Sustainable Bituminous Pavement: A Study on Low-Density Polymer Modified Bituminous Binder

Vishnu Vijayan [,](http://orcid.org/0000-0003-2793-6127) Jeevan Mathew Tharayil [,](http://orcid.org/0000-0001-6113-1803) R. Rakhil Krishna, Jiji Saji, Divya S. Shaji, and G. Lakshmi

Abstract Plastics, especially low-density polyethylene (LDPE), is now being the most effective pollutant on Earth due to its non-degradability and increased demand. The reduced reusability and recyclability of these plastics makes more hurdle in proper disposal and treatment of these wastes. Thus, this problem can be managed only by implementing an alternative method of recycling these polymeric compounds for other beneficial purposes. The aim of this study is to suggest an ecofriendly approach of disposing these wastes by using it as bitumen modifiers. The effect of LDPE on bitumen has been analyzed by different wt% of LDPE from 2 to 8 wt% and was compared with pristine bitumen. Characterization tests mainly viscosity test, standard penetration and softening point, Fourier transform infrared spectroscopy, thermogravimetric analysis, and rheological analysis were done in order to examine the effect of LDPE addition to bitumen. The results show rise in thermal stability and deformation resistance for modified bitumen. However, a rise in polymer content above an optimum limit is not preferable. LDPE addition of 4% shows a better performance and is suggestable for employing in road sectors.

Keywords Low-density polyethylene · Modified bitumen · Eco-friendly · Road pavement

1 Introduction

Owing to the use in many aspects of our lives, plastics produce a significant quantity of waste on Earth such as packaging, building and manufacturing, automobile, electrical, and computer uses. Plastics are polymers derived primarily from fossil fuels by-products and made primarily by variations of carbon, hydrogen, and oxygen [\[1\]](#page-9-0). Though these possess extreme temperature tolerance, resistant to UV irradiation, and often non-biodegradable, these are able to continue polluting the whole Earth for centuries. Plastics appear to detach into smaller, meso-, and micro-structured

V. Vijayan (B) · J. M. Tharayil · R. R. Krishna · J. Saji · D. S. Shaji · G. Lakshmi Department of Civil Engineering, Baselios Mathews II College of Engineering, Kollam, Kerala, India

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nature that have unique and important effects on the environment that may have a detrimental impact on the health of humans and animals involved with the structure. Bioaccumulation of these to the food chain causes severe impacts on humans [\[2\]](#page-9-1). The asphalt industry is extensively involved in polymer improvements in bitumen to boost the properties of bentonite, impacting breaking, breakdown, and rolling pavements in thermal tolerance. Polymers are well known for their usage to significantly improve the performance of bitumen. Polymers are capable of increasing the elasticity and the stability, therewith increasing the durability, versatility, and deformation resistance for asphalt mixtures and reducing their temperature sensitivity [\[3\]](#page-9-2).

Polymer alteration of bitumen has been shown to enhance different properties of bitumen including high-temperature rigidity, moisture tolerance, fatigue life, and the ability to lower temperature stresses. Bitumen modifiers have three types of polymers, according to the chemistry and properties of their materials: plastomers, elastomers, and reactive polymers [\[4\]](#page-10-0). Each serves different functions like increase in stiffness can be attained by addition of plastomers, whereas fatigue properties can be raised using elastomers. Majority of plastomers effect the thermal properties of binder, especially reduces the thermal susceptibility of bitumen [\[5\]](#page-10-1). Plastomers not only just decreases the stiffness parameter but also provides deformation resistance to a greater extent. The major plastomers which we usually deal with are polyethylene and are waste materials after use. Polyethylene has many forms out of which high-density polyethylene (HDPE) and low-density polyethylene (LDPE) are more available. LDPE is commonly the non-reusable waste plastic which has a melting temperature of around 125 °C. For common bitumen binders, the temperature adopted is above 170 °C which makes LDPE to consider as bitumen modifier as its melting temperature is less than that of bitumen [\[6\]](#page-10-2).

Hinislioglua and Agar worked on various concentration of HDPE additions in asphalt concrete HDPE concentrations on four to eight wt. % in asphalt concrete 20. The temperature variations have been done at $145-160$ °C with mixing duration ranging from 5 to 30 min. The results show high-stability and high-marshal quotient, and out of those, the best results were obtained for $4\%/135$ °C/30 min composition. Moreover, this mix was also highly resistant to permanent deformation (rutting) [\[7\]](#page-10-3). Deformation resistance was also notable for this mixture. In a study done by Zoorab et al. [\[8\]](#page-10-3) has done incorporation of polypropylene and low-density polyethylene in bituminous concrete mixture this shows an increase in properties like durability and resistance to deformation. The incorporation of recycled plastic in pavement design will not only increase the basic pavement properties but also give economic as well as environmental advantages [\[8\]](#page-10-4). Nearly, every industry uses plastics and its use is very likely to grow as plastics technologies are developed, causing plastic waste to increase in turn. Because they are found to modify the ecosystem's functionality it is important that instead of being left freely in nature or landfilled, to take into account preventive or reduced plastic waste and to restore this ecologically hazardous waste. As the present waste management concentrates on 3R principle, more changes have to be introduced [\[9\]](#page-10-5). Novel strategies are to be established before these turns out to be nonmanageable crisis. Shredded plastic-reinforced hot mix asphalt is a research field for such a goal that is still in its early stages. It can be used as strengthening components

for pavement asphalt of concrete. In the past literatures, polyethylene waste was typically applied with dry process to the asphalt mixture or used as an aggregate for improving the HMA output in the asphalted mixture (aggregated substitution) [\[10\]](#page-10-6). The literatures has revealed an increase in permanent resistance to deformation, Marshall stability, stiffness, and tiredness in asphalt mixtures, while a reduction in moisture damage was also observed while using shredded plastics in pavements. The past works also highlights a reduction specific gravity for these asphalt mixtures.

Even though there are a few investigations that have been done on using polyethylene as binder modifiers, the incorporation of recycled low-density polyethylene into bitumen was less explored one. Thus, the aim of this work is to investigate the effect of recycled LDPE in modifying bitumen properties. LDPE with different wt% (2, 4, 6, and 8%) were studied, and physical, thermal, chemical, and rheological properties of every samples were analyzed and are compared with pristine hot mix asphalt.

2 Materials

2.1 Fabrication of Shredded Polyethene

The polyethene waste was collected from the dumping yard at Kollam, Kerala. The collected plastic waste includes, low-density and high-density polyethylene derivatives and a few of PET low-grade bottles. The initial steps were sorting of the plastics, cleaning, and then drying. The polyethene bags were shredded into flakes so as to get a size range of 2.36 to 4.75 mm by maintaining an aspect ratio of 2:1 shown in Fig. [1.](#page-2-0) The tensile strength analysis of the flakes has been done with 10 mm/min of elongation. The results obtained were on an average of 0.31KN, thereby providing an elongation rate of 25%.

Fig. 1 Shredded LDPE

2.2 Incorporation of Polyethylene to Bitumen

For preparing bitumen—polyethylene blend—the bitumen was initially heated to a temperature of 170 °C. Then, the shredded low-density polyethylene flakes were added to the bitumen, thereby obtaining bitumen polyethylene blend. The LDPE additive was incorporated to pristine bitumen at 2, 4, 6, and 8% weight ratios and is described in Table [1.](#page-3-0) Uniform polymer–bitumen blended mixture was obtained using shear mixer at a rate of 4000 rotations per minute. The mixing was done for an average of 2 h maintaining the temperature of 180 °C.

3 Methodology and Characterization

3.1 Viscosity

Rotational viscosity tests were conducted according to ASTM D-4402 at 135 °C. The tests were done for pristine as well as modified binders. Brookfield DV III rheometer was employed for testing.

3.2 Penetration and Softening Point

The softening point was measured using ring and ball analysis in accordance with ASTM D36. The penetration test was conducted by following ASTM D-5 [\[11\]](#page-10-7). Both tests were done for pristine and modified binders.

3.3 FTIR Analysis

FTIR tests were done for identifying the functional groups present in both pristine and modified bitumen. Fourier-transform infrared spectroscopy (FTIR in transmission mode, Cary 630, Agilent Technologies).

3.4 TGA Analysis

Thermogravimetric analysis (TGA) of nanoparticle was carried out under a constant flow of nitrogen at a heating rate of 10 °C/min on the Hitachi, STA7200 TGA instrument.

3.5 Rheological Analysis

The rheological tests were done using dynamic shears rheometer (DSR) (Physica MCR302, Anton Paar). Frequency sweep test was conducted for bitumen at linear viscoelastic region at 60 °C, and the frequency range adopted was $0.1-10$ Hz. For this, DSR plates with a radius of 12.5 mm having a thickness of 1 mm. The storage and loss modulus were obtained as output for respective frequency. The shear modulus and phase angle were calculated according to [\[12\]](#page-10-8)

$$
|G| = \sqrt{G_1^2 + G_2^2}
$$
 (1)

$$
\delta = \tan^{-1}(G_2/G_1) \tag{2}
$$

where G_1 is storage modulus; G_2 is loss modulus; *G* is complex shear modulus; δ is the phase angle.

4 Results and Discussion

4.1 Viscosity

Viscosity is considered to be a basic physical property of bitumen which gives the flow resistance of fluid. The rotational viscosity of bitumen and bitumen polyethylene blend has been tested for analyzing the resistance to shear stress. The results showing the effect of varying concentration of LDPE are given in Fig. [2,](#page-5-0) and from the results, it depicts that by increasing LDPE concentration, there shows a significant rise in viscosity in comparison with pristine bitumen [\[12\]](#page-10-8). At 130 \degree C, the viscosity was about 0.59 Pa.s for normal pristine bitumen. The variation in polyethylene content was done for $2\%, 4\%, 6\%,$ and 8 wt% which rises the viscosity to 0.75, 1.21, and 2.98. The obtained results are on agreement with previous literatures [\[13\]](#page-10-9). The rise in viscosity also results in rising the temperature stability of the bitumen polyethene blend. The enhancement in viscosity with rise in polyethene concentration is due to the presence of asphaltenes, which results in the formation of complex internal

Fig. 2 Viscosity of LDPE–bitumen samples

structure. But increased viscosity may effects workability of the mix for 8 wt% sample which has effects on mixing and workability.

4.2 Determination of Penetration and Softening Point

The results of penetration and softening point was according to the expectation that these properties get good results with addition of LDPE and are given in Figs. [3](#page-5-1) and [4.](#page-6-0) The penetration test depicts the flow and deformation properties of the LDPE– bitumen mix. The penetration decreased with rise in the polymer concentration. Pristine bitumen shows penetration value of 55.1 mm, while with rise in concentration of polymer to $2wt\%$, $4wt\%$, $6wt\%$, and $8 wt\%$, the penetration results were 52.3 mm,

Fig. 3 Penetration of LDPE–bitumen samples

Fig. 4 Softening point of LDPE–bitumen samples

44.7 mm, 33.2 mm, and 22.2 mm, respectively. The reduction in the penetration of the bitumen polymer blend shows the increased hardening of the bitumen with addition of polymer [\[14\]](#page-10-10). The effect of polymer in physical properties of bitumen is evident from reduction in penetration results. The reduced penetration results show the ability of modified bitumen to resist temperature effects and rutting [\[15\]](#page-10-11).

Rise in softening point values was observed with rise in polymer concentration. The softening point of the pristine bitumen was 51 °C which gets increased to 54, 76, 94, and 108 °C with 2, 4, 6, and 8 wt% of polyethene additions. The complexation in internal structure caused by addition of polymer results in the rise in softening point of modified mix. From the raised softening point is evident that the modified mix has better resistance to deformation at varying range of temperature. These blended mixtures will be strong enough to resist rutting [\[16\]](#page-10-12).

4.3 FTIR

The FTIR analysis of both pristine and modified asphalt samples was done and is given in Fig. [5.](#page-7-0) It is evident that both pristine and modified bitumen have major similar peaks with only few new peaks for modified bitumen. The peaks at pristine bitumen at 2918.3 cm^{-1} show the presence of CH stretching vibrations, thus confirming the presence of organic hydrocarbons in the bitumen. These peaks are also seen in polymer blended bitumen samples at 2916.21 cm−1. C=C attraction was found to be observed at 1598 cm⁻¹. The addition of LDPE is confirmed by the peaks in range of 1200–750 cm⁻¹. The SO₂ peaks at 1329 cm⁻¹ confirm the addition of polymer of bitumen matrix [\[17\]](#page-10-13).

Fig. 5 FTIR spectra of LDPE–bitumen samples

4.4 Rutting and Fatigue Properties

The rutting and fatigue properties were analyzed by conducting DSR test of the LDPE-added bitumen mixes. The test was done at a wide range of temperatures as 64 and 70 °C. The rutting was analyzed in reference to ASTM D2872. The result of DSR test is given in Table [2.](#page-8-0) The shear modulus (*G*) and phase angles were analyzed by the DSR tests. *G* indicates that the shear modulus depicts the stiffness of asphalt mix samples. The elasticity at varying temperature was determined as phase angles (δ) . As phase angles decreases, the elasticity of the bitumen mix rises which in turn increase the resulting rutting tolerance. From the results, the modified bitumen binders has reduced δ value for both aged and unaged RTFO samples [\[18\]](#page-10-14). Ideally, the bitumen possesses a specific stiffness value to relieve rutting. From Table [2,](#page-8-0) it is evident that a reduction in *G* value can be noted, thus causing reduction of rutting parameter $(G/\sin \delta)$. Referring the pavement criterions, AASHTO: MP1, the value of rutting parameter should be greater than 2.2 kPa; for aged RTFO sample, it must be 1 kPa. From the results, the values of the modified bitumen are favorable, and thus concludes its better tolerance against rutting. *G* Sin θ δ is considered as the fatigue parameter and must be less than 5000 kPa for PAV-pressurized aged vessel bitumen samples [\[19\]](#page-10-15). The results are under the higher limit and fatigue prominent at temperature less than 20 °C, thus concluding that the polymer bitumen mixture has enough rutting and fatigue resistance.

4.5 TGA

Thermal gravimetric analysis was done to test the thermal stability of the prepared samples. The decomposition of the samples with respect to temperature was identified

Bitumen	Temperature	Parameter	Pristine bitumen	2% (w/w) LDPE	4% (w/w) LDPE	6% (w/w) LDPE	8% (w/w) LDPE
Original (Unaged)	64° C	G^* (kPa)	1.4195	1.3564	1.1969	1.1521	1.1103
		δ (\circ)	88.07	86.94	86.18	85.16	84.58
		G^* /sin δ (kPa)	1.4203	1.3583	1.1996	1.0132	0.8331
	70 °C	G^* (kPa)	0.6421	0.62465	0.57001	0.53112	0.51231
		δ (\circ)	88.73	87.99	87.33	86.45	85.88
		G^* /sin δ (kPa)	0.6394	0.6249	0.5821	0.5433	0.5216
RTFOT residue	64 °C	G^* (kPa)	3.0898	3.0333	3.0292	3.0272	3.0188
		δ (\circ)	86.24	85.79	85.36	84.88	84.27
		G^* /sin δ (kPa)	3.0965	3.0395	3.0292	3.0181	3.0012
	70 °C	G^* (kPa)	1.5342	1.3664	1.3499	1.3488	1.3411
		δ (\circ)	87.58	87.19	86.88	86.55	86.32
		G^* /sin δ (kPa)	1.5692	1.3612	1.3558	1.3496	1.3443

Table 2 Dynamic shear test rheometer result

by TGA analysis. The respective weight loss at varying temperature is provided with TGA curve in Fig. [6.](#page-8-1) From the results, it is seen that the weight loss was evidently high during a temperature range of 380–500 °C [\[20\]](#page-10-16). The loss in weight at this temperature

range was because of decomposition of bituminous compounds in mixture. These bituminous compounds include the aromatics and saturates in asphalt [\[21\]](#page-11-0). The pristine bitumen shows a higher decomposition than modified samples. The reduced decomposition of modified samples is due to its reduced evaporation, and thus makes it clear that these modified samples have a greater stability over varying temperature ranges.

5 Conclusion

The performance of the pristine asphalt has been intensified by the polyethene addition. The reusing of polyethene bags as an asphalt additive was done in this work which in turn reduces the plastic contamination of nature. The characteristic analysis shows that there is a notable enhancement in the overall properties of the pristine bitumen. As expected, the physical properties like viscosity and softening point get raised with increase in concentration of polymer. The penetration and softening point results give emphasis that the stiffness gets raised with rise in polymer concentration. The chemical composition was analyzed using FTIR, and it seems not to have drastic changes in absorbance peaks. The temperature stability was in higher side for modified bitumen mixes. From the DSR test, the rise in fatigue and rutting resistance was confirmed as these modified binders have much more rutting and fatigue resistance than pristine bitumen. Discussing about the limitation, the viscosity seems to affect the workability at higher concentration of polymer. A drastic rise in viscosity was observed for LDPE 3 and LDPE 4 mixes which are of polymer 6 and 8 wt%; this heavy rise in viscosity will curb the workability of the mixture. Thus, an additive concentration of 4% can be suggested so as to get an optimum workability along with increased overall performance. As a conclusion, it can be summarized that incorporation of additive gives favorable effects on rheological as well as overall properties of bitumen. Thus, using polyethene waste as bitumen modifier has a great potential in road pavements as well as for a clean environment.

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