



Dynamic Rheological Behavior of Green Alternative Binder for Asphalt 60/70 at Different Temperatures

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Abstract

Using bio-binders is an alternate strategy for reducing the need for and reliance on bitumen/crude petroleum binders. In this paper, bio-binders were used as a bitumen extender to acquire a green, eco-friendly alternate binder (GAB). To achieve this objective, an extensive investigation of the flow properties (dynamic and rheological) for the prepared green alternative binders (GAB) which was obtained by mixing a conventional 60/70 bitumen with different Green binder additives (GBA) - that was formed from waste cooking oil and waste polymers under different conditions- this mixing have been done as 1:1 volume ratio. For this reason, a number of tests were carried out, including standard physical attributes, Dynamic viscosity at different (time, temperature and shear rate) and Dynamic shear rate (DSR) test. The results obtained from this research, show that the prepared GAB samples enhanced the rheological and physical characteristics of asphalt samples and the sample with formula (L+ GBA₃) could be considered as the most promising mixture can be used for paving, as it has the highest rutting factor value with the same physical and rheological properties for the original asphalt sample.

Key Words: Rheology, Dynamic Rheology, Green Alternative Binder, Asphalt, waste oil, waste polymers, Pavement.

1. Introduction

The engineering field of transportation and highways encompasses the design, planning, building, operating, and maintaining of all types of roads, tunnels, and bridges to guarantee the security and comfort of people and products being transported [i, ii]. It is well known that bitumen, which is frequently utilized in pavement engineering, is mostly derived from petroleum. Nevertheless, bitumen use is not sustainable due to petroleum's non-renewable nature. Therefore, developing a workable alternative to traditional petroleum bitumen is essential to ensuring the pavement building industry's sustained growth [iii, iv]. Researchers have suggested bio-bitumen—binding polymers made from renewable biomass resources—as a sustainable alternative to conventional petroleum-based bitumen [v].

"Bio-binders" are the increasingly diverse variety of bio-oils that have been developed recently to supplement or replace certain petroleum-based asphalts [vi]. It would be very advantageous to use bio-oil, which is made from leftover cooking oil, as paving material in order to reduce environmental impact and preserve natural resources. The fact that bio-oil produces far fewer nitrogen oxide (NOx) emissions and a negligible sulfur oxide (SOx) emission when heated is another benefit it has over petroleum asphalt in terms of the environment [vii].

Crude oil is one of the main sources of bituminous binders used in pavement materials. Other materials also contain fossil fuels. In order to identify pavement distresses and subsequently forecast and assess pavement performance, it is imperative that one looks into the rheological characteristics of a pavement binder. To reduce the

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demand for bituminous binders made from fossil fuel, bio-binders were used in three different ways: a) direct alternative binder (100% replacement), b) a bitumen extender (25% to 75% bitumen replacement), and c) bitumen modifier (<10% bitumen replacement) [viii]. The best method to assess bitumen behavior is to look into its rheological characteristics using dynamic shear rheometer tests, which can be used to find complex shear modulus (G^*), phase angle (π), and rutting factor ($G^*/\sin \delta$), as bitumen performance is correlated with loading time and temperature [ix].

Experimental;

2.1. Materials used: -

Asphalt; asphalt 60/70 (L) with the specification shown in Table (1) were used. **Waste polymer;** they are industrial wastes, which are; waste polyethylene (WPE), waste polypropylene (WPP), and waste Poly (styrene-butadiene-styrene) (WSBS) Collected from some industries' waste in 10th Ramadan city, Egypt.

Waste oil: Also known as waste cooking oil (WCO), this oil is usually produced after frying and cooking and is a good regenerator because it mimics the lighter oil component of asphalt. WCO is safe to apply since its flash point is often greater than the temperatures needed to manufacture and compact asphalt mixtures.

2.2. Preparation Methods;

(GBA) Green Bio-Additive preparation.

The Parr-model 7575 batch autoclave reactor, depicted in figure (1), was utilized. The reactor is located in a furnace that has a control system that adjusts the stirrer's speed and temperature remotely. While manual controls are used to regulate the pressure and gas flows.

Reactant feeds are put into the reactor in accordance with the ratio specified in table (2). Once the reactor has been shut down, purge with N₂ to prevent pressure leaks. After being twice purged with the operational hydrogen gas to prevent air or oxygen pollution, the device is fixed in the initial hydrogen atmosphere of the experimental setup. The reactor then experienced about 30 minutes of 80 bar pressure. Once waste oil and feed stock (waste polymers) have been mixed at room temperature, the heating process has begun, and the reactor pressure is subsequently changed to the operational pressure. The stirrer is only activated when the temperature reaches 150 °C to avoid damage.

Table (1): Physical and chemical characteristics of original asphalt 60/70 (L)

Physical Properties (ASTM D-946)	Asphalt 60/70 (L)
Ability to dissolve in organic solvent, %	99.0
Flash point, °F (Cleveland open cup)	450
Kinematic viscosity at 135 °C, C.st.	375
Absolute viscosity at 60 °C, poise	2560
Softening Point, Ring and Ball, °C	47
Infiltration at 77 °F (25 °C) 100 g, 5s, 0.1 mm	65
Durability at 77 °F (25 °C), 5cm/min, cm	+150
Chemical Components, Wt %	
Oil	25.5
Wax Content of oil portion	4.4
Asphaltene	20.8
Resin	53.6

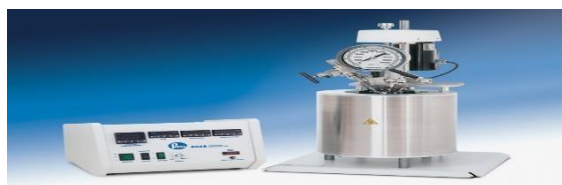


Fig (1): The device applied to the GBA fabrication

Table (2): mixing ratio between waste polymers and waste oil to GBA formation.

	Waste Polymer %	Waste Oil %
GBA 1	WPE	
	a) 80	20
	b) 70	30
	c) 60	40
GBA 2	WPP	
	a) 80	20
	b) 70	30
	c) 60	40
GBA3	WSBS	
	a) 80	20
	b) 70	30
	c) 60	40

Preparation of the Asphalt Modified with Waste Polymers; herein the two types of asphalt A and B was modified with waste polymers (WPE, WPP, and WSBR) in the ratio 3, 6, 9, 12 and 15 % from the asphalt weight at 163 °C, and stirring for 2 h at 1000 rpm^[xi].

Preparation of the (GAB) from asphalt and GBA; A mixture of bitumen binder with a ratio ranging from 50 to 90% and GBA₁₋₃ with a ratio ranging from 50 to 10% was generated in order to prepare the green alternative binders (GAB). As control binders, asphalt types A and B were used. GAB₁ (Asphalt A/B 90: GBA₁₋₃ 10), GAB₂ (Asphalt A/B 80: GBA₁₋₃ 20), GAB₃ (Asphalt A/B 70: GBA₁₋₃ 30), GAB₄ (Asphalt A/B 60: GBA₁₋₃ 40), and GAB₅ (Asphalt A/B 50: GBA₁₋₃ 50) are some of the combinations that were created and examined in this study. Both the green alternative binder and the control asphalt sample were preheated and mixed at a high shear mixer speed of 5000 rpm for 20 minutes at 120 °C^[xii].

2.3. Rheological Characterization of GAB and Polymer Modified Asphalt (PMA)

2.3.1. The dynamic viscosity (η) at different temperature Vs time (min),

The dynamic viscosity (η) of the GAB and blank samples (L) was measured at high and low shear rates over a range of temperatures and times, from 30 to 60°C, for 1 to 15 minutes. The measurements were performed using a stress-controlled rheometer from Austria (Modulated Compacted Rheometer (MCR 52), Anton Paar Instruments). Dynamic oscillatory and steady-shear testing on the steel cone plate CP 50-2, a conventional steel parallel plate shape with a 500 mm gap separation, was used to generate the samples. A universal temperature control system was used to adjust the temperature between 100 °C and 140 °C, with a shear rate of 0.3 cm⁻¹, or 20 rpm, specifically for asphalt samples. 0.7958 Pa was the initial control value for the shear stress. 79.58 Pa was the ultimate control value for the shear stress. Version 5.8.2 of the Data Manager Software was used to assess the rheological data [xii]. The *Brookfield DV-III Ultra Programmable Rheometer* measures two fluid properties at specific shear rates: viscosity and shear stress.

2.3.2. The dynamic viscosity (η), mPa.s. Vs shear rate at different temperature,

Dynamic viscosity (η), for the (L) and GAB at different temperature from 30°C to 60°C carried out against shear rate ranging from 2.34 to 1198.08.

2.3.3. Flow curve measurement

This rotating viscometer/rheometer has preset settings that can be used to create a flow curve. Step-

by-step increases in the shear rate are made, with each measurement point's duration being specified. Other environmental factors, such as the temperature, are constant. The flow curve that results displays the shear stress in relation to the shear rate. Shear rate vs shear stress for (L) and GAB at various temperatures between 30°C and 60°C. The dynamic viscosity η is defined by Newton's Law as the shear stress divided by the shear rate [xiii]. A specific shear stress or shear rate is applied to the sample when measuring viscosity on a rotating viscometer, accordingly.

$$\eta = \tau' / \dot{\gamma} \quad (1) \text{ Newton's viscosity law}$$

Where; η ... dynamic viscosity, τ ... shear stress, $\dot{\gamma}$... shear rate

If an ISO 3219 standard measuring geometry is used for the measurement, the physical attributes of torque and speed can be translated into the rheological values of shear rate and shear stress [xiv]. Using a conversion factor (which varies depending on the measurement geometry employed), the speed of the viscometer is translated into shear rate, and the torque is likewise converted into shear stress.

2.3.4. DSR Test.

The DSR test was designed to identify complex shear modulus G^* and rutting resistance ($G^*/\sin \delta$) across a broad temperature range, thereby characterizing the visco-elastic properties of the manufactured GAB and the polymer-modified asphalt. The Anton Paar RheoCompassTM, V1.20.496-Release rheometer model was used in this investigation to perform the DSR test. A sinusoidal shear stress was applied to the binder specimen in order to calculate the phase angle (δ) and complex shear modulus G^* . A maximum shear strain level of 12% was regulated. Additionally, the impact of waste polymers and GBA on G^* and δ was examined. We next computed the rutting resistance ($G^*/\sin \delta$) at 1.59 Hz to assess the high temperature performance. 58, 64, 70, and 76 °C were chosen as the testing temperatures since this study intends to examine the high temperature performance [xv].

3. Results and Discussion

3.1. Time (min) and its effect on the dynamic viscosity (η), mPa.s. for asphalt (L) and polymer-modified asphalt (PMAs) at high and low shear rates.

Figures (1-4) display the dynamic viscosities for PMA at various temperatures. The findings indicate that the PMA's dynamic viscosity increases by maintaining the same temperature for longer periods of time, the dynamic

viscosities were decreases the dynamic viscosity of (L+ 9% WPP) increased with temperature; for example, at 30 °C, it was 933.8 m.Pa.s.; at 40, 50, and 60 °C, it was 764.2, 401.4, and 302.7 m.Pa.s., respectively.

The PMA was found to have the highest dynamic viscosity in the orders L+9%WPP > L+9%WSBS> L+9%WPE> L. This suggests that the changed sample is more resilient to temperature changes due to the addition of asphalt and waste polymer additives. Because the PMA's resistance to temperature change increases with increasing additive concentration, the dynamic viscosity decreased at a lower rate when using a high share rate and increased when applying a low share rate.

3.2. Time (min) effects on the dynamic viscosity (η), mPa.s., for asphalt (L), PMA, and GAB at high and low shear rates.

The dynamic viscosities for L and GAB at different temperatures are shown in Figures (5-8). These graphs show that the dynamic viscosities of L dropped with temperature, whereas the dynamic viscosities of GAB rose with time at the same temperature. For instance, at 30 °C, the dynamic viscosity of (L+ GBA3) was 832.9; at 40, 50, and 60 °C, it was 691.8, 289, and 239 m.Pa, respectively.

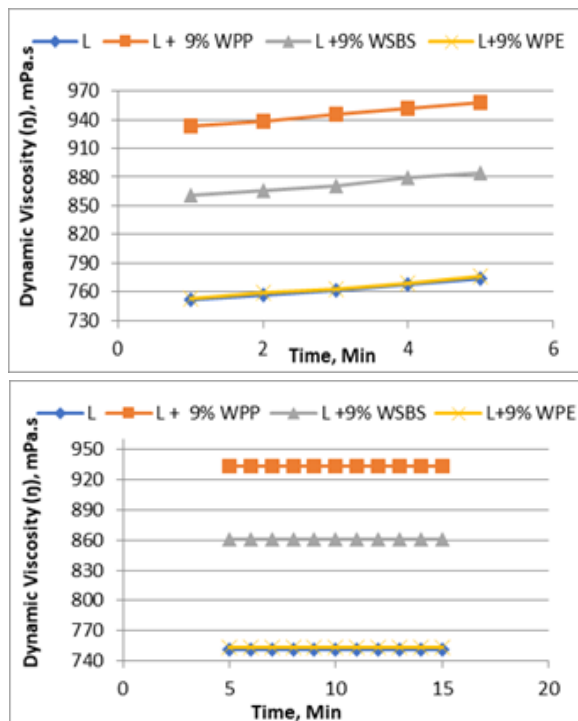


Fig 1: The relationship of (η) to time on asphalt (L) and PMAs at high and low shear rates, and 30 °C

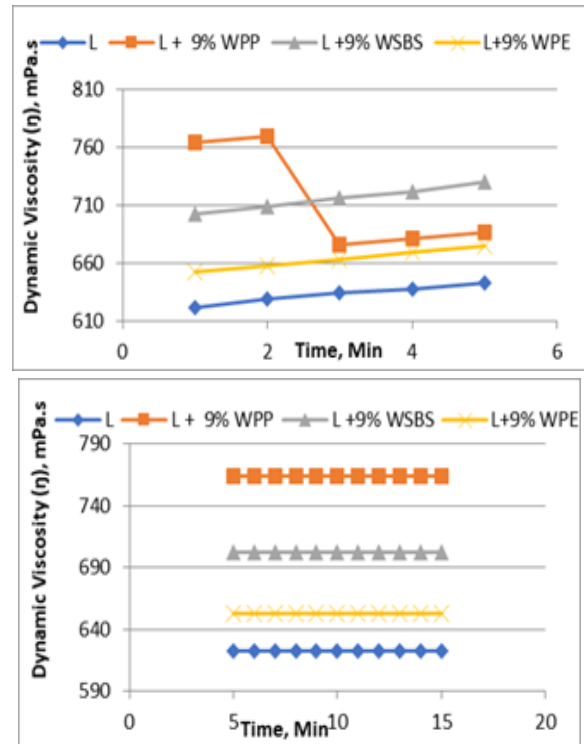


Fig 2: The relationship of (η) to time on asphalt (L) and PMAs at high and low shear rates, and 40 °C

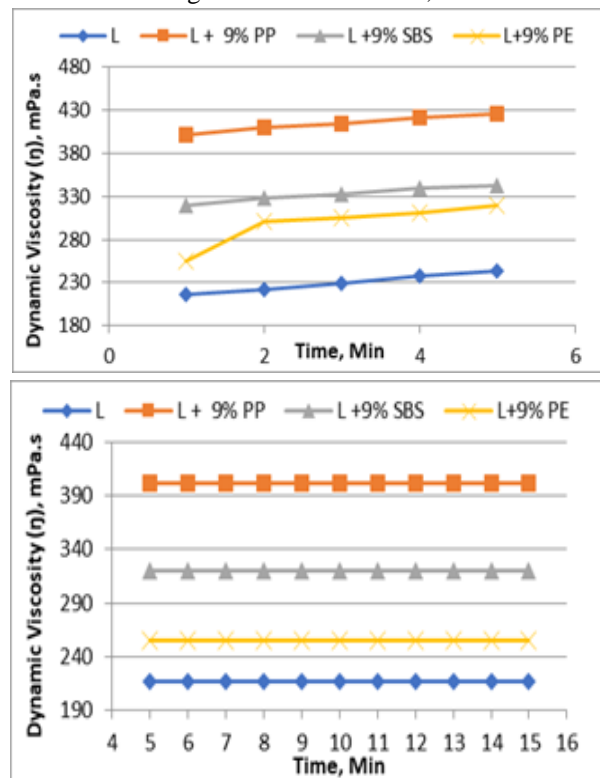


Fig 3: The relationship of (η) to time on asphalt (L) and PMAs at high and low shear rates, and 50 °C

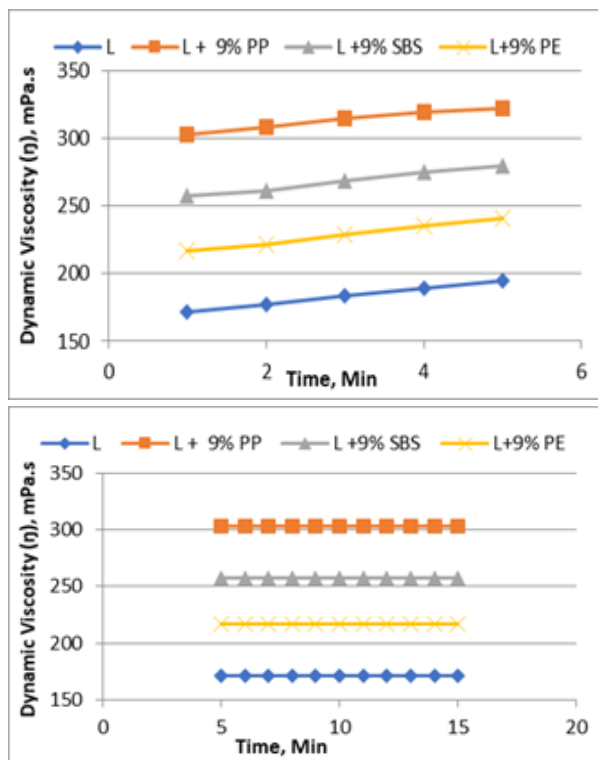


Fig 4: The relationship of (η) to time on asphalt (L) and PMA at high and low shear rates, and 60 °C.

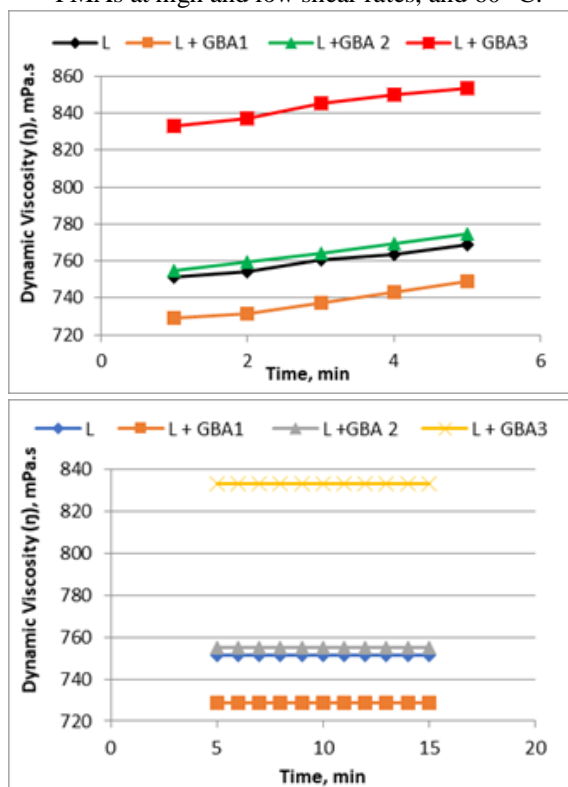


Fig 5: The relationship of (η) to time on asphalt (L), PMA, and GAB [50% asphalt + 50% GBA] at high and low shear rates, and 30 °C.

The results show that the PMA has the maximum dynamic viscosity in the orders $L + GBA_3 > L + GBA_2 > L > L + GBA_1$, indicating that the asphalt and waste polymer additives in the modified asphalt cause it to become harder and less sensitive to temperature variations. By applying a low share rate, the dynamic viscosity decreased at a larger rate than when using a high share rate. This suggests that as the additive content rises, so does the PMA's resilience to temperature change. Compared to the original binder, the alternative binder—which is made up of 50% asphalt and 50% GBA—is less sensitive to temperature fluctuations.

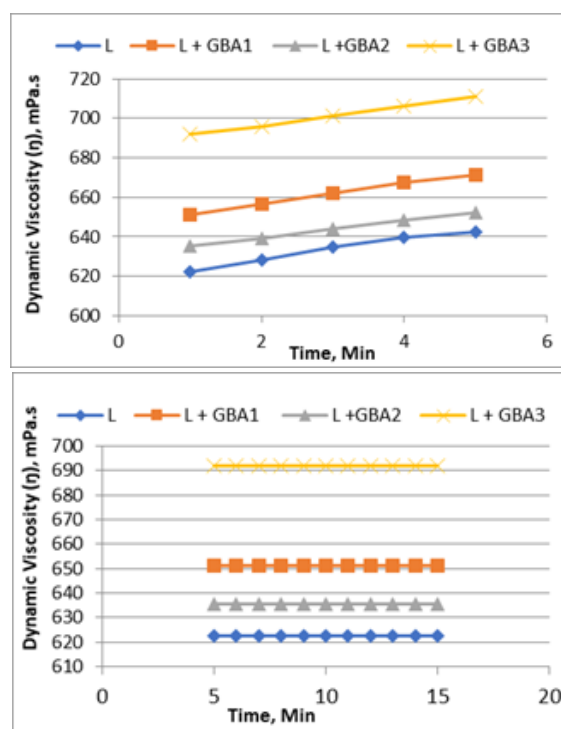
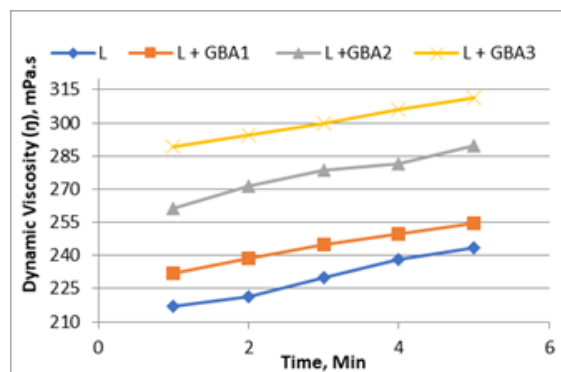


Fig 6: The relationship of (η) to time on asphalt (L), PMA, and GAB [50% asphalt + 50% GBA] at high and low shear rates, and 40 °C.



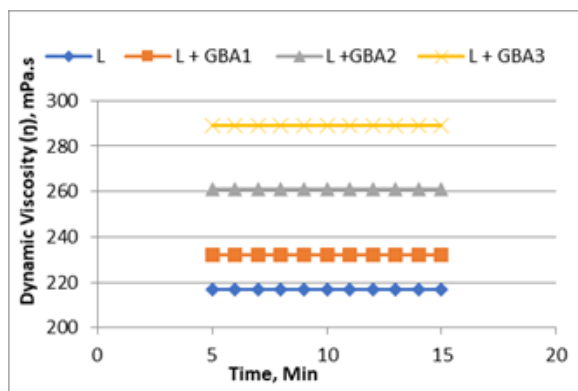


Fig 7: The relationship of (η) to time on asphalt (L), PMA, and GAB [50% asphalt + 50% GBA] at high and low shear rates, and 50 °C.

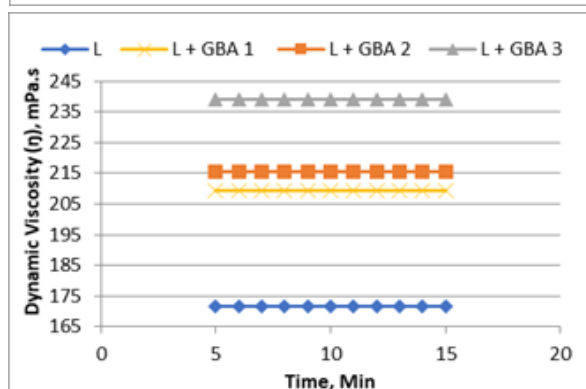
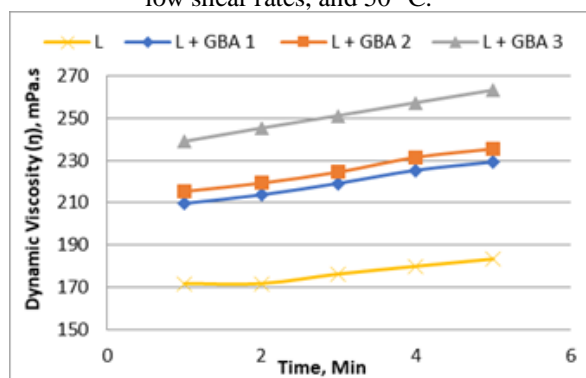


Fig 8: The relationship of (η) to time on asphalt (L), PMA, and GAB [50% asphalt + 50% GBA] at high and low shear rates, and 60 °C.

3.2. The dynamic viscosity (η) of the prepared GAB, mPa.s. Vs shear rate at different temperature.

The dynamic viscosity of PMA at different temperatures and shear rates is shown in **Figures 9–13**. These graphs demonstrate that PMA's dynamic viscosity reduces as temperature rises at the same shear rate. This phenomenon is caused by an increase in the loss of volatile asphalt components

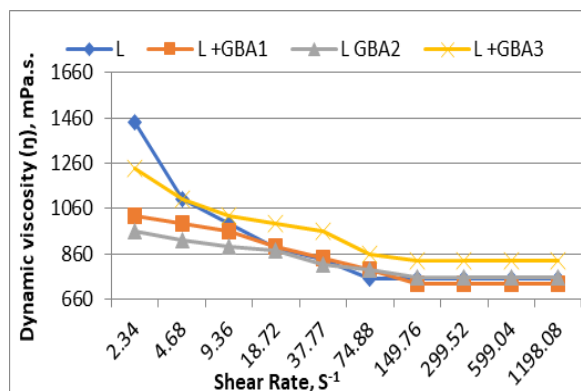


Fig 10: The relationship of (η) with shear rates to (L) and GAB (50% asphalt and 50% GBA) at 30 °C.

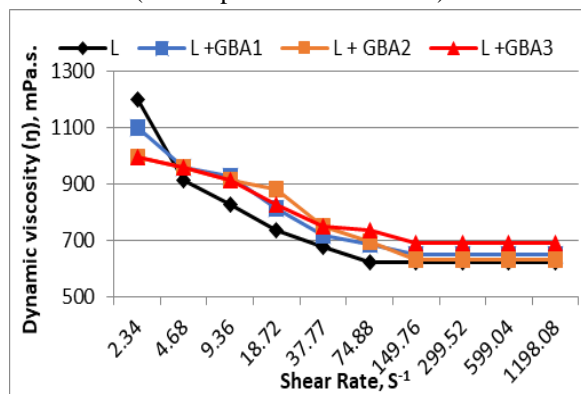


Fig 11: The relationship of (η) with shear rates to (L) and GAB (50% asphalt and 50% GBA) at 40 °C.

At the same temperature, the dynamic viscosity decreases with increasing shearing rate. At 2.34 S⁻¹, it falls with increasing temperature and orders L > L+GBA₁ > L+GBA₃ > L+GBA₂. For instance, the dynamic viscosity of (L+GBA₃) at 30 °C dropped from 1237.1 to 832.9 mPa, at 2.34 to 1198.08 S⁻¹ correspondingly.

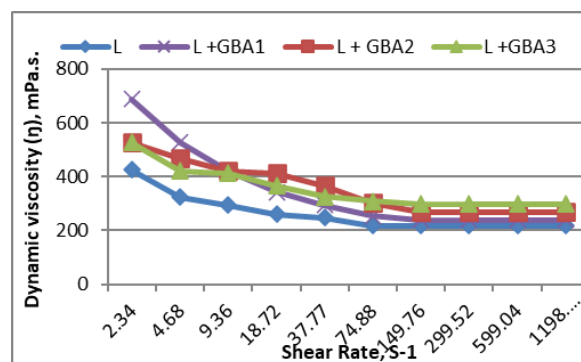


Fig 12: The relationship of (η) with shear rates to (L) and GAB (50% asphalt and 50% GBA) at 50 °C.

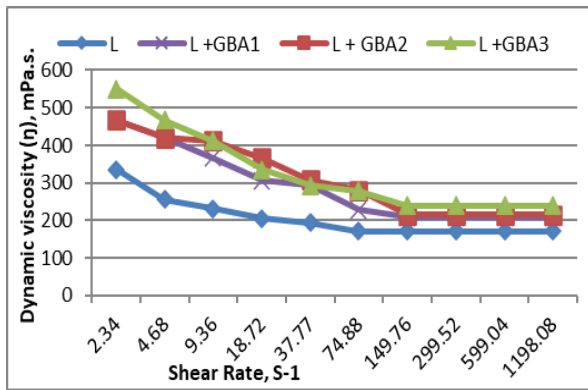


Fig 13: The relationship of (η) with shear rates to (L) and GAB (50% asphalt and 50% GBA) at 60 °C.

3.3. The dynamic viscosity (η) of the PMAs, mPa.s. Vs shear rate at different temperature.

The dynamic viscosity of PMA to varying temperatures and shear rates is depicted in **Figures (14–17)**, studies demonstrate that the increased loss of asphalt's volatile components is what's causing this behavior. At the same shear rate, PMA's dynamic viscosity falls as temperature rises.

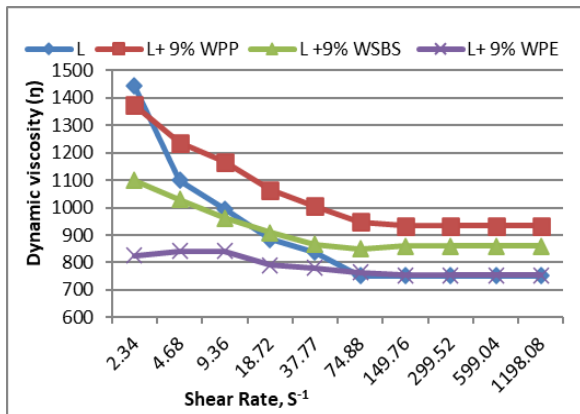


Fig 14: The relationship of (η) with shear rates to (L) and PMAs at 30 °C.

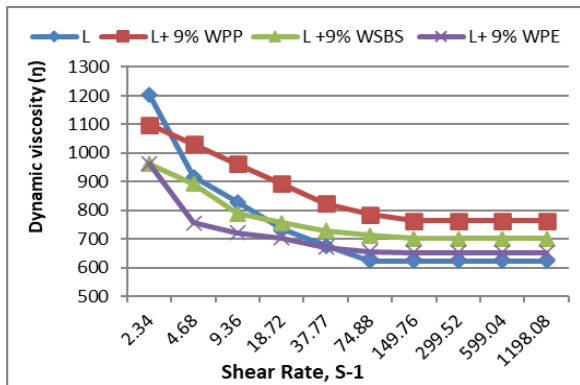


Fig 15: The relationship of (η) with shear rates to (L) and PMAs at 40 °C.

The dynamic viscosity decreases at 2.34 S-1 as the temperature rises and does so in the following order: $L > L + 9\% \text{ WPP} > L + 9\% \text{ WSBS} > L + 9\% \text{ WPE}$, but at the same temperature, a rise in the share rate causes the dynamic viscosity to drop. For instance, the dynamic viscosity of (L+ 9% WPP) at 30 °C dropped from 1347 to 933.8 m.Pa at 2.34 to 1198.08 S⁻¹, respectively.

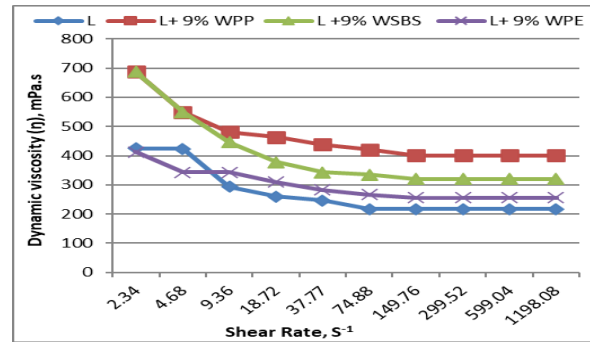


Fig 16: The relationship of (η) with shear rates to (L) and PMAs at 50 °C.

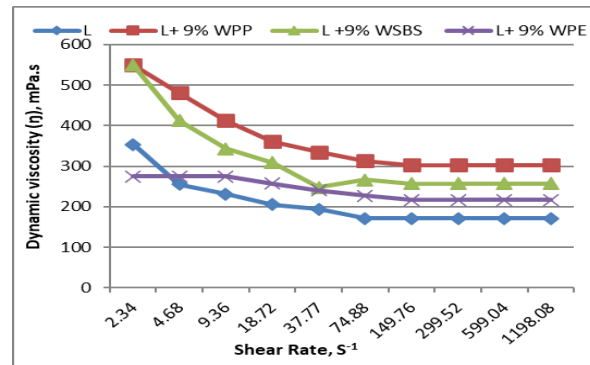


Fig 14: The relationship of (η) with shear rates to (L) and PMAs at 60 °C.

3.2. Flow curve measurement

3.2.1 Shear rate against shear stress Pa at varying temperatures for GAB [50% asphalt + 50% GBA] and asphalt (L).

An evaluation of the shear stress at different shear rates was conducted in order to describe the flow characteristics of the substitute binder that was developed. **Figures (18–21)** show this relationship for the prepared GAB at various temperatures. It has been noted that as the shear rate of the prepared GAB increases, the shear stress increases as well; the maximum shear stress was measured for (L+ GBA3). Shear rate against shear stress Pa at varying temperatures for GAB [50% asphalt + 50% GBA] and asphalt (L). When the shear rate rises, the prepared GAB's shear stress rises in the order: $L + GBA3 > L + GBA2 > L + GBA1 > L$. Shear stress decreases at varying temperatures while maintaining a constant share rate. The physical behavior of GAB (50% asphalt + 50% GBA) is comparable to that of original asphalt.

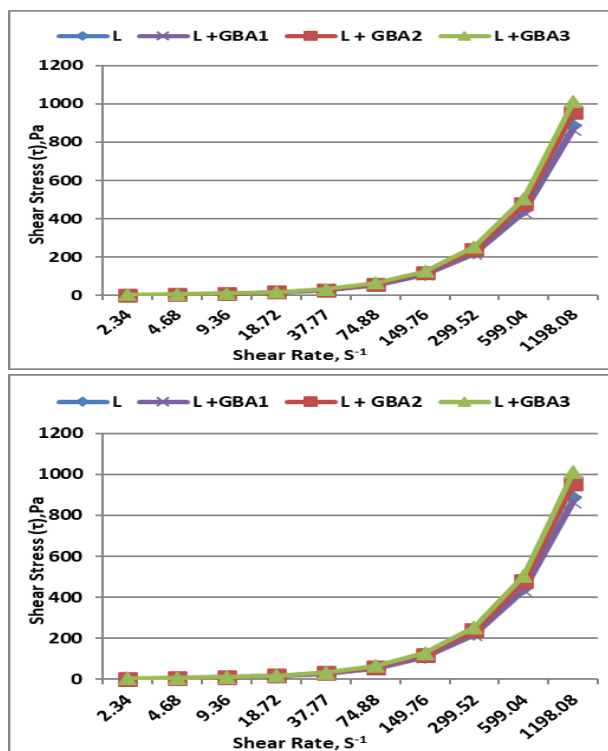


Fig 19: The relationship between Shear Rate and Shear Stress for (L) and GAB [50% asphalt + 50% GBA] at 40 °C.

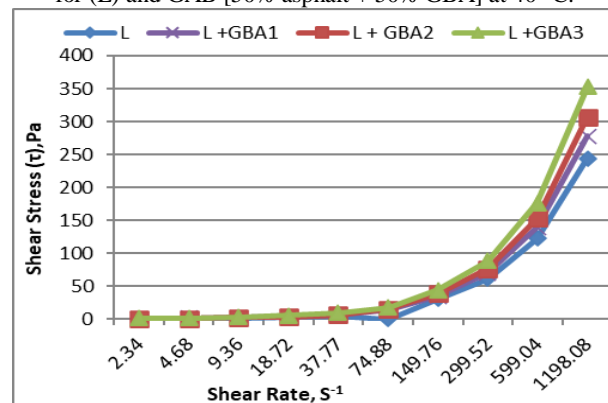


Fig 20: The relationship between Shear Rate and Shear Stress for (L) and GAB [50% asphalt + 50% GBA] at 50 °C.

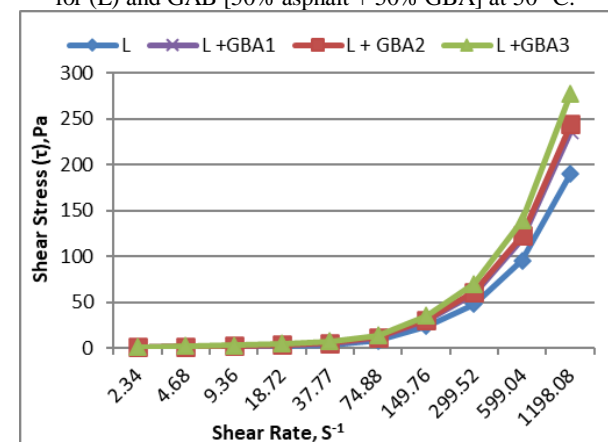


Fig 21: The relationship between Shear Rate and Shear Stress for (L) and GAB [50% asphalt + 50% GBA] at 50 °C.

3.3.1. Shear rate for (L) and PMAs at various temperatures against shear tension Pa

The created alternative binder's flow characteristics were described by evaluating the shear stress at various shear rates. **Figures (22-25)** show the relationship between shear stress and shear rate for the prepared GAB at various temperatures.

The produced GAB exhibits an increase in shear stress as the shear rate increases, with the maximum shear stress being recorded for (L+ 9% WPP). With an increase in shear rate, the prepared GAB's shear stress rises in the following order: L+9% WSPP > A+WPE > L+9% WSBS > L. Shear stress decreases at varying temperatures while maintaining a constant share rate. The physical behavior of GAB (50% asphalt + 50% GBA) is comparable to that of original asphalt.

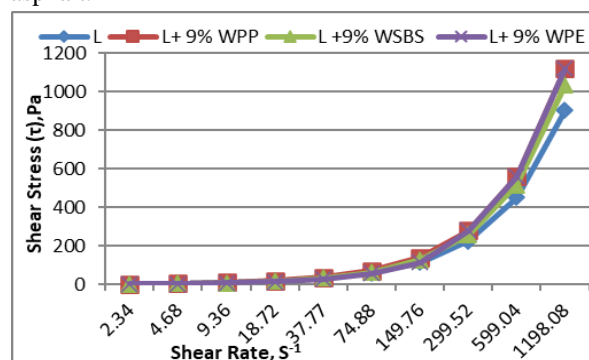


Fig 22: The relationship between Shear Rate and Shear Stress for (L) and PMAs at 30 °C.

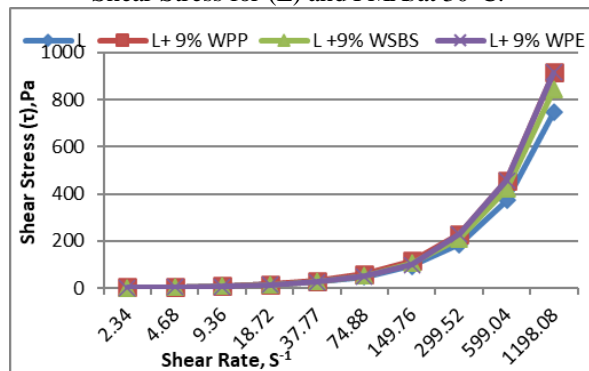


Fig 23: The relationship between Shear Rate and Shear Stress for (L) and PMAs at 40 °C.

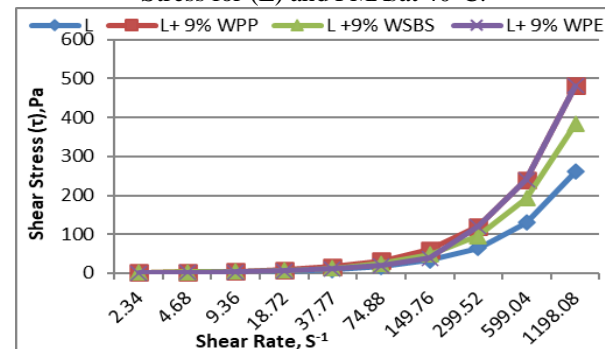


Fig 24: The relationship between Shear Rate and Shear Stress for (L) and PMAs at 50 °C.

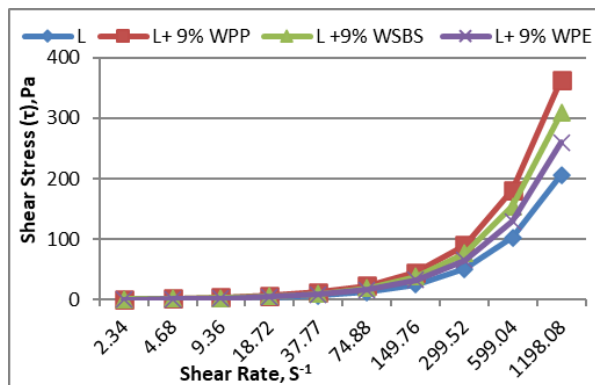


Fig 25: The relationship between Shear Rate and Shear Stress for (L) and PMAs at 60°C.

3.2. Thermal stability of the Prepared GAB.

Figure (26) shows the weight loss percentages along with curing time for virgin asphalt binder and the prepared GAB samples.

As this figure illustrates, all three of the bio binders had lower mass losses than the control at both temperatures, which may have been caused by the bio-oil's decreased volatile fractions following the high-temperature cooking process. The breakdown of the bio-oil molecules and volatilization of the maltenes were the main causes of the mass loss [xvi]. Based on the results, it is clear that (L+GBA2) has almost the same thermal stability as the virgin asphalt sample.

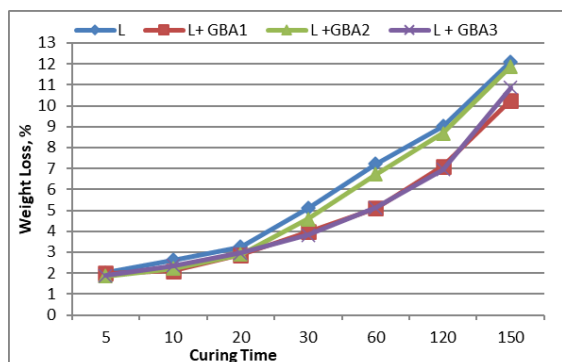


Fig26: Thermal stability for GAB [50% asphalt+ 50% GBA] cured at 250°C.

3.4. Rheological performance

DSR test for the PMA_s and the prepared green alternative binder samples using waste cooking oil and waste polymers at different test temperatures was carried out. The test findings are displayed in the subsections that follow.

3.4.1. Complex Shear Modulus (G^*)

The values of the complex shear modulus (G^*) of virgin asphalt, PMA samples and the prepared GAB samples at different temperatures are shown in figures (27-28).

These Figures show that the PMA samples and the produced GBA have complex modulus results reported. Because bitumen mixture is temperature-sensitive, the dynamic modulus of PMA samples and the generated GAB samples mixture reduces as temperature rises. Higher temperatures cause the physical properties of asphalt to soften, resulting in a drop in stiffness and strength and a fall in dynamic modulus. The modified bitumen has a much higher dynamic modulus than the unmodified [17] asphalt in both hot and low temperature environments. It has been demonstrated that GBA₃ has the largest complex shear modulus when compared to base bitumen and increases in the order of GBA₃ > GBA₂ > L > GBA₁ because of the polymer's increased adhesiveness when combined with bitumen, increasing the bitumen's hardness and viscosity. As a result, the complex modulus rises.

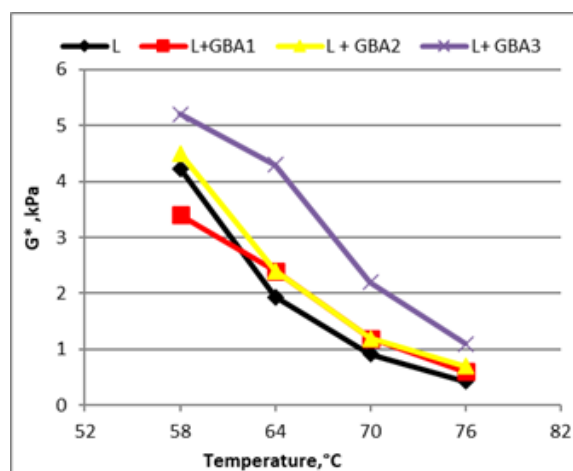


Fig27: Complex shear modulus values (G^*) of both original AC (L) and GAB [50% asphalt + 50% GBA] at different temperatures.

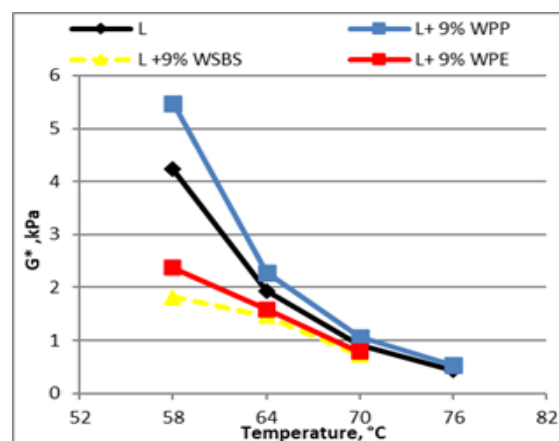


Fig 28: Complex shear modulus (G^*) values of both original AC and PMA using different contents of waste polymers at different temperatures.

3.2.1. Rutting factor

The rheological performance of polymer-modified asphalt, virgin asphalt, and produced GAB binders are displayed in Figures (29–30). The values of $G^*/\sin(\delta)$ were obtained at 52, 58, 64, 70, and 76°C. It was noted that throughout the whole temperature range (between 52 to 76 °C), $G^*/\sin(\delta)$ decreased by increasing temperature. Additionally, it was discovered that the kind of prepared GAB had an impact on the rutting factor; L+GBA₃ had the highest rutting factor, indicating that it would be the most promising prepared GAB for paving.

4. Conclusion

This study examined the rheological and dynamic characteristics of the green alternative binder (GAB) for asphalt 60/70 at various temperatures. The aforementioned analysis allow for the drawing of the following conclusions:

- Standard laboratory tests were used to analyze the physical and rheological properties, and the results were evaluated and compared with the control sample. The modified bitumen shows essentially identical physical qualities as bitumen 60/70, and the results are promising. Furthermore, the rheological characteristics remain within an acceptable range.
- The highest rutting factor was recorded for L+GBA₃ which means it would be the most promising prepared GAB that can be used for paving.
- (L+GBA₂) has nearly the same thermal stability as the virgin asphalt sample.

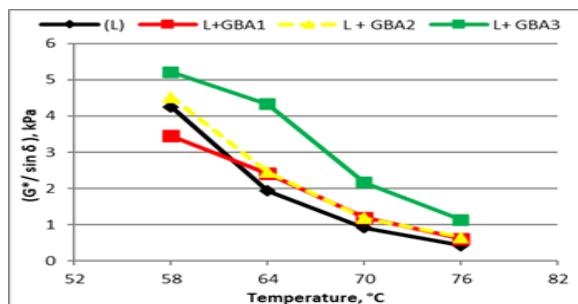


Fig (29): Rutting resistance parameter values ($G^*/\sin \delta$) of both original and GAB [50% asphalt + 50% GBA] at different temperatures.

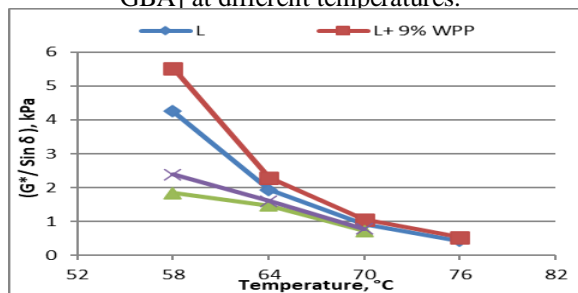


Fig (30): Rutting resistance parameter ($G^*/\sin \delta$) values of both original AC and PMA using different contents of waste polymers at different temperatures.

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