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Sustainable recovery of waste vegetable cooking oil and aged bitumen: Optimized modification for short and long term aging cases

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ABSTRACT

Bitumen ages in the short and long-term due to environmental conditions. As the bitumen ages, it loses its original properties and flexibility. Hardened bitumen causes certain distresses in pavement that may endanger traffic safety and reduce travel comfort. Bitumen is one of recoverable material, however some techniques may not be ecological due to considerable energy, time, and cost. On the other hand, recovering of aged bitumen might be eco-friendlier, energy-efficient, and economic by using bio-based waste materials such as waste vegetable cooking oils (WVCO). In this perspective, this paper was established on the idea of sustainable recovery of aged bitumen and WVCO. Base bitumen was aged in case of short-term (ST) and long-term (LT) in laboratory condition and modified with WVCO ranging from 2 to 10% by weight of bitumen. To determine the effect of WVCO modification on aged bitumen conventional and rheological test methods were utilized. To find the optimum rate of WVCO for full recovery of aged bitumen, an index called Pure Rejuvenation Index (PRI) was specified and applied for each test results. It can be concluded from PRI analyses that WVCO can be used as rejuvenator to recover aged bitumen and approximately 3% and 6% of WVCO are required for ST and LT aged bitumen cases, respectively. However, different test methods yield different optimum rates of WVCO for ST and LT aged bitumen. Recovering of WVCO and aged bitumen by using together may provide environmental protection and conservation of resources.

1. Introduction

Recovering of asphalt pavements is nowadays an inevitable requirement for sustainable and efficient transportation. Additionally, researchers investigate sustainable approaches and green technology solutions to develop new bio-based materials. The common point of these two efforts is recovering of aged bitumen by utilizing bio-based waste materials. The concept of utilizing bio-based materials in asphalt industry is called ***Bio-asphalt*** or ***Bio-bitumen.***

Bitumen is a kind of binder that used for construction of flexible pavements, mostly. It is a valuable and versatile material that remains after refining of crude oil, and its resources scarce and decrease in daily base. Although bitumen is a minor component of asphalt pavement by weight, it plays a vital role in structural and functional performance of pavements (Bocci et al., 2019; El-Shorbagy et al., 2019; Norton, 1992).

Bitumen ages in the short and long-term due to different production processes and various environmental conditions. As the bitumen ages, its original physical and chemical properties changes and it loses its flexibility. Since, rheological characteristics of bitumen has changed significantly due to aging, the performance of pavement under traffic loads and environmental conditions is decreased, accordingly (Garcia et al., 2016; Gökalp et al., 2018; Gökalp and Uz, 2019; Hesp et al., 2007; Jacob, 1989; Lesueur, 2009; Romera et al., 2006).

Aging of bitumen is occurred from production stage to the end of service life of pavement. There are four main factors that contribute to aging: (1) ultraviolet rays light, (2) moisture, (3) time, and (4) temperature (Airey, 2003; Dokandari et al., 2014). These factors cause a change in chemical structure and respectively rheology of them (O'connell and Steyn, 2017; Turk and Tušar, 2016). Generally, aging is classified under two-time duration. Aging occurs from production of hot mix asphalt to opening the pavement for traffic is called short-term (ST), while long term (LT) aging covers the whole service life of pavement. In laboratory condition, aging can be simulated with standard and non-standard methods such as thin film oven test, pressure aging vessel, and oxidative

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aging for short and long-term aging cases (Arias, 2019; Hesp et al., 2007; Lamontagne et al., 2001; Yan et al., 2019; Ye et al., 2019).

The population growth of world increases the demand for vegetable cooking oil (VCO) in daily basis. VCO is mainly consumed for frying and cooking activities at food industry, restaurants, hotels, and residences. As a result, considerable amount of waste vegetable cooking oil (WVCO) is accumulated (Maharaj et al., 2015). Estimated annual WVCO amount for countries such as the United Kingdom, Netherlands, Italy, Portugal, and Spain are reported as 24.3, 18, 16, 7.5, 73, 67 thousand m^3, respectively (Cocchi and Ugge, 2013). Ministry of Environment and Urbanization of Republic of Turkey reported that approximately 1.7 million ton (Mt) of refined oil is produced annually, and approximately 0.35 Mt that of WVCO can be collected (Yaman, 2012). WVCO is a hazardous material and has serious effect on environment, such as contamination of land and potable water resources, and ecosystems inhabited by different creatures (Azahara et al., 2016; Gökalp et al., 2018; Onurlubaş and Kızılaslan, 2007). In these respect, WVCO needs to be disposed safely and/or recycled/recovered for viable purposes. Since, disposal of WVCO by storage is highly expensive it is not a desirable action to follow. Instead of this, recycling/recovering for a new purpose or utilizing them for production of an oil based material is mostly preferred. Considering those aspects, several studies have been done to evaluate the possibility of using WVCO in different areas to produce biodiesel, soap, animal feed and so forth (Azahar et al., 2016; Eriskin et al., 2017; Gökalp et al., 2018; Kulkarni and Dalai, 2006; Su et al., 2015; Wen et al., 2012). However, effectiveness of biodiesel production is questionable due to economic and environmental concerns (Alptekin and Ç anakçı, 2006; Bolat et al., 2016; Kulkarni and Dalai, 2006; Photaworn et al., 2017; Ray and Prakash, 2019).

Recovering of hot mix asphalt needs rejuvenation of aged bitumen to make it convenient for reuse. This rejuvenation is done by commercial additives or waste materials (Romera et al., 2006) and utilizing WVCO is one of the most popular waste in recent (Behnood, 2019). The studies that have been investigated usability of WVCO as rejuvenation agent in asphalt reclamation are briefly presented at the following.

Zargar et al. (2012) evaluated the possibility of using WVCO as an additive material to rejuvenate the aged bitumen by a non-standard test method in laboratory. The authors revealed that addition approximately 4% WVCO (by weight of bitumen) into the aged bitumen make it similar to the base bitumen. Asli et al. (2012) performed traditional tests to evaluate the effect of WVCO as a rejuvenation agent on recycled asphalt pavement (RAP) materials. The authors recommended that WVCO up to 5% can be used in the mixture without compromising the structural performance. However, Ji et al. (2016) offered different rate (8%) of WVCO according to their study investigated rutting resistance of aged bitumen extracted from RAP materials. In the study done by Koudelka et al. (2018) it was highlighted that optimum rate of rejuvenating agents changes according to their origin. In their paper, optimum rate of the agents was found between 5 and 10% by weight of binder. Maharaj et al. (2015) and Gökalp and Uz (2019) investigated the influence of WVCO addition on the rutting and fatigue cracking resistance performance of bitumen with adding up to 10% by two increments. It was concluded that the samples with WVCO has less rutting but better cracking (fatigue and low temperature) resistance compared to base bitumen. Similarly, Gökalp et al. (2018) was found that modification with excessive amount of WVCO changes the chemical configuration of the bitumen and make the bitumen more viscous.

Traditional or rheological tests were utilized for evaluation of WVCO modified bitumen performance at the aforementioned studies. It can be easily concluded from these literatures survey that the method used for evaluation is very important on determining the optimum amount of WVCO. Moreover, rejuvenation effect of WVCO was investigated only for the LT aged bitumen which were extracted from RAP or artificially aged at the laboratory. This study was established to overcome the shortcomings at these previous studies linked with evaluation methods and as well as the aging form of bitumen.

Within the scope of this study, sustainable recovery of WVCO and aged bitumen were investigated. To determine the optimum rate of WVCO for the ST and LT aged bitumen, WVCO was added up to 10% by weight of bitumen with two increments. The ST and LT aged bitumen samples were obtained with the oxidative aging method. Finally, the optimum rate of WVCO for each tests method and for both aging cases was determined with a specified new index called pure rejuvenation index (PRI).

2. Materials

In this study, base bitumen with 70/100 penetration grade and short and long term-aged forms of it were utilized. WVCO samples were collected by the students of Kurttepe Şehit Ali Öztaş Vocational and Technical Anatolian High School within the scope of a social responsibility project carried out by Ministry of Environment and Urbanization of Republic of Turkey. The WVCO were filtered with a membrane (0.45 μm) to remove the impurities. It is almost not possible to know the origin of the WVCO, so chemical analysis was done. Physical and rheological properties of the base bitumen were found by series of tests. The properties of bitumen and chemical analysis results of WVCO are presented in Table 1. According to the chemical analysis, WVCO is identified as sunflower oil.

Since all test results of samples are presented with a code, a nomenclature was formed such as following examples for short and long term aged cases.

- Base Bitumen + Short Term Oxidative Aging + X (%) WVCO: OxiA-ST-XW
- Base Bitumen + Long Term Oxidative Aging + X (%) WVCO: OxiA-LT-XW

where X is rate of WVCO such as 2, 4, 6, 8, and 10.

3. Experimental testing process

In this section of the paper conventional, rheological test procedures, aging and modification processes of bitumen are presented, briefly.

3.1. Conventional testing process

Penetration test is applied on the bitumen samples to determine their consistency or hardness under a certain load and temperature by utilizing a specific needle. In this study, this test is applied on the samples by following EN 1426 (CEN, 2015a) standard. Softening point test is generally utilized to determine the heating temperature of the bitumen to make it flow without deforming its chemical structure. In this test method, softening point of the bitumen is identified by utilizing two certain sized rings and standard steel balls under specifically elevated temperature conditions. To apply the test, the standard codded with EN 1427 (CEN, 2015b) was followed. Because of the fact that bitumen is a thermo-elastic material and its viscosity changes with temperature, it is important to determine viscosity of bitumen. Moreover, viscosity is a kind of scale to determine heating temperature of bitumen for providing workable hot mix asphalt during both mixing and compaction processes. The viscosity of bitumen samples is generally measured under two specific temperatures (135 °C and

Table 1
Properties of the base bitumen and chemical configuration WVCO.

Bitumen			WVCO	
Tests	Standards/Unit	Results	Main Chemical Compositions	Content (%)
Penetration	EN 1426/0.1 mm	77	Linoleic acid (C18:2n6c)	55.54
Softening Point	EN 1427/°C	42	Oleic Acid (C18:1n9c)	30.56
Flashing Point	EN ISO 2592/°C	254	Palmitic Acid (C16:0)	9.94
Ductility	EN 13589/cm	>108	Stearic Acid (C18:0)	3.44
Viscosity	ASTM 4402/cP	425.5 at 135 °C	Palmiotetic Acid (C16:1)	0.14
		120.8 at 165 °C	Elcosatrienoic Acid (C20:3n6) (Cis-8.11.14)	0.69
PG Grade	EN 14770, EN 14771	64–22	Linoleic acid (C18:2n6c) (Cis-9.12.15)	0.64

165 °C). Standard codded with ASTM 4402 (ASTM, 2015) was followed to determine the viscosity values of the bitumen samples at the two temperatures.

3.2. Rheological testing process

3.2.1. Dynamic Shear Rheometer (DSR)

DSR is utilized to determine the viscous and elastic behavior of bituminous materials under different temperature and loading frequency. Base, short-term and long-term aged bitumen can be evaluated within a specific logical framework. DSR consists of a chamber to make a stable environment for the test temperature. Loading and measuring system of the device is controlled by software. At the end of the test, complex shear modulus (G*) and phase angle (δ°) can be determined for the bitumen sample at elevated temperatures, automatically. Utilizing the results, rutting and fatigue resistance factors of the bitumen can be calculated and compared with the limitations. These two factors are explained briefly at below, which the bitumen performance can be estimated.

Rutting Factor: G*/Sin (δ°) is developed for assessment of "high temperature stiffness" that addresses rutting resistance of bitumen. Rutting resistance is referred to permanent deformation of bitumen under traffic loadings and environmental conditions. The value of G*/Sin (δ°) is utilized in evaluation of base and ST aged bitumen (Anderson et al., 1994).

Fatigue Factor: G*. Sin (δ°) is developed for evaluating "intermediate temperature stiffness" of bitumen binder that points fatigue cracking resistance performance of pavement during its service life. The value of G*Sin (δ°) is utilized in evaluation of base and LT aged bitumen (Anderson et al., 1994).

Phase angle is a parameter that gives an idea about viscoelastic properties of the samples. It is actually, the time lag between response with applied force. Its value is in the range of 0–90°. Based on this scale, a perfectly elastic material has a phase angle of 0°, whereas for characterizing it as a fully viscous material the phase angle is expected to be 90°.

In SuperPAVE performance grade system, limit values of complex modulus for the base and ST and LT aged bitumen addresses the rutting and fatigue factors were specified as 1 kPa, 2.2 kPa, and 5000 kPa, respectively. In this study, DSR test was applied on the samples with respect to EN 14770 (CEN, 2012b) standard. The test was conducted on the samples of unaged, short and long-term aged bitumen at different temperatures (46–70 °C for short-term aged bitumen, 16–40 °C for long-term aged samples).

3.2.2. Bending Beam Rheometer (BBR)

BBR test is applied on LT aged bitumen samples to assess their thermal cracking and relaxation characteristics. In this study, EN 14771 (CEN, 2012c) was followed to apply the test. Thermal cracking and relaxation characteristic are evaluated by creep stiffness and m-value, respectively. m-value is the slope of the creep stiffness (St) at 60 s. The limitations for St and m-value of a sample are specified as maximum 300 MPa and minimum 0.300, respectively. For higher St value, it can be indicated that the sample has higher thermal resistance, whereas less ability to relax stresses for a lower m-value. Specifically, BBR test is applied on the long-term aged bitumen and duration of the test approximately 240 s after conditioning finishes. BBR test system consists of a fluid bath for conditioning the sample at desired temperature with the help of temperature controller system. Loading units and deflection measuring systems are controlled by software. In this study, BBR tests were performed on each sample at four different temperatures, −6, −12, −18, −24 °C for at least two repetitions.

3.3. Aging process and WVCO modification of aged bitumen

3.3.1. Aging process

One of the most known and accepted test for evaluation the short term aging effect on bitumen during mixing, transportation and compaction is Rolling Thin Film Oven (RTFO) test. In this test method, certain amount of bitumen sample is poured in a specified open ended glasses and spreading bitumen through the glasses as a thin film is ensured by rotation of the holder plate at a constant speed. During the test, oven temperature is kept constant. at 163 °C and dry air is blown into the sample through the open ending of glasses. Pressure Aging Vessel (PAV) test is one of the most important test method in Superpave design to obtain long-term aged bitumen samples. The test can be carried out at 90, 100, 110 °C under 2.1 kPa pressure for 20 h. Within the scope of this study, 100 °C was used for conditioning.

At first, RTFO and PAV tests were conducted on base bitumen sample according to the standards normed with EN 12607-1 (CEN, 2014) and EN 14769 (CEN, 2012a), respectively. In RTFO and PAV tests, the approximate amount of residue material that can be collected from the glasses/pans at the end of the tests are 30 g and 40 g, respectively. However, to implement all the tests throughout the current study, more than 1000-grams of aged bitumen is required for each aging term. Therefore, a large number of tests are required to provide this amount of residue with standard test methods. In order to save energy and minimize labor a method called oxidative aging was used to obtain short and long term aged bitumen samples in the light of the study done by Zargar et al. (2012), Arias (2019); Yang et al. (2017), and Gökalp and Uz (2019).

In oxidative aging process, the bitumen is continuously stirred under a specified temperature with a propeller mixer for a definite duration. Under the defined condition, the oxygen present at environment is allowed to enter the bitumen sample and loss of volatile components is ensured. In oxidative aging, the temperature, stirring speed, and duration vary depending on the type of bitumen and the desired aging processes. In this study, stirring durations for short and long term aging were decided according to the results of penetration, softening point and viscosity tests performed on aged base bitumen residues of both standard (RTFO/PAV) and Oxi-aging (ST/LT) methods. The confidence intervals for each test performed on the standard aged samples were calculated for 95% confidence level according to Eq. (1). Then the average values of each test performed on the Oxi-aged samples were checked whether they fall within the range. Moreover, the variations between the test results gather from standard aging and Oxi-aging method were calculated according to Eq. (2) to make clear how the results are closed to each other.

$$\mu = \bar{x} \pm z\frac{\sigma}{\sqrt{n}} \quad (1)$$

where μ is unknown population mean, $\bar{x}$ is sample mean, σ is standard deviation, n is number of tests, and z is 1.96 which is a standard score for 95% confidence level.

$$Var\ \ (\%) = \frac{|OxiAging - Standart\ \ Aging|}{Standart\ \ Aging} * 100 \quad (2)$$

where Var is variations.

The mean test results of penetration, softening point and viscosity tests in case of both standard and Oxi-aging methods and calculated intervals for 95% confidence level and the variations value are presented in Table 2.

As it can be clearly seen from the data presented in Table 2 that the results of penetration, softening point and viscosity tests in case of both standard and Oxi-aging methods are matching in 95% confidence level and the variations between them are lower than 10%. After several attempts, stirring duration were found as 5 and 10 h for ST and LT Oxi-aging methods, respectively. Therefore, below procedure was followed to ensure Oxi-aging of bitumen samples in this study.

Bitumen was heated at 140 ± 5 °C for two hours to obtain certain viscosity and 500 g of bitumen was poured on a cylindrical metal container with a diameter of 10 cm and height of 12 cm. The bitumen in metal container was placed on the heater at 140 ± 5 °C and stirred with a propeller mixer at 1000 revolution per minute (rpm) for five hours for short term aging and another five hours for long term aging.

3.3.2. WVCO modification of aged bitumen

In the scope of this paper, the effect of WVCO on the short and long-term aged bitumen was also investigated by means of conventional and rheological tests to consider sustainable recovery of both aged bitumen and WVCO. To achieve this aim, 70/100 penetration graded base bitumen was aged with Oxi-Aging method for short and long term aging cases, and then WVCO was added to the aged bitumen in different ratios (2, 4, 6, 8, and 10%) to rejuvenate it. To modify the aged bitumen with WVCO, aged bitumen was heated at 160 °C for at least 2 h in a metal container. After that WVCO weighed for defined rate was added into the aged bitumen. Finally, the bitumen and WVCO mixture was stirred with a propeller mixer at 100 rpm for 30 min at 160 ± 5 °C.

3.3.3. Pure Rejuvenation Index (PRI) analysis

PRI is a new and simple but an effective analyzing method to determine the contribution rate of additives used for recovered bitumen. PRI (Eq. (3)) method was developed with inspiration of modification, aging, and viscosity indexes used in the literatures (Dokandari et al., 2014; Gökalp and Uz, 2019; Lesueur, 2009; Teltayev et al., 2019; Zhang et al., 2012).

$$\mathrm{PRI}\,(\%) = [(\mathrm{VRB}^t - \mathrm{VBB}^t)/\mathrm{VBB}^t] \times 100 \quad (3)$$

where VRB refers to the test results of rejuvenated bitumen,

VBB refers to the test results of base bitumen, and t refers the test temperature.

PRI values closed to zero "0" indicates that the aging effect is minimized by means of rejuvenation and the contribution rate of WVCO matching zero value of PRI is identified pure rejuvenation contribution rate.

4. Results and discussions

4.1. Physical properties test results

This section of the paper consists of the physical test results of base, aged and WVCO modified bitumen. Penetration, softening point, and viscosity at 135 and 165 °C test results of base, aged bitumen, and WVCO modified bitumen are presented in Fig. 1.

As shown in Fig. 1, penetration value decreases with aging however, increases with including WVCO in the aged bitumen. There is a trend of increasing in softening point as bitumen age. In contrast to penetration values, addition of WVCO caused a significant decrease in softening point of the aged bitumen. As aforementioned before bitumen is a visco-elastic and heat-sensitive construction material and viscosity of bitumen is expected to decrease with heating. On the other hand, aging processes hardened the bitumen and increased its viscosity. However, viscosity of the aged bitumen decreases with increasing WVCO content.

4.2. Rheological test results

The rheological properties of the samples analyzed with DSR and BBR testers according to related EN norm standards (EN 14770 and EN 14771, respectively). Results of the tests performed on the both short and long-term aged bitumen modified with different rates of WVCO are presented in Fig. 2.

As mentioned in method section that G*/Sin (δ°) value is used to evaluate the rutting resistance of bitumen. It can be seen in Fig. 2 that bitumen in form of OxiA-ST has higher G*/Sin (δ°) value and addition of WVCO decrease the rutting resistance performance. The tendency to rutting resistance for the samples marked with Oxia-ST-4W and Oxia-ST-2W are almost the same and closer to the base bitumen performance. As expected, viscous behavior of ST aged samples decreases, since bitumen hardens.

G*Sin (δ°) value is used to evaluate the fatigue cracking resistance of bitumen. Depending on the results, LT aged bitumen has the lowest resistance to fatigue cracking. However, increasing in the rate of WVCO in LT-aged bitumen is increasing the fatigue resistance performance. Comparing to the phase angle (δ°) graphs for both aging cases, LT aged samples has the lowest phase angles, which means LT aged bitumen samples showed more elastic properties.

Decreased St and increased m-value indicate improved performance at low temperature cracking for the pavement surface. From the BBR test results, a tendency of increasing in the low temperature cracking resistance with the decrease of temperature can be observed. All values for St are determined below the specified

Table 2
Comparing the test results for different aging methods.

Tests	RTFO	ST Oxi-aging	Intervals for 95% Confidence level	Var (%)	PAV	LT Oxi-aging	Intervals for 95% Confidence level	Var (%)
Penetration (0.1 mm)	54	57	49–59	5.6	35	38	32–38	8.6
Softening Point (°C)	49	46	45–53	6.2	63	60	59–67	4.7
Viscosity at 135 °C (cP)	553	607	490–616	9.8	729	792	655–803	8.6
Viscosity at 165 °C (cP)	141	152	123–159	7.8	174	189	158–190	8.6

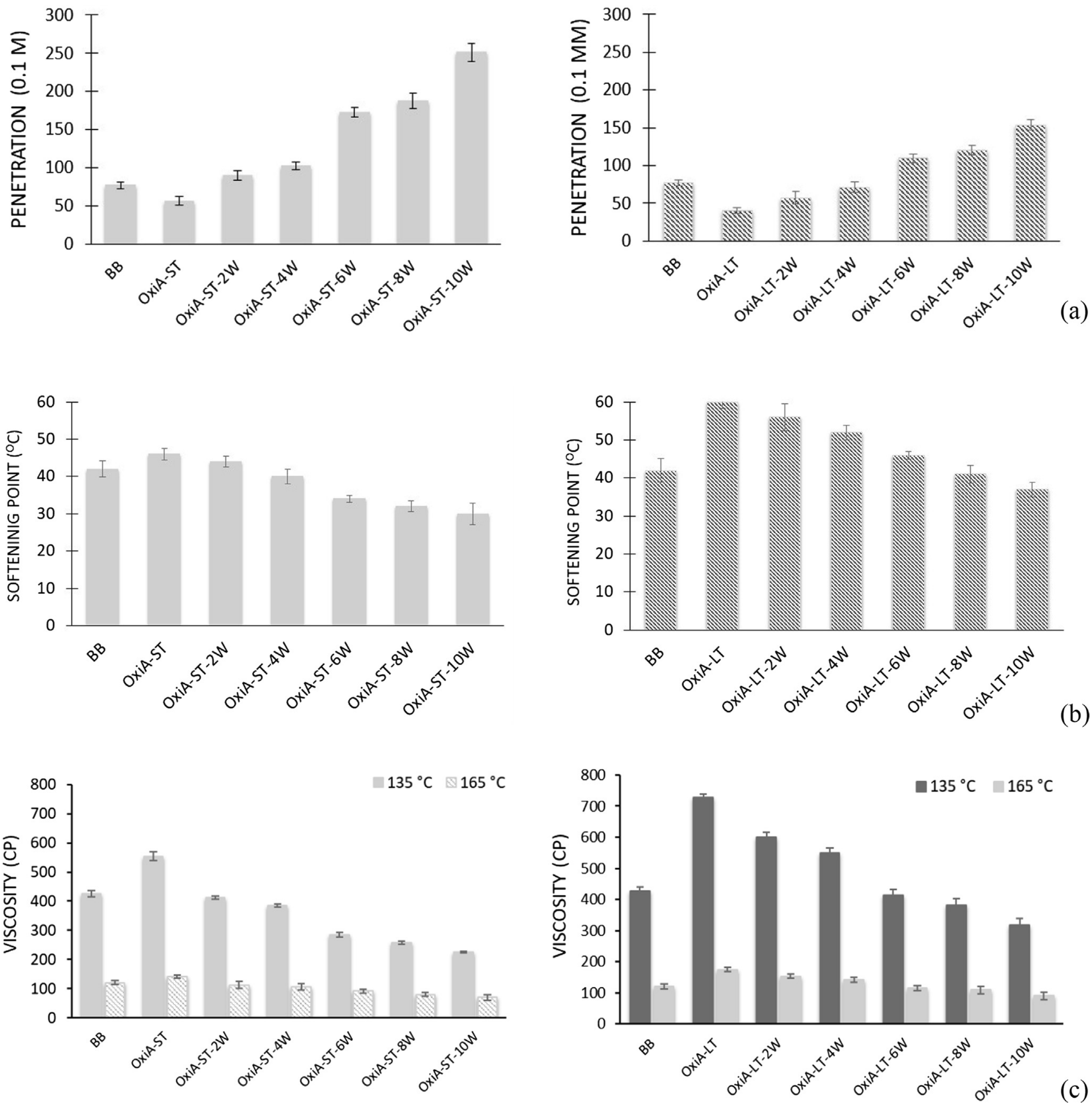

Fig. 1. Penetration (a), softening point (b) and viscosity (c) test results for bitumen samples.

maximum value by the standard. On the other hand, m-value, which must be minimum 0.300, satisfied at −6, −12, and −18 °C temperatures for all samples except of OxiA-LT. The results showed that adding WVCO into the LT-aged bitumen enhances the resistance of creep stiffness of bitumen.

4.3. PRI analysis results

The optimum rates of WVCO for purely rejuvenation of aged bitumen were determined for each test method. The process is based on drawing best fit line for PRI vs WVCO contents and produce the equations with root square in case of ST and LT aging. Then, the optimum WVCO contribution rates for sustainable and purely recover were calculated using the equation. The data for physical test methods are presented at Table 3.

According to the data presented in Table 3, the optimum contribution rates of WVCO for rejuvenation of ST and LT aged bitumen significantly changes with the evaluation method. Depending on the penetration test results, 2.1 and 3.7% WVCO is required to recover the ST and LT aging effect, respectively. When the softening point test results are considered, 2.5 and 7.8% WVCO contribution rates may recover the ST and LT aging effect, respectively. Although the optimum rates of WVCO for ST aging case is close to each other, softening point evaluation requires almost twice compared to that of the penetration test results for LT aging cases. The penetration test is implemented at room temperature whereas the softening point starts when the solution, which is mostly

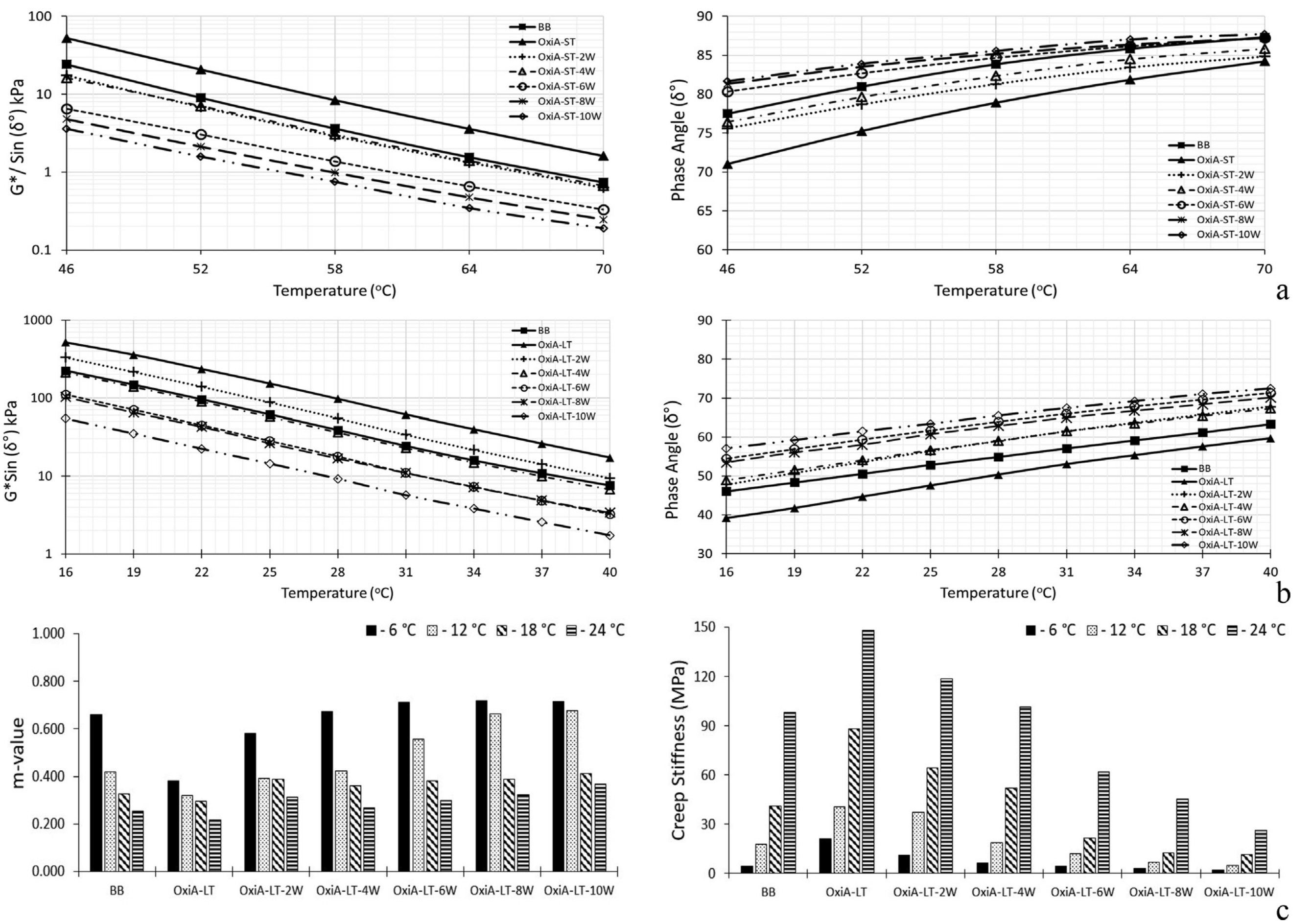

Fig. 2. Results of DSR tests for base, OxiA-ST (a) and OxiA-LT (b) bitumen forms and that of BBR tests (c).

Table 3
PRI results and optimum WVCO content based on physical tests.

Test Methods	OxiA-ST	OxiA-ST-2W	OxiA-ST-4W	OxiA-ST-6W	OxiA-ST-8W	OxiA-ST-10W
Penetration	−26.1	−13.0	33.9	124.4	143.6	226.3
BFLE/R^2/Opt. WVCO (%)	y = 19.39x − 40.11/0.95/2.1					
Softening Point	9.5	4.8	−4.8	−19.1	−23.8	−28.6
BFLE/R^2/Opt. WVCO (%)	y = −4.15x + 10.43/0.97/2.5					
Viscosity at 135 °C	30.0	−2.9	−9.5	−32.8	−39.5	−47.0
BFLE/R^2/Opt. WVCO (%)	y = −7.40x + 20.02/0.93/2.7					
Viscosity at 165 °C	16.7	4.3	−12.4	−24.1	−33.8	−42.1
BFLE/R^2/Opt. WVCO (%)	y = −6.00x + 14.76/0.99/2.5					
	OxiA-LT	OxiA-LT-2W	OxiA-LT-4W	OxiA-LT-6W	OxiA-LT-8W	OxiA-LT-10W
Penetration	−48.1	−25.8	−7.6	42.4	56.3	99.8
BFLE/R^2/Opt. WVCO (%)	y = 14.79x − 54.44/0.98/3.7					
Softening Point	42.9	33.3	23.8	9.5	−2.4	−11.9
BFLE/R^2/Opt. WVCO (%)	y = −5.65x + 44.10/1.00/7.8					
Viscosity at 135 °C	71.4	41.2	29.3	−2.7	−10.1	−25.2
BFLE/R^2/Opt. WVCO (%)	y = −9.55x + 65.07//0.97/6.8					
Viscosity at 165 °C	44.3	18.2	14.2	−4.1	−10.9	−28.5
BFLE/R^2/Opt. WVCO (%)	y = −6.70x + 39.08/0.97/5.8					

where y is PRI value and x is WVCO rate, BFLE is best fit line equations, R^2 is root square, Opt. WVCO is optimum rate of waste vegetable cooking oil.

water, at 5 °C and continue up to test was finished. However, viscosity test is conducted at relatively higher temperatures 135 and 165 °C. The effect of the test temperature on the optimum WVCO contribution rate is more apparent especially for LT aged bitumen. Since, there is 1% difference between WVCO contents for the viscosity test temperatures. Specifically, higher contribution rates were found for lower temperature.

To make a rheological assessment, DSR test by means of rutting and fatigue factors and BBR test by means of creep stiffness and m-value parameters results were analyzed. The optimum contribution rates of WVCO according to the PRI values and test results for each aging cases are presented in Table 4. It is worth to indicate that DSR test was performed for ST aged samples at the temperatures between 46 and 70 °C, while LT aged samples were tested

Table 4
PRI results based on rheological tests.

Temperature (°C)	OxiA-ST	OxiA-ST-2W	OxiA-ST-4W	OxiA-ST-6W	OxiA-ST-8W	OxiA-ST-10W
46	118.4	−27.6	−32.6	−72.9	−80.0	−85.2
52	130.6	−23.7	−21.4	−66.1	−76.4	−82.5
58	131.9	−19.7	−15.7	−62.0	−72.8	−79.3
64	131.2	−14.1	−9.9	−57.6	−69.6	−77.7
70	118.6	−14.2	−9.3	−55.0	−66.6	−74.1
BFLE/R^2/Opt. WVCO (%)	y = −17.63x + 66.98/0.73/3.8					
Temperature (°C)	OxiA-LT	OxiA-LT-2W	OxiA-LT-4W	OxiA-LT-6W	OxiA-LT-8W	OxiA-LT-10W
40	127.38	23.84	−11.70	−57.19	−55.13	−77.15
37	140.57	31.22	−8.41	−55.66	−54.80	−76.34
34	150.95	36.81	−6.44	−54.11	−54.34	−75.86
31	151.31	39.36	−6.25	−55.00	−54.82	−76.65
28	149.83	41.41	−6.99	−54.31	−56.60	−76.23
25	149.34	43.28	−6.51	−54.11	−56.94	−76.45
22	146.12	46.04	−6.11	−53.07	−54.97	−76.52
19	143.79	46.94	−5.51	−52.18	−55.64	−76.14
16	132.17	48.96	−4.49	−50.65	−53.71	−75.45
BFLE/R^2/Opt. WVCO (%)	y = −20.44x + 100.68/0.86/4.9					
Temperature (°C)	OxiA-LT	OxiA-LT-2W	OxiA-LT-4W	OxiA-LT-6W	OxiA-LT-8W	OxiA-LT-10W
−6	377.95	146.50	43.52	5.07	−31.90	−54.21
−12	128.34	110.43	5.96	−31.54	−60.34	−71.42
−18	114.20	57.12	26.79	−47.65	−69.55	−72.18
−24	50.93	20.88	3.33	−36.86	−53.74	−73.00
BFLE/R^2/Opt. WVCO (%)	y = −23.40x + 137.38/0.64/5.9					

where y is PRI value and x is WVCO rate, BFLE is best fit line equations, R^2 is root square, Opt. WVCO is optimum rate of waste vegetable cooking oil.

under the 40–16 °C temperature range. The BBR test was performed only on LT aged samples at the temperatures −6, −12, −18, and −24 °C. However, while determining the best fit line equations for PRI and WVCO contribution rates, all the test temperatures were evaluated together. In other words, temperature sensitivities were not taken into consideration.

According to the results presented in Table 4, it obvious that the optimum rate of WVCO to recover ST aged bitumen is lower than LT aged one. When the rutting and fatigue performance of the bitumen were evaluated with DSR test, 3.8% and 4.9% contribution rates of WVCO may recover the aging effect, respectively. To recover the thermal cracking performance of LT aged bitumen 5.9% of WVCO should be used based on BBR tests.

In order to a make general assessment for optimum WVCO contribution rate for pure rejuvenation of ST and LT aged bitumen based on the PRI analysis, the following Table 5 is prepared.

From Table 5, the importance of evaluation method and the aging term on determination of the optimum rate of WVCO can be obviously seen. Also, it is clear that LT aged bitumen needs more WVCO content than that of ST aged ones. On the other hand, the evaluation temperature is an effective parameter for evaluation the optimum rate of WVCO. Since, temperatures for application of the tests are different from each other. For instance, penetration test is applied at room temperature, whereas the softening test is applied relatively higher temperatures which changes based on the bitumen type. Rotational viscosity test is applied at highest temperatures comparing with that of others. The DSR test is applied to the samples at the temperatures between 40 and 16 °C provided lower WVCO rates than the BBR tests which conducted at the temperatures between (−6) and (−24) °C for the same aging cases.

Table 5
Summary of optimum WVCO contribution rate based on PRI analysis.

Test Types	Test Methods	Optimum WVCO (%)	
		ST Aging	LT Aging
Conventional	Penetration	2.1	3.7
	Softening Point	2.5	7.8
	Viscosity at 135 °C	2.7	6.8
	Viscosity at 165 °C	2.5	5.8
Rheological	DSR Rutting Resistance Test at 70–46 °C	3.8	–
	DSR Fatigue Resistance Test at 40–16 °C	–	4.9
	BBR Thermal Cracking Resistance test at (−)6 – (−)24 °C	–	5.9

5. Summary and conclusion

The usage of WVCO in ST and LT aged bitumen was analyzed by means of recovering of the two waste materials in the scope of this study. In this respect, it has been attempted to reveal the positive or negative effects that might be arisen with usage of WVCO in bitumen. To achieve it, a base (70/100 penetration grade) bitumen was aged in case of ST and LT with an alternative aging process, called Oxidative Aging (OxiA). The produced aged bitumen was modified with certain amount of WVCO from 2 to 10% by weight of base bitumen with two increments. To evaluate the effect of WVCO on properties of bitumen, series of test including physical and rheological ones were implemented. The following consequences can be highlighted based on the results obtained from presented study.

The first finding is WVCO and aged bitumen can be recovered effectively with bitumen rejuvenation. The second is evaluation method and test temperature are two critical parameters to determine the optimum rate of WVCO for rejuvenation of aged bitumen. Thirdly, it can be said that the contribution rate of WVCO changed according to aging terms and higher WVCO rate was found necessary to rejuvenate the LT aged bitumen. The next, approximately 3% and 6% of WVCO is required for rejuvenation of ST and LT aged bitumen, respectively. The optimum contribution rates of WVCO for both short and long-term aging cases were identified based on the PRI analysis for the bitumen that has 70/100 penetration grade. PRI can be used to determine the optimum rate of any rejuvenation agents such as WVCO.

In general, recovering of WVCO as a rejuvenation agent for recovering aged bitumen will provide not only environmental protection, but also conservation of unrecoverable resources. Moreover, both industrialist and public can benefit by using these two materials in circular economy.

6. Recommendation and future studies

The current study fills a gap in the literature by proposing the optimum contribution rate of WVCO for ST and LT aged bitumen by means sustainable recovery of the two waste materials. However, the scope of this study is limited in terms of bitumen type/origin. It is known that the chemical configuration of bitumen may be different due to its origin or production style thus it affect its performance. Moreover, this study is established to evaluate only the bitumen performance not the hot mix asphalt (HMA) performance. It would be worth if HMA samples were prepared with WVCO rejuvenated aged bitumen for performance evaluation. Moreover, the results can be analyzed based on the temperature effect, which is not included throughout the current study.

Declaration of Competing Interest

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

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