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Research Article

Investigation of the rheological properties of low-density polyethylene (LDPE) modified bitumens using two plastic wastes

İremgül BEKTAŞ ATICI[1], Erkut YALÇIN[1,*], Mehmet YILMAZ[1]

[1]*Fırat University, Department of Civil Engineering, Elazığ, 23119, Türkiye*

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ABSTRACT

This study aimed to solve one of the most significant environmental problems and develop the performance properties of bitumen using low-cost materials by utilizing low-density polyethylene (LDPE), one of the plastic wastes. Rheological characteristics of the pure binders and the LDPE modified bitumen prepared using these binders were determined at various temperatures and frequencies. In the study, modified bitumens were prepared by adding 2 different LDPEs in 4 different ratios (1%, 2%, 3%, and 4%) to the binder. The dynamic shear rheometer tests were conducted to examine the rutting resistance parameters and phase angles to examine the elastic treatment of the binders and their performance at high temperatures. Moreover, the rheological behaviors of the bituminous binders under different conditions were evaluated by testing pure and modified bitumens at 4 different temperatures and 10 different frequencies. According to the results of the analyses, the increase in the temperature led to a logarithmic decline in the rutting resistance parameters. The binders' black diagrams revealed that the modified binders prepared using LDPE A demonstrated more elastic behavior compared to the modified binders prepared using LDPE B.

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INTRODUCTION

Globally, one of the most important environmental problems is the waste materials, which increase each passing day. As a waste material, plastic waste has become a crucial issue all over the world. Plastic packaging wastes account for the largest share of plastic waste [1]. A survey on plastic recycling conducted in Australia revealed that the total plastic consumption was 3.513.100 tons in 2016-2017, and only 415.200 tons were recycled in the same period [2]. In recent years, it was reported that the production and utilization of plastics have led to more than 30 million tons of plastic waste annually in recent years in China [3]. According to the estimations, 275 million tons

***Corresponding author.**
*E-mail address: erkutyalcin@firat.edu.tr

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of plastic waste were produced in 192 coastal countries in 2010. Moreover, between 4.8 and 12.7 million tons were estimated to contaminate the ocean [4]. Despite the development of systems to manage plastic waste in the European Union, most of these wastes (41%) are sent for incineration, and approximately 30% are recycled [5]. According to the data of Environmental Protection Agency (EPA) in USA, 35.370 tons of plastics were produced in the US in 2017, of which only 8.4% (2960 tons) were recycled, and 75.8% (26.820 tons) were buried in landfills. Accordingly, it was detected that the leachate samples collected from landfills contained microplastics between 10 and 100 mm, which indicated that plastic wastes threatened the environment and public health [6]. These results suggest that the landfill is not the final pit of plastics, but potential sources of microplastics that pose a threat to drinking water quality and other water resources. Celauro et al. investigated a specially designed polymeric compound to test its effects on a general asphalt mixture [7]. In the study, where various tests were utilized to investigate the effectiveness of the modification, the tests were conducted both in the laboratory and on a real pavement section of the Palermo International Airport in Sicily. The results of the tests indicated that a proper mixture design could lead to a reduction in permanent deformations, lower bitumen ratio, and more significant improvement in the modulus of the material, enabling more economic savings. Additionally, considering fatigue resistance parameters, the modified mixture with low bitumen content provided comparable performance parameters to the control mixture with higher bitumen content. The study suggested that the proposed implementation could improve pavement performance even when using a softer binder, which was convenient in Italy.

Plastic waste is one of the most crucial materials in the last few years [8, 9]. However, the rate of recycling plastic waste in the US is rather lower other countries, which report their recycling rates ranging from 30% to 60%. Of these, Japan has the greatest recycling rate with 78% [10]. Plastic waste mixtures are hard to recycle due to their complex structure and non-efficient mechanical recycling. Rather than exporting waste to developing countries, Australia has taken proactive steps to develop an alternative for using recycled plastics, thus making a significant contribution to waste generation [2]. Moreover, new alternatives were explored to use plastic wastes in the US. In China and India, importing plastic wastes was prohibited [11].

Decreasing the use of plastics may be the best way to decrease plastic waste directly. For instance, a perspective was suggested to move towards zero waste by banning single-use plastics [12]. However, it can be difficult to practice and implement this ban. Therefore, other options should be sought to reduce plastic waste. Researchers and engineers have been investigating the production of wood-plastic composites [13], concrete blocks [14], and mortars [15, 16], which can be used in construction infrastructures by utilizing plastic wastes. Ramli and Tabassi reported that mortars with polymer modifications demonstrated improved engineering properties compared to traditional mortar mixtures [15]. Arulrajah et al. investigated the use of plastic granules with crushed brick and recycled asphalt pavement (RAP) wastes as the main filling material [17].

Utilizing waste materials in bituminous pavements is an environmentally friendly approach that has attracted great attention in recent years. Bitumen, which is the most frequently used binder in pavements, is obtained from petroleum, which is a non-renewable resource [9]. Researchers utilize various wastes, such as used engine and cooking oils, pig manure, and coffee grounds within the scope of environmental studies. Undoubtedly, this can also reduce the harmful environmental impacts of wastes as well as the consumption of raw materials. Additionally, researchers are hesitant to support the use of large amounts of recycled materials if the performances of the pavement infrastructure containing recycled materials are not as satisfactory as those without recycled materials. In the literature, several studies investigated the use of various polymer wastes in road construction [18-21]. However, there are gaps in the literature about fully comprehending performances of asphalt pavements containing various ratios and types of recycled plastics.

Low-density polyethylene (LDPE) is usually used in bitumen modification to improve rutting resistance at high temperatures and reduce temperature sensitivity significantly. The present study investigated the effect of using low-density polyethylene (LDPE) with two different physical contents in bitumen modification on the rheological characteristics of bituminous binders. Four different ratios (1%, 2%, 3%, and 4%) of two different LDPE binders were added to pure bitumen. Dynamic shear rheometer test was conducted on pure and modified bitumen. Reactive dyes are typically azo-based chromophores combined with different types of reactive groups. They are different from all other classes of dye because they are covalently bonded to textile fibers. [26,27] The use of reactive dyes over the last decade is resistant to biodegradation, ie, heat and light under aerobic conditions and biodegradable, which is usually due to poor biodegradation of such dyes (especially those containing azo groups). [28] A wide variety of treatment methods are used to remove dyestuffs from wastewater. [29]. Traditional treatment methods for waste water include biological, chemical and physical methods. [30-32].

MATERIAL AND METHODS

The Penetration Grade Bitumen 50/70 (B 50/70) acquired from TÜPRAŞ (Turkish Petroleum Refineries Corporation) Batman Refinery was utilized as the pure binder in the current study. The pure binder was modified with low-density polyethylene (LDPE) additives with two different physical properties. The LDPE used in the study was obtained from Adanus Plastik company. In the present study, target binders were prepared by adding four different ratios (1%, 2%, 3%, and 4%) of LDPE additives with two different physical properties to pure bitumen. The LDPE-added bitumen preparation process was conducted in the following order.

- First, the pure bitumen was heated in a drying oven at 180 ± 5°C for 30 minutes to make it viscous. Then, 500 grams of viscous bitumen was poured into the metal chamber of the mixer.
- To create a homogeneous heat source, the bitumen poured into the metal chamber was left inside the thermal jacket on the heating source conditioned at 180±5°C. Then, it was left on the heater until reaching the thermal equilibrium at 180±5°C. The LDPE was poured into hot bitumen at ratios determined by the weight of the bitumen. The LDPE-bitumen mixtures were prepared by mixing using a mechanical mixer at 1000 rpm for 60 minutes [22].

The abbreviations used in the study for the pure and binders containing LDPE are presented in Table 1. The physical properties of the LDPE additives are presented in Table 2.

The results of the penetration, softening point, and viscosity tests of pure and LDPE modified bitumen are presented in Table 3. The penetration values of the L1 and L2 modified bitumens were observed to demonstrate a polynomial decrease with the increase in the additive content. The penetration values of the L1 and L2 modified binders were observed to be close to each other. However, the L1 modified bitumen demonstrated a lower penetration value compared to the L2 modified bitumen. Moreover, the penetration values of both LDPE additives were observed to decrease with the increase in the additive content. The LDPE modification was determined to have a significant impact on the softening point. The softening points were determined to increase steadily with the increases in the LDPE content. Both the L1 and L2 modified binders with 1% and 2% additive content demonstrated similar softening points. However, the L1 and L2 modifications with more than 2% additive content demonstrated different softening points. Both additives can contribute to the high-temperature resistance of hot mix asphalt. The viscosity values were determined to increase significantly with the increases in the L1 and L2 contents. Moreover, no processability issue was observed even with the highest LDPE content. L1 modified binders were determined to have higher viscosity values with all additive contents compared to L2 modified binders. The viscosity values were determined to increase with the increase in the additive content.

DYNAMİC SHEAR RHEOMETER TEST

The dynamic shear rheometer (DSR) test method is used to characterize the viscous and elastic treatment of bituminous binders at medium and high temperatures. DSR test is conducted to examine the complex shear modulus (G*) and phase angle (δ) values of the binders. The complex shear modulus represents the total deformation resistance of the binder during a given period of torsion while the phase angle indicates the delay between the implemented shear strain and the resulting shearing deformation. A larger phase angle indicates a more viscous bituminous binder. For the original non-aged binders, the permanent deformation is controlled by limiting the rutting resistance parameter (G*/sinδ) to a value greater than 1.0 kPa [23]. The test was conducted with a strain control on non-aged pure and modified bitumens using Bohlin DSRII rheometer by ASTM D7175 standard. The test was conducted at a frequency of 1.59 Hz on a plate with a diameter of 25 mm and a plate span of 1 mm at the temperatures of 52°C, 58°C, 64°C, 70°C, 76°C, and 82°C. The rutting resistance parameters and phase angles were determined in the test to evaluate the binders' high-temperature performance and elastic properties.

The complex shear modulus includes the elastic and viscous components specified as storage (elastic) modulus (G') and loss (viscous) modulus (G"). These two components are related to the phase angle (δ). The phase angle, which is the delay between the strain and deformation in an oscillation test, is a parameter of the material behavior's viscoelastic equilibrium. The bituminous material can be

Table 2. Physical properties of the LDPE additives

Property	L1	L2
Specific gravity (g/cm^3)	0.913	0.916
Tensile strength (MPa)	20	15
Melting temperature (°C)	120	130
Impact strength (kJ/m^2)	5	5

Table 1. Abbreviations used for bituminous binders in the study

Name of Additive	Low-Density Polyethylene Additive Content (%)				
	0	1	2	3	4
Low-density polyethylene A (L1)	Pure (C)	L1-1	L1-2	L1-3	L1-4
Low-density polyethylene B (L2)		L2-1	L2-2	L2-3	L2-4

Table 3. Results of the conventional binder test of pure binder and LDPE added bitumen

Binder Type	Penetration (mm^{-1})	Softening point (°C)	Viscosity (°C)	
			135	165
C	61.8	53.3	737.5	225
L1-1	48.0	56.5	1000	250
L1-2	39.0	59.0	1375	325
L1-3	33.0	62.0	1945	425
L1-4	30.3	65.0	2763	550
L2-1	46.0	56.0	812.5	250
L2-2	36.0	58.2	1038	287.5
L2-3	30.0	59.8	1525	375
L2-4	27.3	60.7	2213	500

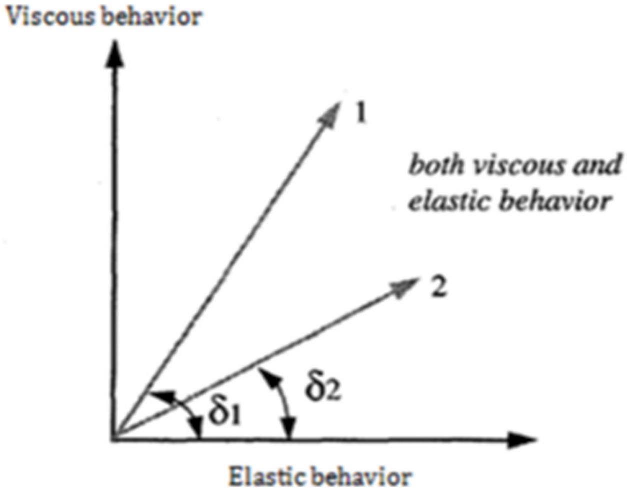

Figure 1. Demonstration of elastic and viscous behavior according to DSR test [24].

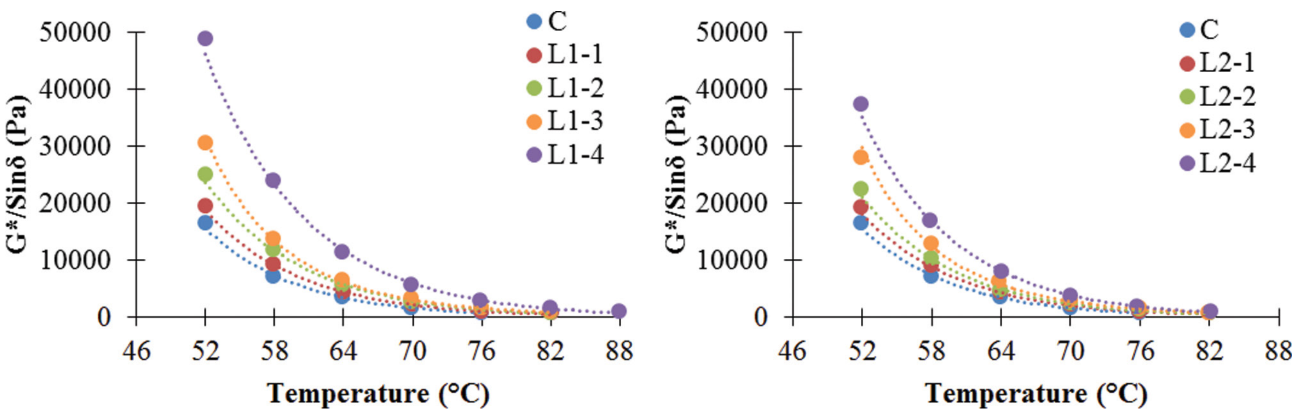

Figure 2. Change in the G*/sinδ values by the temperature.

considered completely viscous when δ=90 degrees. On the other hand, the phase angle of 0 degrees corresponds to the completely elastic material behavior. The material behavior between these two extreme points is regarded to be viscoelastic as combinations of viscous and elastic responses (Figure 1) [24]. In the present study, the specimen geometry was set with a diameter of 25 mm and a height of 1 mm to examine the viscoelastic properties of the binders. Then, the DSR test was conducted on the binders at 4 different temperatures (40°C, 50°C, 60°C, and 70°C) and 10 different frequencies (between 0.01 and 10 Hz).

RESULTS AND DISCUSSION

Complex Shear Modulus and Phase Angle

The test was conducted at a frequency of 1.59 Hz on a plate with a diameter of 25 mm and a plate span of 1 mm at the temperatures of 52°C, 58°C, 64°C, 70°C, 76°C, and 82°C. The rutting resistance parameters and phase angles were determined in the test to evaluate the binders' high-temperature performance and elastic properties.

Figure 2 demonstrates the change observed in the rutting resistance parameters of pure and modified binders due to the increase in temperature. The rutting resistance parameters demonstrate a logarithmic decrease with the increase in temperature. Similarly, this decrease is observed in all binder types. Each temperature increase of 6 degrees causes a decrease of about 50% in rutting resistance parameters. While the G*/sinδ value of the pure binder fell below 1000 Pa at 76°C, the binders of L1-1, L1-2, L1-3, and L1-4 demonstrated the G*/sinδ values of 1053 Pa, 1368 Pa, 1601 Pa, and 2837 Pa, respectively at 76°C. On the other hand, the G*/sinδ values of the binders of L2-1, L2-2, L2-3, and L2-4 at 76°C were determined to be 1015 Pa, 1143 Pa, 1457 Pa, and 1834 Pa, respectively. The test conducted for L1-4 at 82°C revealed that these binders could provide the limit value of 1000 Pa. The change in the rutting resistance parameter of the L1 binder by temperature demonstrates a

Table 4. Increases in the G*/sinδ values

Temperature (°C)	L1-1/Pure	L1-2/Pure	L1-3/Pure	L1-4/Pure	L2-1/Pure	L2-2/Pure	L2-3/Pure	L2-4/Pure
52	1.18	1.50	1.84	2.95	1.17	1.36	1.69	2.26
58	1.29	1.64	1.91	3.35	1.24	1.44	1.81	2.36
64	1.28	1,68	1.94	3.44	1.24	1.42	1.84	2.32
70	1.26	1.71	2.01	3.51	1.25	1.43	1.82	2.36
76	1.31	1.70	1.99	3.53	1.26	1.42	1.81	2.28

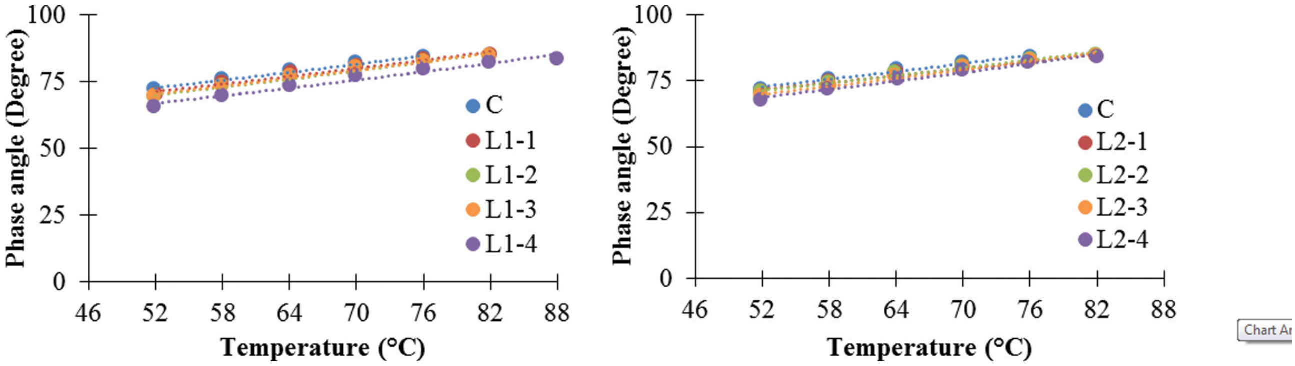

Figure 3. Change in the binders' phase angles by the temperature.

similar trend to that of the L2 binder. However, the G*/sinδ value of the L1 binder was observed to be higher than that of the L2 binder.

The index values of the modified binders were calculated by dividing their G*/sinδ values by the G*/sinδ values of the pure binder at the same temperatures. Table 4 presents the index values of the modified binders at 5 different temperatures. It was observed that 3% and 4% L1 additives to the binder increased the G*/sinδ values of the pure binder about 2 and 3.5 times, respectively, at each temperature. With the inclusion of both additives, the ratios of the increases were elevated as the temperatures increased. This indicated that the L1 modifications up to 4% additive had greater resistances to deformation at high temperatures. The L1 binders were determined to be more resistant to deformations at high temperatures compared to the L2 binders.

Figure 3 demonstrates the changes observed in the binders' phase angles by the temperature. Since the phase angles of all binders increased by the increase in temperature, the binders demonstrated a more vicious behavior. The changes in the phase angles of the L1 and L2 modifications were observed to be similar. The phase angle parameters of the binders with both L1 and L2 additives were observed to increase with the increase in the temperature. The L1 modification demonstrated the greatest change in the phase angle. Considering the values of the phase angles in Figure 3, the change in the L1 binder is observed to be more than that of the L2 binder. In the L2 binder, the increase in the additive is observed to lead to a similar increase in the parameter of the phase angle. In the L1 binders, on the other hand, the greatest change in the parameter of the phase angle is observed in the L1-4 type. The L2 modification was determined to have similar phase angle values with the pure binder. The phase angle of the pure binder at 52°C was determined to be 1.024 and 1.018 times higher than that of L1-1 and L2-1 binders, respectively. The phase angle of the pure binder at 52°C was determined to be 1.100 and 1.019 times higher than that of L1-4 and L2-4 binders, respectively, while this value at 76°C was determined to be 1.052 and 1.026 times higher than that of L1-1 and L2-1 binders, respectively.

Considering the phase angles, the L1 and L2 binders, which have lower phase angles compared to the pure binder, demonstrate a more elastic property. Evaluating the phase angles corresponding to the same rutting resistance parameter to examine the elastic characteristics of the binders seems to be more realistic in terms of comparing the elastic components. Figure 4 demonstrates the changes in the rutting resistance parameter corresponding to the phase angle of the L1 and L2 modifications for different temperatures.

Master Curve Results

In the study, the rheological behaviors of bituminous binders under different conditions were evaluated by conducting dynamic shear rheometer test on pure and modified bitumens at 40°C, 50°C, 60°C, and 70°C and 10

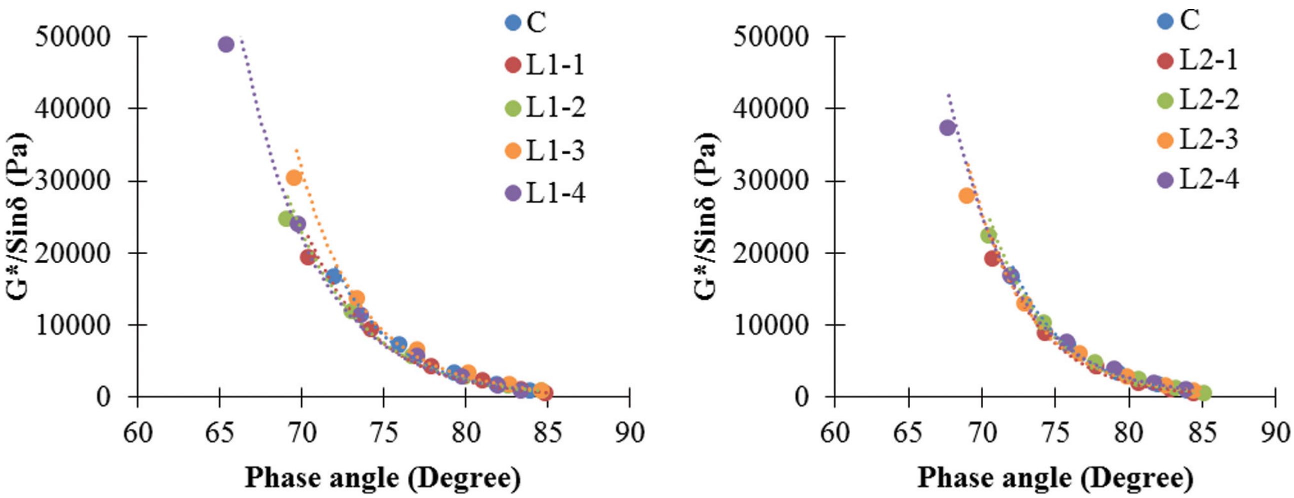

Figure 4. Changes in the rutting resistance parameter corresponding to the phase angle.

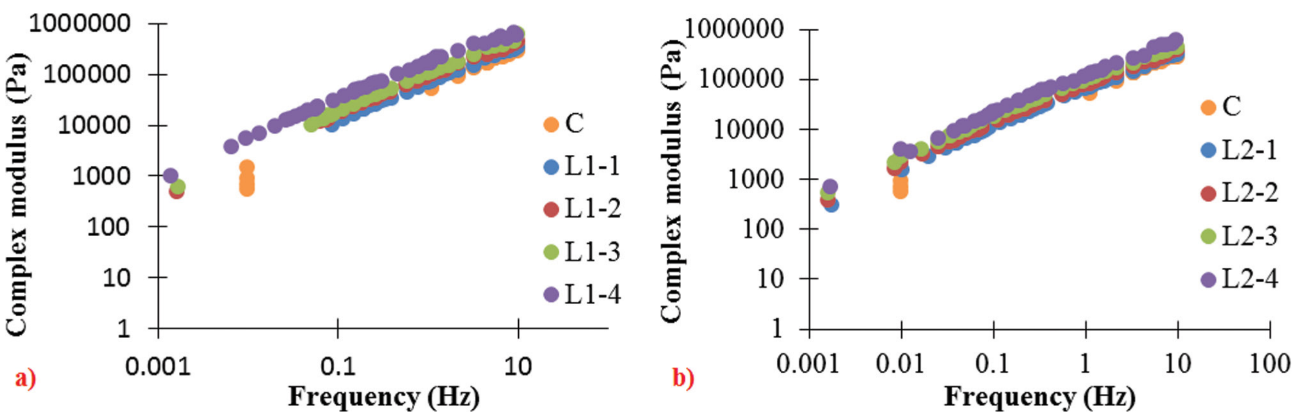

Figure 5. The change in the complex shear modulus values of binders prepared using L1 (a) L2 (b) modified bitumens by the frequency.

different frequencies in the frequency range between 0.01 and 10 Hz. The master curves were plotted by superposing the test results using the Malvern Bohlin DSR software. The master curves and black diagrams of the complex modules of modified bitumen prepared using two different LDPEs are demonstrated in Figure 5.

As seen in Figure 5, the values of the complex shear modulus increased with the increase in frequency (loading speed). The binders' complex modulus values were observed to increase with the increase in the content of the additive which is a waste material. While the effect of additives was more significant at low frequencies, the effect of LDPE content was reduced at high frequencies. Since both the horizontal axis and the vertical axis of the chart have a logarithmic scale, the values seem to be close to each other although the differences between the binders are distinct. Furthermore, the difference in complex modulus values became more significant with the increase in the LDPE content. The L1 additive caused greater change compared to the L2 additive in terms of the complex modulus values. In the figure, the complex modulus values increased with the increase in the additive content. The L1-4 and L2-4 binders were observed to demonstrate the highest increase. The complex modulus values of the modified binders containing L1 additive were determined to be lower than those of the modified binders containing L2 additive. The complex modulus values obtained from the master curves of the binders at the frequencies of 0.01, 0.1, 1.0, and 10 Hz are presented in Figure 5 to evaluate the complex modulus of the binders more easily.

Figure 6 demonstrates the black diagrams of pure and modified binders. The low values of the phase angle and high values of the complex shear modulus indicate that the binders demonstrate more elastic behavior. Figure 6 reveals that the value of the phase angle decreases with the increase in the complex modulus value of the pure bitumen.

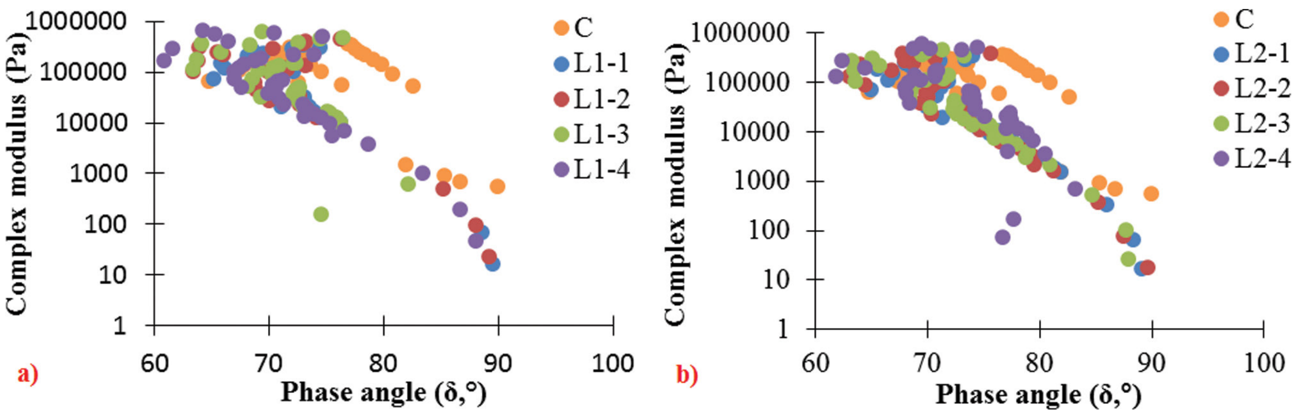

Figure 6. The complex shear modulus - phase angle relationship (black diagrams) of the binders prepared using L1 (a) and L2 (b) modified bitumens.

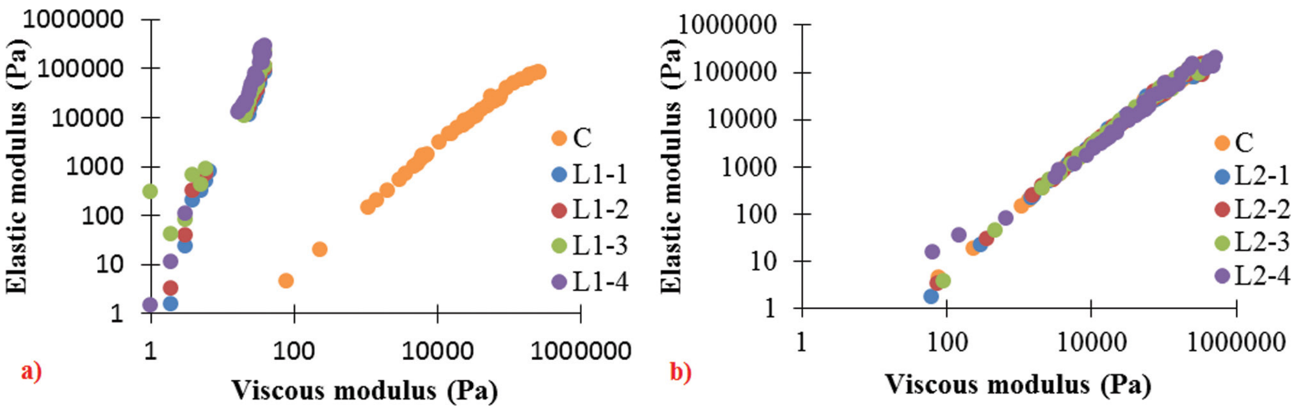

Figure 7. Elastic modulus - viscous modulus relationships of binders prepared using L1 (a) and L2 (b) modified.

Considering modified bitumen, the value of the phase angle reached the lowest level, particularly at the complex modulus values around 1.0E + 5 Pa, with the increase in the additive content; moreover, the value of the phase angle decreased steadily with the increase in the additive content. Both LDPE types were observed to demonstrate the lowest phase angle values with the 4% modified bitumen, and this value ranged between 60° and 65°. In modified bitumens, the decrease in the phase angle values up to the G* value of 1.0E+5 Pa and the increase after this value demonstrates that the boundary shear strain strength values of the modified bitumens prepared using both LDPEs are similar. While the phase angle value of the L1 binder was observed to decrease as low as 60°, the phase angle value of the L1-4 binder, which provided the best result, was around 60.92°. On the other hand, the L2-4 binder provided the lowest phase angle with a value of 61.91° in the L2 binders. This indicates that the binders obtained from the L1 modification have better elastic behavior compared to the binders obtained from the L2 modification in the modified bitumens.

Figure 7 demonstrates the change in the elastic modulus of binders by their viscous modulus (Cole-Cole diagrams). As can be seen in Figure 7, the slope of the Cole-Cole diagram of the modified bitumen prepared using L1 is steeper than that of the modified bitumen prepared using L2. While the L1 binder's viscous modulus value ranges between 1 Pa and 100 Pa, the L2 binder's viscous modulus value ranges between 100 Pa and 1000000 Pa. This demonstrates that modified bitumens prepared using L1 offer a more elastic behavior compared to modified bitumens prepared using L2. The diagrams of the binders containing L1 additive and those containing L2 additive were observed to be similar with the increase in the additive content. The L1 binder demonstrates a more elastic property compared to the pure binder.

RESULTS

In the present study, modified bitumens were prepared by adding 4 different ratios (1%, 2%, 3%, and 4%) of low-density polyethylene (LDPE) with two different physical components to Penetration Grade Bitumen 50/70 (B 50/70) acquired from TÜPRAŞ Batman Refinery. The rutting resistance parameters and phase angles were determined to evaluate the binders' high-temperature performance and elastic properties. Additionally, the rheological properties of the binders and the changes in their rheological properties under different conditions were evaluated by conducting dynamic shear rheometer tests on modified bitumens at 4 different temperatures and 10 different frequencies.

- The rutting resistance parameters demonstrate a logarithmic decrease with the increase in temperature. The change in the rutting resistance parameter of the L1 binder by temperature demonstrates a similar trend to that of the L2 binder. However, the G*/sinδ value of the L1 binder was observed to be higher than that of the L2 binder. The L1 modification demonstrated the greatest change in the phase angle. In the L2 binder, the increase in the additive is observed to lead to a similar increase in the value of the phase angle. In the L1 binders, on the other hand, the greatest change in the value of the phase angle is observed in the L1-4 type. The phase angle values of the L2 modification were determined to be similar to the phase angle values of the pure binder.
- According to the results of the tests, the binders prepared using L1 and L2 demonstrated different complex shear modulus and phase angle values. The binders prepared using L1 were determined to have higher complex modulus values. The pure binder demonstrated lower complex modulus values at all frequencies compared to binders prepared using L1. All binders demonstrated higher complex modulus values compared to the pure binder. Moreover, the complex modulus values were observed to increase with the increase in the additive content in both L1 and L2 binders. The binders' black diagrams revealed that modified binders prepared using L1 demonstrated more elastic behavior compared to modified binders prepared using L2.
- In L1 and L2 modified bitumens, the decrease in the phase angle values up to the G* value of 1.0E+5 Pa and the increase after this value demonstrates that the boundary shear strain strength values of the modified bitumens prepared using both LDPEs are similar. The phase angle values of the binders containing L1 were determined to be higher than those of the binders containing L2. This indicates that the binders containing L1 have a more elastic property compared to the binders containing L2.
- Today, the consistency of bituminous binders is determined by a static test at a single temperature for the penetration classification, while the rutting resistance and fatigue parameters of the binders are determined by conducting the DSR test at a single frequency in the Superpave method. As can be seen in the study, the bituminous binders may exhibit different rheological behaviors at different temperatures and different loading speeds. This is thought to be associated with the chemical structure of the LDPE additives. Therefore, it is thought that it would be more appropriate to conduct rheological tests in a wide range of parameters particularly in the evaluation of the viscosity and rheological behavior of the modified bitumens.

AUTHORSHIP CONTRIBUTIONS

Authors equally contributed to this work.

DATA AVAILABILITY STATEMENT

The authors confirm that the data that supports the findings of this study are available within the article. Raw data that support the finding of this study are available from the corresponding author, upon reasonable request.

CONFLICT OF INTEREST

The author declared no potential conflicts of interest with respect to the research, authorship, and/or publication of this article.

ETHICS

There are no ethical issues with the publication of this manuscript.

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