

## Article

# Asphalt Binder Rheological Performance Properties Using Recycled Plastic Wastes and Commercial Polymers

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## Abstract

Polymer-based product usage in modern society is increasing day by day. Following usage, these inert products and hydrophobic materials contribute to environmental pollution, often accumulating as litter in ecosystems and contaminating water bodies. The rapid socio-economic development in the Kingdom of Saudi Arabia (KSA) has resulted in a significant increase in waste generation. This study was conducted on the utilization of recycled plastic waste (RPW) polymer along with commercial polymer (CP) for the modification of the local binder. The hot environmental conditions and increased traffic loading are the major reasons for the permanent deformation and thermal cracks on the pavements, which require improved and modified road performance materials. The Ministry of Transport and Logistical Support (MOTLS) in Saudi Arabia, along with other related agencies, spends a substantial amount of money each year on importing modifiers, including chemicals, hydrocarbons, and polymers, for modification purposes. This research was conducted to investigate and utilize available local recycled plastic materials. Comprehensive laboratory experiments were designed and carried out to enhance recycled plastic waste, including low-density polyethylene (rLDPE), high-density polyethylene (rHDPE), and polypropylene (rPP), combined with varying percentages of commercially available polymers such as Styrene-Butadiene-Styrene (SBS) and Polybilt (PB). The results indicated that incorporating recycled plastic waste expanded the binder's susceptible temperature range from 64 °C to 70 °C, 76 °C, and 82 °C. The resistance to rutting was shown to have significantly improved by the dynamic shear rheometer (DSR) examination. Achieving the objectives of this research, combined with the intangible environmental benefits of utilizing plastic waste, provides a sustainable pavement development option that is also environmentally beneficial.

**Keywords:** recycled plastic waste; commercial polymers; dynamic shear rheometer; upper performance grade; rutting



Received: 20 August 2025

Revised: 23 September 2025

Accepted: 28 September 2025

Published: 4 October 2025

**Citation:** Abdul Wahhab, H.I.A.; Rafiq, W.; Habib, M.A.; Babalghaith, A.M.; Abdulrahman, S.; Shahzad, S. Asphalt Binder Rheological Performance Properties Using Recycled Plastic Wastes and Commercial Polymers. *Constr. Mater.* **2025**, *5*, 75. <https://doi.org/10.3390/constrmater5040075>

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## 1. Introduction

Plastic waste has long been considered a direct and indirect primary source of pollution related to the environment [1,2]. The nuisance created by plastic waste in the environment is well documented due to its inertness to biodegradation. As far as conserving the marine

ecosystem is concerned, plastics have drastic effects, as they are regarded as biochemically inert materials and hardly react with the endocrine system due to the fact that they have large molecular size. Thereby, penetration through the cell membrane would be inhibited due to this. Chemicals of low molecular weights ( $MW < 1000$ ) are found in marine plastics, which are plastic trash found in the marine environment. These substances have the ability to enter cells, interact chemically with important molecules in the body, and perhaps interfere with the operations of the endocrine system [3,4]. Since recycling reduces the demand for more virgin plastics and keeps PW from damaging the environment, it has been determined to be the best plastic waste (PW) management technique [5,6]. Waste utilization offers a compelling alternative to disposal by reducing or even eliminating disposal costs and potential pollution issues while also promoting resource conservation [7–9]. Among the many benefits of mechanically processing polymer waste are the provision of job opportunities, a decrease in the carbon footprint related to the production of polymer resin, and lower processing costs for plastic converters.

Recent research has examined the possibility of using plastic waste in the manufacturing of building materials, namely for the asphalt and bitumen industries, either by commingling or single-stream processing with very minor adjustments to standard procedures [10–12]. In KSA, it has been shown that adding recycled low-density polyethylene (rLDPE), rHDPE, and polypropylene (rPP) to locally obtained asphalt binders can improve their performance grade (PG) and storage stability [13–15]. The maximum seven-day pavement temperature in KSA ranges between 64 °C and 76 °C, suggesting that locally sourced bitumen performs effectively at pavement temperatures below 64 °C. This has resulted in a significant demand for polymer-modified bitumen binders in flexible pavement construction across the country [16]. Commercial sources of plastic waste that have been recycled and processed have been created since 2015 and used in asphalt for pavement surfacing applications [17–19].

The capacity of the bitumen mixture to withstand rutting and shoving brought on by traffic loads is the main criterion for the wearing course design of hot mix asphalt (HMA) pavements. To meet this requirement, the mixture must possess sufficient stability to endure traffic stresses effectively, while aligning with the specific traffic conditions [20–23]. Studies have indicated that adding polymers or plastic waste to bitumen greatly enhances the asphalt mix's performance. These modifications lessen the material's sensitivity to temperature changes, improve resistance to rutting deformation, and increase stiffness at high temperatures. Additionally, variations in the bitumen's rheological characteristics have been linked to improvements in fatigue resistance, contingent on the type of polymer utilized [24–26].

Plastics are incorporated into bitumen using the dry, wet, or hybrid methods [27,28]. The recycled plastic waste's chemical and physical properties determine the improvement expected from plastic-modified bitumen [25,29]. Pollutant adsorption, along with the leaching of chemicals during the plastic lifespan, significantly influences the property variations in RPW compared to their virgin counterparts, with additional dependence on their mechanical and thermal history [30,31], thereby generating a development that leads to the expected behavior of the modified RPW. It is crucial to establish a homogeneity in the RPW before its addition as a modifier, due to the presence of a variety of grades [32,33]. Expectation for a cost-effective use of RPW is quite high, and combined with its general availability across the region at cheaper rates compared to virgin plastics, this is the tipping point in the large-scale use of RPW in the modification of bitumen binders [34–36]. Additionally, the higher expenses incurred in the case of polymer-based additives when compared to waste thermoplastics have driven the usage of waste thermoplastics for bitumen modification [37,38].

SBS and ethylene vinyl acetate (EVA) polymers have been widely used as bitumen modifiers and have shown great efficacy in creating high-performance polymer-modified asphalt roads [39,40]. Among plastomers, polyethylene is regarded as one of the most effective polymers for modifying asphalt binders. Bitumen binders modified with polyethylene exhibit superior resistance to low-temperature cracking and permanent deformation compared to traditional bitumen binders [41,42]. Polyethylene, a widely used plastomer, is available in multiple forms, including LDPE, medium-density polyethylene (MDPE), HDPE, and linear low-density polyethylene (LLDPE). The melting point temperatures for these forms range from 108 to 120 °C for LDPE, 126 to 129 °C for MDPE, 129 to 149 °C for HDPE, and 124 to 128 °C for LLDPE [43,44].

MOTLS in KSA, along with other associated agencies, spends a significant amount of money annually on importing modifiers, including chemicals, hydrocarbons, and polymers, for modification purposes. Fewer efforts were made to explore the potential of using locally available material that can be recycled. The present study concentrated on utilizing recycled plastic waste for binder modification, reducing the consumption of commercial polymers while supporting sustainability objectives. The research study focused on optimization techniques to determine the modified binder blends mixing conditions and recycled waste polymer utilization in the Kingdom. Furthermore, the performance properties of modified binder blends were assessed with consideration for potential cost reductions compared to conventional polymers.

## 2. Methodology

### 2.1. Materials

The asphalt binder utilized in this study was acquired from the Ras-Tanura refinery in the Eastern Province of Saudi Arabia, among other sources. The recycled waste, including rLDPE, rHDPE, and rPP, was collected from the waste bin of the KFUPM central cafeteria, while some recycled local materials were sourced from relevant identified locations. The waste materials were subsequently processed for easier utilization. The commercial polymers employed in this investigation are comprised of the elastomer (SBS Calprene) and the plastomer (Polybilt 101), manufactured by Dynasol Group, and ExxonMobil Chemical company, respectively locally supplied by Alkabi, Saudi Arabia. The fundamental properties and chemical composition of the recycled and commercially sourced polymers, such as class, chemical composition, melting point, and density, are summarized in Table 1. The RPWs were processed by removing the marked labels and then followed by washing and drying for any impurities. The recycled plastic waste was initially cut into small sizes so that it was suitable to feed into the extrusion machine, as shown in Figure 1. After using the shredding machine, further grounding was conducted to ensure homogeneous mixing and proper melting with the base binder. Figure 2 depicts the raw plastic waste and the shredded plastic dust after processing through the extrusion machine.

**Table 1.** Physical properties and chemical composition of both recycled and commercial polymers.

| Polymer Type | Code | Origin       | Class     | Chemical Composition    | Melting Point (°C) | Density (g/cm <sup>3</sup> ) |
|--------------|------|--------------|-----------|-------------------------|--------------------|------------------------------|
| rLDPE        | L    | Waste        | Plastomer | Branched Polyethylene   | 130                | 0.941                        |
| rHDPE        | H    | Waste        | Plastomer | Line Polyethylene       | 150                | 0.952                        |
| rPP          | P    | Waste        | Plastomer | Isotactic PP            | 160                | 0.943                        |
| SBS          | S    | Commercially | Elastomer | Aromatic styrene blocks | 180                | 1.061                        |
| Polybilt     | B    | Commercially | Plastomer | Ethylene vinyl acetate  | 140                | 0.942                        |

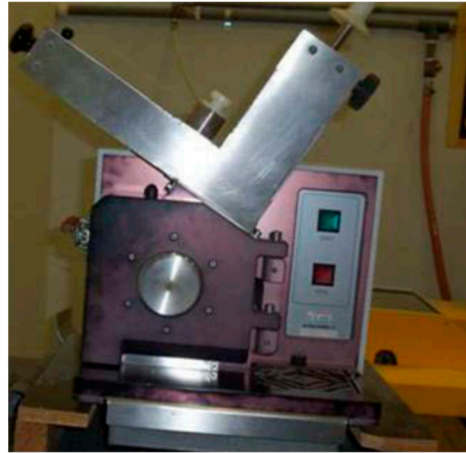


**Figure 1.** Polymer extrusion machine.



**Figure 2.** Polymers used for this study: (a) recycled plastic waste, (b) pre-shredding, (c) post-shredding, (d) commercial polymers ("B" and "S").

## 2.2. Preparation and Testing of Polymer Asphalt Blends

After collection, recycled plastic wastes from multiple sources were initially cleaned and processed to make them into a small size of  $1 \times 1$  mm. The dust was presoaked in the binder for one hour before blending. The shear blending process was subsequently

conducted at a speed of 3500 rpm, as shown in Figure 3, with the temperature maintained at 150 °C. As recycled plastic wastes are difficult to blend with a base binder because of high density and a corresponding high melting point, high shear speed is recommended as compared to virgin polymer. Optimum blending time was determined using recycled waste polymers. The consistency of the complex shear modulus ( $G^*/\sin\delta$ ) parameter at 70 °C was utilized. This was conducted to counter the polymer agglomeration that could happen if blending time exceeds the polymer mixing limit, which also caused oxidation [45,46]. Moreover, this approach is not cost-effective and would negatively impact the rheology of the modified polymer binder blend [47]. Complex shear modulus values were recorded at time intervals ranging from 10 to 70 min. The ideal blending duration for a particular recycled polymer was determined based on the stabilized ( $G^*/\sin\delta$ ) value. For achieving the optimum mixing time for specific waste material with binder, many mixing durations were considered. Adjustments to the mixing time, temperature, and speed were made as needed to ensure proper blending whenever inadequate blending was observed. Hence, under optimal conditions, the risk of aging and oxidation is eliminated while reducing mixing energy.



**Figure 3.** High-speed shear mechanical blender.

After preparing the RPW and fresh binder blends, physical characterization was conducted to compare them with the standard specifications outlined by the MOTLS, KSA. A viscosity test, performed in accordance with ASTM D 4402 [48], was carried out on all blended samples using a Brookfield viscometer (Brookfield Engineering Laboratories, Inc., Middleboro, MA, USA) to assess the binders' viscous behavior at 135 °C. The ageing qualities of the blends were assessed using the rolling thin film test in accordance with ASTM D2872-12 [49]. This test assesses how heat and air affect a thin layer of both new and modified binder mixtures. The resulting binder after the performed test was evaluated for the characteristics of bitumen before and after the test. To assess the viscoelastic behavior of RPW binder blends, the DSR test was performed complex shear modulus, rutting, and fatigue were the major parameters evaluated from the results. Following the Superior Performing Asphalt Pavement System (Superpave) modified binder blends using recycled plastic waste, "S" and "B" RPWs were classified.

### 2.3. Experimental Matrix

The experimental matrix was designed for the determination of the physical, rheological, and viscoelastic behavior of aged and unaged binder blends. Table 2 shows the various combinations of RPW and commercial polymers, viz., "S" and "B".

**Table 2.** Experimental matrix for polymer combination.

| Recycled Plastic Waste | RPW Percentage | “S” |    |      |    | “B” |    |      |    |
|------------------------|----------------|-----|----|------|----|-----|----|------|----|
|                        |                | 0%  | 1% | 1.5% | 2% | 0%  | 1% | 1.5% | 2% |
| “L”                    | 0%             | ✓   | ✓  | ✓    | ✓  | ✓   | ✓  | ✓    | ✓  |
|                        | 2%             | ✓   | ✓  | ✓    | ✓  | ✓   | ✓  | ✓    | ✓  |
|                        | 4%             | ✓   | ✓  | ✓    | ✓  | ✓   | ✓  | ✓    | ✓  |
|                        | 6%             | ✓   | ✓  | ✓    | ✓  | ✓   | ✓  | ✓    | ✓  |
| “H”                    | 0%             | ✓   | ✓  | ✓    | ✓  | ✓   | ✓  | ✓    | ✓  |
|                        | 2%             | ✓   | ✓  | ✓    | ✓  | ✓   | ✓  | ✓    | ✓  |
|                        | 4%             | ✓   | ✓  | ✓    | ✓  | ✓   | ✓  | ✓    | ✓  |
|                        | 6%             | ✓   | ✓  | ✓    | ✓  | ✓   | ✓  | ✓    | ✓  |
| “P”                    | 0%             | ✓   | ✓  | ✓    | ✓  | ✓   | ✓  | ✓    | ✓  |
|                        | 2%             | ✓   | ✓  | ✓    | ✓  | ✓   | ✓  | ✓    | ✓  |
|                        | 4%             | ✓   | ✓  | ✓    | ✓  | ✓   | ✓  | ✓    | ✓  |
|                        | 6%             | ✓   | ✓  | ✓    | ✓  | ✓   | ✓  | ✓    | ✓  |

### 3. Design of Experiments Based on RSM

For experimental design, optimization, statistical analysis, and result validation, the Response Surface Methodology (RSM), an intuitive statistical tool, was employed. RSM has wide-ranging applications in the concrete and asphalt industry in the context of developing experimental design, generating statistical models, and determining the association between independent variables (factors) and dependent variables (responses) [50,51]. Optimal custom design for the experimental design involving factors and responses was opted in this research for effective utilizing of time and resources [52–54]. In the current RPW and commercial polymer (CP) are variables affecting the performance properties of binder blends, which include the viscosity and RPW binder’s upper performance grade as responses. Table 3 shows the factors, viz., recycled plastic waste and commercial polymer level, and the codes used.

**Table 3.** RSM-based design of factors and their corresponding codes.

| Factors                      | Units | Code           | Levels |     |    |
|------------------------------|-------|----------------|--------|-----|----|
|                              |       |                | −α     | 0   | +α |
| Recycled Plastic Waste (RPW) | %     | X <sub>1</sub> | 0      | 4   | 6  |
| Commercial Polymer (CP)      | %     | X <sub>2</sub> | 0      | 1.5 | 2  |

On the basis of the model’s significance, quadratic models were proposed for the prediction of the performance properties of modified binder blends. The predicted responses were validated using second-degree polynomial equations, as demonstrated in Equation (1) [55].

$$Y = C + A_1(X_1) + A_2(X_2) + A_3(X_1^2) + A_4(X_2^2) + A_5(X_1 * X_2) \quad (1)$$

where  $Y$  is the obtained response from the experimental results (viz., viscosity and upper performance grade),  $X_1$  and  $X_2$  are recycled plastic waste and commercial polymers, respectively,  $C$  represents the model equation constant, while  $A_1$  to  $A_5$  denote the model coefficients. After assigning the factor levels as low and high, the experimental design resulted in 16 runs. Five-time replication of the central points was carried out to enhance reliability during the experimental and analytical phases. The modified binder blends, which include raw plastic waste and commercial polymers, were prepared in line with the outcome of the RSM experimental matrix, and corresponding performance property results were inserted in the Design-Expert software. The analysis steps were performed

for the model development, followed by optimization and validation. The evaluation of the model, based on performance properties, was carried out using ANOVA for the specified responses. Regression analysis was further examined using lack of fit,  $F$ -values, and  $p$ -values. To visualize the variables, 3D diagnostic plots were generated.

## 4. Results and Discussion

### 4.1. Recycled Plastic Waste Blending Temperature

The melting temperature was needed to specify the polymer blending temperature. The melting temperature of commercial temperature, viz., “S” and “B”, was easily identified, but for the recycled plastic waste polymer, the melting point was figured out by the Differential Scanning Calorimetry test, as shown in Figure 4a–c for “H”, “L”, and “P”, respectively. The melting temperatures of RPW (160–190 °C), compared to commercial polymers, which are SBS and PB, were approximately 180 °C and 145 °C. These values confirm why higher blending temperatures were required for RPW compared to virgin elastomers. The literature shows that excessive blending above these temperatures can induce oxidation and lumps. Table 4 shows the obtained melting temperature for recycled polymers.

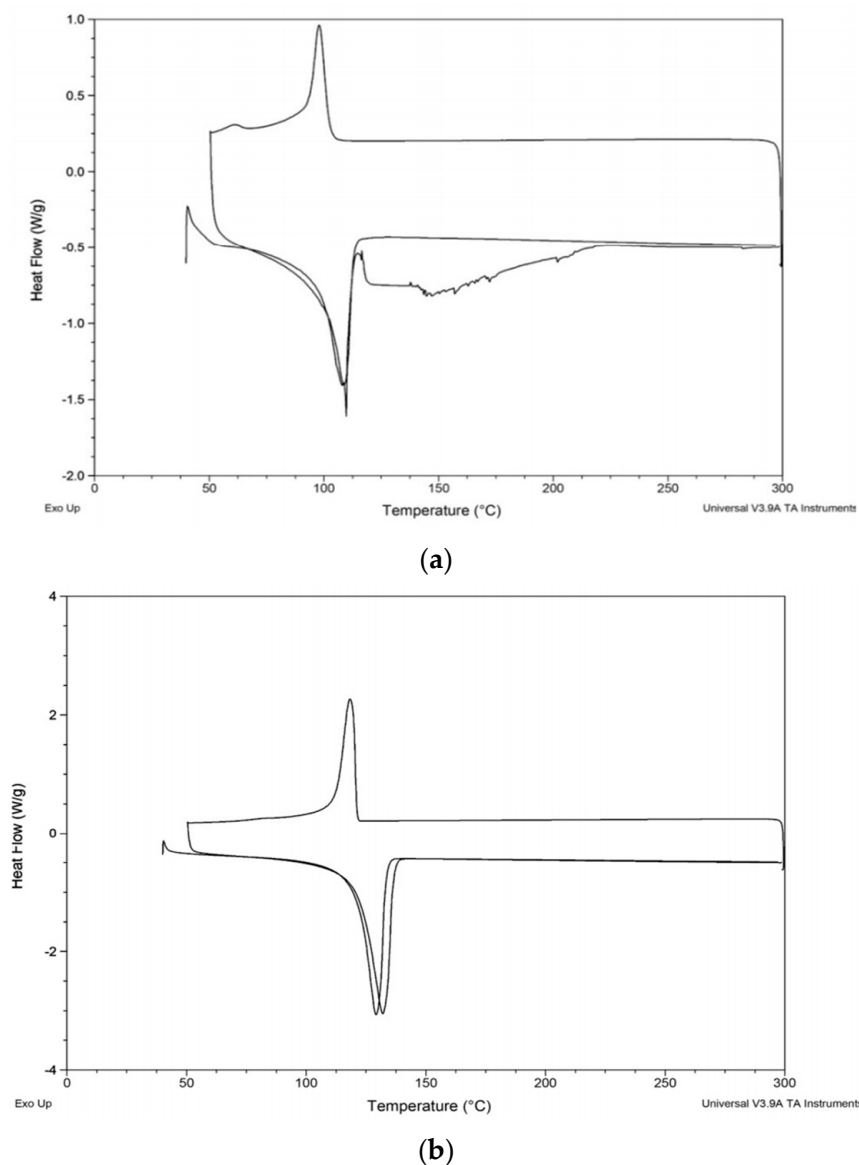


Figure 4. Cont.

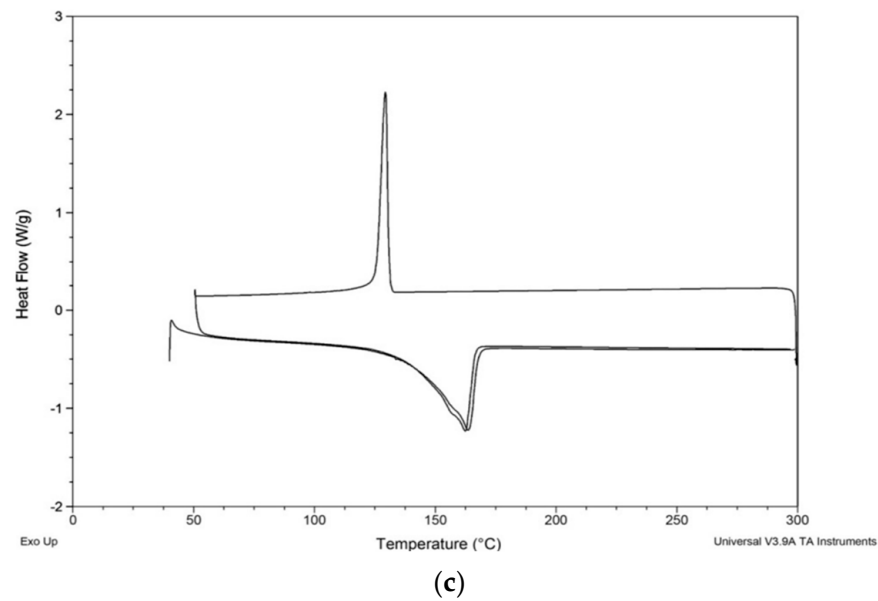


**Figure 4.** Differential scanning calorimetry result for (a) “H”, (b) “L”, and (c) “PP”.

**Table 4.** Recycled polymer blending temperature.

| Recycled Plastic Waste | Blend Temperature | Symbolic Representation                                                               | Code |
|------------------------|-------------------|---------------------------------------------------------------------------------------|------|
| rLDPE                  | 160 °C            |   | L    |
| rHDPE                  | 180 °C            |  | H    |
| rPP                    | 190 °C            |  | P    |

#### 4.2. Optimum Polymers Blending Duration

The optimum blending time for recycled plastic waste polymer was calculated using 4% of the RPW polymer  $G^*/\sin\delta$  value for the time interval from 10 to 70 min. The complex shear modulus value was determined for each recycled waste polymer with respect to time and is presented in Figure 5. The observation had been made that by the addition of recycled waste polymer to fresh binder, the dispersion of the molecular association becomes stable as the value of  $G^*/\sin\delta$  becomes uniform. The “H” optimum blending time, after close monitoring, came out to be 60 min. Similarly, the required blending time for “L” was observed at 30 min. The blending time for “P”, where the constant value of complex modulus was found, came out to be 50 min. From the available literature and from the manual provided by the suppliers, the mixing temperature and time of commercial polymers were easily identified and are shown in Table 5, along with the calculated mixing temperature and time for recycled plastic polymer waste [56–58]. Also, the same table presents the blending temperature and time for modified polymers, in which critical temperature and time were selected based on the individual polymer. The critical values of blending temperature and time were used for the modified blend of recycled plastic waste and commercial polymer.

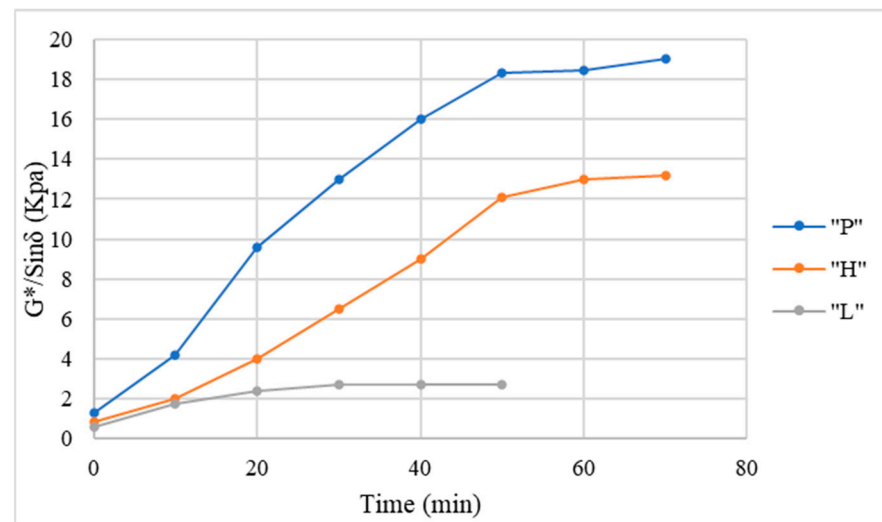


Figure 5. "H" optimum blending duration.

Table 5. Temperature and blending duration of RPW and commercial polymers.

| Polymers  | Temperature (°C) | Time (min) |
|-----------|------------------|------------|
| "L"       | 160              | 30         |
| "H"       | 180              | 60         |
| "P"       | 190              | 50         |
| "S"       | 180              | 60         |
| "B"       | 145              | 60         |
| "S" + "L" | 180              | 60         |
| "S" + "H" | 180              | 60         |
| "S" + "P" | 190              | 60         |
| "B" + "L" | 160              | 60         |
| "B" + "H" | 180              | 60         |
| "B" + "P" | 190              | 60         |

#### 4.3. Performance Properties and Superpave Upper Performance Grade (UPG)

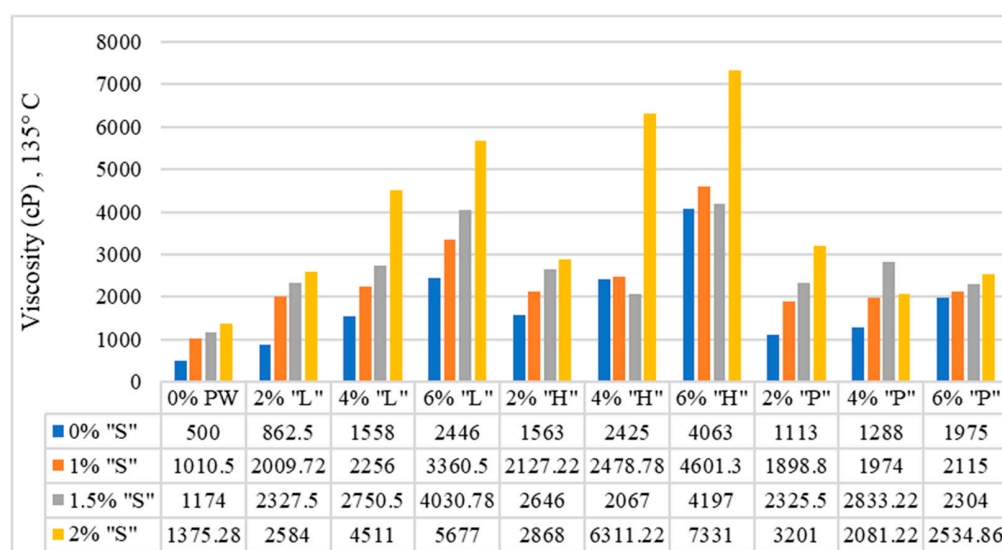
The performance properties, including the viscous behavior, rheological parameters, and Superpave upper performance grade for the modified binder blends and fresh binder, were measured and will be discussed in the sub-sections. Viscosity and rheological changes to binder structure are linked to the polyethylene-based RPW, which tends to stiffen the binder through the formation of a crystalline network, while on the other side, elastomers like SBS impact the elasticity.

#### 4.4. Rotational Viscosity Test Result

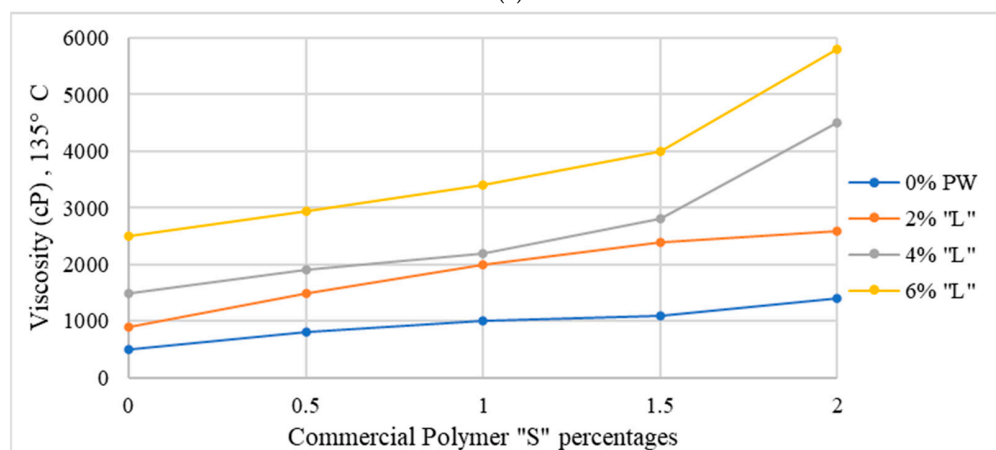
Viscoelastic behavior refers to the binder's ability to exhibit viscous properties at high temperatures and elasticity at low pavement temperatures, ensuring appropriate mixing and compatibility with aggregates. In this study, different binder blends of modified recycled plastic waste polymer with commercial polymers were tested with different compositions. Figure 6a shows the viscosity results conducted at 135 °C. For the control binder addition of "S" content from 0 to 2% a nominal change in the viscosity values was observed, viz., 500, 1010, 1173, and 1374 cP, respectively. With the addition of "L" from 2 to 6%, along with "S" from 0 to 2%, the viscosity values enhanced quite significantly, as shown in Figure 6b. This showed the increase in the molecular resistance to flow at max 6% "L" content by the creation of the network in between the modified binder. The viscosity behavior of "H", with the utilization of the "S" polymer, was high, as shown in Figure 6c.

The more dominant was the “H” binder network, which showed an increasing viscosity trend, as opposed to the “S”, and the binder network can be seen in 0% PW and the 6% “H” line. The 1.5% “S” polymer and binder network seems to be more prevalent than the binder and “H” network, which resulted in a reduction in the extra friction in the blend between particles. However, at 2%, the “S” addition to the increase in the viscosity was found, which was evident from the thickening effect of the “S” polymer. Viscosity in this current scenario can be the function associated with networking and polymer structure formation inside the binder matrix blend [59–61].

Figure 7a shows the commercial polymer, i.e., “B”, with the viscosity results of the recycled plastic waste polymers. The pattern shows that adding “B” enhanced the viscous behavior of the binder blend, as evident from the 0% PW line. The “B” blend with “P” from 2 to 6% showed a continuous increment of viscosity; however, “H” and “B” showed a decreasing trend in viscosity. This is because of the increase of “H” and binder network to resist flow and intermolecular friction, and along with this, “B” and binder seem to shear thin and result in a reduction in intermolecular friction, to an extent. So, the “H” and binder flow resistance dominated over the “B” binder blend. The results shown in Figure 7b for viscosity showed that a few blends, viz., L4S2, H6S1, H6S1.5, H6S2, H6B1.5, and H6S2 with “S” polymer and P6S1.5, and P6S2 with “B”, do not pass the SHRP specification for viscosity that should be below 3000 cP at 135 °C.



(a)



(b)

Figure 6. Cont.

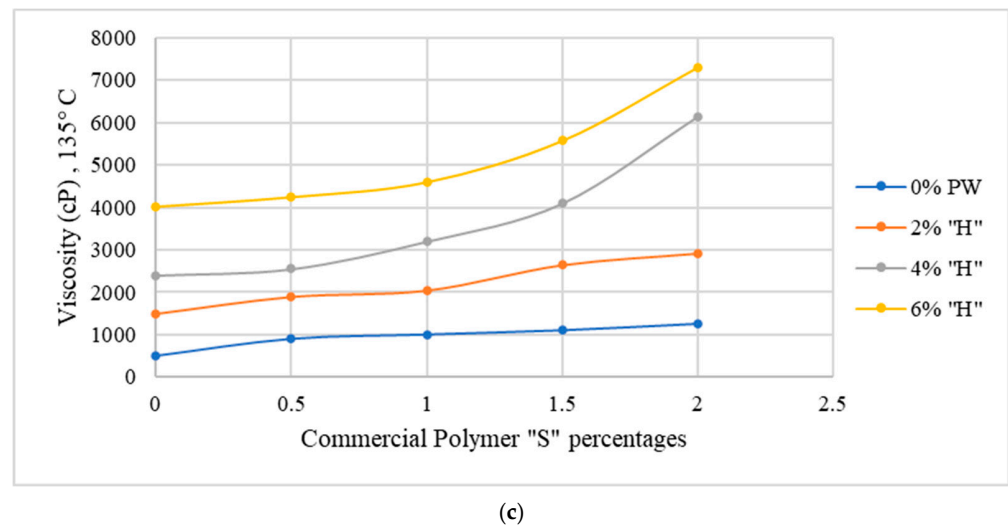


**Figure 6.** Viscosity results of (a) RPW and “S”, (b) recycled “L” and “S”, (c) recycled “H” and “S”.

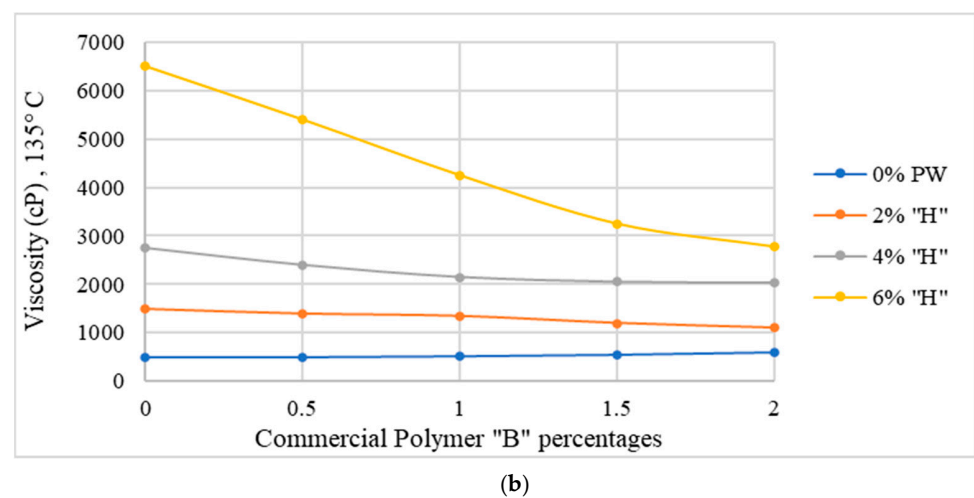
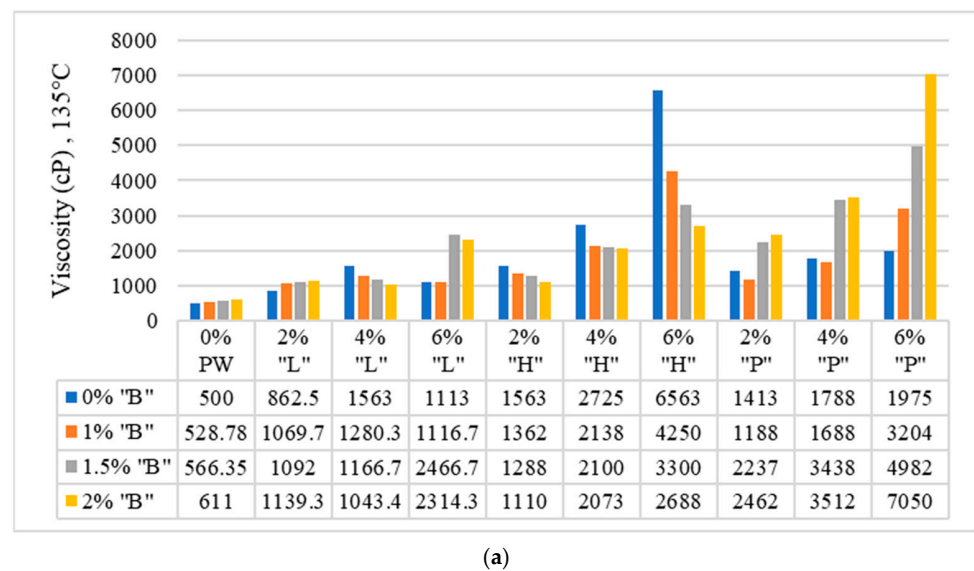
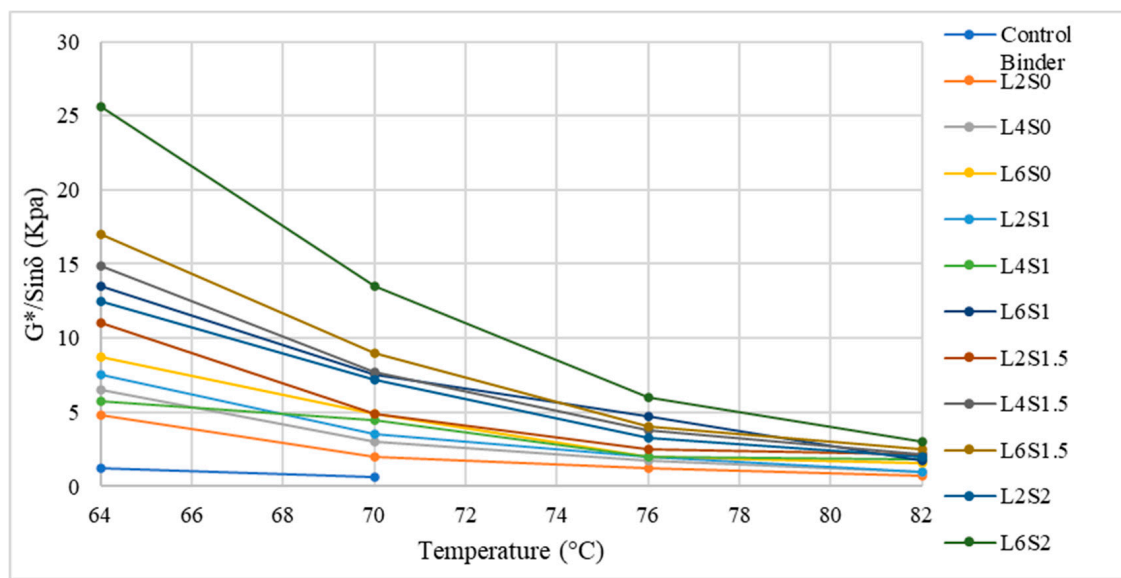


**Figure 7.** Viscosity results of (a) recycled polymer and “B”, (b) “H” and “B” modified binder.

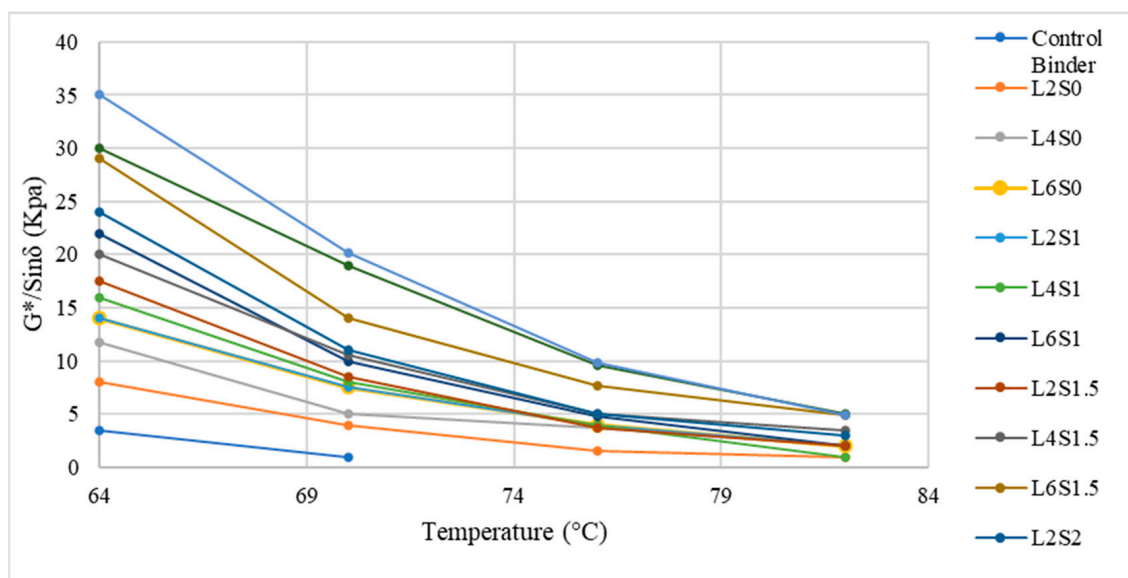
#### 4.5. Modified Binder High Temperature Rutting Resistance

The improvement in rutting resistance was assessed for both aged and unaged binders by plotting the values of  $G^*/\sin\delta$  against the test temperature. Figure 8a,b present the

plots for “L” and “S” modified binder blends under both aged and unaged conditions. The results revealed a significant improvement in both aged and unaged samples’ rutting resistance. When compared to the control bitumen at higher temperatures, the stiffness of modified binder blends improved with the inclusion of waste and commercialized polymer. Looking closely at the graph lines of the recycled and commercial formulations, namely L2S0, L4S0, and L6S0, the results show that 2% “L” improves the rutting resistance compared to the control binder, indicating that the network formation of the binder with “L” is somewhat stiffer, and adding 2% more waste polymer to L4S0 improves the rutting even more, and the peak of the graph line moves away from L2S0. The significant improvement was observed by the further addition of polymer to L6S0, and a large gap of improvement in rutting resistance can be seen in the graph plots for L4S0 and L6S0. The noteworthy improvement in the permanent deformation resistance is reliant on the “L” molecules’ thickening effect. The comparison between 0% “S” and 1% “S” addition in “L” showed that the improvement in rutting is high at lower temperatures and the improvement is slight at higher temperatures.



(a)



(b)

**Figure 8.** PMB high temperature rutting resistance: (a) fresh condition, (b) RTFO aged condition.

#### 4.6. Viscoelastic Component Analysis from DSR Result

Dynamic Shear Rheometer (DSR) results are calculated by determining the complex shear modulus ( $G^*$ ), which quantifies the binder's total resistance to deformation under repetitive shear loading.  $G^*$  is further divided into two components: elastic and viscous. With the increase in  $\delta$  at constant  $G^*$  values, the dominant part will be viscous. From Figure 9,  $\tan \delta$  was expressed as the viscoelastic property of the binder mixture, and with increasing temperature, the viscous component became dominant, compared to its elastic counterpart. The steepness of the graph lines at the low percentage of "L" was higher in comparison to the high percentages. For instance, L2S0 is steeper than L4S0, and so on. The L2S1 and L2S1.5 graph lines indicated stiffer blends, and thereby, flatter lines can be seen as a result of the polymeric network formation of blends with lower viscosity values at a high temperature. Similarly, higher percentages with "L" and "S" showed less viscoelastic change at high temperature in comparison to lower "L" and "S" contents.

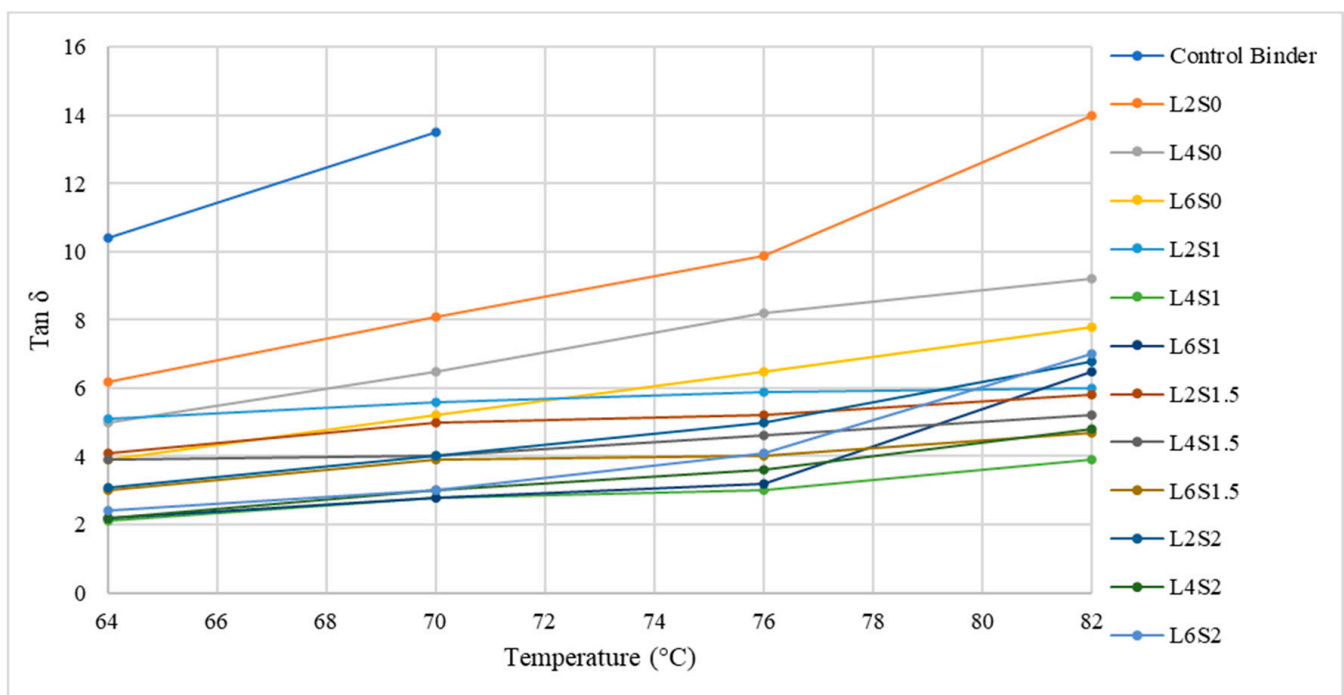


Figure 9.  $\tan \delta$  (viscous to elastic) vs. temperature graph.

#### 4.7. Actual PG at High Service Temperature

Following the Superpave specification for determining the actual PG grade, a sample calculation presented in Figure 10a,b displays the  $G^*/\sin \delta$  result of the original condition, the aged RTFO binder results of L4S1 (4% "L" and 1% "S"). The results showed that the actual temperature of the fresh modified binder was 79.6 °C, which is in line with the climatic requirements of the Kingdom of Saudi Arabia, which requires paving temperatures of 76 °C. On the other hand, the temperature of the fresh modified binder was 79.4 °C. The results showed that RPW and polymers can extend binder usage beyond the conventional limits. The outcome is also compared to SHRP-based specifications, which are adopted for Gulf countries. Therefore, the lowest, considered as a critical one, was 79.4 °C, and has been selected as an upper service temperature grade for rL4S1. Similarly, for all selected modified binders, high PG temperature was calculated and visualized in Figure 11.

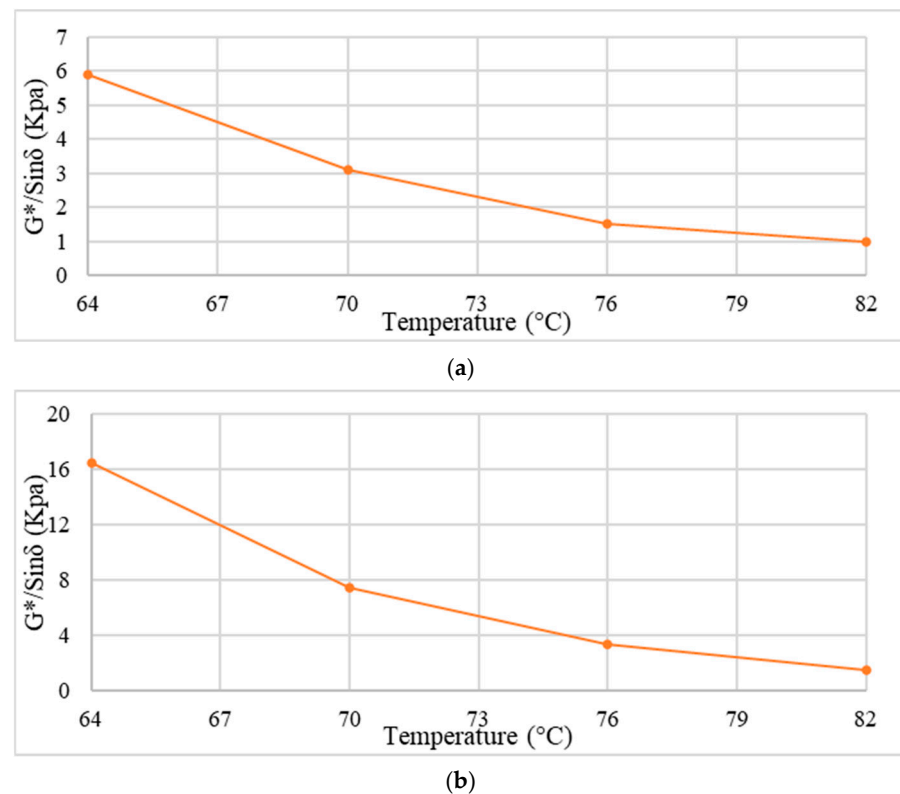


Figure 10. L4S1 modified blend plot of  $G^*/\sin\delta$  vs. temp for (a) unaged  $G^*/\sin\delta$  vs. temp. (b) aged.

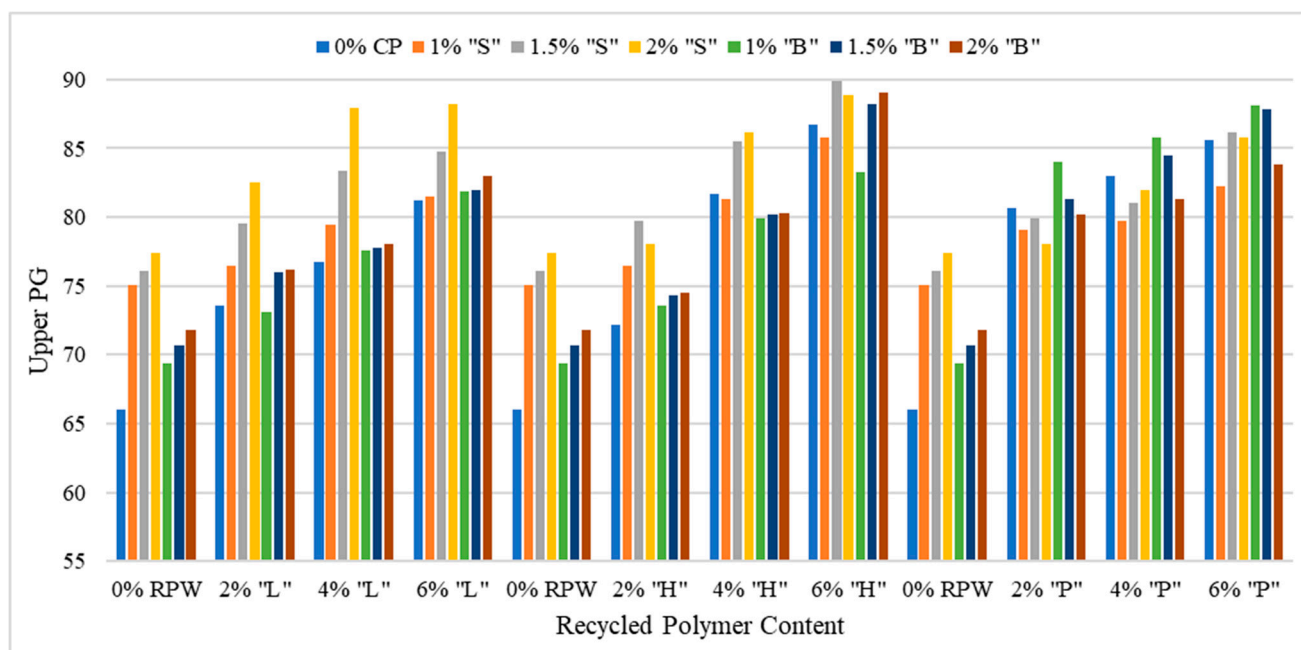
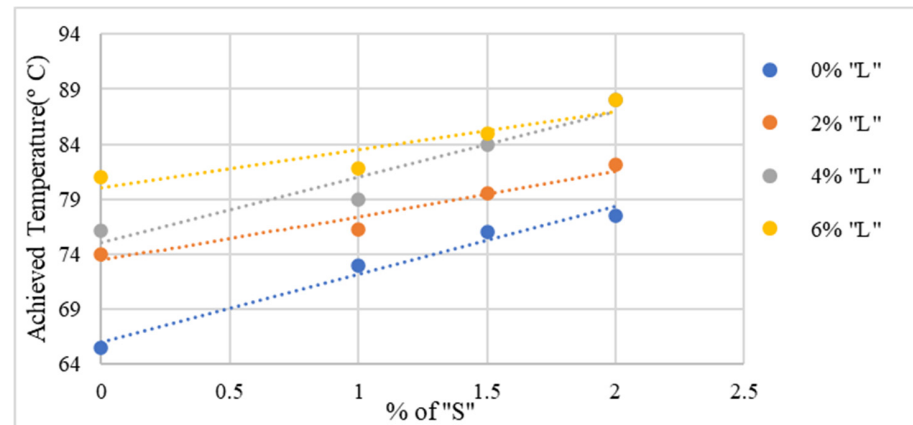


Figure 11. Recycled plastic waste and commercial polymer blends—upper PG.

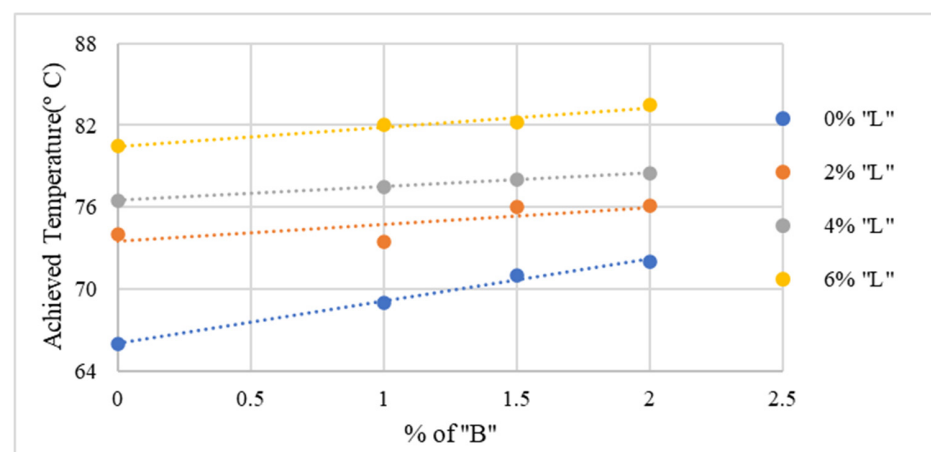
#### 4.8. Effect of Recycled LDPE on Upper Performance Grade of SBS and Polybilt

The performance grade of "L" with commercial "S" improved sharply as compared to the addition of the only commercial "S". Figure 12a shows the addition of 1.5% of commercial "S", where the modified binder can pass up to 76  $^{\circ}\text{C}$ , and another addition of 4% "L" where the PG is accelerated up to 82  $^{\circ}\text{C}$ . This showed that the elimination of the costly commercial modifier can be conducted by the addition of the "L". Similarly,

the “L” and “S”, 6% and 1% addition, respectively, can provide the required targeted temperature of 82 °C. Figure 12b showed the performance grade plots for the different percentages of the commercial polymer “B”; prominent changes in the temperature have been found for the performance grade. The achievement of 82 °C temperature with 1% “B” and 6% “L” can be possible; similarly, the use of 2% “L” with 2% “B” can be used to attain a performance-grade temperature of 73 °C. The graph is used to obtain the required polymer and “L” percentages shown in Table 6 to obtain the PG grade of polymer blends.



(a)



(b)

**Figure 12.** Modified binder effect on UPG for (a) “L” and “S” (b) “L” and “B”.

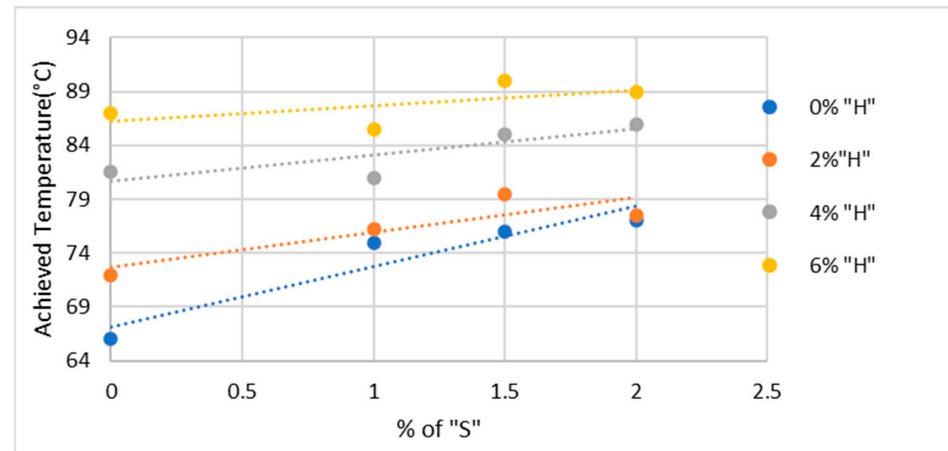
**Table 6.** Determination of amount of “L” and “S” to attain expected temperature.

| Upper PG | “L” | “S”  | “L” | “S”  | “L” | “S”  |
|----------|-----|------|-----|------|-----|------|
| 82 °C    | 6%  | 0.6% | 4%  | 1.2% | 2%  | 2%   |
| 76 °C    | 4%  | 0%   | 2%  | 0.6% | 2%  | 1.5% |

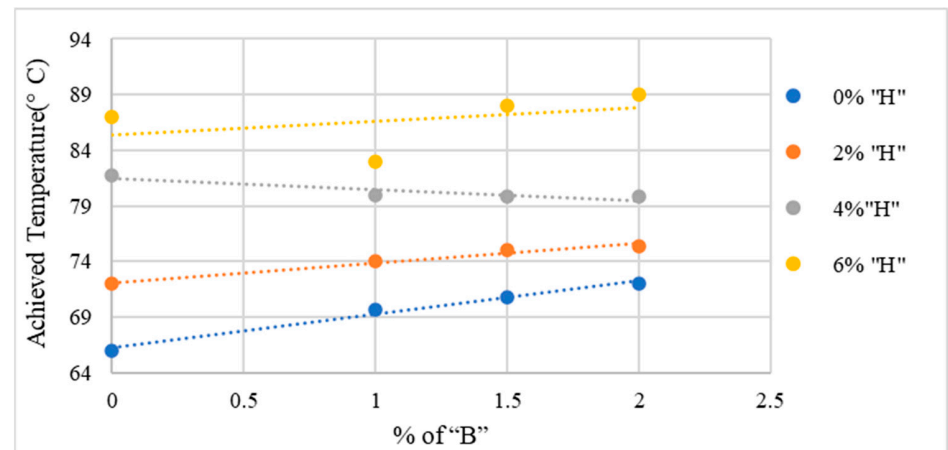
#### 4.9. Effect of HDPE on Upper Performance Grade of SBS and Polybilt Modified Binder

The effect of “H” with “S” was significantly improved in terms of high-performance-grade temperature. The higher crystalline and linear structure of “H” creates stiffness in the binder but requires elastomeric reinforcement to counter this brittleness. This explains why the results of “S” are a better match because of elastic recovery as compared to the “B”. Figure 13a shows the effect of adding commercial elastomer “S” with “H”, which has uplifted the binder performance grade. The effect of “H” with “B” was significantly improved in terms of high-performance-grade temperature. From Figure 13b, “H” and

“B”, 6% and 2%, respectively, showed good improvement; however, the “H” 4% trend was unusual, as this could be due to poor compatibility between the waste plastic and the commercial polymer. The optimal percentage of “S” with “H” was determined and presented in Table 7.



(a)



(b)

**Figure 13.** Modified binder effect on UPG for (a) “H” and “S” (b) “H” and “B”.

**Table 7.** Determination of amount of HDPE and “S” to attain expected temperature.

| Upper PG | “H” | “S”  | “H” | “S”  | “H” | “S”  |
|----------|-----|------|-----|------|-----|------|
| 82 °C    | 4%  | 0.5% | 2%  | 1%   | -   | -    |
| 76 °C    | 4%  | 0%   | 2%  | 0.6% | 2%  | 1.5% |

## 5. Statistical Analysis

The effect of plastic waste polymers and commercial polymer usage on performance properties was analyzed; plastic waste polymer and commercial polymer were independent variables, and the effect on the viscosity and upper performance grade were dependent variables. ANOVA was performed using RSM. The performance properties of the blends were predicted by the quadratic polynomial equation suggested based on the significance of the model. The modified Equation (2), derived from the generalized Equation (1) and its selected factors, is hereby presented.

$$Y = C + A_1(RPW_1) + A_2(CP_2) + A_3(RPW_1^2) + A_4(CP_2^2) + A_5(RPW_1 * CP_2) \quad (2)$$

Y stands for the experimentally determined viscosity and upper performance grade temperatures. C is the intercept constant, while  $A_1$  through  $A_5$  are the equation coefficients. RWP (L, H, P) and CP (S, B) are independent variables. Tables 8 and 9 display the created models' significance and fit attributes as established by the ANOVA results.  $F$ - and  $p$ -values are used to assess the significance of the model; a  $p$ -value of less than 0.05 suggests a strong correlation between the independent and dependent variables. Furthermore, the accuracy of the expected responses in the established models is confirmed by an  $R^2$  value greater than 0.80 and a numerical difference between the adjusted and predicted  $R^2$  values of less than 0.20. The quality and fitness of the model are shown by the coefficient of determination ( $R^2$ ). Greater  $R^2$  values ( $>0.80$ ) indicate that the models fit the data well and show a high degree of agreement between the expected and actual responses [61,62].

Furthermore, the model adequacy and lack of fit can be evaluated by analyzing the variation in the data within the fitted model. The model's fitness can be confirmed by a lower lack of fit value and an insignificant  $p$ -value greater than 0.05 [63,64]. ANOVA analysis indicated smaller  $F$ -values and insignificant lack of fit, demonstrating highly significant and well-fitted models for the establishment of correlation between independent and dependent variables [65]. In addition, the results obtained from the combination of polymer blends, viz., recycled plastic waste and commercial polymers, showed noteworthy influence on the performance properties, viz., viscosity and the upper performance grade of the modified binder. Empirical equations from (3) to (14) were developed based on the independent variables, viz., RPW and CP, for the response prediction of the dependent variables [66].

$$\text{Viscosity (L)} = 62.04 + 23.93_1(L_1) + 1.585_2(S_2) - 11.03_3(L_1^2) + 19.3_4(S_2^2) + 26.26_5(L_1 * S_2) \quad (3)$$

$$\text{UPG (L)} = 66.22 + 3.95_1(L_1) + 0.975_2(S_2) - 0.316_3(L_1^2) + 2.200_4(S_2^2) - 0.065_5(L_1 * S_2) \quad (4)$$

$$\text{Viscosity (H)} = 82.05 + 32.01_1(H_1) - 12.01_2(S_2) + 11.94_3(H_1^2) + 76.9_4(S_2^2) + 30.77_5(H_1 * S_2) \quad (5)$$

$$\text{UPG (H)} = 67.41 + 2.732_1(H_1) + 2.511_2(S_2) + 0.128_3(H_1^2) + 1.37_4(S_2^2) - 0.884_5(H_1 * S_2) \quad (6)$$

$$\text{Viscosity (P)} = 44.90 + 40.83_1(P_1) + 95.67_2(S_2) - 46.95_3(P_1^2) - 22.2_4(S_2^2) + 17.68_5(P_1 * S_2) \quad (7)$$

$$\text{UPG (P)} = 66.9 + 5.66_1(P_1) - 2.22_2(S_2) - 0.33_3(P_1^2) + 3.71_4(S_2^2) - 1.201_5(P_1 * S_2) \quad (8)$$

$$\text{Viscosity (rLDPE)} = 46.90 + 36.55_1(rLDPE_1) - 12.8_2(PB_2) - 31.8_3(rLDPE_1^2) + 11.24_4(PB_2^2) - 51.9_5(rLDPE_1 * PB_2) \quad (9)$$

$$\text{UPG (L)} = 67.10 + 2.61_1(L_1) - 0.08_2(B) - 0.043_3(L_1^2) + 1.26_4(B_2^2) - 0.080_5(L_1 * B_2) \quad (10)$$

$$\text{Viscosity (H)} = 51.9 + 42.8_1(H_1) - 16.07(B_2) + 31.9_3(H_1^2) + 21.05_4(B_2^2) - 20.06_5(H_1 * B_2) \quad (11)$$

$$\text{UPG (H)} = 67.5 + 2.74_1(H_1) - 2.21_2(B_2) + 0.113_3(H_1^2) + 1.93_4(B_2^2) - 0.42_5(H_1 * B_2) \quad (12)$$

$$\text{Viscosity (P)} = 10.56 - 63.04_1(P_1) - 16.73_2(B_2) + 34.9_3(P_1^2) + 75.3_4(B_2^2) + 39.7_5(P_1 * B_2) \quad (13)$$

$$\text{UPG (P)} = 66.5 + 5.76_1(P_1) + 8.62_2(B_2) - 0.33_3(P_1^2) - 1.51_4(B_2^2) - 1.45_5(P_1 * B_2) \quad (14)$$

To further illustrate the relationship between the independent and dependent variables, three-dimensional graphs were created. Figure 14a–c show the recycled plastic waste and commercial polymer “S” interaction; the dependent variables, viscosity and upper performance grade, have significant effects based on the different percentages used in the modification of binder blends. An increase in viscous and higher upper performance-grade properties has been found with an increase in waste polymer percentages, which showed

that plastic waste addition, along with commercial polymer, has significant bonding, which affected the behavior of the binder blend. Figure 15a–c shows the recycled plastic waste and commercial polymer “B” interaction; the dependent variables, viscosity and upper performance grade, showed prominent and improved results with the different percentages of the polymers. The optimal values of the waste and commercial polymers achieving the required performance grade can be identified by the response variables, viz., viscosity and upper performance grade.

**Table 8.** ANOVA and model validation for RPW and “S”.

| Responses                    | Viscosity<br>("L" and "S") | UPG<br>("L" and "S") | Viscosity<br>("H" and "S") | UPG<br>("H" and "S") | Viscosity<br>("P" and "S") | UPG<br>("P" and "S") |
|------------------------------|----------------------------|----------------------|----------------------------|----------------------|----------------------------|----------------------|
| R <sup>2</sup>               | 0.97                       | 0.82                 | 0.95                       | 0.91                 | 0.91                       | 0.84                 |
| Adjusted R <sup>2</sup>      | 0.96                       | 0.80                 | 0.93                       | 0.85                 | 0.86                       | 0.76                 |
| Predicted R <sup>2</sup>     | 0.95                       | 0.76                 | 0.88                       | 0.74                 | 0.72                       | 0.62                 |
| Adequate Precision           | 38.93                      | 18                   | 23.5                       | 14.6                 | 15.6                       | 9.82                 |
| F-value                      | 143                        | 31.22                | 44.36                      | 19.19                | 19.57                      | 10.57                |
| p-value                      | <0.0001                    | <0.0001              | <0.0001                    | <0.0001              | <0.0001                    | <0.0001              |
| Model                        | Significant                | Significant          | Significant                | Significant          | Significant                | Significant          |
| Lack of fit (F- and p-value) | Not-significant            | Not-significant      | Not-significant            | Not-significant      | Not-significant            | Not-significant      |
|                              | 4.27                       | 0.56                 | 4.74                       | 2.18                 | 3.15                       | 0.79                 |
|                              | 0.0645                     | 0.77                 | 0.056                      | 0.206                | 0.06                       | 0.49                 |
| Final model                  | Quadratic                  | Quadratic            | Quadratic                  | Quadratic            | Quadratic                  | Quadratic            |

**Table 9.** ANOVA and model validation for RPW and “B”.

| Responses                    | Viscosity<br>("L" and "B") | UPG<br>("L" and "B") | Viscosity<br>("H" and "B") | UPG<br>("H" and "B") | Viscosity<br>("P" and "B") | UPG<br>("P" and "B") |
|------------------------------|----------------------------|----------------------|----------------------------|----------------------|----------------------------|----------------------|
| R <sup>2</sup>               | 0.86                       | 0.91                 | 0.86                       | 0.88                 | 0.94                       | 0.77                 |
| Adjusted R <sup>2</sup>      | 0.79                       | 0.89                 | 0.84                       | 0.86                 | 0.92                       | 0.71                 |
| Predicted R <sup>2</sup>     | 0.61                       | 0.86                 | 0.80                       | 0.80                 | 0.74                       | 0.61                 |
| Adequate Precision           | 11.4                       | 23.6                 | 8.81                       | 18.07                | 21.7                       | 13                   |
| F-value                      | 12.74                      | 65.71                | 8.66                       | 47.68                | 36.44                      | 13.84                |
| p-value                      | <0.0001                    | <0.0001              | <0.0001                    | <0.0001              | <0.0001                    | <0.0001              |
| Model                        | Significant                | Significant          | Significant                | Significant          | Significant                | Significant          |
| Lack of fit (F- and p-value) | Not-significant            | Not-significant      | Not-significant            | Not-significant      | Not-significant            | Not-significant      |
|                              | 2.39                       | 1.22                 | 2.13                       | 2.63                 | 4.64                       | 4.39                 |
|                              | 0.180                      | 0.43                 | 0.46                       | 0.15                 | 0.058                      | 0.06                 |
| Final model                  | Quadratic                  | Quadratic            | Quadratic                  | Quadratic            | Quadratic                  | Quadratic            |

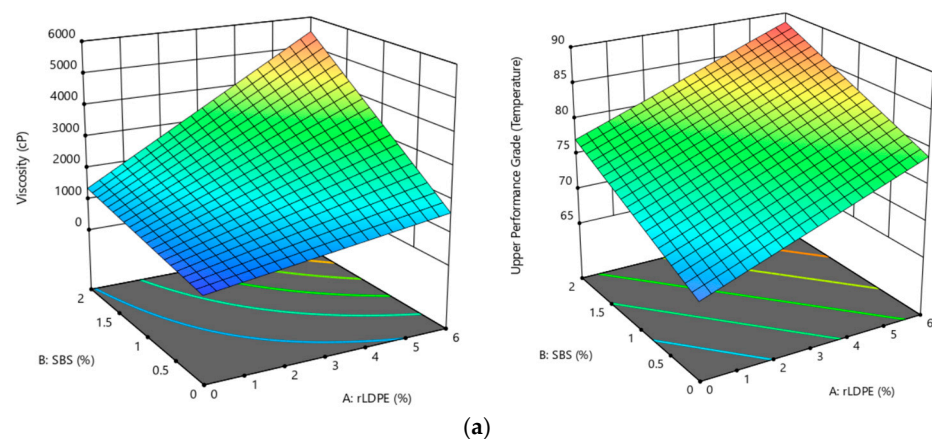


**Figure 14.** Cont.

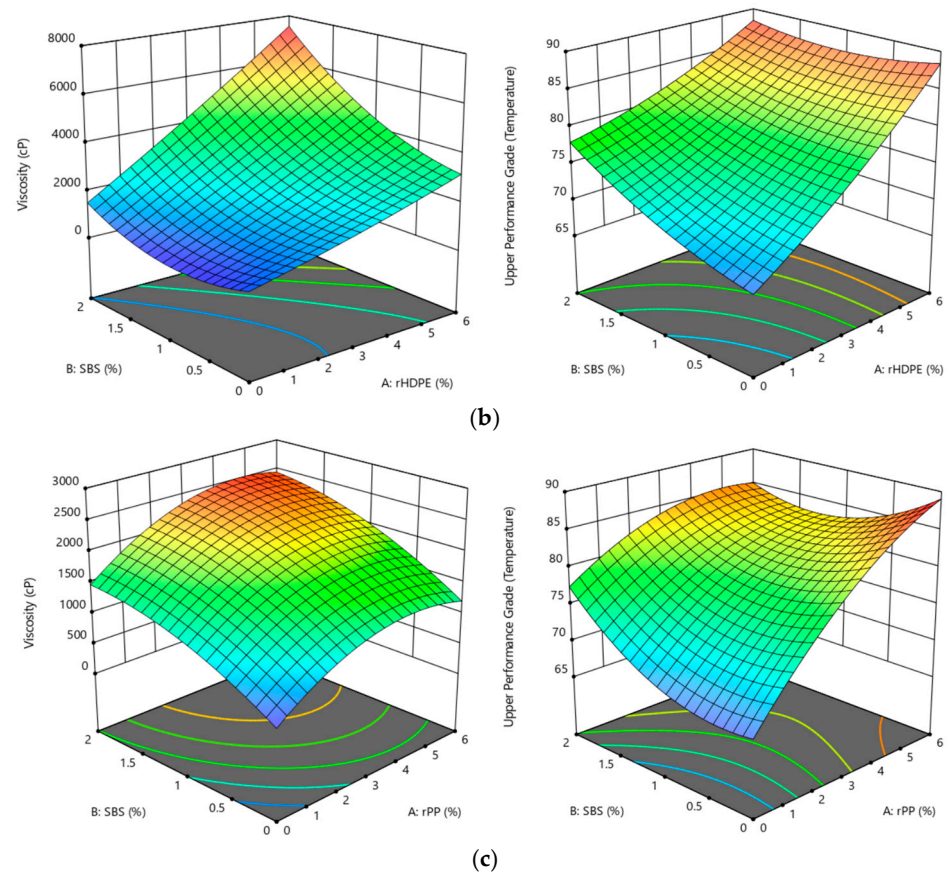


Figure 14. Upper performance grade using "S" for (a) "L", (b) "H", (c) "P".

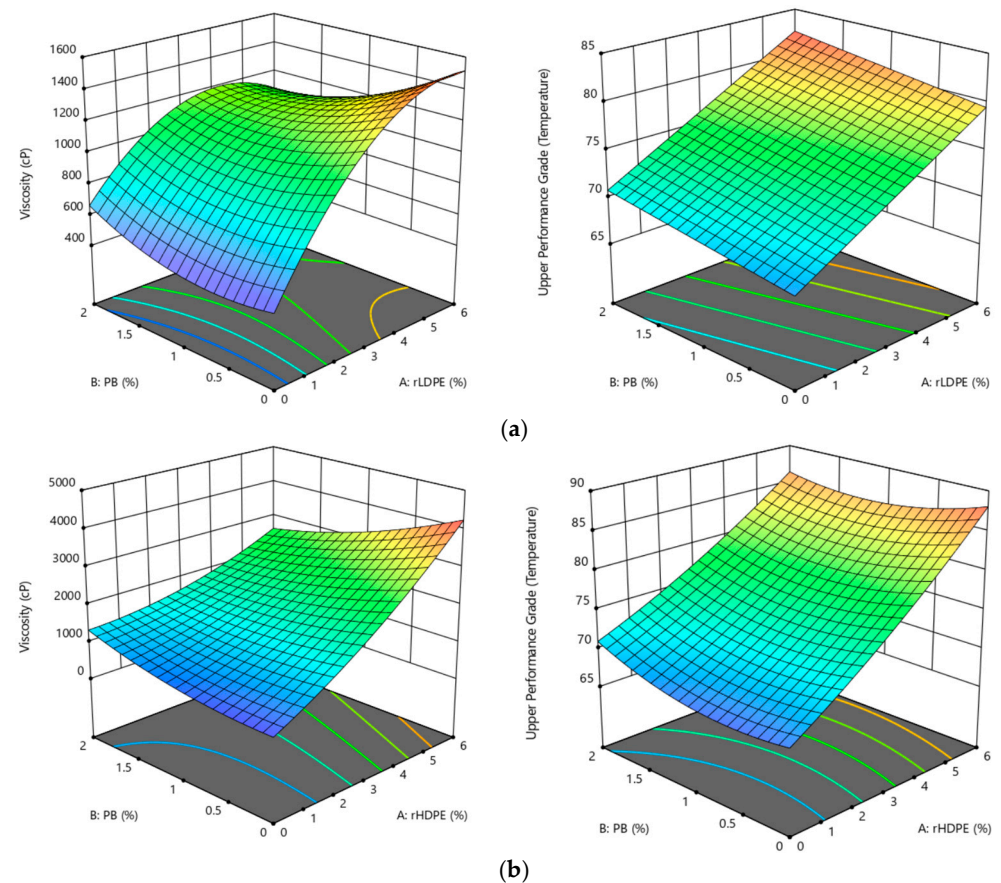


Figure 15. Cont.

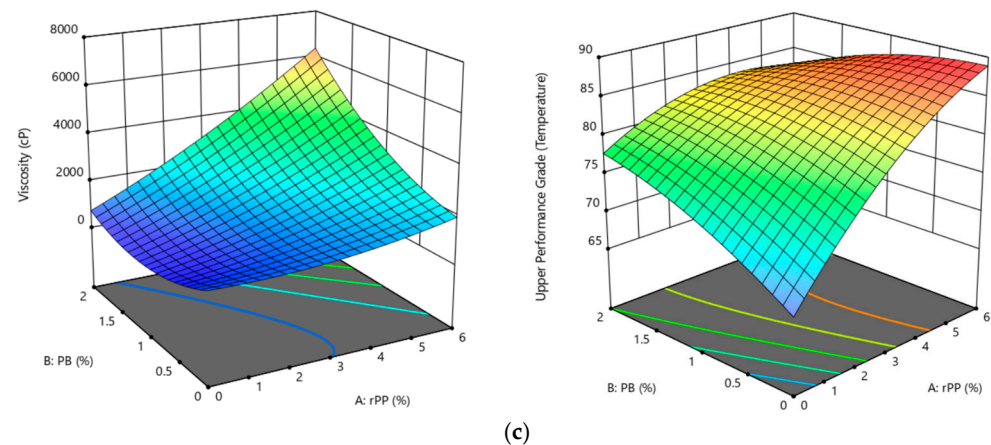


**Figure 15.** Upper performance grade using “B” for (a) “L”, (b) “H”, (c) “P”.

## 6. Conclusions

The influence of recycled plastic waste and commercial polymers was investigated for use in binder modification. The binders’ performance characteristics, specifically viscosity and dynamic shear rheometer analysis for the upper performance grade, were evaluated. The following conclusions were derived:

The locally available fresh binder can survive pavement service temperatures up to 64 °C, which necessitates a modification of the binder for the upper performance grade in KSA from 76 °C to 82 °C. Hence, the modification of the base binder with commercial and RPW polymers up to PG 82 °C was the major objective achieved by this study.

With the addition of “L” from 2 to 6%, along with “S” from 0 to 2%, the viscosity values were enhanced quite significantly; this showed the increase in the molecular resistance to flow at max 6% “L” content by the creation of the network in between the modified binder. The “H” binder network was the most dominant, showing an increasing trend in viscosity compared to the “S” binder network. The binder network can be seen in the 0% PW and 4% “H” line. The addition of “B” enhanced the viscous behavior of the binder blend, as evident from the 0% RPW. The “B” blend with “P” from 2 to 6% showed continuous increment of viscosity.

The noteworthy improvement in the permanent deformation resistance is reliant on the “L” molecules’ thickening effect. The comparison between the 0% “S” and 1% “S” addition to “L” showed that the improvement in rutting is high at lower temperatures and the improvement is slight at higher temperatures.

The higher “L” with “S” percentages showed less viscoelastic change at high temperature in comparison to low “L” and “S” contents. Similarly, the “L” and “S” (6% and 1%) additions, respectively, can provide the required targeted PG temperature of 82 °C. The effect of “H” with “S” was significantly improved in terms of high-performance-grade temperature. “H” and “B” (6% and 2%), respectively, showed good improvement. The lower percentages of 2 and 4% PP displayed poor performance, and this interpreted the poor compatibility of “S” and PP. This performance was from the structural incompatibility of plastomer and elastomer, causing poor dispersion and reduced elastic network formation. In comparison, PP and PB, both of which are plastomers, showed better compatibility, which resulted in improved performance-grade values.

This study demonstrated the binder characteristics for viscosity and dynamic shear rheometer analysis under controlled conditions. The future study for long-term performance, which includes cyclic fatigue loading, rutting, and thermal and moisture conditions for environmental effects, needs to be assessed. Structural characterization incorporation will further strengthen the interpretation, providing molecular interaction, morphology,

and loss of mass behavior. These findings will be vital for mechanical performance, durability, and viability for the adoption of recycled plastic waste in pavement applications.

**Author Contributions:** Conceptualization, H.I.A.A.W., M.A.H. and W.R.; methodology, H.I.A.A.W. and M.A.H.; software, W.R.; validation, W.R., S.A. and S.S.; formal analysis, A.M.B.; investigation, S.S. and W.R.; resources, H.I.A.A.W. and S.S.; data curation, M.A.H. and A.M.B.; writing—original draft preparation, W.R. and M.A.H.; writing—review and editing, S.S., S.A. and A.M.B.; supervision, H.I.A.A.W.; project administration, H.I.A.A.W. All authors have read and agreed to the published version of the manuscript.

**Funding:** This research received no external funding.

**Data Availability Statement:** The original contributions presented in this study are included in the article. Further inquiries can be directed to the corresponding authors.

**Acknowledgments:** The authors would like to acknowledge the Department of Civil & Environmental Engineering, King Fahd University of Petroleum and Minerals, Saudi Arabia, for supporting this research. Authors acknowledge the lab in-charge of highway materials Mohammed Khaliludin for his continuous support in this research.

**Conflicts of 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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