# Construction and Building Materials 220 (2019) 228-237

Contents lists available at ScienceDirect

# **Construction and Building Materials**

journal homepage: www.elsevier.com/locate/conbuildmat

# Investigation of waste oils as rejuvenators of aged bitumen for sustainable pavement

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# HIGHLIGHTS

• Waste cooking and engine oils (WCO, WEO) were investigated as bitumen rejuvenators.

• Optimum percentages of the waste oils were 3.5% for the WCO and 5.5% for the WEO.

• Rejuvenated bitumen showed less tendency to short-term aging based on DSR results.

• Rejuvenated bitumen by both WCO and WEO had a performance grade of PG 64-28.

• Rejuvenated 100% RAP mixes cope with Egyptian limits for stability, flow, and ITS.

#### ARTICLE INFO

Article history: Received 27 November 2018 Received in revised form 26 May 2019 Accepted 29 May 2019 Available online 10 June 2019

Keywords:

Waste Cooking Oil (WCO) Waste Engine Oil (WEO) Reclaimed Asphalt Pavement (RAP) Dynamic Shear Rheometer (DSR) Bending Beam Rheometer (BBR) Fourier Transform Infrared Spectroscopy (FTIR) Scanning Electron Microscopy (SEM) and Energy Dispersive X-ray (EDX) Indirect Tensile Strength (ITS)

# ABSTRACT

For sustainable pavement construction, this research paper aims to investigate the feasibility of using Waste Cooking Oil (WCO) and Waste Engine Oil (WEO) as rejuvenators on the rheological properties of aged bitumen extracted from Reclaimed Asphalt Pavement (RAP). The aged bitumen was extracted from milled RAP recruited from an old pavement. The rheological characteristics of the rejuvenated bitumen were determined by penetration, softening point, Brookfield viscosity, Dynamic Shear Rheometer (DSR), and Bending Beam Rheometer (BBR) tests. In addition, the chemical composition of virgin (control), aged and rejuvenated bitumen by WCO, and WEO was investigated using Fourier Transform Infrared Spectroscopy (FTIR) and Energy Dispersive X-ray (EDX). The quality of the rejuvenated bitumen was also evaluated by the Scanning Electron Microscopy (SEM) imaging technique. Based on penetration and softening point testing results, the optimum percentages of the waste oils were found to range from 3.5 to 4.0% for the WCO and from 5.5 to 6.0% for the WEO. The aged bitumen properties were significantly improved by rejuvenators as evidenced by the chemical analysis (FTIR and SEM/EDX) along with the ratio of asphaltenes to maltenes. Furthermore, the surface morphology was renovated as well as the fundamental physical properties of the rejuvenated aged bitumen. Additionally, the rejuvenated bitumen showed less tendency to short-term aging as confirmed by the DSR results. BBR results of rejuvenated aged bitumen samples exhibited comparable performance to those of the control samples at different low temperatures having a performance grade of PG 64-28. Moreover, rejuvenated 100% RAP mixes were found to cope with Egyptian requirements for heavy traffic as binder courses and medium traffic as wearing courses in terms of Marshall stability and flow. Finally, Indirect Tensile Strength (ITS) results were within the specification limits.

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# 1. Introduction

Despite the limited resources, about 110 million tons of bitumen are used annually in the highway industry worldwide [1].

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Bitumen is an organic hydrocarbon material with high thermal sensitivity. Exposure to elevated temperatures during mixing and placement of the Hot Mix Asphalt (HMA), and the environmental conditions throughout the pavement service life, lead to aging of the asphalt binder. The loss of volatiles and oxidation of bitumen (as bitumen is a hydrocarbon material) are the main causes of aging, which result in higher viscosity and stiffer bitumen [2]. Aged bitumen tends to lose its flexibility and turns into a brittle material that is prone to crack easily [3].







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There has been an enormous increase in the use of Reclaimed Asphalt Pavement (RAP) worldwide as an economic and environmentally sound alternative for sustainable pavement construction [4–6]. However, some road authorities are hesitant in encouraging the use of RAP in pavement construction on a large scale due to some limitations. The barriers that limit the use of high RAP percentages (more than 15%) are the high variability in milling processes, mixing RAP from different sources, different properties of aged bitumen than required for virgin bitumen, in addition to one more additional material to care about during HMA manufacturing [6,7]. These factors may lead to early pavement distresses when using high percentages of RAP due to low content of bitumen, inferior quality of bitumen due to aging, mixture degradation, and oversized RAP. Therefore, rejuvenating the hard bitumen in the RAP has become an attractive option to the use of RAP in asphalt mixes This is especially true in case of incorporating higher RAP percentages [7].

Several investigations i.e., [8] showed that the binder and aggregate from an old HMA are still valuable even if the life cycle of HMA has reached its end. Nevertheless, aging of bitumen remains the problem that hindered the application of RAP for years. In order to restore some of the mechanical properties of the aged bitumen, rejuvenators may be used. Rejuvenators are classified into rejuvenating and softening agents [1]. They recover the original ratio of asphaltenes to maltenes in aged binder, soften the aged bitumen and replenish the volatiles, and disperse oils while promoting adhesion [3,9]. In recent years, the application of rejuvenators in RAP was found to considerably increase in HMA mixtures, and chip-seals to rejuvenate the aged bitumen [10].

Recently, owing to the high demand on rejuvenators, application of waste products such as Waste Cooking Oil (WCO) and Waste Engine Oil (WEO) as rejuvenators, has been sought [8]. WCO can be collected from different places such as, restaurants, food industry, and house-hold disposable and recycling centers [11]. There are different uses of WCO such as bio-diesel yellow grease, animal food, and soaps [12,13]. The problem is that only small amounts of WCO are properly collected and recycled, and the majority are illegally dumped into rivers and landfills causing pollution and environmental problems [14].

On the other hand, WEO is removed from the vehicle during a routine oil change in local auto repair shops. Small amounts of WEO if well blended with RAP may become beneficial reducing the hardening and stiffness of aged bitumen. As a common practice, automobile repair shops collect WEO residues from different vehicles, which usually contain contaminants from the wear of engine, heating, and oxidation of lubricating oil during the engine operation [15]. The properties of WEO depend on the combustion process, operation temperature, and contaminant sources such as moisture, soot, diluents, rust, detergents and engine wear metal particles.

Most of the previous studies i.e. [16,17-19] were based on routine testing to determine the physical properties of the rejuvenated bitumen rather than advanced testing to better assess the rheological and chemical properties. In addition, these studies recommended significantly different amounts of WCO in the range of 1-4%, while for the WEO, the recommended amounts were in the range of 4-8%. Thus, the rejuvenator amount depends on the type and condition of the aged bitumen.

### 2. Objectives and scope of work

The main objective of this research is to investigate the effect of adding WCO and WEO as rejuvenators to aged bitumen from RAP and find out the optimum percentage of both rejuvenators based on the local materials. A secondary objective is to find the best potential alternative from the two rejuvenators that improves the characteristics of the aged bitumen giving the best performance.

In this paper, WCO and WEO were evaluated as rejuvenators to local aged bitumen extracted from an old pavement in order to restore its virgin properties. These properties were compared side by side to the virgin (control) bitumen. The testing program includes conventional binder testing such as penetration and softening point temperature and advanced rheological testing such as Brookfield viscosity, Dynamic Shear Rheometer (DSR), and Bending Beam Rheometer (BBR). In addition, advanced chemical and physical characterization tests were conducted in order to understand the role of rejuvenator. These tests are Fourier Transform Infrared (FTIR) spectroscopy, Scanning Electron Microscopy (SEM), and Energy Dispersive X-ray (EDX). Finally, in order to study the effect of rejuvenator on the mix properties, Marshall stability, flow and



(a) Extraction equipment



(b) Distillation equipment

Fig. 1. Extraction and distillation equipment.

Indirect Tensile Strength (ITS) tests were conducted on a100% RAP mix and a rejuvenated 100% RAP mix.

#### 3. Materials

In this study, a 60/70 penetration-grade virgin bitumen sourced from Suez Oil Processing Company (SOPC) as control bitumen was used. RAP samples were collected from Mansoura-Damietta road with a service life of about 16 years. RAP samples were extracted by the recovery method using the methylene chloride solvent according to ASTM D2172 [20] and distilled to remove any remnants of the solvent. Fig. 1 shows the extraction and distillation equipment. The WCO was sourced from a food factory located along Port Said-Damietta road, whereas the WEO was collected from local auto service shops.

# 4. Methodology and experimental work

After extraction and recovery of the aged bitumen (AB), WCO was applied to the extracted AB at different percentages of 2.0, 3.0, 3.5 and 4% by the total weight of bitumen, while for WEO

the percentages were 2.0, 3.0, 4.0, 5.0, 5.5, and 6.0%. The chemical, rheological, and physical properties of control bitumen, AB, and rejuvenated bitumen were determined through: penetration, softening point temperature, Brookfield viscometer, ratio of asphaltenes to maltenes, DSR, BBR, FTIR, and SEM/EDX. In addition, three asphalt mixes, 100% original RAP, and two rejuvenated 100% RAP mixes by WCO and WEO at optimum percentages, were prepared for testing in Marshall to determine stability, flow, and bulk specific gravity. Furthermore, the ITS test was conducted to determine Tensile Strength Ratio (TSR) and fracture energy. Fig. 2 summarizes the details of the experimental program conducted in this research.

DSR tests were conducted on control and rejuvenated aged bitumen by WCO and WEO to measure the bitumen complex shear modulus (G<sup>\*</sup>) and phase angle ( $\delta$ ) at intermediate and high temperatures in accordance with the ASTM D7175 [21]. For control and rejuvenated aged binders by WCO and WEO, DSR tests were performed on unaged, short-term aged bitumen specimens using the Rolling Thin Film Oven (RTFO) in accordance with the ASTM D2872 [22] and long-term aged bitumen specimens using the Pressure Aging Vessel (PAV) according to ASTM D6521 [23]. For the extracted AB, the DSR samples were not subjected to any RTFO or PAV aging and were tested as-is in its current state for compar-



Fig. 2. Experimental program flowchart.

ison only with other investigated binders. For the unaged and RTFO aged bitumen, 1 mm thick and 25 mm diameter specimens were prepared, while for PAV aged samples, 2 mm thick by 8 mm diameter specimens were used. G\* and  $\delta$  were measured for the unaged and RTFO aged bitumen specimens at high temperatures starting from 52 °C until specimen failure. For PAV specimens, the DSR test started at intermediate temperature of 31 °C, then the temperature was decreased until specimen failure. The DSR oscillation rate was 10 rad/s (1.59 Hz).

BBR tests were used to measure the bitumen flexural creep stiffness and relaxation properties at low temperatures according to the ASTM D6648-08 [24]. BBR tests were conducted on control and rejuvenated aged bitumen by WCO and WEO on long term aged samples using PAV to determine the bitumen performance after exposure to service life aging. After RTFO then PAV aging, bitumen was heated with minimal time to prevent any hardening, then poured into aluminum beams with dimension of  $127 \times 6.35 \times 12.7$  mm. Specimens were conditioned and tested at four different low temperatures of 0, -6, -12, and -18 °C before applying a mid-point constant load of 0.98 N at the center of the beam, which was supported at two points 102 mm apart. The center deflection was measured over 4 min, then the creep stiffness and creep (m-value) were calculated.

Mixture testing was also conducted in order to quantify the effect of the rejuvenator on the mixture mechanical properties as well as moisture damage. Three different mixtures, 100% RAP, 100% RAP with 3.5% WCO by bitumen weight and 100% RAP with 5.5% WEO by bitumen weight were prepared. Nine Marshall specimens (three replicates for each mixture) were prepared to determine stability, flow, and the bulk specific gravity. Before testing, the samples were placed into a water bath at 60 °C for 30 min [25]. In addition, twelve more specimens were prepared at about 7% air voids for the ITS testing in order to evaluate the asphalt mixes sensitivity to moisture according to the AASHTO T283 [26]. One set of samples was tested dry, while the other set was tested in wet condition. The dry set samples for each mix were kept in environmental chamber at 25 °C before the beginning of the test. The wet set samples were placed in a water bath at 60 °C for 24 h, and then they were left for 2 h in a water bath at 25 °C. Both sets of specimens were tested at a loading rate of 50 mm/minute by loading the specimens with Marshall apparatus using a specific head parallel to the vertical diametric plane of specimen.

#### 5. Results and discussion

#### 5.1. Bitumen traditional testing

The effect of adding WCO and WEO to aged bitumen on the penetration values is illustrated in Fig. 3-a, and Fig. 3-b, respectively. It is observed from both figures that the penetration increased linearly as the amount of WCO and WEO increased to reach the target penetration grade of 60/70. The increase in penetration is due to the decrease in the ratio of asphaltenes to maltenes [7,27] as illustrated in Table 1. Asphaltenes to maltenes ratio was determined in accordance with the ASTM D4124 [27] by the solvent extraction method using n-heptane. The maltenes was then fractionated by the liquid-solid chromatography method [27]. Based on the penetration testing with a target grade of 60/70, the optimum percentage of WCO was found to range from 3.5 to 4%, while it was 5.5–6% for WEO.

The softening point test was conducted in accordance with ASTM D36 [28]. Fig. 4-a, and Fig. 4-b show the softening point temperatures for control, AB, and WCO and WEO rejuvenated bitumen. The softening point temperature of AB decreased with the addition



a) Control bitumen and different WCO contents



b) Control bitumen and different WEO contents

Fig. 3. Penetration of control bitumen compared to WCO and WEO rejuvenated bitumen.

#### Table 1

Asphaltenes to maltenes of control bitumen, aged bitumen and rejuvenated bitumen by WCO and WEO.

Material	Asphaltenes (%)	Maltenes (%)	Asphaltenes/ Maltenes (%)
Aged bitumen (AB)	43.4%	56.6%	76.67%
Control bitumen	23.0%	77.0%	29.87%
AB + 3.5%WCO	21.0%	79.0%	26.58%
AB + 5.5% WEO	22.2%	77.8%	28.53%

of both WCO and WEO contents to resemble control bitumen (minimum required is 45 °C) as presented in both figures. The addition of rejuvenators decreased the asphaltenes proportion with high molecular weight leading to softer bitumen. Based on penetration and softening point testing results, the optimum contents that achieved the Egyptian requirements and resembled the physical properties of control bitumen were 3.5% for WCO and 5.5% for WEO.

Fig. 5-a, and Fig. 5-b show the Brookfield viscosity results of the investigated bitumen with different percentages of WCO and WEO at different temperatures. As expected, the aged bitumen showed the highest viscosity values, while the addition of 3.5% WCO and 5.5% WEO into the aged bitumen led to similar viscosity values to that of the control bitumen. Hence, the same workability was accomplished by the renovation process.



a) Control bitumen and different WCO rejuvenated bitumen



b) Control bitumen and different WCO rejuvenated bitumen

Fig. 4. Softening point temperature of control bitumen and different WCO and WEO rejuvenated bitumen.

Table 2 summarizes the ASTM Ai and VTSi values for the control, aged and rejuvenated aged bitumen at the optimum contents of the rejuvenators according to the ASTM D4402 [29]. The Ai-VTSi values of the rejuvenated aged bitumen samples were lower than the AB and in fact they were comparable to the control bitumen indicating higher temperature susceptibility and lower viscosity values compared to the aged bitumen.

#### 5.2. DSR testing results

Fig. 6-a shows that the rutting parameter ( $G^*/\sin \delta$ ) of the AB at tested temperatures was much higher than those for control bitumen owing to aging over the pavement service life (about 16 years). By adding 3.5% WCO (the optimum percentage) to AB, the  $G^*/\sin \delta$  values were improved (softened) and became almost identical to the  $G^*/\sin \delta$  values of the control bitumen. On the other hand, with the addition of the 5.5% WEO, the  $G^*/\sin \delta$  values were lower than those of the control bitumen and became more susceptible to rutting.

Fig. 6-b emphasizes the same trend as in Fig. 6-a with a slight change for RTFO sample of AB + 3.5% WCO. The G\*/sin  $\delta$  values of the AB + 5.5 WEO were lower than those obtained for control bitumen indicating that this waste can be used only for lower traffic volumes. To determine the PG of the bitumen, the rutting parameter, G\*/sin  $\delta$  must be greater than 1.0 kPa and 2.2 kPa for original and RTFO samples, respectively. Based on that, the PG grade was found to be PG 64-xx for control, AB + 3.5% WCO, and AB + 5.5% WEO. Fig. 6-c shows that by adding 3.5% WCO, the rutting failure temperature of the rejuvenated bitumen became similar to that



a) Control bitumen, aged bitumen and WCO rejuvenated bitumen



Fig. 5. Brookfield viscosity of control bitumen, aged bitumen and rejuvenated aged bitumen with different contents of WCO and WEO.

of the control bitumen. Fig. 6-d shows that the WCO rejuvenated aged bitumen exhibited the lowest values of the fatigue parameter (G\*sin  $\delta$ ) indicating better resistance to fatigue cracking.

#### 5.3. BBR testing results

Table 3 summarizes the low temperature performance in terms of creep stiffness and creep m-value for the control and rejuvenated bitumen by WCO and WEO at the four tested low temperatures (0, -6, -12, and -18 °C). It can be noticed from the data that the creep stiffness of all tested rejuvenated bitumen was generally low, and the creep rate was higher than or equal to 0.30. All rejuvenated bitumen samples tested at all temperatures as well as the control bitumen passed the Superpave binder specification criteria (stiffness < 300 MPa and m-value > 0.3) [24]. Consequently, the

Table 2

ASTM Ai-VTSi values for the control, aged and rejuvenated aged bitumen at optimum contents.

	Regression intercept (Ai)	VTSi
AB	10.187	-3.4111
Control Bitumen (60/70)	8.799	-2.960
AB + 3.5% WCO	8.827	-2.921
AB + 5.5% WEO	8.925	-2.942



 a) G\*/Sin δ values for original samples of control, aged, and rejuvenated aged bitumen with different contents of WCO and WEO



rejuvenated aged bitumen by WCO and WEO



c) Failure temperature of control and aged rejuvenated bitumen by WCO and WEO



d) Fatigue Parameter (G\*. Sinδ) for PAV samples of control and rejuvenated aged bitumen by WCO and WEO

Fig. 6. DSR results of control and rejuvenated aged bitumen with WCO and WEO.

#### Table 3

Average creep stiffness and m-value from BBR tests.

	Creep stiffness at 60 s (MPa)				m-value at 60 s		
	Temperature (°C)	Measured value	Specification	Pass or Fail	Measured value	Specification	Pass or Fail
Control bitumen	0 °C	15.53	$\leq$ 300	pass	0.56	≥0.30	pass
AB + 3.5% WCO		16.43		pass	0.58		pass
AB +5.5% WEO		13.25		pass	0.68		pass
Control bitumen	−6 °C	15.15		pass	0.47		pass
AB + 3.5% WCO		15.10		pass	0.45		pass
AB +5.5% WEO		16.38		pass	0.39		pass
Control bitumen	−12 °C	25.08		pass	0.35		pass
AB + 3.5% WCO		20.07		pass	0.36		pass
AB + 5.5% WEO		17.78		pass	0.30		pass
Control bitumen	−18 °C	69.88		pass	0.34		pass
AB + 3.5% WCO		127.46		pass	0.37		pass
AB + 5.5% WEO		83.88		pass	0.35		pass

Superpave Performance Grade of the rejuvenated aged bitumen by WCO and WEO was found to be PG 64-28 indicating that the performance of rejuvenated bitumen by WCO and WEO is comparable to the control bitumen.

# 5.4. Scanning Electron Microscopy (SEM/EDX)

SEM/EDX is a useful tool for characterizing the surface morphology and the fundamental physical properties of the aged and rejuvenated aged bitumen. A JEOL JSM 65101v Microscope and Oxford X-Max 20 equipment were used for SEM and EDX analyses, respectively. Scanning electron micrographs of the aged and rejuvenated aged bitumen by WCO and WEO are shown in Fig. 7, whereas the EDX data are formulated in Table 4. As seen in Table 4, the main mineral phases for aged bitumen are CaCO<sub>3</sub> (93.71%), SiO<sub>2</sub> (4.34%), FeS $_2$  (1.67%), and Al $_2O_3$  (0.12%). The addition of 3.5% WCO to the aged bitumen led to the increase in the of CaCO<sub>3</sub> (97.68%) and FeS<sub>2</sub> (2.27%) along with the presence of very low content (0.04%) of new mineral element, copper (Cu), with no aluminum appeared in the sample. While, the mineral phases of aged bitumen rejuvenated with 5.5% WEO were CaCO<sub>3</sub> (95.64%), FeS<sub>2</sub> (2.85%), and Al<sub>2</sub>O<sub>3</sub> (1.51%). Noteworthy, in the case of WCO, the percentage of CaCO<sub>3</sub> was 97.68% because the source of oil was vegetarian. In the WEO case, the percentage of Al<sub>2</sub>O<sub>3</sub> because the source of oil was the petroleum derivatives in addition to the engine body, which is basically made of aluminium material and led to an increase the aluminium content. Fig. 7-a exhibited spread bores with different sizes on the surface morphology of the aged bitumen with some unshaped fragments. The reason for that is due to the presence of SiO<sub>2</sub> (Table 4), which was completely dissolved by the rejuvenation of WCO as seen in Fig. 7-b, since pockets were disappeared. This means that the application of WCO as a rejuvenator for aged bitumen was successful and can be reused as an environmental and economical alternative. However, for the rejuvenated bitumen with WEO as shown in Fig. 7-c, some fragments are still seen confirming that WEO did not properly

#### Table 4

Energy dispersive x-ray (EDX) results.

Sample	Source	Abbreviation	Weight	Atomic
			%	%
Aged Bitumen (AB)	CaCO <sub>3</sub>	С	89.31	93.71
	SiO <sub>2</sub>	0	5.34	4.20
	$Al_2O_3$	Al	0.26	0.12
	SiO <sub>2</sub>	Si	0.32	0.14
	FeS <sub>2</sub>	S	4.24	1.67
	Ca	Ca	0.38	0.12
	Fe	Fe	0.16	0.04
Rejuvenated Bitumen by 3.5%	CaCO <sub>3</sub>	С	93.94	97.68
WCO	FeS <sub>2</sub>	S	5.83	2.27
	Cu	Cu	0.23	0.04
Rejuvenated Bitumen by 5.5%	CaCO <sub>3</sub>	С	89.68	95.64
WEO	FeS <sub>2</sub>	S	7.14	2.85
	$Al_2O_3$	Al	3.18	1.51

renovate the aged bitumen as required. This may be because waste engine oil contains some mineral elements such as aluminium [30] as previously illustrated, contributing to high content of Al<sub>2</sub>O<sub>3</sub>, which hindered the improvement.

# 5.5. Fourier Transform Infrared (FTIR) Spectroscopy test

The FTIR technique was used to determine the surface function groups of the dye adsorbent. Samples of the investigated materials



(a) Aged bitumen





(c) WEO rejuvenated

Fig. 7. SEM images of aged and rejuvenated bitumen at (x 2000).

were analysed using a Thermo Fisher Nicolet IS10. Fig. 8 shows the FTIR analysis of the control and aged bitumen as well as the FTIR results of the rejuvenated aged bitumen by both WCO and WEO. Sample (AB) shows the main characteristic vibration bands as given in Table 5 without any shift in their position (wave number). Adding WCO to the aged bitumen, results in de-intensifying of all bands due to the lower content of vibration groups. For WEO, extra intensified bands are found in the spectral range of 500–1000 cm<sup>-1</sup>, which may be attributed to the metallic content that results from high friction and heat in the engine while it was in use for its first life.

The FTIR spectrum of AB shows the presence of OH broad stretching absorption band at wave length,  $\upsilon$  3384 cm<sup>-1</sup>, and two absorption bands at 2924 and 2852 cm<sup>-1</sup> due to CH alkane. It also shows strong absorption band at 1688 cm<sup>-1</sup> attributed to the presence of carbonyl group (C=O), and two absorption peaks stretching frequency at  $\upsilon$  1603 cm<sup>-1</sup> and other is bending frequency at 742 cm<sup>-1</sup> confirming the presence of C = C function group in AB. Also, presence of stretching frequency at  $\upsilon$  1459 cm<sup>-1</sup> and bending frequency at 812 cm<sup>-1</sup> indicates the presence of aromatic rings in AB.

The FTIR spectrum of both AB + WCO and AB + WEO shows the presence of OH broad stretching absorption band at  $\upsilon$  3432 and 3349 cm<sup>-1</sup>, respectively. The deviation of 3384 cm<sup>-1</sup> obtained by AB was due to mixing of AB with both rejuvenators. Fig. 8 also shows two absorption bands at 2923 and 2852 cm<sup>-1</sup> for WCO and 2924 cm<sup>-1</sup> for WEO due to the presence of CH aliphatic, which is present in both rejuvenators. As shown in Fig. 8 and Table 5, two absorption peaks were observed; stretching frequency at  $\upsilon$ 



Fig. 8. FTIR analysis of control, aged bitumen and rejuvenated aged bitumen by WCO and WEO.

Table 5						
FTIR compounds	and functional groups.					

1600 cm<sup>-1</sup> for both rejuvenators and other was bending frequency at 740 cm<sup>-1</sup> and 726 cm<sup>-1</sup> for WCO and WEO, respectively, confirming the presence of C=C function group in AB. The presence of stretching frequency at v 1459 cm<sup>-1</sup> for both rejuvenators refers to the aromatic rings, which is shown previously in AB (Fig. 8). Infrared spectrum of both rejuvenators shows two absorption peaks at approximately 1029, and 1301 cm<sup>-1</sup> due to C–O. In conclusion, the addition of WCO or WEO to the aged bitumen increased the conjugated system with C=O function, which led to the slight decrease in its stretching frequency (C=C). This slight decrease in the C=C bonds after the addition of the WCO and WEO to aged bitumen, could decrease the asphaltenes content. However, the naphthalene aromatic content increased in the rejuvenated bitumen by both rejuvenators as well as the carbonyl of ester (C=O), which means an increase in the maltenes content. As a result, the asphaltenes to maltenes ratio decreased in the rejuvenated bitumen rather than the aged bitumen but not restored as the original values of the control sample.

# 5.6. Marshall testing results

Marshall test was applied on three prepared mixes, 100% RAP, 100% RAP with 3.5% WCO by bitumen weight, and 100% RAP with 5.5% WEO by bitumen weight. The intent of this simple study is just to evaluate the effect of the rejuvenator on the mechanical properties of the mix as directly compared to a mix with no rejuvenator. Table 6 shows the stability, flow, and bulk unit weight of the three investigated RAP mixes. A significant improvement was observed for rejuvenated RAP mixes by both rejuvenators

Table	6
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Properties/Mix	Aged Bitumen (AB)	AB + 3.5% WCO	AB + 5.5% WEO
Stability (kg) Flow (mm) Bulk unit weight (gm/cm <sup>3</sup> )	220 1.13 1.800	776 2.16 2.345	542 2.03 2.260

T	able	7		
-				

Indirect tensile strength results.

Indirect Tensile Strength	Aged Bitumen	AB + 3.5%	AB + 5.5%
	(AB)	WCO	WEO
ITS (Dry) (kPa)	188.1	480.1	474.5
ITS (Wet) (kPa)	102.1	393.7	380.1
Tensile Strength Ratio	54.0	82.0	80.0
(TSR), %			

Control bitumen		Aged bitumen (A	.B)	AB + 3.5%WCO		AB + 5.5%WEO	
Wave length (cm <sup>-1</sup> )	Function group	Wave length (cm <sup>-1</sup> )	Function group	Wave length (cm <sup>-1</sup> )	Function group	Wave length (cm <sup>-1</sup> )	Function group
3291 broad	—ОН	3384	—ОН	3432 broad	—ОН	3349	-OH stretching
2923	CH-Alkane stretching	2924	CH-Alkane stretching	2923	CH-Alkane stretching	2924	CH-Alkane stretching
1732	C=0	2852	CH-Alkane stretching	2852	CH-Alkane stretching	1744	C=0
1663	C=0	1688	C=0	1705	C=0	1600	C=C
1604	C=C	1603	C=C	1600	C=C	1460	C=H bending
1459	C—H bending	1459	C—H bending	1459	C—H bending	1376	-OH bending
1376	OH bending	1376	–OH bending	1376	—OH bending	1301	C-0
1030	C—0	1027	C—0	1029	C—0	-	-

compared to the aged bitumen mix. Marshall results suggest that the WCO can be used as a rejuvenator in wearing surface courses (750 kg minimum stability for medium traffic) or binder courses (700 kg minimum stability for heavy traffic) [31]. The WEO can be used as a rejuvenator in binder courses for light traffic volumes as the minimum stability requirement is 500 kg [31]. In addition, the minimum requirement for flow was also achieved for both



Fig. 9. Stress versus strain curves for 100% RAP and rejuvenated 100% RAP by WCO and WEO mixes.

rejuvenated mixes (2–4 mm) [31]. Moreover, the bulk unit weight of the aged bitumen mixes significantly increased by about 27.8% in average for both rejuvenated mixes. It should be noted that, the long-term behaviour of the rejuvenated mixtures was not studied in this paper.

# 5.7. Indirect tensile strength testing results

The ITS test was applied on three prepared mixes, 100% RAP, 100% RAP with 3.5% WCO by bitumen weight, and 100% RAP with 5.5% WEO by bitumen weight. The test was conducted to evaluate the moisture damage resistance and fracture energy of the rejuvenated mixes as compared to the 100% RAP mix with no rejuvenator. Table 7 summarizes the ITS results for all mixtures. A significant improvement was noted for rejuvenated RAP mixes by both rejuvenators compared to the aged bitumen mix. It is evident from the results that the TSR of the 100% RAP with 3.5% WCO was higher than 80%, which is the minimum requirement [31]. Moreover, the fracture energy of the investigated asphalt mixes was determined based on the ITS testing results by computing the area under the stress-strain curve up to failure [32,33]. Fig. 9 shows the stress-strain curves of the three investigated mixtures. The specimens of the 100% RAP + 3.5% WCO mix in both dry and wet conditions exhibited the highest energy to fracture, which agreed with the TSR testing results. While, the 100% RAP exhibited the lowest fracture energy for both conditions indicating lower fatigue resistance.

#### 6. Conclusions

The purpose of this study was to investigate the impact of waste cooking oil and waste engine oil as rejuvenators on aged bituminous pavement by evaluating the physical, rheological and chemical characteristics of the investigated binders. Also, mechanical testing was conducted on laboratory prepared mixes. Results of this study reflected the following conclusions:

- A 3–4% WCO and 5 to 6% WEO content can rejuvenate the local aged bitumen in Egypt and restore its original properties. The SEM images and EDX analysis demonstrated the positive effect of the WCO and WEO to rejuvenated aged bitumen.
- By adding the optimum percentage, 3.5% of the WCO, the rutting parameter (G\*/sin  $\delta$ ) of the aged bitumen was improved and became almost identical to the (G\*/sin  $\delta$ ) of the control bitumen.
- The WCO rejuvenated aged bitumen exhibited the lowest values of the fatigue parameter ( $G^* \sin \delta$ ) indicating better resistance to fatigue cracking.
- The rejuvenated bitumen showed better tendency to short-term aging, connected with physical, rheological, and chemical properties, compared with control bitumen.
- With the addition of WCO and WEO to aged bitumen, the creep stiffness and rate of rejuvenated bitumen was within the Superpave specification criteria, which demonstrates that WCO and WEO improved the low temperature thermal cracking of aged bitumen with a PG 64-28.
- The investigated rejuvenators were found to improve the mechanical properties in terms of stability as well as the moisture resistance of rejuvenated mixes compared with the 100% RAP mix.
- The application of WCO and WEO as rejuvenating agents for aged bituminous pavement was successful and introduce an environmental and economical alternative for recycling these wastes in pavement construction.

#### **Declaration of Competing Interest**

None.

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