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Orginal Article

Investigation into the effect of waste engine oil and vegetable oil recycling agents on the performance of laboratory-aged bitumen

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Abstract: The pavement recycling method is one of the practical ways to integrate sustainable development into transportation infrastructure, and it has been adopted worldwide. The use of reclaimed asphalt pavement (RAP) in new asphalt highways is limited due to the ageing effects caused by UV damage and weathering on the asphalt binder. To address this issue, waste vegetable oil (WVO) and waste engine oil (WEO) have been proposed as potential rejuvenating agents to enhance the recyclability of pavements containing RAP. This study evaluated the effectiveness of WEO and WVO as chemical rejuvenating agents through various tests on both aged and virgin asphalt binders. The tests included measurements of ductility, fire, and flash points, softening points, and penetration. The results indicate that the addition of WEO and WVO can improve the low-temperature properties of asphalt binders when mixed with RAP, resulting in reduced stiffness. The mixture containing WEO and WVO exhibited improved stability compared to the control, suggesting enhanced flow with increasing waste oil content in comparison to aged binder, albeit with slightly reduced flow compared to the virgin binder. This study demonstrates the potential of WEO and WVO as recycling agents to enhance the performance of bituminous mixes incorporating RAP.

Keywords: recycling, reclaimed asphalt pavement, waste vegetable oil, waste engine oil, rejuvenating agents, asphalt binder, low-temperature properties

1. Introduction

The inclusion of rejuvenators, commonly derived from natural resources, can reduce the stiffness, viscosity, and brittleness of reclaimed asphalt pavement (RAP) binder [1-3].

A wide range of organic oils has been studied as potential recycling agents to rejuvenate aged asphalt, restoring its viscosity and flexibility [4]. Substituting virgin petroleum-based asphalt binder with waste cooking oil (WCO) can improve the low-temperature performance of asphalt pavement [5-7]. The RAP binder is highly aged and brittle; hence, it could crack easily, especially at low temperatures. As a result, low-temperature performance is a key property to evaluate when using recycled asphalt [8]. Recycling asphalt pavement also involves creating a bituminous mixture using conventional materials. The milling result, also known as RAP, is often an old and deteriorated pavement. If successful, RAP will produce a substance with properties resembling those of a conventional asphalt mixture [9]. Substantial research is being conducted to improve the sustainability and durability of pavement structures using RAP [10]. The use of non-renewable resources, such as minerals and oil, as well as the greater emphasis on environmental conservation and sustainable development, has gradually been limited [11]. The use of WEO as a rejuvenator has been noted to improve the mechanical properties and moisture resistance of the reclaimed blends [12]. Following proper rejuvenation and mixture design, RAP can be employed in asphalt mixtures as a partial replacement for both asphalt and aggregate [13]. The use of softer virgin asphalt improves aged asphalt's properties, making them similar to those of virgin asphalt [14]. To attain a target performance grade (PG), blending charts were utilized to calculate the optimum rejuvenator concentrations for the blends. Due to the performance-based characteristics of the mixes, it was shown that the mean value of concentrations could be more reliably predicted when used as a design parameter for recycling. The rejuvenator contains a low proportion of asphaltene and is a soft binder (less than 2% by weight) [15]. RAP is anticipated to crack more easily than virgin material since it is stiffer due to aging. The mechanical performance of the RAP binder at low temperatures improves when bio-based rejuvenators are added. Rejuvenators can recover toughness and mechanical properties at low temperatures. Chemical modifications at the functional group level did not cause mechanical improvements; hence, physio-chemical oxidation did not reverse despite the rejuvenators' existence [16]. The application of rejuvenators has improved the workability of RAP-containing asphalt mixes. This result is based on asphalt binder viscosity measurements. However, it has not been proven using recycled asphalt mixtures. The emulsified rejuvenator reduces the air void content and mixing torque of reclaimed mixes by 4% and 6%, respectively, compared to the ordinary rejuvenator [17]. The aging of asphalt binder can be classified into two stages: short-term and long-term aging. Short-term aging occurs during mixing, silo storage, transportation, and laying operations, where the asphalt is subjected to high temperatures. Long-term aging takes place over the service life of the pavement, where the asphalt undergoes a slow aging process [18]. Six rejuvenators were tested to lower the Superpave PG of the aged binder. At 25°C, all six rejuvenators lowered binder viscosity to levels similar to virgin binder. Waste oil was also investigated as a potential rejuvenator due to its significant impact on binder properties [19]. Applying heated RAP binder to WVO has the greatest impact on the softening point and penetration value. Temperature, waste oil volume, and RAP all have an effect on the performance properties [20].

Despite extensive studies on the influence of waste engine oil (WEO) and waste vegetable oil (WVO) on the low-temperature properties of asphalt binders, their potential as rejuvenating agents in asphalt paving mixes incorporating RAP has not been widely explored

[21-23]. Therefore, investigating the potential of WEO and WVO as recycling agents to reduce RAP stiffening is a significant research area that requires further exploration [24, 25].

After reviewing the existing literature, it becomes evident that the influence of the rejuvenator is examined in two distinct stages within the laboratory setting. Firstly, the rejuvenated binder undergoes a series of tests. Secondly, in rejuvenated mixture testing, the results from the binder experiments are utilized.

Previous studies have shown that rejuvenators significantly improve the performance of aged asphalt binders and mixes. In a study by Shen et al. [26], rutting resistance was investigated using a wheel tracking device, and fracture properties at low temperatures were examined by a thermal stress-restrained specimen test. At lower dosages of rejuvenators, organic products functioned similarly to petroleum-based products. The introduction of rejuvenating agents into the asphalt paving mixes resulted in a decrease in the critical temperature, thereby improving resistance to low-temperature cracking for all mixtures. It was proposed that optimizing the rejuvenator dose would enhance mix performance [18]. The incorporation of a blend of Evotherm and Sylvaroad has been effective in producing warm mix asphalt containing high proportions of RAP. The viscosity of the binder decreased upon adding both components, with greater reductions observed at higher dosages. This approach allows for an improved binder content while maintaining adequate fatigue resistance [27]. Using excessive waste oil reduces asphalt viscosity. The recommended waste oil dosage is 1 to 4%. During road testing, the compound rejuvenate was found to have a 45% RAP application [28]. The rejuvenator Paraffinic Oil exhibited the greatest efficiency in lowering the PG of the aged binder used in the RAP at both high and low temperatures. Furthermore, the rejuvenators led to enhance fatigue resistance without adversely impacting rutting performance [29]. Incorporating a rejuvenator at a 0.75% weight of bitumen concentration resulted in significant improvements in viscosity, fatigue resistance, and low-temperature properties of the tested asphalt [30]. The addition of Styrene Butadiene Rubber (SBR) latex to RAP mixes has been shown to enhance their viscoelastic properties, improve resistance to moisture damage, reduce rutting, and mitigate low-temperature cracking [31]. Results have shown that increasing CR and SBR improves strength, with an optimal mix containing 5% CR and 4% SBR [32]. The properties of the bituminous concrete mixture can be increased even more by including waste plastic [33]. The results show that using 4% shear thickening fluid (STF) by weight of the binder (an asphalt binder with an 80/100 penetration grade) improves high-temperature viscoelastic properties and bitumen performance grading [34]. The use of asphalt rejuvenators and polymer-modified asphalt binders was observed to enhance the performance of surface-layer mixtures that incorporated 50% RAP and 50% PMA [35]. Rejuvenators enhanced the cracking resistance of recycled mixtures. However, they decreased rutting resistance and moisture susceptibility in recycled materials [36]. Rejuvenators refer to a group of organic compounds employed to replenish the physical and chemical properties of aged asphalt binders, enabling them to reach a satisfactory state for reuse in pavement construction [37].

The addition of WEO and WVO to RAP has the potential to reduce stiffening and enhance the low-temperature properties of the resulting pavement. RAP serves as the primary material for constructing road surfaces, while waste oil refers to oil that has been removed from vehicles during oil changes. Mixtures of virgin materials and these waste types can yield comparable pavement performance. RAP can be utilized as a substitute for raw materials when repairing or replacing highways.

The main purpose was to evaluate the properties of laboratory-aged bitumen (Pen 60/70) modified by WEO and WVO additives. The rheological characteristics of virgin, aged, and rejuvenated binders will be evaluated using a dynamic shear rheometer (DSR). Fourier transform infrared (FTIR) spectroscopy will be employed to analyse the infrared absorption spectrum for a better understanding of the rejuvenation mechanisms on the aged asphalt binder. Moreover, laboratory experiments will be conducted to determine the permissible range of high and low service temperatures, or PG, of the asphalt binder.

2. Research methodology

The study conducted various experiments to determine the optimal percentage of rejuvenator necessary to make aged asphalt binder workable. Initially, virgin bitumen was obtained from NRL (National Refinery Limited) in Karachi, and comprehensive tests were carried out to assess its properties. The bitumen then underwent aging through the Rolling Thin Film Oven Test (RTFOT) and Pressure Aging Vessel (PAV) test. These tests were subsequently repeated on the aged bitumen, following the sequence performed on the virgin bitumen, to evaluate the properties post-aging.

To rejuvenate the aged bitumen, different proportions of WEO and WVO were introduced, and subsequent tests were conducted on the rejuvenated binder. The properties of this rejuvenated bitumen were compared with those of the original virgin bitumen. Additionally, Marshall mix design procedures were used to compare the properties of virgin bitumen with those of aged bitumen containing rejuvenators. This section outlines the research methodology for determining the optimal dosage of three rejuvenators to restore the PG of the blend of virgin and recycled binders. The study is divided into two parts, and a comprehensive laboratory testing plan with control binders is depicted in Fig. 1.

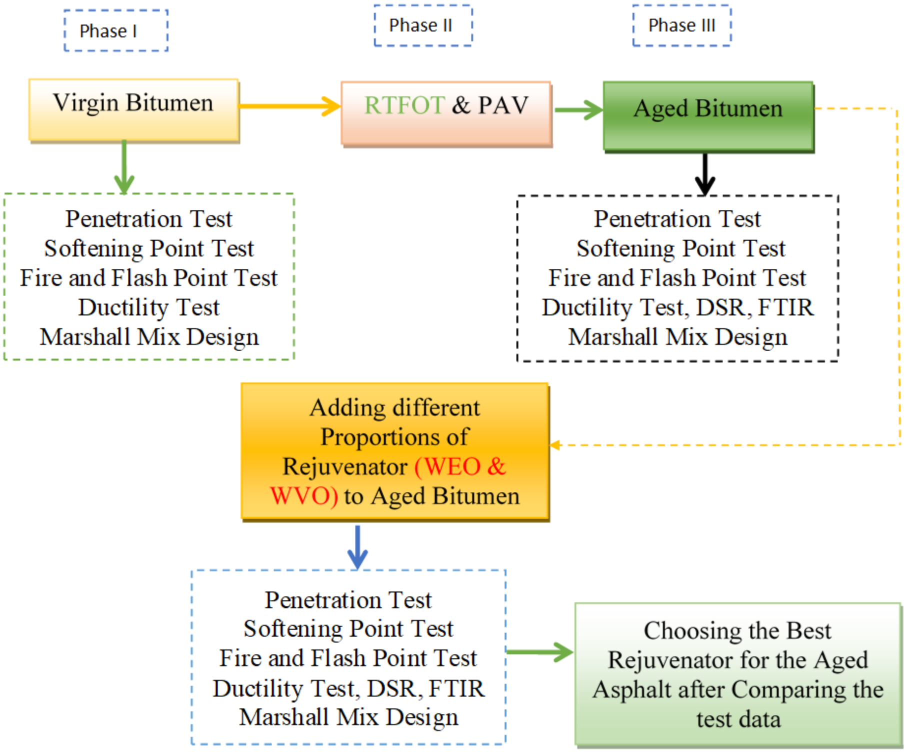

Fig. 1. Laboratory testing plan using control binders

The study requires various materials, such as asphalt binder and rejuvenators, as well as equipment like a rolling thin film oven, pressure aging vessel, DSR, and FTIR spectroscopy, to investigate theoretical aspects including the source and properties of the asphalt binder, the source of rejuvenators, bitumen aging, and rheological properties.

3. Materials

3.1. Aggregates

Figure 2 displays the selected aggregate grading, meeting the standard for Class B roads with a maximum size of 19.5 mm. The percentage of aggregates passing through each sieve is within the AASHTO T-27 limits, according to the sieve analysis [38]. The curve also illustrates the AASHTO lower and upper limits for each sieve size. The graph clearly shows that the actual passing for each sieve occurred between the AASHTO T-27 lower and upper limits. Figure 2 presents the plotted gradation with percentage passing versus sieve size.

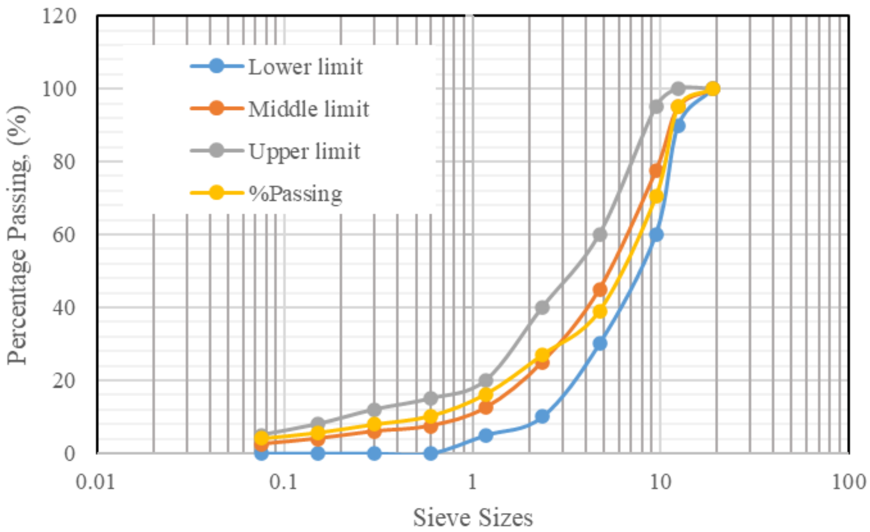

Fig. 2. NHA Class-B gradation

The coarse aggregate for the experiment was sourced from Margalla [39-42], utilizing aggregate sizes of 0-5 mm, 5-10 mm, and 10-20 mm. Aggregate provides most of the permanent deformation resistance (up to 95%), while the asphalt binder contributes only 5%. A compacted stone skeleton with coarse and fine aggregates is used to provide strength against repetitive load applications. The properties of asphalt concrete are influenced by the aggregate's gradation, surface roughness, and morphology. Rough and angular aggregates, as opposed to round and smooth-surfaced ones, tend to offer greater strength. According to the NHA (1998 requirements for dense-graded surfaces, the aggregate was graded using the NHA Class B classification. As per the Marshall mix design, the nominal maximum aggregate size for NHA Class B gradation was 19 mm.

Aggregate grading refers to the percentages of various-sized fractions that constitute the overall material after sieving. To determine the particle size distribution of the aggregate sample, it undergoes a series of sieving operations using sieves with specified aperture sizes.

The weight of each size fraction of the retained aggregate particles is then determined by weighing the material on the sieves. This process is referred to as "grading", or "identifying the material's particle size distribution". The appropriate sieve sizes for a particular substance are specified in the material's specification. Table 1 shows the upper, middle, lower limits, and percent passing of the aggregates.

Table 1. Upper, middle, lower limits, and percentage passing of the aggregates

Sieve No.	%Passing	Lower limit	Middle limit	Upper limit
19	100	100	100	100
12.5	95	90	95	100
9.5	70.4	60	77.5	95
4.75	39.1	30	45	60
2.36	27.1	10	25	40
1.18	16.3	5	12.5	20
0.6	10.3	0	7.5	15
0.3	8	0	6	12
0.15	5.7	0	4	8
0.075	4.1	0	2.5	5

The aggregate samples were dried for approximately 18 hours at 105°C to 110°C in the oven. To categorize the aggregate into different sieve sizes, it was dry sieved. To obtain the average stockpile gradation delivered from the field with the sample, sieve sizes matching the criteria for the "type" of the individual aggregate fractions in the correct proportions were selected. The trial-and-error method described below was used. After combining the trial percentages for each size, the mixture was sieved and compared to the average stockpile gradation. The process was repeated, adjusting the proportions of each size, until the desired gradation was achieved. Specimens were then made using the final percentages of each size as required later in the method.

3.2. Reclaimed Asphalt Pavement materials

This research aims to investigate the feasibility of utilizing RAP in the construction of new pavements. The aged asphalt binders used in the mixtures were produced in the laboratory through an accelerated aging process on a virgin straight-run asphalt binder with a penetration grade of 60-70, like those commonly used in Pakistan.

3.3. Virgin asphalt binders

The asphalt binder, with a penetration grade of 60/70, was obtained from Attock Refinery Limited (ARL), Rawalpindi. Its popularity and compatibility with cold to moderate temperatures made this penetration grade asphalt binder an appropriate choice for use in Pakistan. In this study, three distinct types of asphalt binders were employed for experimentation and analysis. The virgin asphalt binder used in the virgin mixes had a penetration grade of 60-70 (1/10 mm) as per the National Highway Authority (NHA) guidelines. The selection of Pen 60-70 VAB for the virgin mixes was based on its targeted grade, which was suitable for all the blends used in the mixtures.

3.4. Rejuvenators

Rejuvenators assist in restoring the physical and/or chemical characteristics of aged asphalt binders to acceptable levels by recovering the asphaltene-to-maltene ratio, as well as other properties that have been degraded due to oxidation. WEO and WVO are among the widely used rejuvenators. In this research, a soft binder with a low asphaltene content and a viscosity of 202 pascal-seconds at 60ºC was employed as a rejuvenator for aged binder blends. The rejuvenator percentages used in the study were 0%, 6.0%, 18%, and 24% by weight of the aged binders. These percentages were used to create blending charts, which helped determine the appropriate amounts of rejuvenator required for rejuvenating aged binders. The determined percentages were obtained at both high and low limits of 6% and 24%, respectively.

4. Test methods

According to AASHTO specifications, three properties of binding materials should be considered for road construction and engineering design purposes: binder purity, consistency, and safety [43]. The fluidity of bitumen is influenced by temperature, making it essential to establish a standard temperature for comparing the diverse consistencies of asphalt as a binding component. The rejuvenated binders were prepared by mixing preheated aged bitumen with various compound rejuvenators of different viscosity grades and dosages at 150 °C under a mixing speed of 1500 rpm for 30 minutes. To determine the consistency of bitumen in its role as a binding agent, a penetration test is conducted. Additionally, the properties of the asphalt binder were evaluated using various laboratory tests, including penetration, softening point, ductility, flash point, and fire point tests.

4.1. Experimental progress

In this study, a range of conventional and specific tests were conducted to assess the rejuvenation properties of aged asphalt. The experimental setup included both conventional tests, such as penetration, softening point, flash and fire point, and ductility, as well as performance tests.

Various performance tests were conducted, including the RTFOT, PAV, DSR, and FTIR. These tests were performed to determine the proportion of rejuvenator required to make the aged asphalt binder usable again.

The study involved several tests to assess the properties of virgin bitumen. This virgin bitumen underwent two aging tests, RTFOT and PAV. Subsequently, the experiments were repeated using virgin bitumen to evaluate the properties of the aged bitumen. The effectiveness of the rejuvenators in improving the properties of aged bitumen was evaluated by comparing it with virgin bitumen, after incorporating different concentrations of WEO and WVO. The properties of both virgin and rejuvenated bitumen were also evaluated using the Marshall Mix design.

4.2. Rolling Thin Film Oven Test and Pressure Aging Vessel Test

The initial step in obtaining the aged binder involved conducting two essential tests at the University of Engineering and Technology in Lahore.

4.2.1. Rolling Thin Film Oven Test

The Rolling Thin Film Oven Test (RTFOT) simulates the short-term aging of asphalt binders that occurs during the hot-mixing process. This test measures the percentage loss of volatiles during the heating of asphalt binders under controlled conditions. A 35-gram sample of melted bitumen is poured into a cylindrical glass bottle, which is then placed in a rotating oven carriage. To age the bitumen, it is heated to 163ºC for 85 minutes. The test is crucial for determining pavement fatigue and rutting resistance at high temperatures.

4.2.2. Pressure Aging Vessel Test

The Pressure Aging Vessel (PAV) uses hot compressed air to simulate the long-term oxidative ageing of asphalt binders. It provides an early result that may be acquired after the bitumen has been in use for 10 to 15 years. The RTFOT samples of aged bitumen were placed in stainless steel PAV pans. The RTFOT sample was heated to 100ºC for 20 hours under 2.1 MPa pressure. The test was conducted in compliance with AASHTO R 28 standards [44].

4.2.3. Laboratory aging of asphalt binder

To assess the laboratory ageing of asphalt binder, the Superpave technique was employed, which utilises two machines, namely the RTFOT and the PAV. The RTFOT, specified in AASHTO T 240, simulates short-term ageing of the asphalt binder, akin to the ageing that occurs during the mixing and transportation phase to the construction site. In this method, 35 grams of asphalt binder are spun in glass bottles in a 163°C oven for 75 minutes. On the other hand, to simulate long-term ageing, the PAV, outlined in AASHTO R 28 [44], is utilised. The RTFOT aged binder is placed in shallow steel pans, each containing 50 grams of binder, and then sealed inside the PAV, where temperature and pressure are maintained at 100°C and 2.10 MPa (305 psi), respectively. Following 20 hours of heating and compression under high pressure in an oxygen-rich environment, the resulting binder exhibits a simulated field age of 5 to 10 years.

4.3. Mixing of aged binder and rejuvenators

The WEO and WVO were chosen as rejuvenators for the study. The rejuvenators were sourced from Hayatabad, Peshawar, an industrial state.

To ensure uniform dispersion of the rejuvenators in the bitumen, both the rejuvenated and aged bitumen samples were heated in an oven at 145°C for 30 minutes and mixed for 5 minutes using an electric stirrer operating at 500 rpm. Rejuvenators were used in proportions of 6, 18, and 24%.

4.4. Dynamic Shear Rheometer

The Dynamic Shear Rheometer (DSR) is a tool that measures the viscoelastic properties of an unaged bitumen sample, specifically the phase angle and G* complex shear modulus. The DSR test involves placing a bitumen sample between two rotating plates, one oscillating while the other remains stationary and subjecting it to shear action at a frequency of 10 Rad/sec or 1.59 Hz. The ASTM D 7175 standard was used to determine the rheological properties of the asphalt binder through DSR analysis [45].

Addressed by:

$$G^*/\sin\delta \text{ on unaged binder} > 1.00 \text{ kPa} \quad (1)$$

$G^*/\sin\delta$ on RTFOT aged binder ≥2.20 kPa (2)

$G^* x \sin\delta$ on PAV aged binder ≤5000 kPa (3)

4.5. Fourier Transform Infrared spectroscopy

The primary method for infrared spectroscopy is Fourier Transform Infrared (FTIR). Infrared spectroscopy involves passing infrared radiation through a material. During this process, some of the infrared rays pass through the sample, while others are absorbed by it (transmitted). The resulting spectrum creates a molecular fingerprint, indicating how the sample's molecules absorb and transmit light. This technique can distinguish between familiar and unknown materials, assess the quality or consistency of a sample, and determine the quantity of each component in a mixture. Potassium bromide (K-Br) is a powder that can be stabilized by applying 1000 pounds per square inch of pressure. To prepare the sample, the K-Br pellet is placed in a holder and coated with hot bitumen using a brush. Initially, the K-Br pellets are inserted into the machine without the bitumen to create a K-Br spectrum. Subsequently, a bitumen pellet is inserted into the machine, which operates within a wavelength range of 0 to 600 nm. The bitumen spectra are then drawn and subtracted from the K-Br spectrum.

4.6. Marshall Mix design method

Determining the optimum bitumen content is a crucial step in mix design. The performance of the mix is evaluated through the Marshall Mix design approach, which utilises the Marshall Stability and Flow Test. The maximum load that the test specimen can withstand during the stability phase of the test, at a loading rate of 50.8 mm/minute, is recorded to assess the mix's performance. The specimen is loaded until it cracks, and the load required to cause cracking determines stability. During loading, a dial gauge attached to the specimen measures plastic flow (deformation). Throughout the Marshall Stability and Flow Test, the maximum load and flow value are simultaneously recorded in increments of 0.25 mm (0.01 inch). The Marshall Method is a widely adopted approach for designing Hot Mix Asphalt (HMA) that is both cost-effective and capable of meeting specified gradation requirements. The Marshall Mix design process consists of six stages, namely: (1) Aggregate selection, (2) Asphalt binder selection, (3) Sample preparation, (4) Compaction test, (5) Stability determination using the Marshall Stability and Flow Test, and (6) Selection of optimum asphalt binder content.

4.6.1. Compaction with Marshall hammer

In a virgin mix design, an optimal asphalt content (OAC) of 4.5% by weight dry aggregate was achieved using HMA with virgin asphalt binder Pen 60–70. The OAC for each sample size was determined using the Marshall Test under compaction conditions of 75 blows. This value was applied to all asphalt mixture samples with the same types and gradations of aggregate. A total of three samples of recycled mixture and one sample of virgin mixture were prepared. The samples were then heated to the required temperature for compaction before being compacted with a Marshall hammer, which presses down on the sample with a tamper foot. Some hammers are electrically powered, while others are operated by hand.

The compactor used in this study features a cylindrical sample with a diameter of 102 mm (4 inches) and a height of 64 mm (2.5 inches), with provisions for adjustments in sample height. The tamper foot is flat and circular, with a diameter of 98.4 mm (3.875 inches) and an area of 76 cm² (11.8 in²). Compaction is achieved through a 457.2 mm (18 inches) free fall drop of a hammer assembly with a 4536 g (10 lb.) sliding weight. The compaction process involves a specified number of blows (35, 50, or 75) on each side, depending on expected traffic loading. The tamper foot strikes the sample on the top, covering the entire top area, and after a designated number of blows, the sample is inverted for repetition. This method, conforming to AASHTO T 245 standards, assesses the resistance to plastic flow of bituminous mixtures using the Marshall Apparatus. The compaction temperature, set between 135-165°C, varies according to factors such as asphalt binder type, mix design, and desired pavement characteristics.

5. Results and discussion

The study involves various asphalt samples, each with a distinct composition. These include Virgin Bitumen (VB), Aged Bitumen (AB), and combinations of Aged Bitumen with different percentages of Waste Engine Oil (06EO, 18EO, 24EO) or Vegetable Oil (06VO, 18VO, 24VO). These compositions represent different blends aimed at assessing the impact of these oils on the properties of aged bitumen. The percentages indicate the proportion of waste engine oil or vegetable oil mixed with the aged bitumen in each sample. The type/composition of asphalt samples is shown in Table 2.

Table 2. Type or composition of asphalt samples

S.No.	Type or composition	Sample
01	Virgin bitumen	VB
02	Aged bitumen	AB
03	06% waste engine oil + aged bitumen	06EO
04	18% waste engine oil + aged bitumen	18EO
05	24% waste engine oil + aged bitumen	24EO
06	06% vegetable oil + aged bitumen	06VO
07	18% vegetable oil + aged bitumen	18VO
08	24% vegetable oil + aged bitumen	24VO

5.1. Penetration test

Figure 3 provides a side-by-side comparison of the penetration indices. The penetration tests indicated that both WEO and WVO influenced the stiffness of bitumen. According to the graph, each 1% increase in oil corresponds to nearly a 10 mm increase in the penetration value. It was observed that the binder containing 24% WEO exhibited the maximum penetration result, in proportion to the substantial amount of WVO.

A higher penetration number signifies a softer consistency, whilst a lower number indicates the opposite. The effect of modified binders, such as waste oils, on the ageing process can be assessed through the penetration ageing ratio. This ratio compares the properties of aged bitumen with those of unaged bitumen, thereby providing a measure of the variations in bitumen properties occurring during the ageing process [46].

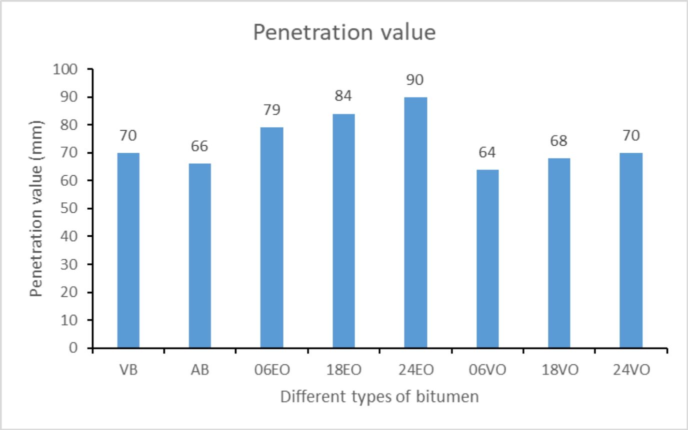

Fig. 3. Penetration test results for varying bitumen contents

5.2. Softening point test

The softening point of a substance, or the temperature at which it softens to a specific degree, determines its thermal sensitivity. This test ascertains whether the material will flow at higher temperatures. The temperature at which the two rings allow each 9.5mm-diameter, 3.5g bitumen-coated ball to fall 25mm is referred to as the softening point [47].

The results of the softening point test are illustrated in Fig. 4. The modified binders displayed lower softening points compared to their virgin counterparts, signifying the effectiveness of the modification process. The softening point of the virgin binder was determined to be 51 °C. Owing to the inclusion of WEO and WVO, the binder's softening point decreased, rendering it softer than the virgin binder.

Bitumen with a higher softening point is generally preferred for asphalt mixtures, as it is stiffer and more resistant to deformation and fatigue cracking under high-temperature conditions. Figure 4 demonstrates the impact of blending WVO and WEO on the softening point of bitumen samples. The softening point of the modified bitumen was observed to diminish with an increase in the concentration of WEO and WVO. The WVO-modified bitumen took longer to solidify compared to the WEO-modified bitumen.

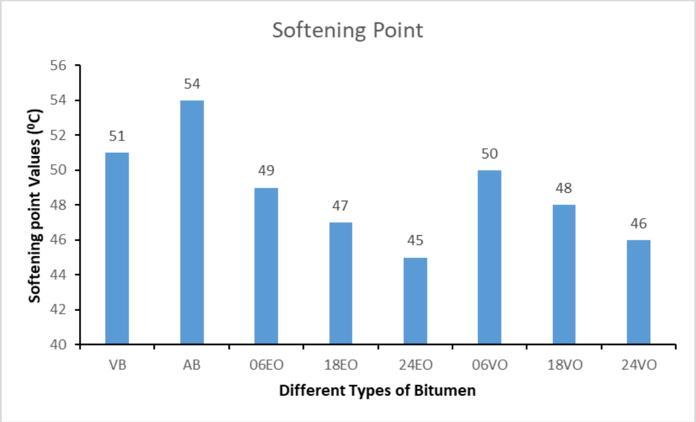

Fig. 4. Softening point test results for varying bitumen contents

The measurement of the Penetration Index (PI) provides quantitative information about the thermal susceptibility of asphalt binders, as indicated in the literature [48]. PI values typically range from -3 to +7, reflecting the varying thermal susceptibilities of the binders. Equations (4) and (5) were employed to calculate the PI and thermal susceptibility (A) values, adhering to the methodology outlined in previous studies [49]. Figure 5 presents the Penetration Index results of asphalt binder samples. Analysis of these results reveals that an increase in the content of waste-oil modified binder leads to a reduction in PI values. For instance, a 24% concentration of waste-oil modified binder in virgin binder corresponds to the lowest observed PI value. The diminishing PI values and the rising trend of susceptibility (A) values suggest that the addition of waste-oil modified binder enhances the thermal susceptibility of the treated binders. Nonetheless, the PI values remain within the permissible range defined by prior research [50], which stipulates -2 to +2 as the acceptable limit for paving binders. Consequently, waste-oil modified binders are deemed suitable for paving applications, aligning with earlier expectations. However, a primary limitation in predicting thermal susceptibility from PI estimations is that the calculation of A values involves only two temperatures (specifically, 20°C and 25°C for softening point and penetration, respectively). This limitation underscores the need for a comprehensive study of the rheology of both virgin and modified binders across a broad range of temperatures and frequency levels. Dynamic Shear Rheometry (DSR) is identified as the most appropriate method for such testing. Figure 5 illustrates the correlation between Penetration Index and Thermal Susceptibility in the tested asphalt binders.

$$PI=\frac{20\text{-}500A}{1+50A} \tag{4}$$

$$A=\frac{\log(800)-\log(\text{Pen at T})}{SP-25} \tag{5}$$

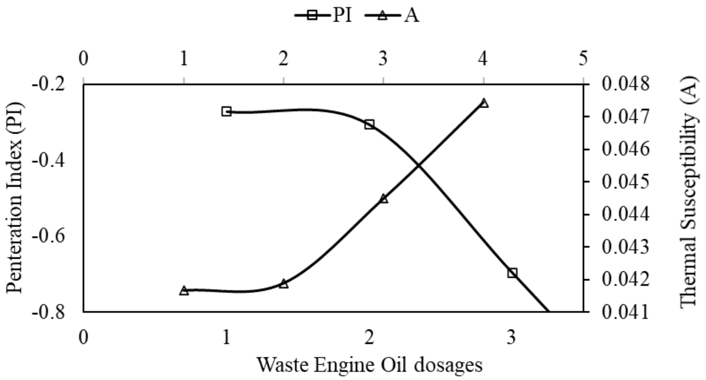

Fig. 5. Correlation between Penetration Index and Thermal Susceptibility of tested asphalt binder

5.3. Flash and fire point

Figure 6 illustrates the impact of WVO and WEO on the flash point of both aged and virgin asphalt binders. The results demonstrate that incorporating WVO and WEO into the asphalt binders enhances their flash point. Notably, WVO was found to have a more significant effect in reducing the flash point than WEO. Despite this reduction, all measured values remained above the minimum acceptable threshold, suggesting that the rejuvenated binders retain their safety for use in construction.

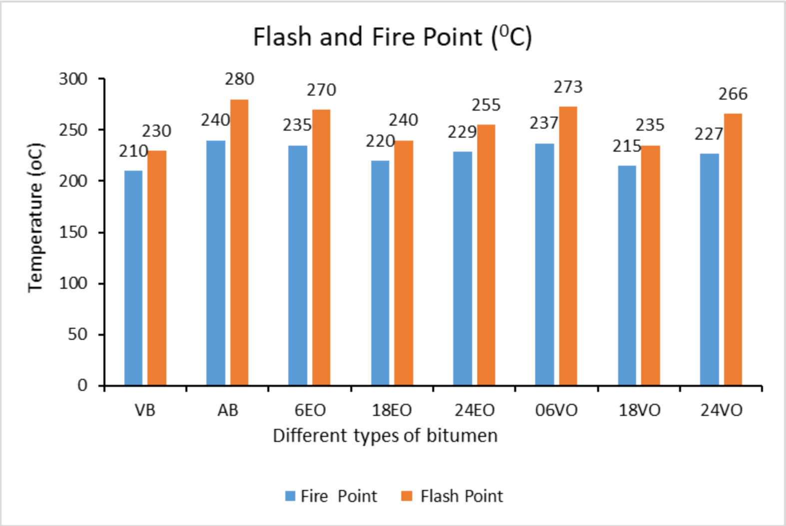

Fig. 6. Flash and fire point test results for varying bitumen contents

5.4. Ductility test

The ductility of bitumen, indicative of its stickiness and cohesiveness, is illustrated in Fig. 7, which demonstrates the effect of waste oil addition on the ductility of asphalt binders. An increase in the amount of waste oil in the mixture correlates with a decrease in ductility, which is associated with the softness of the bitumen. Compared to the unmodified bitumen, with a ductility value of 40 cm, all binders modified with WVO and WEO exhibited ductility values of less than 100 cm. Additionally, an increase in the oil content within the samples resulted in higher ductility values for the modified bitumen. The study indicated that bitumen modified with WEO possessed better ductility compared to that modified with WVO, suggesting that WEO might enhance the resistance of aged bitumen to cracking and deterioration after use.

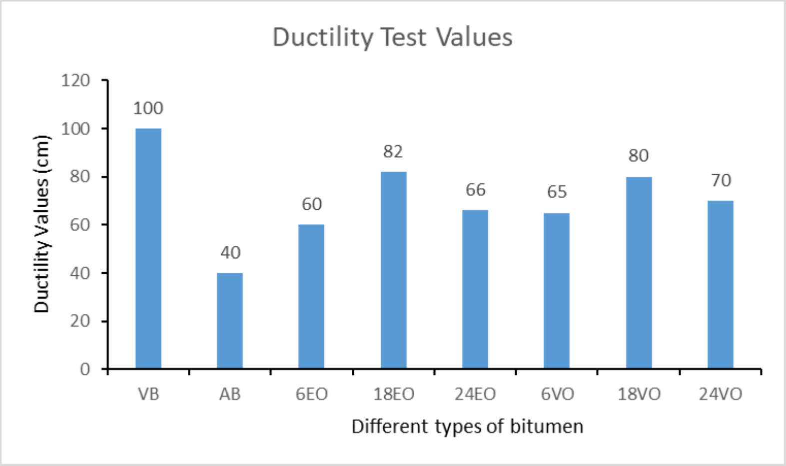

Fig. 7. Ductility test results for varying bitumen contents

5.5. Dynamic shear rheometer

Phase angle and complex modulus are essential parameters influencing the rheological characteristics of asphalt binders. The phase angle represents the ratio of stress to strain, whereas G* signifies the binder's stiffness at different temperatures. Phase angle values range from 0 to 90 degrees: materials demonstrating viscosity manifest a 0-degree angle, and those exhibiting elasticity have a 90-degree angle [51]. Figure 9 shows that the virgin binder possesses the highest G* value in comparison to the modified binders. Results from the complex modulus and phase angle tests are depicted in Fig. 8.

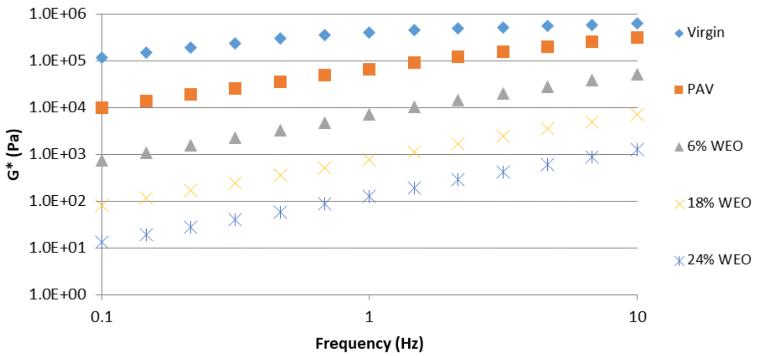

Fig. 8. Results of complex modulus and frequency (Hz) of WEO

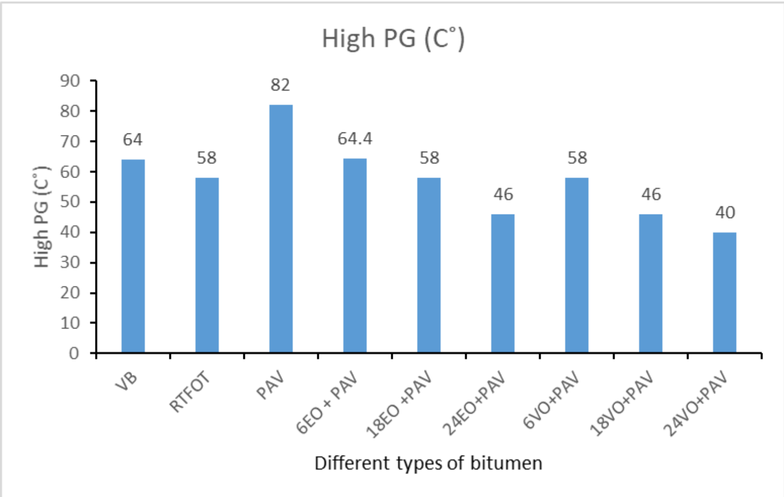

Fig. 9. Results of high PG(°C) of the binder state

5.6. Fourier Transform Infrared results

The structural indices, specifically S=O and C=O, are significantly higher in aged bitumen, as depicted in Fig. 10. To restore its properties to those of virgin bitumen and decrease the structural index percentage, we treated it with the appropriate proportion of rejuvenators.

The FTIR analysis was conducted in transmission mode to measure the transmittance of wavelengths not absorbed by the sample, allowing them to pass through. The main goal of this study was to assess the functional properties of both aged and rejuvenated asphalt binders, as well as to explore the physical and chemical interactions between the waste oils (WVO and WEO) and the recovered aged asphalt. The intensities of the sulfoxide and carbonyl peak areas serve as indicators for monitoring the aging and rejuvenation of asphalt. The presence of asphaltenes reinforces the strength of the S=O and C=O bands [52, 53].

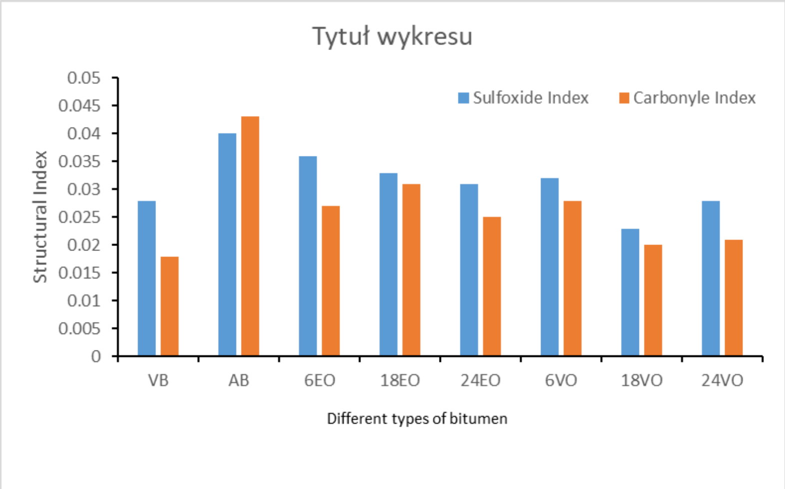

Fig. 10. Percentages of structural indices for varying bitumen contents

Figure 11 presents the infrared spectroscopic analysis curves for both aged and WVO/WEO-rejuvenated binders, spanning wavenumbers from 0 to 650 cm^{-1}. The curves indicate a decrease in the intensity of the (S=O and C=O) peak areas upon adding WVO and WEO, in comparison to aged asphalt. This suggests that waste oils can mitigate the aging process and rejuvenate aged asphalt by reducing asphaltene concentration. Notably, there was no new peak height observed with the addition of the waste oils. Physical testing of both aged and rejuvenated binders produced distinct results, implying that the interaction of WVO and WEO with aged asphalt is more physical than chemical.

The structural index is a technique developed to quantify the types and strengths of bonding in base and oil-modified bitumen samples. To assess the effect of varying oil doses on the absorption band intensities of each sample, the band maxima values were analysed. A single absorbance value from a specific group was examined to establish the relative baseline or tangential approach for a particular band. Employing the maximum band method aids in reducing errors that might occur from integration methods at the tangential ends [54].

5.6.1. Fourier Transform Infrared spectrum

Scanning Electron Microscopy analysis showed successful dispersion of WEO and WVO in proportions of 6%, 18%, and 24%. Additionally, FTIR was employed to investigate the chemical changes in the oil-modified bitumen and macromolecules [55, 56]. The resulting FTIR spectrum was used to identify the various functional groups in the bitumen mixture, including single, double, and triple bonds between carbon and other groups introduced during the mixing process [57]. Data on wavenumber and infrared absorbance peaks from the graphs were used to identify the functional groups in the bitumen sample through FTIR analysis [56]. Figure 11 displays the FTIR spectra of five samples, including base bitumen, aged bitumen, and three oil-modified bitumen samples with 6%, 18%, and 24% waste oil by weight, showing wave numbers on the x-axis and absorbance on the y-axis. The spectrum reveals peaks corresponding to various functional groups such as hydrocarbons, aromatic hydrocarbons, aromatics, carbonyls, and saturated hydrocarbons. The addition of waste oil led to an increase in the intensity of these functional groups, which were absent in the base bitumen. It was expected that the intensities of these functional groups would rise with increasing waste oil doses, without surpassing the intensity of the waste oil itself, indicating effective incorporation and dispersion within the base bitumen [58]. The infrared (IR) spectra show that none of the modified samples (ranging from 6% to 24%) exhibited new peaks or any disappearance of existing peaks, closely resembling the base bitumen peak. However, the intensities in the waste oil-modified bitumen samples were higher than in the base bitumen, suggesting successful functionalization of the bitumen molecules [57]. These findings are consistent with previous research in the field [58, 59].

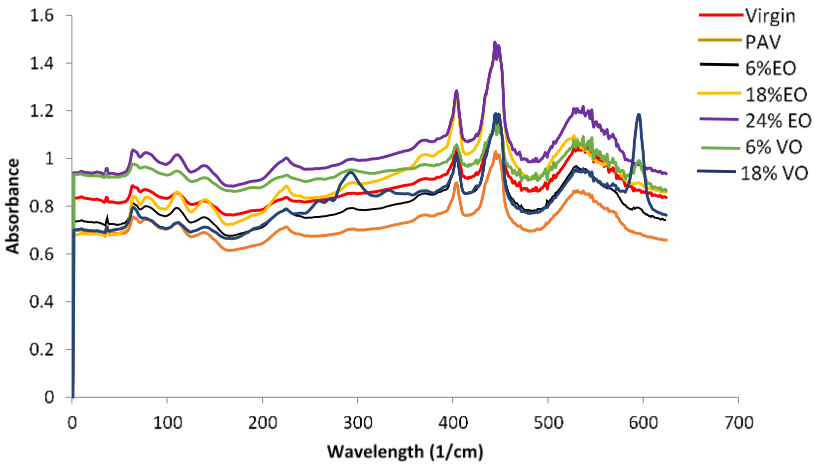

Fig. 11. FTIR spectral analysis results of virgin binder, aged asphalt, and rejuvenated asphalt

5.7. Marshall Stability test

The Marshall Stability test was conducted to assess the stability of asphalt mixes, both with and without waste oil modifications. Results indicated that samples lacking waste oil additions displayed the highest Marshall Stability values, especially after immersion in a water bath for 30 minutes. Samples containing 18% WEO and 18% WVO showed marginally lower Marshall Stability values in comparison to the unmodified samples. However, the Marshall Stability results were not significantly altered by varying waste oil concentrations.

When benchmarked against a control mix (comprising both virgin and 0% waste oil aged asphalt), mixes containing 18% waste oil exhibited enhanced Marshall stability. Nevertheless, in this study, the control mixture achieved the highest Marshall stability, surpassing the modified asphalt mixes. This suggests that a higher concentration of waste oil in the asphalt concrete mixture may have contributed to decreased stability. Nine Marshall specimens were prepared for this test, encompassing three different types: virgin, aged, and optimal oil percentage, all with a bitumen content of 4.5%.

5.7.1. Marshall stability

Figure 12 presents the results of stability tests, indicating that the optimal bitumen content was established at 4.5%. Mixes incorporating waste engine oil (WEO) and waste vegetable oil (WVO) exhibited improved stability compared to the control sample. At a bitumen level of 4.5%, the stability values for mixes with WEO and WVO were 6.24 kN and 7.43 kN, respectively, surpassing the AASHTO requirement of 3.5 kN. These results imply that the modified asphalt mixtures, particularly those with 18% WEO and WVO, demonstrate enhanced stability in comparison to the control samples, which include both virgin and aged asphalt with 0% waste oil.

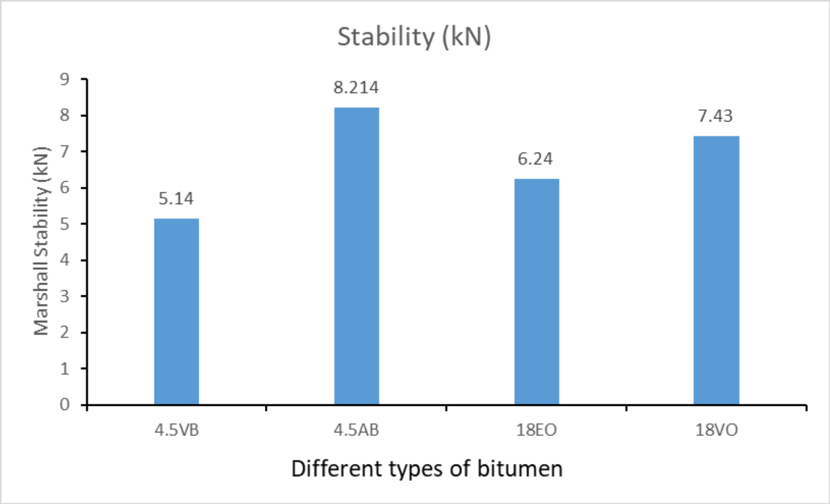

Fig. 12. Stability test results for asphalt mixtures

5.7.2. Marshall flow

Figure 13 displays the flow values of both modified and control asphalt samples. The results indicate that as the amount of waste oil increases, there is an improvement in flow compared to the aged binder, but a decrease when compared to the virgin binder. With an optimum binder content of 4.5%, the flow value of the mix containing WEO was 3.2 mm, while the flow value of the mix with 0% waste oil was 4.1 mm. This modest decrease in flow value may be attributed to the quantity of waste oil used in the aged mixture. Nonetheless, both mixtures met the AASHTO road specifications. It is important to note, however, that the flow value does not precisely represent the resistance of the asphalt mixes to permanent deformation. The incorporation of waste oil into the mixture might have contributed to this reduction in flow value.

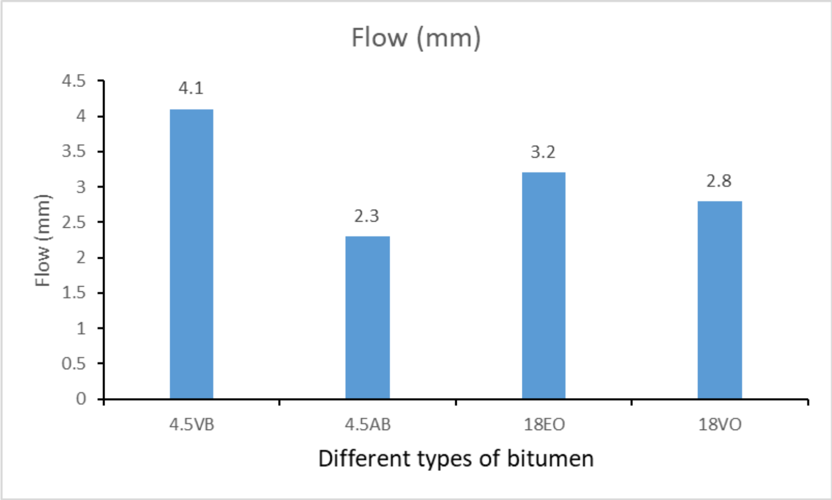

Fig. 13. Flow test results for asphalt mixtures

6. Conclusions

Long-term aging leads to a reduction in the softening point and ductility of virgin binder while increasing penetration. However, the addition of rejuvenators to aged asphalt results in a lower softening point, along with increased penetration and ductility, indicating a significant enhancement in the performance of the aged asphalt. The following conclusions can be drawn from the experimental findings and analysis of different aged asphalts:

- Experimental results suggest that incorporating WEO leads to a notable decrease of up to 24% in the complex modulus of the virgin binder.
- FTIR analysis reveals significant chemical changes in the aged binder due to the addition of rejuvenators. Specifically, there is a decrease in the structural index, reflecting modifications in the carbonyl and sulfoxide functional groups associated with asphaltenes. Additionally, new peaks in the FTIR spectrum indicate the effectiveness of the rejuvenators in altering the chemical composition of the aged binder.
- The introduction of cooking oil and engine oil into an aged binder significantly reduces the complex shear modulus and PG of the binder. This reduction in stiffness suggests a degradation of the aged binder, thereby enhancing its suitability for reclamation and reuse.
- Adding rejuvenating agents to aged RAP mixtures at high temperatures can potentially restore the properties of the aged asphalt and improve the performance of the RAP mixtures.
- An optimal rejuvenating agent dosage content of 18% has been determined. This dosage balances rejuvenation benefits with potential drawbacks, leading to enhanced asphalt mixture properties, including stiffness and Marshall Stability.

Given the limitations of this case study, it is recommended to adopt a cost-effective and environmentally friendly approach by applying these findings on rejuvenators to enhance the performance-based properties of asphalt binder. This involves using rejuvenating agents to improve the stiffness and durability of aged asphalt and RAP mixtures, thus enhancing their overall performance. Such a cost-effective approach reduces the need for expensive asphalt replacement and minimizes environmental impact through asphalt reuse and waste reduction. Further research should focus on optimizing the use of rejuvenating agents in asphalt mixtures for maximum effectiveness and cost savings, as well as analysing the short and long-term aging resistance of binders rejuvenated by WEO and WVO.

Declaration of competing interest

The authors declare no conflict of interest.

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