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Investigation of the Performance of Bio-Oils from Three Different Agricultural Wastes as Rejuvenators for Recycled Asphalt

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ABSTRACT

The aim of this study was to investigate the possibility of using bio-oil obtained from pinecones, olive mill pomace, and wheat straw as rejuvenators for the reuse of aged asphalt binders. Additionally, the biomass used for bio-oil production was selected from waste materials. Therefore, it makes great contributions to both the environment and the economy. B50/70 bitumen was selected as the neat binder. The bio-oils used in the study were obtained as a result of pyrolysis. Bio-oil rejuvenators at 5%, 10% and 20% by the weight of the binder were added to the aged binder obtained from recycled asphalt mixtures to obtain bioregenerated asphalts. The physical and rheological properties of bioregenerated asphalts were investigated and not compared on neat and aged binders through penetration, softening point, rotational viscometer and dynamic shear rheometer tests. In addition, the effects of temperature and biooil content on complex modulus properties were examined using response surface methods. It was found that while the bio-oils increased the penetration values of the aged binders, they also decreased the softening point and viscosity values. The bio-oils significantly modified rutting resistance of the aged binder. The addition of bio-oil improved the viscous components and can rejuvenate the viscoelastic properties of aged asphalt binders to that of almost the original level. In addition, response surface methods results showed that the interactions between both independent variables were effective. Finally, high coefficient of determination (R^2) values indicated good agreement between the actual and predicted values. It was recommended as a result of the study that 20% concentration of bio-oil should be used to rejuvenate the aged asphalt binder for reuse in pavement construction.

Keywords: Bio-oil, rejuvenator, aged asphalt, rheological properties, response surface methodology.

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1. INTRODUCTION

The consumption of bitumen used in road construction and maintenance has paved the way for prices of bitumen obtained from petroleum crude to significantly increase [1-4]. In addition, a trend to sustain environmentally friendly coating has started [2, 5-8]. Environmentally friendly pavement is the reuse of recycled pavement. Thus, instead of traditional bitumen, alternative binders have been investigated. In this sense, reclaimed asphalt pavement (RAP) is among the effective ways to decrease bitumen consumption [9]. Aging of asphalt binders during construction and service time poses a significant problem in pavement engineering [10-12]. Oxidation and loss of volatile substances are the primary reasons for aging of asphalt binders. They lead asphalt binders to increase in viscosity and to be harder than fresh asphalt binders [13, 14]. In order to overcome these problems, many types of rejuvenating RAP binders have been used [9, 15-19].

Of a number of renewable energy sources, bio-oil draws a considerable attention as a result of its advantages such as wide range of sources, high efficiency and low prices [2, 20, 21]. Therefore, agricultural and natural biomass resources are of crucial importance. Although grain, legumes, fruits and vegetables are the primary foods in human nutrition, they also lead to a large amount of waste. Among them, wheat is one of the most common grain produced in the world and in Turkey. The wastes of grain such as stem, straw, and stubble are called agricultural waste [22]. These wastes produced as a result of their harvesting and processing or left in the cultivated area cannot be utilized appropriately [23, 24]. The use of these wastes for fuel is a widely practiced method. However, their burning results in greenhouse gases which cause global warming. It is reported that only in Spain, 11 Tg (1 Tg=109 kg) of carbon dioxide, 23 Gg (1 Gg=106 kg) of nitrogenous compounds and 80 Gg particulate matter are released into the atmosphere annually due to the burning of grain wastes [25]. In addition, many animals will be burned as a result of the burning of stubble. Therefore, the use of straw waste as a rejuvenator will offer great benefits to the economy, environment and ecosystem. Wheat straw is obtained as a plentiful by-product of wheat, which is produced 529 million tons/year worldwide. Asia provides 43% of global wheat production and is the largest production region. The typical harvest of wheat straw is 1.3-1.4 kg per kg of wheat grain. Having an area of 78 million hectares, Turkey has a rich ecological diversity. Forests have a remarkable place in this richness in terms of species and composition. As of 2020, forest areas covered 29.4% of the country's area. In addition, the pine forests in Turkey cover 54,000 ha^2. The General Directorate of Forestry reported that the total pine cone production was approximately 3500 tons in 2006. In the world, extensive amounts of cone are produced every year in pine fields grown especially for the pulp and paper industry [26]. Olive mill pomace is a by-product from olive oil factories and is an important biomass in Mediterranean countries. It is a solid waste which is composed of olive seeds and pulp as a result of olive oil production. In olive oil production, “oil-free olive mill pomace” is obtained by removing the 2-12% of oil in “crude olive mill pomace”. In world olive oil production, Turkey follows Spain, Italy and Greece. In Turkey, 1.000.000 tons of olives and olive oil are produced annually and approximately 450,000 tons of olive mill pomace is obtained. Similar to other wastes, olive mill pomace is generally used as fertilizer or for fuel in industry [27]. Bio-oil is mainly produced through biomass pyrolysis process. Pyrolysis are divided into two types as slow pyrolysis and fast pyrolysis. The difference between slow and fast pyrolysis is the residence time in the pyrolysis process. Although the residence time of the latter is less than 10 seconds and the residence time of the former differs from 5 minutes to 12 hours [28]. As a result of pyrolysis, three main components are produced, including biochars, gases and liquids. The liquid is considered bio-oil [29-31]. Bio-oils have been increasingly used to modify or partially replace asphalt binders [2, 32, 33].

Previous studies revealed that bio-oils are able to soften asphalt binders and contribute to enhancing their low temperature performance [34, 35]. Furthermore, some researchers focused on the potential of reclaimed asphalt pavement to be restored to its original condition. Several types of bio-oil such as waste vegetable oil, organic oil and distilled oil can be used as rejuvenators. The rejuvenation effects differ depending on the types of resource. In particular, it rejuvenated the aged binder by reducing the performance grade of the aged binder with a waste vegetable oil content of 12 wt% from PG 94-12 to PG 64-22. Therefore, the cracking resistance of the binder decreased [17]. By adding 1.75–2% by weight to the aged binder, the bio-oil rejuvenator obtained from biodiesel residue increases the crack resistance at low temperature by compensating for the loss of light components of the aged binder [36]. Waste cooking oil with a concentration of 3-4% by weight can rejuvenate the physical and rheological properties of bitumen with a penetration degree of 40/50 by approximating those with a penetration degree of 80/100 [37]. In their study, Rzek et al. obtained rejuvenator through the waste tires pyrolysis. They added the rejuvenators to the recycled asphalt mixtures at three different rates. They concluded that the addition of a recently developed alternative rejuvenator at the rate of 20% increased the utilization rate of the reclaimed asphalt. The standard mechanical tests and rheological tests applied to the asphalt mixtures showed that the addition of this alternative rejuvenator to the asphalt mixtures could increase the recycled asphalt ratio up to 60% [38]. Avsenik et al. investigated the effect of bio-oils produced as a result of the pyrolysis of waste tires on aged bitumen. They used lab-aged bitumen with a penetration degree of 50/70 using reference short- and long-term aging procedures. They evaluated the effect of rejuvenating addition at four different rates (3%, 5%, 10% and 20%) on aged and unaged bitumen through standard mechanical and rheological tests. The mechanical and rheological experiments indicated that the rejuvenator was suitable for the modification of aged bitumen [39]. Nizamuddin et al. used bio-oil produced from hydrothermal liquefaction of waste plastic films (linear low-density polyethylene - LLDPE) for the rejuvenation of laboratory aged bitumen. The neat binder caused the aged binder to harden; however, they determined that bio-oil rejuvenator significantly softened the aged binder [40]. Mirhosseini et al. conducted a laboratory-based research study evaluating the performance of asphalt mixtures prepared with binders modified with palm kernel oil (DSO) containing different components. They determined that while the rutting performance of the mixtures deteriorated, the addition of DSO increased the fatigue life of samples containing 20% RAP by up to 15% [41].

Mostly, the properties of bio-rejuvenators rely on the biomass. To the best of the researchers knowledge, there are no studies on rejuvenation of RAP binder in which a bio-oil produced from wheat straw, pine cone and olive mill pomace is used as a rejuvenator. The usage of bio-oil as a rejuvenator to recycle the aged binder allows the reuse of waste biomass resources and waste construction materials, which contributes to the environment and sustainable development. In this study, the physical and rheological properties of bio-rejuvenated binders were examined. To do this, bio-based rejuvenators obtained from wheat straw, pine cone and olive mill pomace were used to rejuvenate the aged binder of RAP. In addition, the effect of these three different biorejuvenators on the aged binder was evaluated by comparison. In addition, the effect of these three different biorejuvenators on the aged binder was compared.

2. MATERIAL AND METHODS

2.1. Neat binder and reclaimed asphalt pavement

B50/70 grade bitumen obtained from TÜPRAŞ Batman Refinery was used in the study. The general properties of the bitumen are shown in Table 1. In addition, single-source reclaimed asphalt pavement (RAP) obtained from Elazig Municipality, Turkey was used. It was stored in sealed containers to prevent environmental effect. The reason for using this RAP material was that B50/70 grade bitumen was used in the initial production phase. The aim of this study was to add rejuvenators to the RAP binder and restore the initial hardness of bitumen. Therefore, B50/70 grade bitumen was preferred as neat bitumen. In order to choose HMA mixing and compaction temperatures, equiviscous temperature ranges are also established using the rotational viscometer [42]. Viscosity-temperature graphs are used to calculate mixing and compaction temperatures.

Table 1 - General features of bitumen

Properties	Unit	Standard	Results
Penetration	dmm	EN 1426	56
Softening point	˚C	EN 1427	53.3
Flash Point	˚C	EN ISO 2719	245
Density	g/cm^3	ASTM D70 - 18a	1.015
Elastic Recovery	%	EN 13398	30
Viscosity 135˚C /165˚C	cP	ASTM D4402	737.5/225
Mixing temperature range	˚C	-	159-165
Compaction temperature range	˚C	-	145-151

2.2. Biomass

In the study, ground wheat straw, ground pine cone and olive mill pomace were used as biomass sources. The materials were obtained from Çanbaylar company located in the province of Elazig. In order to use material with the same granule size, the materials passing through No. 30 sieve were used. Thus, it was aimed that the size of the biomass did not have an influence on the experimental results.

2.3. Properties of the Reclaimed Asphalt Pavement (RAP) Material

The RAP used in the study was made on the RAP taken from the Elazığ central highway. This RAP material is milled from a highway that has been used for 10 years. The bitumen properties of this RAP material used 10 years ago were taken from the 8th Regional Directorate of Highways. For this reason, the reference 50/70 bitumen was chosen. Sampling of the RAP material was carried out in line with the EN 932-1 standard [43]. An extraction experiment was performed to examine the gradation of the reclaimed asphalt pavement material (Figure 1a). As a result of the extraction experiment, the amount of bitumen in the

RAP material was determined as 4.6%. The aged binder separated from the reclaimed asphalt binder through extraction was recovered using a rotary evaporator consistent with the TS EN 12697-3+A standard [44] (Figure 1b). The bitumen content was determined by performing 50 extraction experiments. Throughout the study, reclaimed aged binder was used.

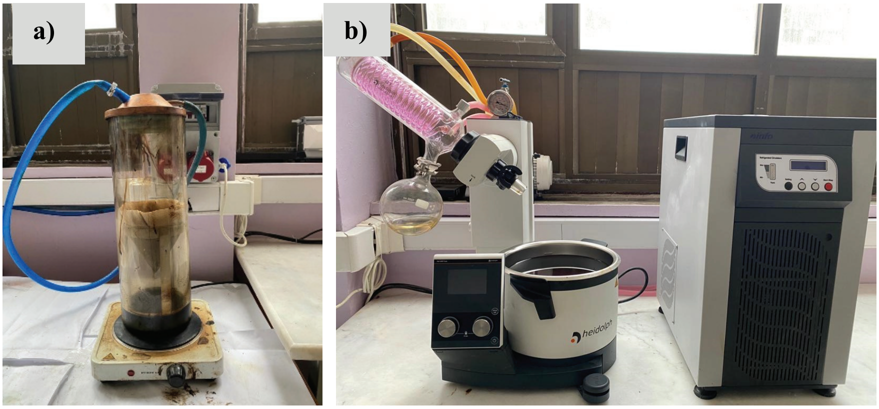

Figure 1 - a) Extraction device b) Rotary evaporator

2.4. Bio-oil Production

Three different rejuvenators obtained from bio-based agricultural products were used. Accordingly, wheat straw, pine cone and olive mill pomace bio-oil were used in the study. In order to use material with the same granule size, they were sieved through No. 30 sieve. Thus, the size of the biomass was selected so that it prevents affecting the experimental results. A slow device was used for the pyrolysis of biomass. The slow pyrolysis experimental setup consisted of a high temperature resistant cylindrical vessel with an inner diameter of 150 mm and a height of 240 mm in which the biomass was placed, a programming device box where the test temperature could be adjusted, a water-cooling system and a chamber where the biogas was condensed into oil after cooling. The slow pyrolysis experimental setup is shown in Figure 2a.

In the study, 1000 grams of biomass was placed in the chamber of the device each time in order to eliminate the effect of biomass amount on the carbonization. Accordingly, 1000 grams of dried biomass sample was exposed to pyrolysis at 500°C in a fixed bed slow pyrolysis setup shown in Figure 2b. This temperature was preferred on the basis of the studies in the literature [45]. Liquid pyrolysis product (bio-oil) was obtained as a result of condensing the pyrolysis vapors in a water-cooled condenser. In addition, non-condensable pyrolysis gases were burned and disposed of in a fume hood at the exit of the setup. The pyrolysis process was continued until the gas output stopped. The obtained liquid pyrolysis product and pyrolysis residue biochar were cooled and then weighed. By subtracting the total weight of the products from the amount of dry biomass used initially, the amount of non-condensed gas product was calculated. The results are presented in Table 2. The liquid products (Figure 2) were subjected to a vacuum evaporation (Heidolph rotary evaporator) process at 80°C and 200 mmHg pressure in order to separate the water and organic acids from the oil phase in a vacuum rotary evaporator since they contained a high percentage of water (Figure 1). As a result, high viscosity oil products were obtained for pine cone, olive mill pomace and wheat straw, in which organic acids such as water and acetic acid were removed at a rate of 12.75%, 11.56% and 10.95% by weight, relative to the amount of dried biomass.

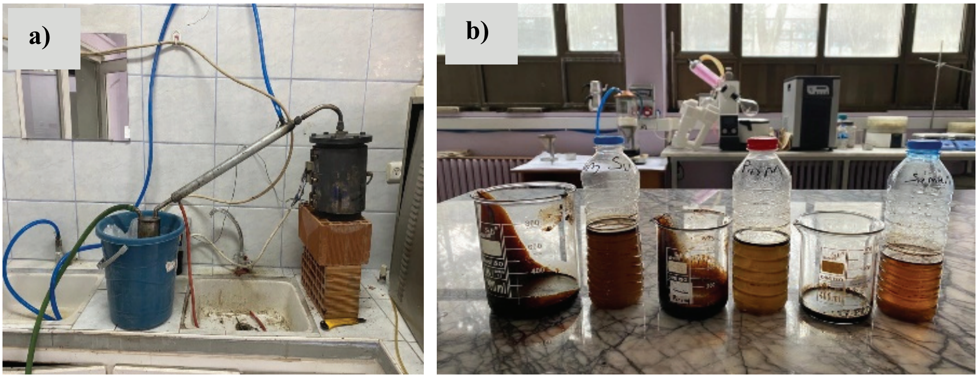

Figure 2 - a) The slow pyrolysis experimental setup, b) Bio-oil and water samples obtained as liquid product as a result of pyrolysis

Table 2 - Yields of liquid bio-oil, bio-char and non-condensed gas obtained as a result of pyrolysis of biomass

Biomass	% Bio-oil yield	% Bio-char yield	% Gas yield
Pine cone	37.92	32.09	29.99
Olive pomace	31.83	32.52	35.65
Wheat straw	27.13	31.57	41.30

2.5. Preparation of Modified Bitumen

In the study, it was aimed to bring the traditional and rheological properties of aged bitumen closer to the traditional and rheological properties before aging. The purpose of using the three ratios in the study is to capture the traditional and rheological properties of the aged binder and the traditional and rheological properties of the unaged binder. Three different ratios (5%, 10% and 20%) were used for the bio-oils. The modified bitumen used in the study was prepared as stated below.

First, the aged binder obtained from RAP was heated in a vacuum furnace at 140±5°C for 30 minutes to become fluid. The fluidized bitumen was poured into the metal chamber of the mixer as 400 grams and placed inside the thermal jacket on the heater source conditioned at 140±5°C to provide a homogeneous heat source, and then waited until the heater reached thermal equilibrium. Rejuvenators at different percentages by weight of the determined bitumen were added into the hot bitumen and mixed with a mechanical mixer operating at 500 rpm for 40 minutes, and modified bitumen was obtained. The same procedure was followed for all rejuvenators. This modification was preferred on the basis of the studies in the literature [13].

After the mixing process was completed, the obtained modified bitumen was transferred to the glass beaker. The same procedure was adopted for mixing each time in order to avoid its interference the test results [45]. Two sets of modified bitumen were prepared for each modification process. Three samples were prepared for each experiment. The abbreviations used for bituminous binders are shown in Table 3.

Table 3 - The abbreviations used for bituminous binders

Biomass	**Bio-oil content (%)**			
	0	5	10	20
Pine cone		K5	K10	K20
Olive pomace		P5	P10	P20
Wheat straw		S5	S10	S20
Neat binder	SB			
Recycled aged binder	RAP			

2.6. Conventional Properties Test

Conventional physical properties of the asphalt binder were determined, including penetration (ASTM D5) and softening point (ASTM D36) at 25°C.

2.7. Rotational Viscometer Test

The rotational viscometer (RV) test is used to determine the viscosity characteristics of bituminous binders at high temperatures. The rotational viscometer values of bituminous binders were determined using a Brookfield DV-III device according to ASTM D44402 standards. In the study, viscosity values of neat binder samples and samples of binders with waste oils were measured at two different temperatures (135°C and 165°C).

2.8. Dynamic Shear Rheometer Test

Determination of complex shear modulus (G*) and phase angle (δ)

Dynamic shear rheometer (DSR) is a test method used to characterize viscous and elastic behaviors of bituminous binders and moderate and high temperatures. In the DSR test, the complex shear modulus (G*) and phase angle (δ) of binders are determined. Permanent deformation is controlled by limiting them to 1.0 kPa for unaged original binders [46]. The tests were conducted by using Bohlin DSR-II rheometer on neat and modified bitumen according to ASTM D7175 standards. The tests were also conducted with a plate with 25 mm diameter and 1 mm plate clearance at 1.59 Hz frequency value, and at 52°C, 58°C, 64°C, 70°C, 76°C, 82°C and 88°C.

Multiple stress creep recovery (MSCR) test

In this study, the multiple stress creep recovery (MSCR) test was conducted according to AASHTO T350 standards on neat binders and binders with waste oils, which were prepared according to T315 [47] standards, by using 25 mm plates at 64°C, 70°C, 76°C, 82°C and 88°C. The test is conducted by applying 10 cycles of 0.1 kPa stress and 10 cycles of 3.2 kPa stress at different temperatures (Figure 3). Each cycle consists of 1 second of shear stress application and 9 seconds of the recovery period. However, this characterization contradicts the purpose of the MSCR test, where non-recoverable strain accumulates gradually at both loading rates. Therefore, 0.1 kPa cannot represent the LVE behavior of the material. With this test, whether the rejuvenators in neat binders affect the elastic recovery and rutting characteristics is determined. With the MSCR test, two parameters are obtained according to AASHTO M332. These are the percentage of recovery (R) and permanent creep compliance (J_{nr}) values. For bituminous binders, the mean recovery rates (R) of binders are calculated according to formulas 1 and 2 at 0.1 ($R_{0.1}$) and 3.2 ($R_{3.2}$) kPa shear stress levels [48].

$$R_{0.1} = \frac{\sum_{N=11}^{20} [\varepsilon_r(0.1, N)]}{10} \tag{1}$$

$$R_{3.2} = \frac{\sum_{N=1}^{10} [\varepsilon_r(3.2, N)]}{10} \tag{2}$$

Here, ε_r (0.1, N) and ε_r (3.2, N) represent the percentage of recovery at N number of cycles and 0.1 and 3.2 kPa stress levels, respectively while N represents the number of cycles at any level of stress. It was determined that the J_{nr} parameter provided a better correlation with rutting resistance according to Superpave PG criteria [49, 50]. The calculations of the fit values for permanent creep for 0.1 kPa (J_{nr} 0.1) and 3.2 kPa (J_{nr} 3.2) were presented in Formulas 3 and 4 while the calculations of the J_{nrdiff} value were presented in Formula 5.

$$J_{nr0.1} = \frac{\sum_{N=11}^{20} [Jnr(0.1, N)]}{10} \tag{3}$$

$$J_{nr3.2} = \frac{\sum_{N=1}^{10} [Jnr(3.2, N)]}{10} \tag{4}$$

$$J_{nrdiff} = \frac{(J_{nr3.2} - J_{nr0.1})}{J_{nr0.1}} * 100 \tag{5}$$

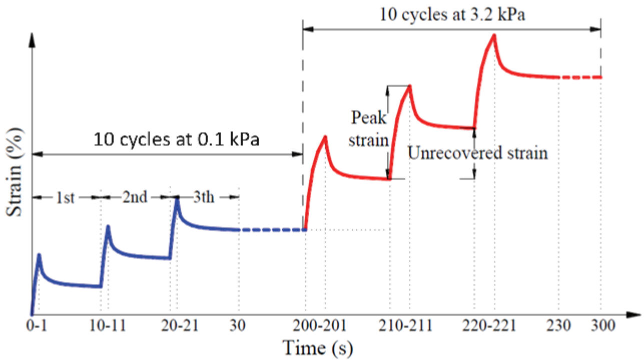

Figure 3 - Schematic representation of the MSCR system

2.9. Method of Analysis and Design of Experiments Using RSM

Response surface methodology (RSM) is a statistical instrument used in the design of experiments to increase relationships between a number of independent factors and one or more responses via mathematical concepts. In addition, it can be used to optimize the variable of response within factors in the experiment. Although RMS choice is based on the number of experimental factors and the variation level of each factor, central composite design (CCD) is largely used for its appropriateness in sequential experiments and providing the best quality predictions through the whole design space [51, 52]. In order to develop an experimental design, to provide mathematical models and statistical analysis of responses and to obtain optimal solutions for the parameters, Design-Expert v12 was used. Finally, it is calculated depending on the input parameters using the RSM, which is then evaluated for the choice of the most appropriate model that fits the correlation between the input and output parameters [36]. In this study, the effects of two input parameters, namely bio-oil in the range of 5-20% and temperature between 52°C and 88°C, were analyzed. It was calculated based on central composite design (CCD). Related studies and preliminary results in the literature were applied to reveal input parameters and corresponding domains of significance [20,21,29,37-41]. CCD is the most frequent and effective method used to statistically evaluate the interaction between independent variables and responses over the experimental range [26,27,42]. Based on the input parameters, five center point replicates were performed to allow robust evaluation of errors, with the RSM running in random order for the individual responses analyzed. Linear models developed through RSM were used for data design and analysis. The numerical variables of the experiments are converted into coded form using the following equation.

$$X_i = \frac{x_i - x_0}{\Delta X} \qquad (6)$$

Here, X_i is the ith independent factor coded value, X_i, X_0, are the actual values of the center point; here ΔX, represents the step change for the ith variable. The appropriate quadratic polynomial model suggested in the literature was used for the prediction of optimal conditions as shown in the following equation [53, 54]:

$$Y = \beta_0 + \sum_{i=1}^{n} \beta_i x_i + \sum_{i=1}^{n} \beta_{ii} x_i^2 + \sum_{i=1}^{n} \sum_{j=1}^{n} \beta_{ij} x_i x_j + \varepsilon \tag{7}$$

Y is the calculated response, β_0 is the constant value. Independent variables in coded form are defined as x_i and x_j. The coefficients β_i and β_{ii} are linear and quadratic terms. β_{ij} is the coefficient of interaction term, ε is the random error and the number of factors is defined as n. In addition, analysis of variance (ANOVA) was run to examine the appropriateness of the proposed model. The coefficients of determination (R^2 and R^2_{adj}) express the goodness of fit to the suggested model. These values can be determined using the equations below [43]:

$$R^2 = 1 - \frac{SS_{residual}}{SS_{model} + SS_{residual}} \tag{8}$$

$$R^2_{adj} = 1 - \frac{SS_{residual} / DF_{residual}}{(SS_{model} + SS_{residual}) / (DF_{model} + DF_{residual})} \tag{9}$$

where R_2 and R^2_{adj} represent the determination coefficients; SS_{res}, SS_{mod}, and DF_{res}, DF_{mod} represent the sum of squares and degrees of freedom for residual and models, respectively. The F-test was used at a 95% confidence level for the proposed models that were statistically appropriate and represented based on the p-value. Also, the F test was used to verify the adequate precision (AP) of the model, and the data-set variability was controlled through Standard Deviation (SD).

3. RESULTS AND DISCUSSION

3.1. Conventional Properties

Figure 4 shows the penetration and softening point values of the binders. Penetration values of bio-oil modified bitumen increased linearly with increasing additive content. The neat binder provided the highest penetration value and RAP binder provided the lowest penetration value. All the bio-oils rejuvenated the aged bitumen. It was found that the most effective bio-oil was the one obtained from the pine cone. Compared to the RAP binder, 5% bio-oil content did not significantly increase the penetration value. Penetration values of K5, K10 and K20 binders increased by 1.5, 2.1 and 3 times, respectively, compared to the penetration value of RAP binder. It was also found that the penetration values of P5, P10 and P20 binders increased by 1.3, 1.7 and 2.5 times, respectively, compared to the penetration value of RAP binder. Finally, compared to the penetration value of the RAP binder, the penetration values of S5, S10 and S20 binders were found to increase by 1.4, 1.9 and 2.4 times, respectively. As a result, it was found that the aged bitumen softened due to the addition of rejuvenators to the aged bitumen obtained from the recycled bitumen mixture.

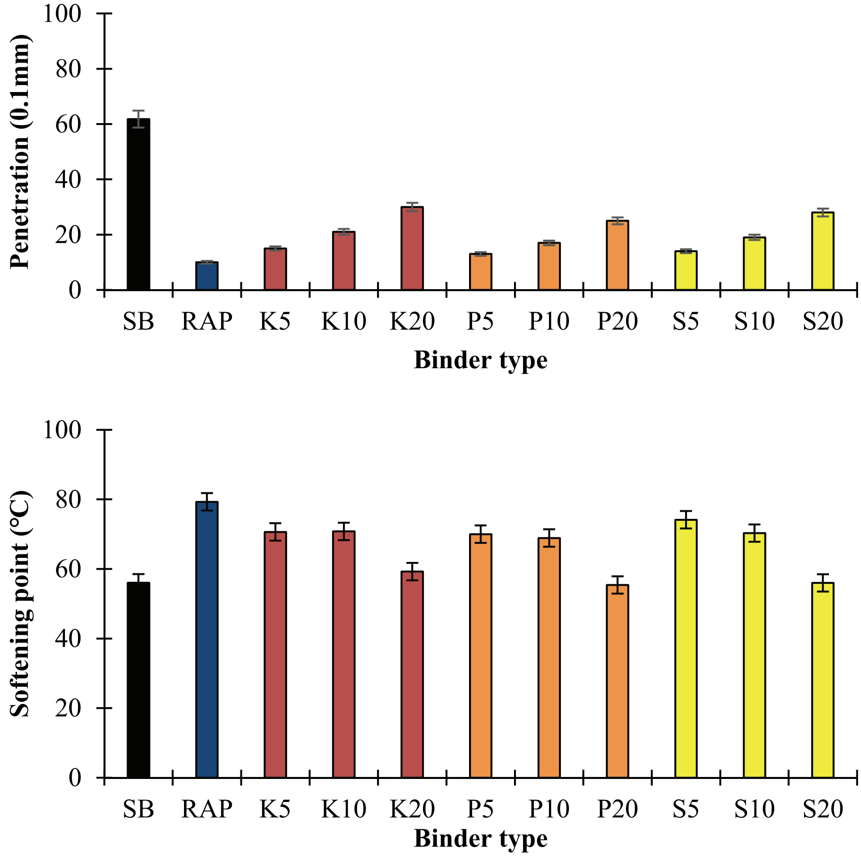

Figure 4 - Conventional properties of asphalt binders

There was a linear relationship between softening point values and biooil content. It was revealed that the softening point values of the RAP binder decreased with the addition of rejuvenators to the RAP binder. Additive contents of 20% provided almost similar softening point values. Especially after 10% additive content, softening point values of bio-oil modifications differed. The softening point value of the bitumen obtained from the recycled asphalt mixture was 1.41 times higher than that of the neat binder. This value decreased because of the fact that the rejuvenators had the feature of softening the bitumen. With the addition of 5%, 10% and 20% of the bio-oil additive obtained from pine cone to the bitumen, the softening point values decreased by 10.8%, 10.72% and 25.2%, respectively, compared to the RAP binder. The softening point values of P5, P10 and P20 binders decreased by 11.72%, 13.11% and 30.14%, respectively, compared to those of the RAP binder. Finally, softening point values of S5, S10 and S20 binders decreased by 6.43%, 11.34% and 29.38%, respectively, compared to the RAP binder. Among the three bio-oils, the most effective bio-oil in the softening point values was the one obtained from olive mill pomace. As a result, it was revealed that all three bio-oils had softening properties on the aged binder. When added to the aged binder (RAP binder) at a rate of approximately 20% from all three bio-oils, it was seen that it reached the softening value before aging (neat binder).

3.2. Viscosity and Workability Requirement

The variation in the viscosities of the neat and modified binders at 135 °C and 165 °C are shown in Figure 5, respectively. As the additive content of biooil modified bitumen increased, a decrease was observed in viscosity values at both 135 °C and 165 °C. It was found that the viscosity value of the RAP binder at 135°C was 6338 cP which is 2.11 times the standard condition of 3000cP. In addition, negative effect on workability was revealed. Although the viscosity value of the unaged binder at 135°C was 675 cP, the viscosity value of the binder as a result of aging was 6338 cP, which indicated the effect of aging. The change in viscosity trend was similar for 135°C and 165°C. The addition of rejuvenators to the aged binder removed this problem. As the amount of bio-oil added to the aged binder increased, the viscosity values of the aged binder decreased and reached almost the pre-aging viscosity value. Compared to the aged binder at 135ºC, bitumen modified with the biooil obtained from pine cone at the rate of 5%, 10% and 20% provided 52.67%, 54.43% and 83.83% lower viscosity values, respectively. At 165 °C, it provided 35.52%, 39.47% and 69.74% lower viscosity values than the RAP binder, respectively. Bitumen modified with the biooil obtained from 5%, 10% and 20% olive mill pomace had 46.54%, 57.98% and 87.18% lower viscosity, respectively, than the aged binder at 135ºC. At 165 °C, it gave 30.26%, 43.42% and 72.37% lower viscosity values than the RAP binder, respectively. Bitumen modified with the biooil obtained from 5%, 10% and 20% wheat straw provided lower viscosity values of 38.86%, 57.59% and 86.59%, respectively, compared to the aged binder at 135ºC. At 165°C, it gave 28.95%, 42.11% and 71.84% lower viscosity values than the RAP binder, respectively. As a result, it was revealed that the use of bio-oil obtained from pine cone, olive mill pomace and wheat straw as a rejuvenator decreased the viscosity value of the aged binder and almost restored the original viscosity value. Among the bio-oils, the most effective bio-oil for viscosity values was pine cone at 5%, and olive mill pomace at 10% and 20%.

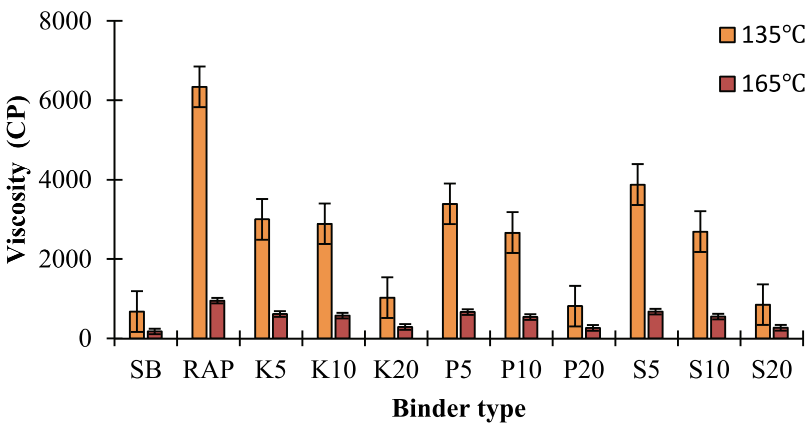

Figure 5 - Viscosity values of binders

The enhanced fluidity of the biooil and the higher content of light compounds in the biooil may play a role in the reduced viscosity of the aged binder. All the viscosity tests indicated that the biorejuvenators can reduce the stiffness of the aged binder. It was concluded that a concentration of 20% bio-rejuvenator is sufficient to rejuvenate the aged asphalt binder to be reused in pavement construction. The findings in the literature is in line with this conclusion [55-57].

3.3. Complex Shear Modulus (G*) and Phase Angle (δ)

Figure 6 shows the change in shear modulus of neat and modified binders with increasing temperature. As the temperature increased, the shear modulus decreased logarithmically, and this decrease showed a similar trend for all connector types. Accordingly, the shear modulus decreased by about 50% for every 6 degrees of temperature increase, while the G*/sinδ value of the SB binder decreased below 1000 Pa at 76 ºC, the K20, P20 and S20 binders provided 1623 Pa, 1092 Pa and 1658 Pa G*/sin δ values at 76 ºC, respectively. On the other hand, the RAP binder gave a G*/sinδ value of 21270 Pa at 76°C. Except for the neat binder, all binders do not fall below the limit condition of 1000 Pa at 88°C. Except for the neat binder, all binders did not decrease below 1000 Pa at 88°C which is the limit condition. After 76°C, the G*/sinδ values approximated to each other. It was determined that all three bio-oils reduced the high temperature performance level of bituminous binders, which is an indication of rejuvenating effect. The rejuvenation rate of the aged binder increased with the increase in the amount of bio-oil. The most effective results were obtained from pine cone at 5%, and olive mill pomace at 10% and 20%.

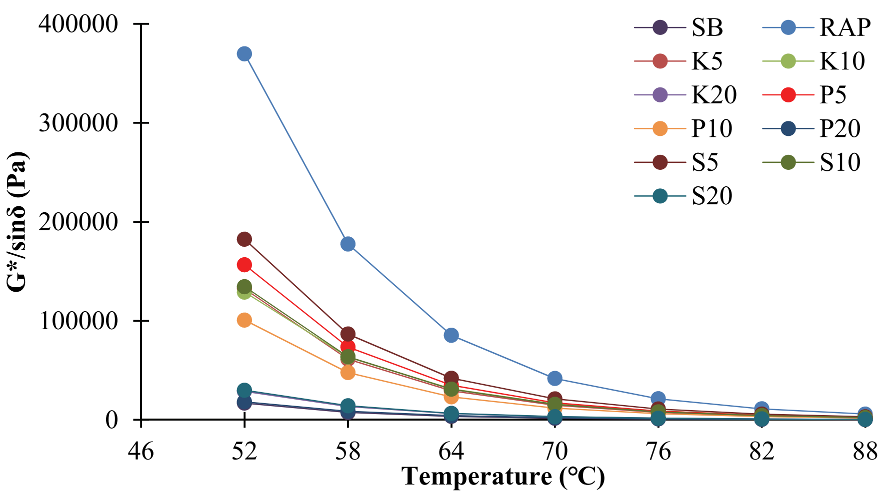

Figure 6 - Change in the G/sinδ considering temperature of the bituminous binders*

The results revealed that the properties of unaged bitumen were acquired by adding bio-oils to the aged binder, which suggests that bio-oils can be used as rejuvenating agents. The rutting parameter (G*/sinδ) is used to determine the asphalt binder's resistance to permanent deformation at high temperature. As shown in Figure 6, after the addition of all three bio-oils to the aged binder G*/sinδ values decreased compared to the aged binder. As the amount of added biooil increased, the decrease in G*/sinδ values increased. G*/sinδ values of RAP, K20, P20, S20 and SB binders at 88°C were found to be 41750, 3167, 2108, 3267 and 1658, respectively. The higher light components and viscous content in the bio-oil are capable of stabilizing the chemical components of the aged binders and thus restoring rutting resistance to a great extent. The results of this study showed that bio-oils rejuvenated the aged bitumen and approached the G*/sinδ value of the bitumen to that of unaged bitumen (50/70). In addition, the results also indicated the positive results of the bio-oils obtained from the three waste products. The studies in the literature support the results of this study [56, 57].

Figure 7 shows the change in the phase angles of the binders with temperature. The phase angle is used to characterize the viscoelastic properties of asphalt binders. The larger the phase angle is, the more viscous components present in the asphalt binder. The phase angles of all binders increase with the increase in temperature and they exhibit a more viscous behavior. The changes in the phase angle values of the three bio-oil modifications were comparable. It was observed that the phase angle values of the bio-oil rejuvenators increased as the temperature increased. A further increase in phase angle values was observed after 10% biooil content. Oxidation and evaporation of some light compounds paved the way for the aged asphalt binder to have less viscous components than the untreated asphalt binder. As the bio-oil content in the aged binder increased, the phase angle value also increased. The phase angle value of the aged bitumen obtained from the recycled asphalt mixture was 62.01 at 70°C and the phase angle value before aging was 81.97. The aged binder provided 1.32 times higher phase angle value than the unaged binder due to the effects such as environmental, climate and traffic in asphalt mixtures. Phase angle values of SB, RAP, K5, K10, K20, P5, P10, P20, S5, S10 and S20 binder at 52°C were 72.06, 52.96, 59.01, 59.12, 67.49, 58.38, 59.87, 71.08, 56.51, 59.00 and 63.52, respectively. 58.38, 59.87, 71.08, 56.51, 59.00 and 63.52phase angle values at 88℃ were 88.7, 71.13, 76.72, 76.65, 82.00, 75.95, 76.58, 84.53, 73.44, 75.99 and 80.11, respectively. The examination of the values indicated that binders with 20% bio-oil content provided approximately the phase angle value of the

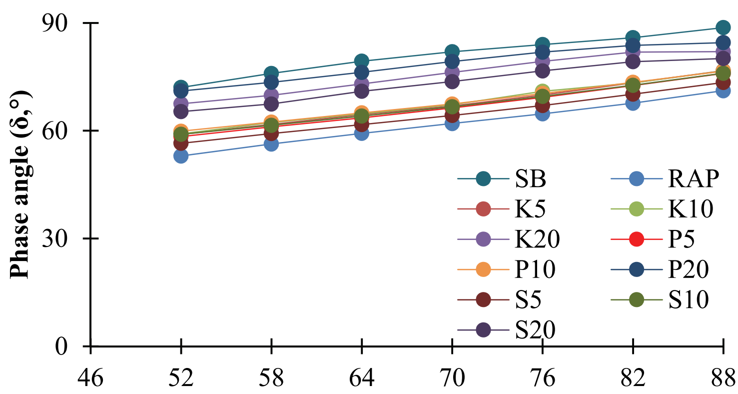

Figure 7 - Change in the phase angles considering temperature of the bituminous binders

unaged neat binder. As a result, it was found that the addition of bio-oil improved the viscous components and could rejuvenate the viscoelastic properties of aged asphalt binders to the similar level as that of the original bitumen. This is also one of the reasons why the viscosities of binders rejuvenated with bio-oils are much lower than that of aged asphalt binders. The results were in line with rotational viscosity results. Similar results were reported by Zhang et al. [57]. It was found that the most effective bio-oil was the one obtained from olive mill pomace.

Figure 8 shows the changes in the shear modulus corresponding to the phase angle of the three bio-oil modifications for different temperatures. As the amount of bio-oil used in the aged binder increased, the phase angle values increased and the G*/sinδ values decreased. P20 binder provided the closest values to that of the unaged binder (SB). These results suggest that bio-oils can be used as rejuvenators in aged bitumen.

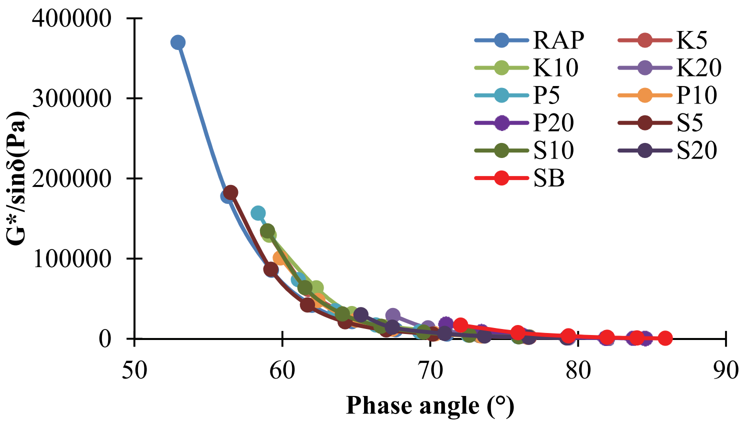

Figure 8 - Change in shear modulus versus phase angle

3.4. MSCR Test and Results

Figure 9 shows the variation of $J_{nr0.1}$ values of K, P and S added binders with temperature. The $J_{nr0.1}$ values of the binders increased with the increase in the temperature. The increase was significant after 76ºC. In the study, the J_{nr} values of the S20 binder increased the most. The RAP binder had the lowest J_{nr} value. However, with the addition of bio-oils to the RAP binder, J_{nr} values increased. Adding 10% bio-oils approached the J_{nr} values of the RAP binder to that of the SB binder. As a result of the addition of 5% and 10% bio-oil to the RAP binder, J_{nr} values provided comparable results. S20 was the fastest increasing binder after 76ºC. At 64°C, J_{nr} values of all binder were close to each other. The J_{nr} values of RAP binders with 5% and 10% content have different values, especially at high temperatures. In all three modifications, it was observed that the J_{nr} values increased significantly with the increase in the additive content. Especially after 10% additive content, there was a significant increase in J_{nr} value. At low additive contents such as 5% K, 5% P and 5% S, J_{nr} values did not change significantly. $J_{nr0.1}$ values increased with the increase in the temperature. The highest increase in $J_{nr0.1}$ values occurred in wheat straw bio-oil. The J_{nr} values of binders with 20% S additives increased by 5.22, 4.75, and 5.71 times at 64°C, 70°C and 76°C, compared to the SB binder.

The J_{nr} values increased by 4.20, 4.10, and 4.99 times in 20% P added binder and 1.91, 1,88, and 2.90 times in 20% K added binder. The highest $J_{nr0.1}$ value was observed at 88ºC for all added binders. K-added binders had the lowest $J_{nr0.1}$ values.

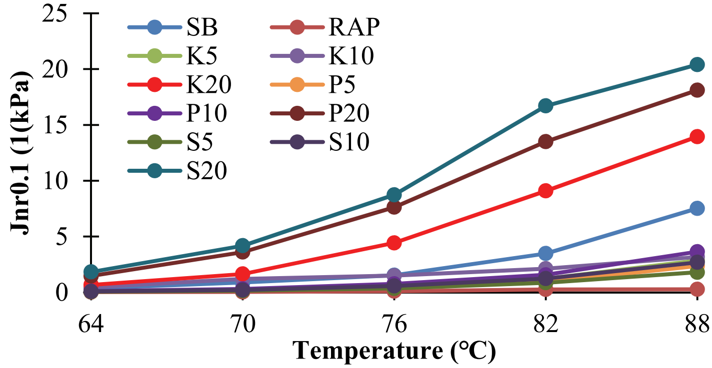

Figure 9 - Change in the $J_{nr0.1}$ considering temperature of the bituminous binders

The change in the additive content and J_{nr} values of the binders at the high shear stress level (3.2 kPa) at 64ºC is shown in Figure 10. At this stress level, a rapid increase in Jnr values was observed with the increase of bio-oil content. As the load applied to the binder increased, the Jnr values decreased. The J_{nr} values of binders containing 20% bio-oil were close to each other until 82°C. With the increase in temperature, the $J_{nr3.2}$ values also increased. S20 binder had the highest $J_{nr3.2}$ value and RAP binder had the lowest $J_{nr3.2}$ value. Binders containing 10% bio-oil had $J_{nr3.2}$ value close to that of unaged binder (SB). Different $J_{nr3.2}$ values were observed in all three additive types at 88ºC. Although the additive content did not have a considerable effect on $J_{nr3.2}$ at low temperatures, it produced an effect at high temperatures (82°C).

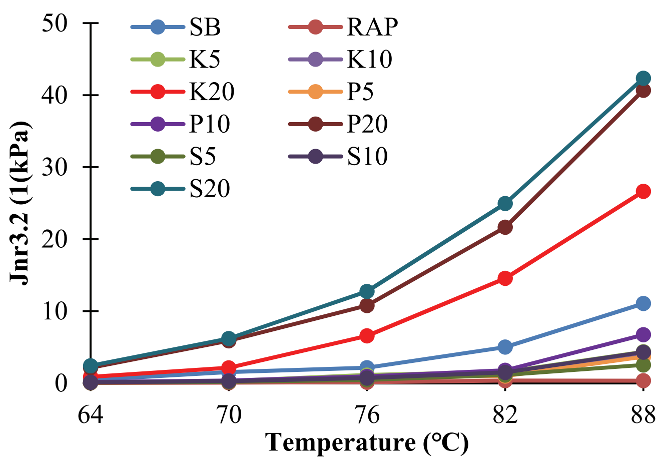

Figure 10 - Change in the $J_{nr3.2}$ considering temperature of the bituminous binders

Binders with 5% and 10% additive content provided comparable $J_{nr3.2}$ values at all temperatures except 88°C. There was a sudden increase in $J_{nr3.2}$ values in the contents after 10% biooil content. $J_{nr3.2}$ values increased at high temperatures. Accordingly, the effect of additive ratios on $J_{nr3.2}$ was greater at high temperature in all three modifications. With the increase in temperature, both $J_{nr3.2}$ values and the difference between $J_{nr3.2}$ values increased significantly.

The elastic recovery values of all binders decreased with the increase in the temperature (Figure 11). The elastic recovery values of the RAP binder decreased as the amount of biooil added to the RAP binder increased. RAP binder provided the highest elastic recovery value and S20 binder gave the lowest elastic recovery value. Especially, the elastic recovery values of P20 and S20 binders were close to those of the unaged neat binder (SB). The elastic recovery values of RAP binders containing 5% and 10% bio-oil were between 50% and 60%. It was found that the elastic recovery value of the RAP binder at 76°C increased 5.3 times compared to the neat binder (SB). These results revealed that there was an increase in the elastic recovery values of the aging bitumen and that the added bio-oil rejuvenated the aged binder. The addition of 20% wheat straw bio-oil to the RAP binder at 76°C decreased the elastic recovery value from 53% to 8%. Similarly, addition of 20% P bio-oil to the aged binder decreased the elastic recovery value 50% to 8%, restoring it to that of SB binder. These results verified the rejuvenation of the aged binder. At 82ºC, the elastic recovery value of the K20, P20 and S20 binders decreased by 68.2%, 86.6% and 86.6%, respectively, compared to the $R_{0.1}$% values of the RAP binder.

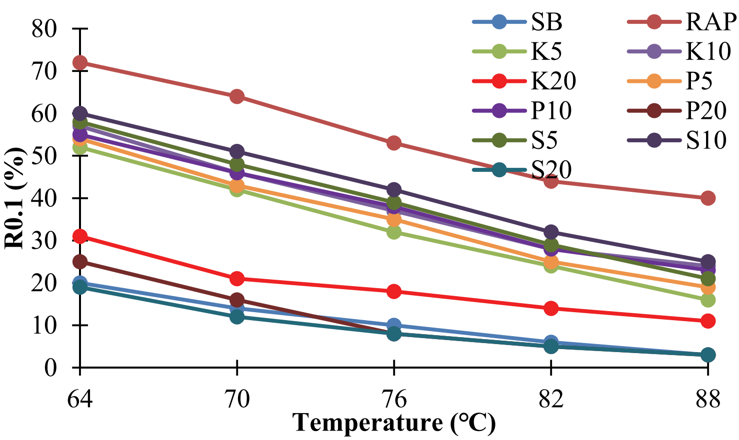

Figure 11 - %R – temperature relationship at 0.1 stress level

Figure 12 shows the change in elastic recovery values of K, P and S modified binders at 3.2 stress level with temperature. The decrease in the %R values with the increase in temperature was more evident in P and S modification. Although there was a rapid decrease until 76ºC, the rate of decrease declined after 76ºC. RAP binder had the highest elastic recovery value and the P20 and S20 binders had the lowest values. When the temperature increases from 64ºC to 76ºC, binders with 20% K, 20% P and 20% S lost 54.5%, 100% and 100% of their flexibility, respectively. As at low stress level, elastic recovery values of all modified binders decreased with the increase in temperature at high stress level. In all three modifications, the elastic recovery values decreased when the stress increased from 0.1 kPa to 3.2 kPa. High additive ratios were more effective at high stresses than at low stresses. In contrast, the elasticity values decreased with increasing stresses. Although the elasticity of the P20 binder at 0.1 kPa stress level was 38% at 76 ºC, it decreased to 19% at 3.2 kPa stress level. Under the same conditions, the elasticity of K20 and S20 binders decreased from 37% and 42% at 0.1 kPa stress level to 12% and 27% at 3.2 kPa stress level.

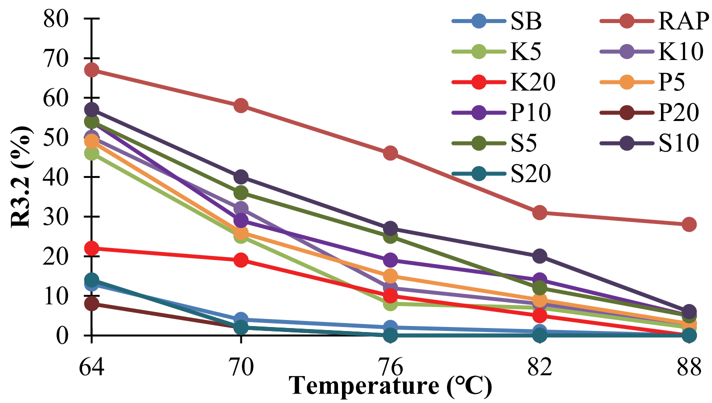

Figure 12 - %R – temperature relationship at 3.2 stress level

The fact that the difference (J_{nrdiff}) between the creep recovery of the binders at 0.1 and 3.2 kPa stress levels was greater than 75% indicated sensitivity to rutting. The change in J_{nrdiff} values of the binders is given in Figure 13. There was not a significant change in the J_{nrdiff} values for all modifications. The J_{nrdiff} values increased as the biooil content increased. With the increase in temperature, J_{nrdiff} values increased in all three modifications and when the additives were used together. The binders, except for P20, did not satisfy the 75% limit requirement at 82°C. At 88°C, K10, K20, P10, P20 and S20 met the limit requirement. RAP had the lowest J_{nrdiff} value and P20 had the highest value.

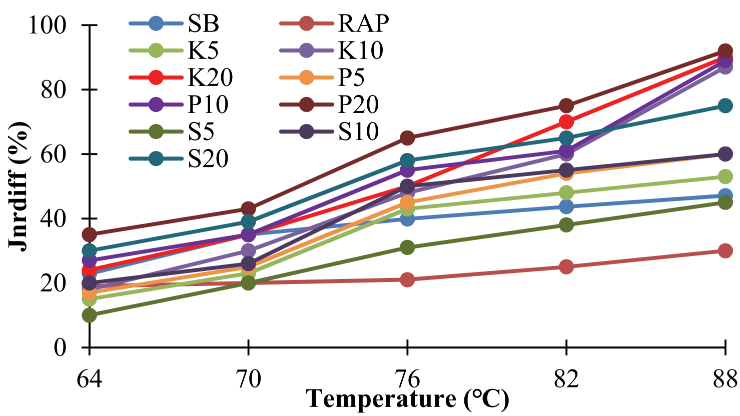

Figure 13 - Change in J_{nrdiff} values of binders

Table 4 - Overview of the binder grading according to PG and MSCR systems

Binder Type and Test Temperature (ºC)		G*/sinδ (kPa) Aged with RAP	AASHTOT315 PG	$J_{nr3.2}$ (1/kPa)	AASHTO T350 J_{nrdiff}	AASHTO T350 PG
Neat	64	85310	88	0,02	19	64-E
	70	41750		0,05	20	70-E
	76	21270		0,12	21	76-E
	82	11100		0,34	25	82-E
	88	5815		0,33	30	88-E
K5	64	29310	82	0,11	15	64-E
	70	14670		0,28	23	70-E
	76	7450		1,12	43	76-H
	82	3882		1,56	48	82-H
	88	2083		4,36	53	88-S
K10	64	31270	88	0,1	18	64-E
	70	15860		0,25	30	70-E
	76	8928		0,9	48	76-V
	82	4626		1,31	60	82-V
	88	2492		4,58	87	-
K20	64	6395	70	0,87	24	64-E
	70	3167		2,12	35	70-S
	76	1623		6,56	50	-
	82	871		14,55	70	-
	88	420		26,63	90	-
P5	64	34890	88	0,08	17	64-E
	70	17120		0,24	25	70-E
	76	8760		0,75	45	76-V
	82	4522		0,98	54	82-V
	88	2458		3,66	60	88-S
P10	64	23170	82	0,14	27	64-E
	70	11810		0,35	35	70-E
	76	6144		0,92	55	76-V
	82	3277		1,78	61	82-H
	88	1781		6,72	89	-
P20	64	4223	70	2,14	35	64-S
	70	2108		5,88	43	-
	76	1092		10,78	65	-
	82	606		21,64	75	-
	88	347		40,68	92	-
S5	64	42010	88	0,06	10	64-E
	70	21120		0,17	20	70-E
	76	10940		0,45	31	76-E
	82	5751		1,12	38	82-H
	88	3073		2,53	45	88-S
S10	64	30940	88	0,1	20	64-E
	70	15550		0,27	26	70-E
	76	8016		0,7	50	76-V
	82	4252		1,48	55	82-H
	88	2295		4,3	60	88-S
S20	64	6469	70	2,41	30	64-S
	70	3267		6,19	39	-
	76	1658		12,73	58	-
	82	912		24,94	65	-
	88	448		42,34	75	-

An overview of the binder grades based on conventional and MSCR systems is shown in Table 4. The J_{nrdiff} values obtained using the T350 approach showed that the non-recoverable creep compliance performed well. The results of the investigation demonstrated that the additions considerably improved the binders' capacity to withstand high temperatures. Some changes can withstand high traffic conditions at this temperature, according to T350. The asphalt binders' resistance to rutting rises with the amount of elastomer they include.

3.5. Statistical Analysis and Model Development

The Quadratic model was the recommended regression model for all dependent variables and the models were in good agreement with the examined parameters. The numerical variables (A, B) of the predicted models were determined using Equation 10-12. Only significant impact variables were included in the equations. The synergistic and antagonistic effects of each factor on the dependent variables are specified with a sign (negative or positive) in front of the terms in the equation. A represents temperature and B represents bio-oil content.

$$K_{Complex\ modulus} = 7.73 * 10^5 - 18073.83A - 8429.96B + 146.86AB + 102.61A^2 - 138.29B^2 \quad (10)$$

$$P_{Complex\ modulus} = 7.96 * 10^5 - 17648.40A - 15821.64B + 182.80AB + 97.20A^2 + 34.96B^2 \quad (11)$$

$$S_{Complex\ modulus} = 9.49 * 10^5 - 21409.66A - 15920.59B + 203.46AB + 119.25A^2 - 28.69B^2 \quad (12)$$

In a situation where the model fit is not satisfactory, the model functions will result in inappropriate responses. Therefore, it is of crucial importance to check the model adequacy as part of the data analysis [58-60]. Hence, ANOVA was performed to examine the adequacy and appropriateness of the proposed models. The degree of appropriateness of the proposed models was investigated through the coefficient of determination (R^2). The R^2 values for the dependent variables $K_{Complex\ modulus}$, $P_{Complex\ modulus}$ and $S_{Complex\ modulus}$ were 0.9267, 0.9254 and 0.9349, respectively. A higher R^2 value suggests stronger agreement between the expected and actual values. High R^2 values in this study indicated stronger agreement between the predicted and actual values. In addition, the predicted coefficients of determination were in acceptable agreement with the adjusted coefficients of determination which refers to an inconsistency of less than 0.2. Sufficient precision reveals the signal-to-noise ratio for comparison between the various quantities calculated at the design points and the average prediction error. The signal-to-noise ratios for the dependent variables of $K_{Complex\ modulus}$, $P_{Complex\ modulus}$ ve $S_{Complex\ modulus}$ were 20.0660 20.8163 and 22.0569, respectively. All of them were greater than 4, indicating that they provide an adequate signal. The results showed that all bio-oils can be used to navigate the design space. In the analysis, the confidence level was at 95% with a p-value less than 0.05. P-values less than 0.05 for the models also indicated that there was only a 0.0001 probability of an F-value of this magnitude due to noise [61].

2D contour and 3D graphical diagnostic plots of the relationship between independent variables and complex modulus values are shown in Figure 14. The figure shows that there is a perfect interaction between the variables and response of the bio-oil-added asphalt mixtures. The addition of bio-oil to aged bitumen decreased the complex modulus values. The binders' complex modulus values decreased with the increase of bio-oil content. As the rate of rejuvenation affecting the aged binder increased with the increase in the amount of bio-oil, a further decrease was observed in the complex modulus values. In addition, as the temperature applied to the binders increased, the complex modulus values decreased. Especially at high temperatures, the complex modulus values decreased significantly. Among the bio-oils used in the study, olive mill pomace was found to be the most effective bio-oil. The decrease in the complex modulus values with the increase of bio-oil in the aged binder can be seen in both 2D contour and 3D graph.

In Figure 14, the intensity red color in the 2D and 3D graphics changes, which indicates that the relationship between the variables is good. The effect of bio-oils on complex modulus values was determined. Accordingly, the redness in the graphs decreases as the bio-oil increases, which shows the positive effect on the complex modulus. As the amount of bio-oil used in the aged binder increases, the oils decrease the viscosity of the bituminous binders as they reduce the gravity force between the molecules. In addition, a decrease in the complex modulus value was observed with the increase in temperature, showing that the rising temperature increased the fluidity of the binders. This is due to the fact that bio-oils contain light components (i.e. aromatic solvents) that can dissolve asphaltenes, and as a result make the binders softer.

Graphs of predicted values and actual quantities were used to obtain a clear interpretation of the satisfaction of the proposed models. Figure 15 displays the plots of the predicted against the actual values of all the responses. It shows that almost all the points are distributed fairly close to the equality line. This means that the proposed models have a good agreement with the data. In addition, the distributions of all the points on the straight line showed a strong correlation between the predicted values and the experimental values. Finally, the results confirmed that the proposed prediction models can adequately navigate the design space defined by CCD.

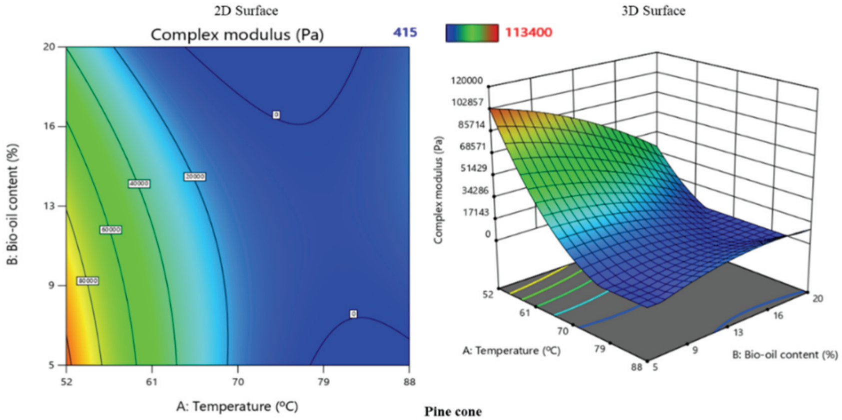

Figure 14 - Effect of fat content and temperature on complex modulus

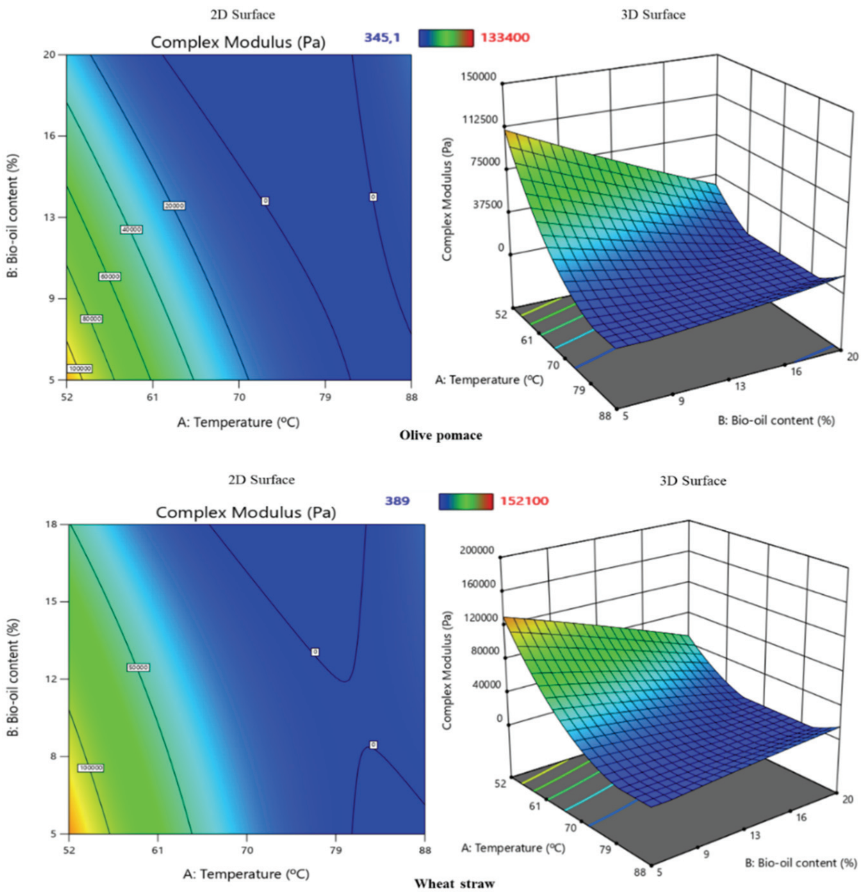

Figure 14 - Effect of fat content and temperature on complex modulus (continue)

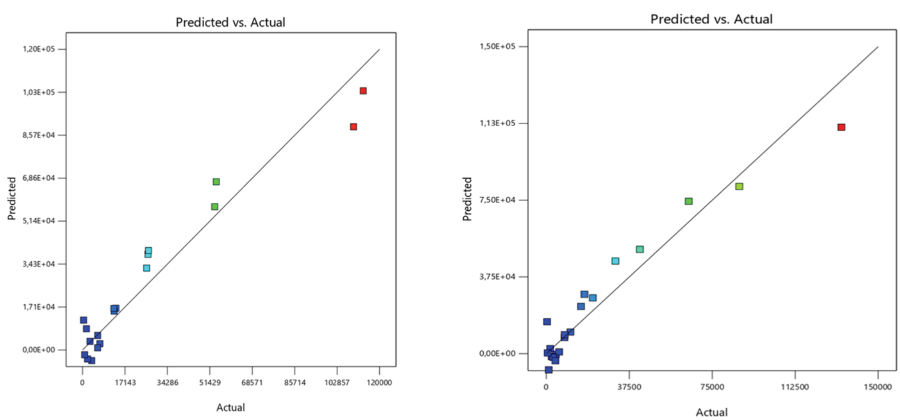

Figure 15 - Trace plot of complex modulus

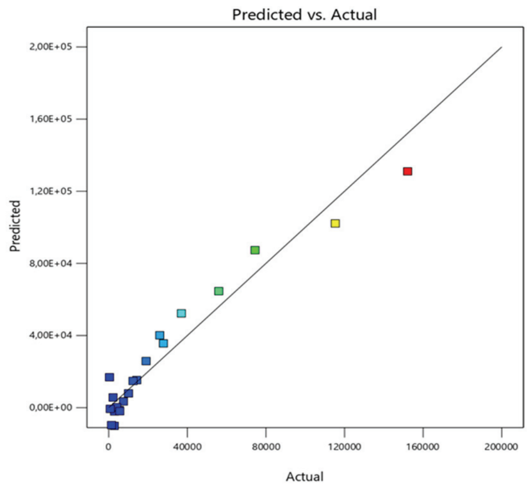

Figure 15 - Trace plot of complex modulus (continue)

4. CONCLUSIONS

In this study, physical and rheological properties of binders were investigated by adding three different bio-oils obtained as a result of pyrolysis to aged bitumen obtained from recycled asphalt mixture. The following conclusions can be drawn:

- The addition of rejuvenators to the aged bitumen obtained from the recycled bitumen mixture increased the penetration value of the aged bitumen. It was determined that all three bio-oils had softening properties on the aged binder. The addition of approximately 20% of all three bio-oils to the aged binder (RAP binder) restored the softening value to the initial value of unaged (neat binder).
- Viscosity tests showed that biorejuvenators can alleviate the stiffness of aged binder. A concentration of 20% bio-rejuvenator was considered to be approximately sufficient to rejuvenate the aged asphalt binder to be reused in pavement construction.
- The higher light components and viscous content in the bio-oil are able to stabilize the chemical components of aged binders and consequently restore rutting resistance to a great extent. The results revealed that bio-oils rejuvenated the aged bitumen and approached its $G^*/\sin\delta$ value to that of the unaged bitumen (50/70).
- In terms of permanent creep compliance, it was observed that the J_{nr} values increased significantly with the increase in the additive content in all three modifications. Especially after 10% additive content, there was a significant increase in J_{nr} value. The elastic recovery values of the RAP binder decreased with the increase in the amount of biooil added to the RAP binder. Although RAP binder provided the highest elastic recovery value, S20 binder provided the lowest value. There was no significant change in the J_{nrdiff} values for all modifications. As the bio-

oil content increased, the J_{nrdiff} values increased. With the increase in temperature, J_{nrdiff} values increased in all three modifications and when additives were used together.

- High R^2 values indicated stronger agreement between predicted and actual values. The intensity of the red color in 2D and 3D graphics changed which indicated that the relationship between the variables was good. The distributions of all the points on the straight line in the graphs showed a strong correlation between the predicted and the experimental values.

This study recommends that 20% concentration bio-oil should be used to rejuvenate the aged asphalt binder for reuse in pavement construction. In addition, future studies should investigate the mechanical properties of the regenerated asphalt mixtures.

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Credit authorship contribution statement

Beyza Furtana Yalcin: Methodology, Writing – original draft, Investigation, Data curation, Result presentation, Resources. **Mehmet Yilmaz:** Writing – review & editing, Supervision, Result presentation, Validation, Conceptualization.

Data Availability

Some or all data, models, or code that support the findings of this study are available from the corresponding author upon reasonable request.

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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