

Use of maleic anhydride grafted recycled polyethylene treated by irradiation in bitumen modification

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ABSTRACT

Most of the polymers currently used as additive in bitumen are not economically attractive. Thus, employing recycled materials in flexible highway instead of virgin ones has become a fundamental case for researchers especially in terms of cost efficiency and environmental awareness. To date, many studies have been made to investigate using of recycled polymers in bitumen and generally improved mechanical properties of bitumen were determined.

This paper aims to present the modification of bitumen by maleic anhydride (MAH) grafted recycled low density polyethylene treated by irradiation (g-LDPEr-g-MA). Maleic anhydride was dissolved and mixed with recycled polyethylene which was proved by the Fourier infrared (FT-IR) spectrum. Subsequently, pre-treated LDPEr granules were subjected to gamma irradiation doze of 10kGy to provide formation of free radicals and some functional groups which can contribute to creation of strong chemical bonds between polymer modifier and bitumen. Six different binders (one base and five modified) were prepared to investigate the effects of g-LDPEr-g-MA modifier on properties of bitumen.

The dispersion of g-LDPEr-g-MA modifier in bitumen was examined by florescent microscope. Conventional tests and rotational viscosity test was used to determine physical properties of bitumens. Complex shear modulus (G^) and phase angle (δ) of base and modified bitumens were measured by means of dynamic shear rheometer (DSR) for different traffic speed and in-service pavement temperature.*

Tests results reveal that a gradually increment in g-LDPEr-g-MA modification leads to a decrease in penetration and an increase in softening point. According to DSR test results, g-LDPEr-g-MA modifier provides an increased rutting parameter. Thus, flexible pavement could become more resistant against permeant deformations by g-LDPEr-g-MA modification.

Keywords: Chemical properties, Complex Modulus, Modified Binders, Polymers, Rheology

1. INTRODUCTION

Polymer modification of bitumen is a widespread phenomenon around the world mainly on the purpose of acquiring better performance properties by decreasing temperature dependency of bitumen which leads to diminish permeant deformation such as rutting [1]. Styrene butadiene styrene (SBS), ethylene vinyl acetate (EVA) are the main polymers using as modifier in bitumen satisfactorily [2,3]. In addition to SBS (thermoplastic elastomer) and EVA (plastomer), polyolefins such as polypropylene (PP), high density polyethylene (HDPE) and low density polyethylene (LDPE), have been used as a modifier which generally provide enhanced mechanic properties of bitumen [4,5,6].

Despite presence of polymers using for bitumen modification, employing waste polymers has been gained great importance with an aim of decreasing environmental pollution and reducing manufacturing cost of flexible pavement [7,8,9]. In respect to this, it was aimed to use recycled low density polyethylene (LDPE_r) as modifier in bitumen in this study. However, normally there is no interaction between bitumen and recycled materials which leads to separation after modification. Hence, LDPE_r was grafted with maleic anhydrite (LDPE_r-g-MA) that is known as compatible with bitumen and with this it was exposed to gamma irradiation which can enable strong polymer/bitumen bonding by forming free radicals on surface of the recycled polymer particles [10]. Gamma irradiated and maleic anhydrite (MAH) grafted LDPE_r (γ -LDPE_r-g-MA) polymer was subjected to Fourier transfer spectroscopy (FTIR) to examine MAH grafting process. Bitumen having penetration grade of 160/220 was selected as binder and, 6 samples (one base and five modified) having different type of γ -LDPE_r-g-MA modifier were prepared with this binder. Base and modified bitumens undergo a series of tests to investigate morphological and physical properties of the binders. The dispersion of γ -LDPE_r-g-MA modifier in bitumen was observed by florescent microscope. Conventional test and rotational viscosity (RV) was employed to examine stiffness and consistency of binders. Dynamic shear rheometer (DSR) was employed to determinate high temperature performance grade (PG) of base and modified binder by determining fundamental parameters such as complex shear modulus (G^*) and phase angle (δ) while bending beam rheometer (BBR) was used to determinate low temperature performance grade (PG) of bitumen.

2. EXPERIMENTAL

2.1 Materials

The base bitumen having a penetration of 160/220 was employed as binder throughout the test program. The recycled LDPE polymers were obtained from Sicily, Italy. The LDPE_r materials were made from greenhouse films which are exposed to U.V. from sunlight and environmental factors such as wind, rainfall, dust etc. as well. In manufacturing process, the waste films were washed, dried and cut to pieces by an industrial scale and finally extruded. The chemical composition of the LDPE_r recycled LDPE used was $T_m = 109^\circ\text{C}$, $E = 180\text{MPa}$, $TS = 16\text{MPa}$, $EB = 500\%$, $\text{MFI } 190/2.16 = 0,29 \text{ g/10 min}$ and $\text{MFI } 230/2.16 = 0,95 \text{ g/10 min}$.

2.3 Maleic Anhydrite grafting of LDPE_r

The grafting reaction was performed using single-screw extruder having a diameter 12 cm and a length of 205 cm. The dicumyl peroxide (DPO) as initiator and maleic anhydrite (MAH) (chemical formula was given in Figure 1) were dissolved in acetone and mixed with LDPE granules. After volatilizing the acetone, MAH and DPO adhered onto the granules homogeneously. The temperature profile of the extruder from the feed zone to the die varied at 140/150/160 °C, and the rotation speed of the screws was constant at 60 rpm. The extrudate was obtained in the form of a strip, which was pelletized.

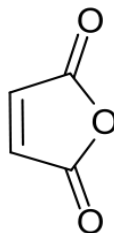


Figure 1: Chemical formula of maleic anhydride

2.4 Gamma-irradiation on grafted LDPE_r

The DPO and MAH were dissolved in acetone and mixed with LDPE_r granules. After volatilizing the acetone, MAH and DPO adhered onto the granules homogeneously. Then pre-treated LDPE_r granules were subjected to gamma ray irradiation from ^{60}Co for 28 min. The doze of γ -irradiation used was 10 kGy.

2.2 Preparation of modified bitumen samples

High shear mixer (Silverson L5M) was employed for mixing of γ -LDPE_r-g-MA polymers in bitumen. Before mixing process, pellet shaped γ -LDPE_r-g-MA were milled and crumbled polymer particles sieved with 0.6 mm sieve in order to provide a homogenous mixture. Bitumen of 160/220 was heated for 90 minutes at 163 °C, and then poured into the flask of the high shear mixer adjusted to 1000 rpm. 1, 3, 5, 7 and 9 % of total weight of the binder were used as modified content for the samples. γ -LDPE_r-g-MA polymer was added to bitumen in duration of 15 minutes and subsequently the mixing rate was adjusted to 4000 rpm for 45 minutes.

The binders were coded as follows:

base bitumen – “B”;

base bitumen + 1% γ -LDPE_r-g-MA – “B-1-P”;

base bitumen + 3% γ -LDPE_r-g-MA – “B-3-P”;

base bitumen + 5% γ -LDPE_r-g-MA – “B-5-P”;

base bitumen + 7% γ -LDPE_r-g-MA – “B-7-P”;

base bitumen + 9% γ -LDPE_r-g-MA – “B-9-P”;

3. TESTING PROGRAM

3.1 Fourier transform infrared (FT-IR) spectroscopy

The MAH content was determined by Fourier transform infrared (FTIR) spectroscopy. The samples were prepared using a Carver laboratory press at 180°C with 10 tons force between alumina sheets to form films of 80-100 μ m. The spectrum was recorded in the region of 2000-600 cm^{-1} . Grafting degree of MAH on LDPE_r was determined in comparison with a peak responsible for asymmetric and symmetric stretching vibrations of a C=O in MAH at a wave number 1853, 1780 cm^{-1} in comparison to methylene group C-H scissoring vibrations in LDPE_r at 1371 cm^{-1} , which was used as an internal reference for the normalization of spectra [11]. The incorporated MAH content was then calculated from the integrated absorbances as follows [12]:

$\% \text{MAH} = 0,032 + 0,411 \left(\frac{A_{1785} + A_{1710}}{A_{1371}} \right)$ in case of LDPE_r-g-MA

$\% \text{MAH} = 0,032 + 0,411 \left(\frac{A_{1853} + A_{1708}}{A_{1371}} \right)$ in case of γ -LDPE_r-g-MA

where A_{λ} is the absorbance at λ cm^{-1} measured above a baseline drawn to the valley between absorptions attributable to carbonyls of grafted maleic acid (1710 cm^{-1}) and MAH (1784 cm^{-1} and 1853 cm^{-1} , symmetric and antisymmetric C-O stretching, respectively).

3.2 Morphology

Observing the dispersions of the polymers within the bitumen is fundamental to estimate behavior of the modified bitumen in applications [13]. To this end, fluorescent microscopy was used. Florescent microscopy allows studying the morphology of bitumen by using a principle in which polymers become swollen after absorbing some of the constituents of the original bitumen. The method of sample preparation for fluorescent microscopy followed the regular procedure consisted of a heating and cooling process. The samples were examined under a Carl Zeiss Primo Star generated from a 40 W halogen lamp and able to magnify up to 1000X. Images taken with three different magnifications by means of different objective lenses were evaluated.

3.3 Conventional tests

Physical effects of polymer modifier on bitumen were investigated by means of conventional tests, such as penetration test (ASTM D5), softening point test (ASTM D36), ductility test (ASTM D 113) [14, 15, 16]. In addition to this, penetration index of binders were calculated by using penetration and softening point test results which is an indicator of the temperature susceptibility of bitumen.

3.4 Rotational viscosity (RV) test

Viscosities of base and γ -LDPE_r-g-MA modified bitumens were determined by a Brookfield viscometer DVRV-II Pro Extra in accordance with ASTM D4402 [17]. RV tests were conducted at 135 °C and 165 °C and the results were depicted in a typical log-viscosity vs. temperature graph which also enabled to calculate mixing and compacting temperature of base and

modified hot asphalt mixtures. Hence, it can be investigated whether binders fluid sufficiently in mixing and compacting process.

3.5 Dynamic shear rheometer test

The dynamic shear rheometer (DSR) was employ to determine high performance grade of binder. DSR test were done by Anton Paar Smartpave plus device on both original and RTFOT aged samples by using 25 mm spindle and 1 mm gap between plates as testing geometry in accordance with specification D7175 [18]. Before measuring fundamental rheological parameters namely, complex shear modulus (G^*) and phase angle (δ), viscoelastic range (LVE) of the samples was determined in order to remain in this range in DSR PG test. Accordingly, the strain controlled test was fixed to 10% of deformation for all samples. Rutting parameter ($G^*/\sin \delta$) is the main parameter for determination of high temperature grade which is expected to be above the 1.0 kPa for original samples and, 2.2 kPa for RTFOT aged samples.

3.6 Bending beam rheometer test

Low temperature performance grades (PG) of binders were determined by means of bending beam rheometer (BBR), Applied Test Systems (ATS) device. In BBR test, long term aged rectangular beam sample having a long of 125 mm, a wide of 12.5 mm and a thick of 6.25 mm was applied a constant load of 980 mN, and creep stiffness (S) and creep rate (m -value) of the binders were measured at the time of loading ranging from 8 to 240 s in accordance with ASTM D6648-01 [19]. According to SUPERPAVE specification creep stiffness (S) must be less than 300 MPa and the m -value must be greater than 0.3 at the test temperature of -6, -12 and -18 °C which are equivalent PG X-16, PG X-22, PG X-28 respectively in low temperature performance grade of binders.

4. TESTING PROGRAM

4.1 Fourier transform infrared (FT-IR) spectroscopy

In Figure 2 the normalized FTIR spectra of initial $LDPE_r$, $LDPE_r$ -g-MA and γ - $LDPE_r$ -g-MA were depicted. The spectra in Fig. 2 evidence of grafting of MAH onto $LDPE_r$. It can be seen the two new bands at 1780 cm^{-1} and 1708 cm^{-1} appear in spectrum of $LDPE_r$ -g-MA. As for γ - $LDPE_r$ -g-MA, the two new bands at 1853 cm^{-1} and 1710 cm^{-1} appear. This facts evidence that grafted MAH exists in an anhydride and acid forms both in $LDPE_r$ -g-MA and γ - $LDPE_r$ -g-MA For $LDPE_r$ -g-MA grafted MAH content was 1.15 wt.% and for γ - $LDPE_r$ -g-MA content of grafted MAH was equal to 0.25 wt.%.

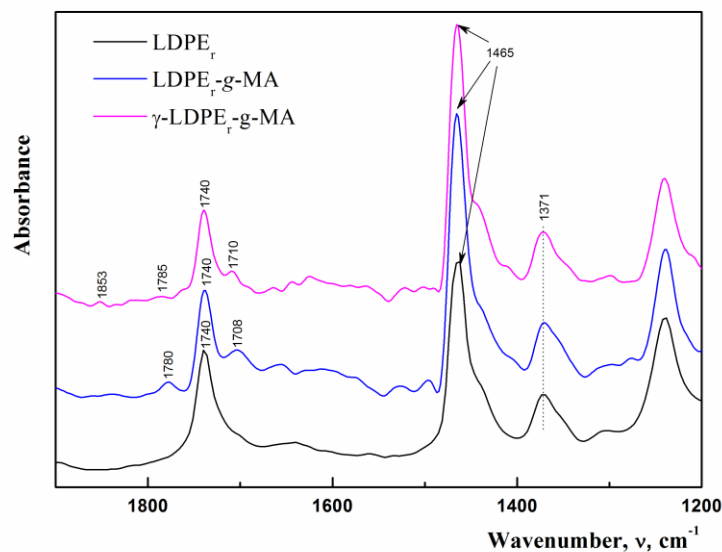


Figure 2: Fourier-transmission infrared spectroscopy (FTIR) of $LDPE_r$, $LDPE_r$ -g-MA and γ - $LDPE_r$ -g-MA

4.2 Morphology results

Florescent microscope images of base and B-9-P modified bitumen obtained at a magnification of 200X were given in Figure 3.a and 3.b, respectively. In Figure 3.a, only bitumen phase can be seen as brown color since there is no polymer modification whereas bitumen phase (brown) and polymers (yellow) appeared in Figure 3.b where polymers mostly dispersed in bitumen phase. It can be said that, polymers remains in bitumen as large and small particles around 5 to 50 microns. There is no dominant polymer phase appeared in bitumen even modification level reaches to 9% whereas other type of polymer modifications such as SBS modified bitumens where polymer phase becomes dominant over bitumen phase at a modification level of 5% [20]. Hence, it is more likely that the polymer particles present in bitumen as molten and mixed with bitumen and behave as only one mixed phase rather than two distinct phases work separately.

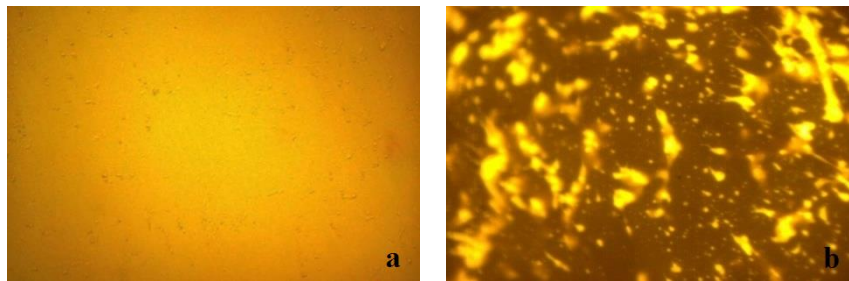


Figure 3: Florescent microscope images of base (a) and B-9-P modified bitumen (b)

4.3 Conventional test results

The results of conventional tests namely, penetration, softening point and ductility on binders were given in Table 1. There is an increasing trend in penetration whereas softening point values decreases with increasing γ -LDPE_r-g-MA modifier content. The increment in penetration is more significant at high modification level (7-9%) while the binder having 1% modifier has slightly lower than that of base bitumen. Penetration and softening point results indicates that bitumen is stiffer after γ -LDPE_r-g-MA modification at middle temperature ranges. According to penetration index (PI) values, temperature susceptibility of binders decreases as higher PI values obtained after modification. This result indicates that modified binder can be applied a wider temperature range than base bitumen. Nevertheless, after performance tests, namely DSR and BBR, suitable temperature range for binders can be determined in more detailed. Increasing modifier content was reduced to ductility which means elongation ability without corruption. Hence, it can be said that γ -LDPE_r-g-MA polymer modifier has a slightly adverse effect on elongation ability of binders.

Table 1: Conventional test results of base and γ -LDPE_r-g-MA modified bitumens

Properties	Binder type					
	Base	Modified bitumens				
		B-1-P	B-3-P	B-5-P	B-7-P	B-9-P
Penetration (25 °C; 0.1mm)	195.5	186	138	102	49	36
Softening point (°C)	38.7	40.2	43.5	50.7	74.3	101.5
Ductility (cm) at 25 °C	-0.73	-0.28	-0.21	0.94	3.45	5.96
Penetration index (PI)	103	95	93	90	87	84

4.4 Rotational viscosity test results

Viscosity of base and γ -LDPE_r-g-MA modified binders at 135 °C and 165 °C were depicted in Figure 4. There is a gradual increase in viscosity as modifier content increases which signifies that modifier has also stiffening effect at high temperature levels. It can be noted that, increment in viscosity was not very significant modification level of 1% while it was very drastic at level of 9%. The increment rate in viscosity is very similar at 135 °C and 165 °C that indicates temperature susceptibility is low at even elevated temperatures. Mixing and compacting temperature ranges, another important criteria for binders calculated from the equations, seem to be slightly increased at low and medium modification levels (1-5%). However, at high modification level of 9%, mixing and compacting temperature ranges rise to 199-205 °C and 186-191°C respectively from 135-142 and 122-128 °C (mixing and compacting temperature range calculated for original binders).

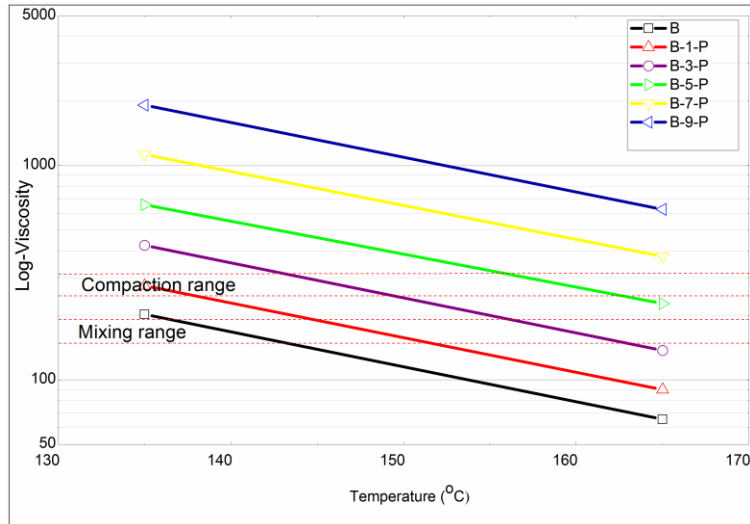


Figure 4: Rotational viscosity test results of base and γ -LDPE_{r-g}-MA modified bitumens

4.5 Dynamic shear rheometer test results

High temperature PG of base and modified bitumen which were determined after DSR tests were given in Table 2. Parameters such as G^* , δ and $G^*/\sin \delta$ belong to original and aged binders were also included in this table. As can be seen, PG of base bitumen was determined as PG 52-Y as neither original nor aged base binders provide a $G^*/\sin \delta$ above the limitations. As for γ -LDPE_{r-g}-MA modified bitumens, it should be noted that there is a remarkable increase in high temperature PG as modifier content increases. Moreover, binder having 9% polymers has reached maximum high temperature grade of PG 82-Y. At lower modification degree, (1-3%), there is still enhancement in high temperature grades as these binders have one level above PG (PG 58-Y) than base bitumen. The increment determined in PG is mainly attributed to increase in G^* as modifier content in bitumen goes up. Increase in G^* is a clear indication of stiffening effect of modifier, which also reduce temperature susceptibility of binder. Hence, binders having higher γ -LDPE_{r-g}-MA modifier content were affected less from temperature rising and provide higher G^* that leads to improved PG grades. Generally, δ considered as a better indicator than G^* that able to indicate elastic and permanent type of deformations. These is also a decreasing trend in δ as γ -LDPE_{r-g}-MA polymer modifier increases which indicate elastic deformation part of the total deformation is increases. Decreasing in δ as polymer modifier content increase also contribute to improved high temperature PG.

Table 2: High temperature PG of base and γ -LDPE_r-g-MA modified bitumens

	Binder type	Temp. (°C)	G* (kPa)	δ (°)	G*/sin δ (kPa)	Limits (kPa)	Grade
B	Original	46	4.64	86.6	4.65	≥ 1.00	PG 52-Y
		52	1.85	87.4	1.85		
		58	0.766	88.2	0.767		
	Aged	52	3.06	87.1	3.07	≥ 2.20	
		58	1.39	87.2	1.39		
		58	2.26	86.5	2.27		
B-1-P	Original	58	1.05	87.2	1.05	≥ 1.00	PG 58-Y
		64	0.523	87.6	0.523		
		52	4.97	84.4	4.99		
	Aged	58	2.22	85.6	2.22	≥ 2.2	
		64	1.03	86.5	1.03		
		52	4.08	83.7	4.10		
B-3-P	Original	58	1.89	84.5	1.90	≥ 1.00	PG 58-Y
		64	0.928	85.0	0.931		
		52	8.31	81.5	8.40		
	Aged	58	3.69	83.1	3.72	≥ 2.20	
		64	1.75	84.2	1.76		
		52	6.51	78.6	6.64		
B-5-P	Original	58	3.13	78.9	3.19	≥ 1.00	PG 64-Y
		64	1.64	78.8	1.67		
		70	0.905	78.0	0.925		
	Aged	52	10.1	80.3	10.3	≥ 2.20	
		58	4.51	81.8	4.56		
		64	2.23	82.8	2.25		
B-7-P	Original	70	1.69	74.8	1.75	≥ 1.00	PG 76-Y
		76	0.794	73.3	0.83		
		52	23.3	70.8	24.7		
	Aged	58	11.1	71.7	11.7	≥ 2.20	
		64	5.71	71.7	6.01		
		70	3.09	71.1	3.26		
B-9-P	Original	76	2.28	70.6	2.42	≥ 1.00	PG 82-Y
		82	1.80	69.8	1.92		
		52	56.2	46.0	40.4		
	Aged	58	26.1	43.7	37.8	≥ 2.20	
		64	17.5	41.1	26.7		
		70	12.2	38	19.9		
B-9-P	Original	76	8.93	34.6	15.7	≥ 1.00	PG 82-Y
		82	6.49	31.2	12.5		
		52	36.6	58.8	42.8		
	Aged	58	19.8	57.3	23.5	≥ 2.20	
		64	11.4	55.4	13.8		
		70	6.84	53.5	8.52		
B-9-P	Original	76	4.36	51.2	5.60	≥ 2.20	PG 82-Y
		82	2.87	48.4	3.84		

3.6 Bending beam rheometer test results

Creep stiffness and m -values obtained at temperatures of -6 °C, 12°C, and -18°C were given in Table 3. Creep stiffness of γ -LDPE_r-g-MA modified binders is generally higher than that of the base bitumen at all testing temperatures. Besides, the increment in creep stiffness is almost gradual with increasing polymer modifier content. Although, the expectation from a modifier at low temperature is to reduce creep stiffness of binder to avoid thermal cracking, it should be noticed that all of the modified binders except B-9-P remain same low temperature PG level with base bitumen's as none of creep stiffness exceeds limitation of 300 MPa. The binder having 9% polymer modifier content is the only one binder of which low temperature PG reduces one level below due to its m -value of 0.278 at -12°C which should be higher than 0.300 according to SUPERPAVE limitations. Hence, the γ -LDPE_r-g-MA polymer modifier has a slightly adverse effect on low temperature performance on bitumen as creep stiffness increases. Nevertheless, when it was compared with the increment in high temperature performance grade of binder, it must be noted that approval temperature range of binders has significantly increased after modification.

Table 3: Low temperature PG of base and γ -LDPE_r-g-MA modified bitumens

Binder types	Creep stiffness (MPa)			m -value			Performance grading (PG)
	-16 °C	-22 °C	-28 °C	-16 °C	-22 °C	-28 °C	
B	42.13	99.50	199.05	0.479	0.432	0.295	PG X-22
B-1-P	71.14	93.19	203.77	0.334	0.327	0.290	PG X-22
B-3-P	64.38	107.65	231.80	0.321	0.314	0.281	PG X-22
B-5-P	68.95	98.38	238.79	0.325	0.318	0.288	PG X-22
B-7-P	78.30	123.49	256.73	0.311	0.321	0.269	PG X-22
B-9-P	85.51	143.44	Terminated	0.307	0.278	Terminated	PG X-16

CONCLUSION

The following conclusions can be reported based on the results of the study:

- The examination by means of florescent microscope indicates that γ -LDPE_r-g-MA polymer particles present as molten and mixed with bitumen and, there is no dominant polymer phase appeared in bitumen even modification level reaches to 9%.
- The decrement in penetration and increase in softening point of binder as modifier content increase signifies that bitumen became stiffer at medium temperature range after γ -LDPE_r-g-MA modification. Moreover, PI index calculated by means of penetration and softening point indicate improved temperature susceptibility of binders after modification. However, ductility tests results shows that the modifier has a slightly adverse effect on elongation property of bitumen.
- Rotational viscosity test conducted at 135 °C and 165 °C was another indicator of the stiffening effect of γ -LDPE_r-g-MA as viscosity of modified bitumen remarkably increases. On the other hand, the increment in viscosity reaches ten times more than that of the original bitumen when modification level is 9%. This drastic increase in viscosity leads to rise of the mixing and compacting temperature of mixtures. Considering the energy consumption of producing, γ -LDPE_r-g-MA should not be exceeded medium modification levels of 5% in bitumen.
- The rheological test results such as DSR and BBR indicate that convenient temperature range of the modified binder is much wider than base bitumen as high temperature performance grade was improved remarkably after γ -LDPE_r-g-MA modification although low temperature performance of binders decreases slightly. Increment in complex shear modulus and decreases in phase angle was determined by means of DSR tests which signifies the binders having γ -LDPE_r-g-MA are more resistant to rutting as stiffness and elastic ability of bitumen increases.
- Based on the results, it can be noted that using γ -LDPE_r-g-MA polymer with a modification level of 5% can reduce temperature susceptibility and increase stiffening of binders which lead to reduce permanent deformation of binders at high temperature region.

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