Cooking Oil as an Alternative Rejuvenator for Aged Asphalt. *Sustainability* **2021**, *13*, 4373. [\[CrossRef\]](#)
22. Tsai, W.-T. Mandatory Recycling of Waste Cooking Oil from Residential and Commercial Sectors in Taiwan. *Resources* **2019**, *8*, 38. [\[CrossRef\]](#)
23. Azahar, W.N.A.W.; Bujang, M.; Jaya, R.P.; Hainin, M.R.; Ngadi, N.; Abdullah, M.M.A.B. Performance of Waste Cooking Oil in Asphalt Binder Modification. *Key Eng. Mater.* **2016**, *700*, 216–226. [\[CrossRef\]](#)
24. Asli, H.; Ahmadinia, E.; Zargar, M.; Karim, M.R. Investigation on physical properties of waste cooking oil—Rejuvenated bitumen binder. *Constr. Build. Mater.* **2012**, *37*, 398–405. [\[CrossRef\]](#)
25. Azahar, W.N.A.W.; Jaya, R.P.; Hainin, M.R.; Bujang, M.; Ngadi, N. Chemical modification of waste cooking oil to improve the physical and rheological properties of asphalt binder. *Constr. Build. Mater.* **2016**, *126*, 218–226. [\[CrossRef\]](#)
26. Ben Zair, M.; Jakarni, F.; Muniandy, R.; Hassim, S. A Brief Review: Application of Recycled Polyethylene Terephthalate in Asphalt Pavement Reinforcement. *Sustainability* **2021**, *13*, 1303. [\[CrossRef\]](#)
27. Ansari, A.H.; Jakarni, F.M.; Muniandy, R.; Hassim, S. A review on the application of natural rubber as asphalt modifier. *IOP Conf. Ser. Mater. Sci. Eng.* **2021**, *1075*, 012031. [\[CrossRef\]](#)
28. Rodrigues, C.; Capitão, S.; Picado-Santos, L.; Almeida, A. Full Recycling of Asphalt Concrete with Waste Cooking Oil as Rejuvenator and LDPE from Urban Waste as Binder Modifier. *Sustainability* **2020**, *12*, 8222. [\[CrossRef\]](#)
29. Niu, D.; Xie, X.; Zhang, Z.; Niu, Y.; Yang, Z. Influence of binary waste mixtures on road performance of asphalt and asphalt mixture. *J. Clean. Prod.* **2021**, *298*, 126842. [\[CrossRef\]](#)

30. Khan, J.; Hussain, A.; Haq, F.; Ahmad, K.; Mushtaq, K. Performance Evaluation of Modified Bitumen with Replaced Percentage of Waste Cooking Oil & Tire Rubber with Bagasse Ash as Modifier. *Civ. Eng. J.* **2019**, *5*, 587. [\[CrossRef\]](#)
31. Adesina, P.A.; Dahunsi, B.I. Blended waste utilization in road construction: Physical characteristics of bitumen modified with waste cooking oil and high-density polyethylene. *Int. J. Pavement Res. Technol.* **2021**, *14*, 98–104. [\[CrossRef\]](#)
32. Jamil, F.; Al-Muhtaseb, A.; Myint, M.T.Z.; Al-Hinai, M.; Al-Haj, L.; Baawain, M.; Al-Abri, M.; Kumar, G.; Atabani, A. Biodiesel production by valorizing waste Phoenix dactylifera L. Kernel oil in the presence of synthesized heterogeneous metallic oxide catalyst (Mn@MgO-ZrO₂). *Energy Convers. Manag.* **2018**, *155*, 128–137. [\[CrossRef\]](#)
33. Safari, A.; Salamat, R.; Baik, O.-D. A review on heat and mass transfer coefficients during deep-fat frying: Determination methods and influencing factors. *J. Food Eng.* **2018**, *230*, 114–123. [\[CrossRef\]](#)
34. Koh, E.; Surh, J. Food types and frying frequency affect the lipid oxidation of deep frying oil for the preparation of school meals in Korea. *Food Chem.* **2015**, *174*, 467–472. [\[CrossRef\]](#) [\[PubMed\]](#)
35. Cárdenas, J.; Orjuela, A.; Sánchez, D.L.; Narváez, P.C.; Katryniok, B.; Clark, J. Pre-treatment of used cooking oils for the production of green chemicals: A review. *J. Clean. Prod.* **2021**, *289*, 125129. [\[CrossRef\]](#)
36. Daud, M.S.M.; Ngadiman, N.I.; Suliman, M.S. The awareness of recycling the used of cooking oil. *J. Crit. Rev.* **2020**, *7*, 30–32. [\[CrossRef\]](#)
37. Choe, E.; Min, D.B. Chemistry of Deep-Fat Frying Oils. *J. Food Sci.* **2007**, *72*, R77–R86. [\[CrossRef\]](#)
38. Tomasevic, A.; Siler-Marinkovic, S. Methanolysis of used frying oil. *Fuel Process. Technol.* **2003**, *81*, 1–6. [\[CrossRef\]](#)
39. Wen, Z.; Yu, X.; Tu, S.-T.; Yan, J.; Dahlquist, E. Biodiesel production from waste cooking oil catalyzed by TiO₂-MgO mixed oxides. *Bioresour. Technol.* **2010**, *101*, 9570–9576. [\[CrossRef\]](#)
40. Demirbas, A. Biodiesel from waste cooking oil via base-catalytic and supercritical methanol transesterification. *Energy Convers. Manag.* **2009**, *50*, 923–927. [\[CrossRef\]](#)
41. Phan, A.N.; Phan, T.M. Biodiesel production from waste cooking oils. *Fuel* **2008**, *87*, 3490–3496. [\[CrossRef\]](#)
42. Ivanov Jordanov, D.; Stoyanov Petkov, P.; Kirov Dimitrov, Y.; Kunev Ivanov, S. Methanol transesterification of different vegetable oils. *Pet. Coal* **2007**, *2*, 21–23.
43. Shahidi, F. Quality characteristics of edible oils. *Adv. Exp. Med. Biol.* **2004**, *542*, 239–249. [\[CrossRef\]](#)
44. Naz, S.; Siddiqi, R.; Sheikh, H.; Sayeed, S.A. Deterioration of olive, corn and soybean oils due to air, light, heat and deep-frying. *Food Res. Int.* **2005**, *38*, 127–134. [\[CrossRef\]](#)
45. Zhang, Q.; Saleh, A.S.; Chen, J.; Shen, Q. Chemical alterations taken place during deep-fat frying based on certain reaction products: A review. *Chem. Phys. Lipids* **2012**, *165*, 662–681. [\[CrossRef\]](#)
46. Ahmed, R.B.; Hossain, K. Waste cooking oil as an asphalt rejuvenator: A state-of-the-art review. *Constr. Build. Mater.* **2020**, *230*, 116985. [\[CrossRef\]](#)
47. Azahar, W.N.A.W.; Jaya, R.P.; Hainin, M.R.; Bujang, M.; Ngadi, N. Mechanical performance of asphaltic concrete incorporating untreated and treated waste cooking oil. *Constr. Build. Mater.* **2017**, *150*, 653–663. [\[CrossRef\]](#)
48. Zhang, D.; Chen, M.; Wu, S.; Liu, J.; Amirkhanian, S. Analysis of the Relationships between Waste Cooking Oil Qualities and Rejuvenated Asphalt Properties. *Materials* **2017**, *10*, 508. [\[CrossRef\]](#)
49. Zargar, M.; Ahmadiania, E.; Asli, H.; Karim, M.R. Investigation of the possibility of using waste cooking oil as a rejuvenating agent for aged bitumen. *J. Hazard. Mater.* **2012**, *233–234*, 254–258. [\[CrossRef\]](#)
50. Oldham, D.; Rajib, A.; Dandamudi, K.P.R.; Liu, Y.; Deng, S.; Fini, E.H. Transesterification of Waste Cooking Oil to Produce A Sustainable Rejuvenator for Aged Asphalt. *Resour. Conserv. Recycl.* **2021**, *168*, 105297. [\[CrossRef\]](#)
51. Lam, M.K.; Lee, K.T.; Mohamed, A.R. Homogeneous, heterogeneous and enzymatic catalysis for transesterification of high free fatty acid oil (waste cooking oil) to biodiesel: A review. *Biotechnol. Adv.* **2010**, *28*, 500–518. [\[CrossRef\]](#)
52. Cvengroš, J.; Cvengrošová, Z. Used frying oils and fats and their utilization in the production of methyl esters of higher fatty acids. *Biomass-Bioenergy* **2004**, *27*, 173–181. [\[CrossRef\]](#)
53. Yuan, X.; Liu, J.; Zeng, G.; Shi, J.; Tong, J.; Huang, G. Optimization of conversion of waste rapeseed oil with high FFA to biodiesel using response surface methodology. *Renew. Energy* **2008**, *33*, 1678–1684. [\[CrossRef\]](#)
54. Ladhe, A.R.; Krishna Kumar, N.S. *Application of Membrane Technology in Vegetable Oil Processing*, 1st ed.; Elsevier Ltd.: Amsterdam, The Netherlands, 2010. ISBN 9781856176323. Available online: https://www.researchgate.net/publication/286013694_Application_of_Membrane_Technology_in_Vegetable_Oil_Processing (accessed on 14 October 2021).
55. Vaisali, C.; Charanyaa, S.; Belur, P.; Regupathi, I. Refining of edible oils: A critical appraisal of current and potential technologies. *Int. J. Food Sci. Technol.* **2014**, *50*, 13–23. [\[CrossRef\]](#)
56. Koris, A.; Vatai, G. Dry degumming of vegetable oils by membrane filtration. *Desalination* **2002**, *148*, 149–153. [\[CrossRef\]](#)
57. Yang, Z.; Peng, H.; Wang, W.; Liu, T. Crystallization behavior of poly(ϵ -caprolactone)/layered double hydroxide nanocomposites. *J. Appl. Polym. Sci.* **2010**, *116*, 2658–2667. [\[CrossRef\]](#)
58. Clark, J.G.; Mountain, S.; Us, T.N. Filtration and Filtration Method for Cooking Oil Used in Frying Process. U.S. Patent US20070289927A1, 20 December 2007.
59. Ma, F.; Hanna, M.A. Biodiesel production: A review. *Bioresour. Technol.* **1999**, *70*, 1–15. [\[CrossRef\]](#)
60. Enweremadu, C.; Mbarawa, M. Technical aspects of production and analysis of biodiesel from used cooking oil—A review. *Renew. Sustain. Energy Rev.* **2009**, *13*, 2205–2224. [\[CrossRef\]](#)

61. Atapour, M.; Kariminia, H.-R. Characterization and transesterification of Iranian bitter almond oil for biodiesel production. *Appl. Energy* **2011**, *88*, 2377–2381. [CrossRef]
62. Babcock, R.E.; Clausen, E.C.; Popp, M.P.; Schulte, W.B. Yield Characteristics of Biodiesel Produced from Chicken Fat-Tall Oil Blended Feedstocks. 2008. Available online: <https://trid.trb.org/view/850767> (accessed on 14 October 2021).
63. Talebian-Kiakalaieh, A.; Amin, N.A.S.; Mazaheri, H. A review on novel processes of biodiesel production from waste cooking oil. *Appl. Energy* **2013**, *104*, 683–710. [CrossRef]
64. Jeromin, L.; Peukert, E.; Wollmann, G. Process for the Pre-Esterification of Free Fatty Acids in Fats and Oils. U.S. Patent US4698186A, 6 October 1987.
65. Zhang, Y.; Dubé, M.; McLean, D.; Kates, M. Biodiesel production from waste cooking oil: 1. Process design and technological assessment. *Bioresour. Technol.* **2003**, *89*, 1–16. [CrossRef]
66. Sahoo, P.; Das, L.; Babu, M.; Naik, S.N. Biodiesel development from high acid value polanga seed oil and performance evaluation in a CI engine. *Fuel* **2007**, *86*, 448–454. [CrossRef]
67. Crabbe, E.; Nolasco-Hipolito, C.; Kobayashi, G.; Sonomoto, K.; Ishizaki, A. Biodiesel production from crude palm oil and evaluation of butanol extraction and fuel properties. *Process. Biochem.* **2001**, *37*, 65–71. [CrossRef]
68. Baldwin, D.; Barr, V.; Briggs, A.; Havill, J.; Maxwell, B.; Walker, H.M. Variables Affecting the Yields of Fatty Esters from Transesterified Vegetable Oils 1. *J. Am. Oil Chem. Soc.* **1984**, *61*, 677–678. [CrossRef]
69. Wang, Y.; Ou, S.; Liu, P.; Xue, F.; Tang, S. Comparison of two different processes to synthesize biodiesel by waste cooking oil. *J. Mol. Catal. A Chem.* **2006**, *252*, 107–112. [CrossRef]
70. Zhang, H.; Ding, J.; Zhao, Z. Microwave assisted esterification of acidified oil from waste cooking oil by CERP/PES catalytic membrane for biodiesel production. *Bioresour. Technol.* **2012**, *123*, 72–77. [CrossRef]
71. Lertsathapornasuk, V.; Pairintra, R.; Aryasuk, K.; Krisnangkura, K. Microwave assisted in continuous biodiesel production from waste frying palm oil and its performance in a 100 kW diesel generator. *Fuel Process. Technol.* **2008**, *89*, 1330–1336. [CrossRef]
72. Supraja, K.V.; Behera, B.; Paramasivan, B. Optimization of process variables on two-step microwave-assisted transesterification of waste cooking oil. *Environ. Sci. Pollut. Res.* **2020**, *27*, 27244–27255. [CrossRef]
73. Refaat, A.A.; El Sheltawy, S.T.; Sadek, K.U. Optimum reaction time, performance and exhaust emissions of biodiesel produced by microwave irradiation. *Int. J. Environ. Sci. Technol.* **2008**, *5*, 315–322. [CrossRef]
74. Palanisamy, K.; Idlan, M.K.; Saifudin, N. Preliminary evaluation of the effectiveness of moisture removal and energy usage in pretreatment module of waste cooking oil for biodiesel production. *IOP Conf. Ser. Earth Environ. Sci.* **2013**, *16*, 12053. [CrossRef]
75. Dias, J.M.; Alvim-Ferraz, M.C.; Almeida, M.F. Comparison of the performance of different homogeneous alkali catalysts during transesterification of waste and virgin oils and evaluation of biodiesel quality. *Fuel* **2008**, *87*, 3572–3578. [CrossRef]
76. Felizardo, P.; Correia, M.J.N.; Raposo, I.; Mendes, J.F.; Berkemeier, R.; Bordado, J. Production of biodiesel from waste frying oils. *Waste Manag.* **2006**, *26*, 487–494. [CrossRef]
77. Issariyakul, T.; Kulkarni, M.G.; Dalai, A.K.; Bakhshi, N.N. Production of biodiesel from waste fryer grease using mixed methanol/ethanol system. *Fuel Process. Technol.* **2007**, *88*, 429–436. [CrossRef]
78. Su, N.; Xiao, F.; Wang, J.; Cong, L.; Amirkhanian, S. Productions and applications of bio-asphalts—A review. *Constr. Build. Mater.* **2018**, *183*, 578–591. [CrossRef]
79. Su, J.-F.; Qiu, J.; Schlangen, E.; Wang, Y.-Y. Investigation the possibility of a new approach of using microcapsules containing waste cooking oil: In situ rejuvenation for aged bitumen. *Constr. Build. Mater.* **2015**, *74*, 83–92. [CrossRef]
80. Wang, H.N.; Gao, J.F.; You, Z.P.; Tian, Y.Y. Advances in bio-binder application on road pavement. *Wuhan Ligong Daxue Xuebao/J. Wuhan Univ. Technol.* **2014**, *36*, 55–60. [CrossRef]
81. Luo, W.; Zhang, Y.; Cong, P. Investigation on physical and high temperature rheology properties of asphalt binder adding waste oil and polymers. *Constr. Build. Mater.* **2017**, *144*, 13–24. [CrossRef]
82. Somé, S.C.; Pavoine, A.; Chailleux, E. Evaluation of the potential use of waste sunflower and rapeseed oils-modified natural bitumen as binders for asphalt pavement design. *Int. J. Pavement Res. Technol.* **2016**, *9*, 368–375. [CrossRef]
83. Al-Omari, A.A.; Khedaywi, T.S.; Khasawneh, M.A. Laboratory characterization of asphalt binders modified with waste vegetable oil using SuperPave specifications. *Int. J. Pavement Res. Technol.* **2018**, *11*, 68–76. [CrossRef]
84. Wang, C.; Xue, L.; Xie, W.; You, Z.; Yang, X. Laboratory investigation on chemical and rheological properties of bio-asphalt binders incorporating waste cooking oil. *Constr. Build. Mater.* **2018**, *167*, 348–358. [CrossRef]
85. Eriskin, E.; Karahancer, S.; Terzi, S.; Saltan, M. Waste frying oil modified bitumen usage for sustainable hot mix asphalt pavement. *Arch. Civ. Mech. Eng.* **2017**, *17*, 863–870. [CrossRef]
86. Rasman, M.; Hassan, N.A.; Hainin, M.R.; Jaya, R.P.; Haryati, Y.; Shukry, N.A.M.; Abdullah, M.E.; Kamaruddin, N.H.M. Engineering properties of bitumen modified with bio-oil. *MATEC Web Conf.* **2018**, *250*, 02003. [CrossRef]
87. Çavdar, E.; Kumandaş, A.; Oruç, Ş. Effect of waste cooking oil use in the modification of sbs modified asphalt binder. In Proceedings of the 3rd International Conference on Advanced Engineering Technologies, Bayburt, Turkey, 19–21 September 2019.
88. Maharaj, R.; Ramjattan-Harry, V.; Mohamed, N. Rutting and Fatigue Cracking Resistance of Waste Cooking Oil Modified Trinidad Asphaltic Materials. *Sci. World J.* **2015**, *2015*, 1–7. [CrossRef] [PubMed]
89. Bilema, M.; Aman, M.Y.; Hassan, N.A.; Abdullah, N.F.A. Investigation on rheology and physical properties of asphalt binder blended with waste cooking oil. *IOP Conf. Ser. Mater. Sci. Eng.* **2019**, *527*, 012045. [CrossRef]

90. Nordiana, M.; Aifa, W.A.W.N.; Hainin, M.R.; Naquiuddin, M.W.M.; Norhidayah, A.H.; Haryati, Y.; Juraidah, A.; Ramadhansyah, P.J. Rutting resistance of untreated and treated waste cooking oil in bitumen after aging condition. *IOP Conf. Ser. Earth Environ. Sci.* **2019**, *244*, 012041. [\[CrossRef\]](#)
91. Joni, H.H.; Al-Rubae, R.H.A.; Al-Zerkani, M.A. Characteristics of asphalt binder modified with waste vegetable oil and waste plastics. *IOP Conf. Ser. Mater. Sci. Eng.* **2020**, *737*, 012126. [\[CrossRef\]](#)
92. Rahman, T.; Hainin, M.R.; Abu Bakar, W.A.W. Use of waste cooking oil, tire rubber powder and palm oil fuel ash in partial replacement of bitumen. *Constr. Build. Mater.* **2017**, *150*, 95–104. [\[CrossRef\]](#)
93. Saboo, N.; Sukhija, M.; Singh, G. Effect of Nanoclay on Physical and Rheological Properties of Waste Cooking Oil–Modified Asphalt Binder. *J. Mater. Civ. Eng.* **2021**, *33*, 04020490. [\[CrossRef\]](#)
94. Liu, S.; Zhou, S.; Peng, A.; Li, W. Investigation of physiochemical and rheological properties of waste cooking oil/SBS/EVA composite modified petroleum asphalt. *J. Appl. Polym. Sci.* **2019**, *137*, 1–13. [\[CrossRef\]](#)
95. Dahunsi, B.I.; Adesina, P.A. Properties of Bitumen Modified with Waste Cooking Oil and High Density Polyethylene for Applications in Flexible Pavement. In Proceedings of the 1st International Conference on Construction Materials for Sustainable Future, Zadar, Croatia, 19–21 April 2017.
96. Mahrez, A.; Karim, M.R.; Ibrahim, M.R.; Katman, H.Y. Prospects of using waste cooking oil as rejuvenating agent in bituminous binder. *Proc. East. Asia Soc. Transp. Stud.* **2009**, *7*, 1751–1766.
97. Sun, Z.; Yi, J.; Huang, Y.; Feng, D.; Guo, C. Properties of asphalt binder modified by bio-oil derived from waste cooking oil. *Constr. Build. Mater.* **2016**, *102*, 496–504. [\[CrossRef\]](#)
98. Cao, W.; Wang, Y.; Wang, C. Fatigue characterization of bio-modified asphalt binders under various laboratory aging conditions. *Constr. Build. Mater.* **2019**, *208*, 686–696. [\[CrossRef\]](#)
99. Gong, M.; Yang, J.; Zhang, J.; Zhu, H.; Tong, T. Physical–chemical properties of aged asphalt rejuvenated by bio-oil derived from biodiesel residue. *Constr. Build. Mater.* **2016**, *105*, 35–45. [\[CrossRef\]](#)
100. Zhu, H.; Xu, G.; Gong, M.; Yang, J. Recycling long-term-aged asphalts using bio-binder/plasticizer-based rejuvenator. *Constr. Build. Mater.* **2017**, *147*, 117–129. [\[CrossRef\]](#)
101. Hong, W.; Mo, L.; Pan, C.; Riara, M.; Wei, M.; Zhang, J. Investigation of rejuvenation and modification of aged asphalt binders by using aromatic oil-SBS polymer blend. *Constr. Build. Mater.* **2020**, *231*, 117154. [\[CrossRef\]](#)
102. Nassar, I.M.; Abdel-Monem, R.M.; Sayed, M.A.; Ragab, A.A. Eco-friendly green alternative binder for asphalt from waste oil and waste polymer. *Polym. Bull.* **2020**, 1–23. [\[CrossRef\]](#)
103. Jia, X.; Huang, B.; Bowers, B.F.; Zhao, S. Infrared spectra and rheological properties of asphalt cement containing waste engine oil residues. *Constr. Build. Mater.* **2014**, *50*, 683–691. [\[CrossRef\]](#)
104. Zahoor, M.; Nizamuddin, S.; Madapusi, S.; Giustozzi, F. Recycling asphalt using waste bio-oil: A review of the production processes, properties and future perspectives. *Process. Saf. Environ. Prot.* **2021**, *147*, 1135–1159. [\[CrossRef\]](#)
105. Guarin, A.; Khan, A.; Butt, A.A.; Birgisson, B.; Kringos, N. An extensive laboratory investigation of the use of bio-oil modified bitumen in road construction. *Constr. Build. Mater.* **2016**, *106*, 133–139. [\[CrossRef\]](#)
106. Zaumanis, M.; Mallick, R.B.; Poulidakos, L.; Frank, R. Influence of six rejuvenators on the performance properties of Reclaimed Asphalt Pavement (RAP) binder and 100% recycled asphalt mixtures. *Constr. Build. Mater.* **2014**, *71*, 538–550. [\[CrossRef\]](#)
107. Fini, E.H.; Samieadel, A.; Rajib, A. Moisture Damage and Its Relation to Surface Adsorption/Desorption of Rejuvenators. *Ind. Eng. Chem. Res.* **2020**, *59*, 13414–13419. [\[CrossRef\]](#)
108. Hung, A.M.; Pahlavan, F.; Shakiba, S.; Chang, S.L.Y.; Louie, S.M.; Fini, E.H. Preventing Assembly and Crystallization of Alkane Acids at the Silica–Bitumen Interface To Enhance Interfacial Resistance to Moisture Damage. *Ind. Eng. Chem. Res.* **2019**, *58*, 21542–21552. [\[CrossRef\]](#)
109. Chen, M.; Leng, B.; Wu, S.; Sang, Y. Physical, chemical and rheological properties of waste edible vegetable oil rejuvenated asphalt binders. *Constr. Build. Mater.* **2014**, *66*, 286–298. [\[CrossRef\]](#)
110. Azahar, W.N.A.W.; Bujang, M.; Jaya, R.P.; Hainin, M.R.; Ngadi, N.; Mohamad, M. Chemical Identification of Waste Cooking Oil as Additive in Bitumen. *Key Eng. Mater.* **2016**, *700*, 207–215. [\[CrossRef\]](#)