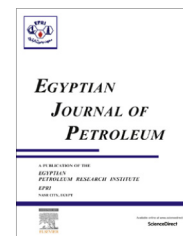




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FULL LENGTH ARTICLE

Thermo-mechanical properties improvement of asphalt binder by using methylmethacrylate/ethylene glycol dimethacrylate



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Abstract Various polymer-modified asphalt compositions for paving and roofing applications are known since several years ago. The degree to which a polymer improves the asphalt's properties depends on the compatibility of the polymer and the asphalt. Highly compatible polymers are more effective in providing property improvements. In this research, the influence of in situ polymerization of methylmethacrylate monomer with asphalt in presence of ethylene glycol dimethacrylate (EGDM) as a crosslinker on the rheological and thermal properties of asphalt binder of type penetration grade 60/70 was studied. To achieve this aim, MMA/EGDM(MC) in different ratios as 5, 10 and 15% (w/w) were used to modify the thermo-mechanical properties of asphalt via forming chemical bond, and the changing in mechanical and thermal properties, of the mixes as well as the storage stability were studied. Also, the morphology (SEM), thermal characterization (TGA), dynamic mechanical analysis (DMA), bending and rheological tests were detected. The obtained experimental results revealed that the addition of MC causes both the rheological and thermal properties of the binder to improve and the prepared PMAs has high temperature susceptibility and low curing time. The improvement in the properties of the virgin asphalt will be effective in using this soft type in coating applications instead of highly expensive oxidized one.

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1. Introduction

Raw asphalt is an interesting material which can undergo different physical states with variation in the temperature. At

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room temperature and below 0 °C, asphalt is bright, rigid, and brittle. When heated above 25 °C, asphalt begins to soften (between 60 and 80 °C). However, the softening point depends on the nature and composition of the asphalt. At 120 °C, asphalt behaves like a Newtonian liquid, and finally, at 200 °C, asphalt starts decomposition, generating residues [1].

Modification of bitumen is one of the approaches to improve the asphalt performance when the asphalt produced

Table 1 Characteristics of Virgin Asphalt.

Characteristics	AC	SP*
Physical characteristics		
• penetration (@ 25 °C, 100 g, 5 s) 0.1 mm	62	60/70
• Softening point (ring and ball) °C	50.6	45/55
• Specific gravity (@ 25 °C) using a pycnometer	1.02	NS**
• Flash and fire points (Cleveland Open Cup) °C	+ 250	+ 250
• Ductility (@ 25 °C, 5 cm/min.) cm	+ 150	+ 150
• Dynamic viscosity (at 60°C) c.p	599186	NS**
• Penetration Index (P.I)	-0.51	-2: + 2
Separation of polymer, (163 C°), 48 h		
Difference in softening point from top and bottom, C° (***)	1	
Chemical composition		
Maltene (wt%)	77.2	
Asphaltene (wt%)	22.8	

N,B

* Standard specification for “General Authority for Roads, Bridges and Land Transportation in Egypt”. Item No 102.1.

** Not specified.

*** According to literature, maxim difference is 2.

does not meet the climatic, traffic and other applications structure requirement, as reported by Fitzegerald [2] and Kim [3]. The concept of modifying asphalt binders and mixtures is not new. In its earliest stages, asphalt modification consisted of mixing two or more asphalt binders of different grades from different sources. The problem with this technique, however, lies in the possibility that the asphalt cement will be chemically incompatible [4]. This incompatibility cannot always be effectively predicted, and it can lead to premature asphalt distresses. Today, all forms of asphalts are usually modified. The modified binders are used for a wide range of applications [5]. Abiola [6] classifies asphalt modifiers as fillers, extenders, polymers, fibers, oxidants and antioxidants, anti-stripping agents, waste materials and hydrocarbons.

Polymer modified asphalts have better mechanical properties and higher durability than the non-modified ones. Since 30 years ago in some European countries, polymers have been added into asphalt [7].

The purpose of bitumen modification with polymers is to achieve the desired engineering properties, such as increased shear modulus and reduced plastic flow at high temperatures and/or increased resistance to thermal fracture at low temperatures.

Polarity of the polymer can enhance its solubility and compatibility with base bitumen [8]. Polar groups present in polymer molecules can react with polar constituents of bitumen. Subsequently, phase separation is prevented, which in turn enhances the material's consistency, and decreases oxidative aging [9,10]. Among polar polymers, a very limited number of studies discuss the fundamental properties of modified bitumens with acrylate polymers. Most frequently used acrylates as bitumen modifying agents in road applications are ethylene vinyl acetate (EVA) glycidyl methacrylate (GMA) terpolymer, ethylene butyl acrylate (EBA) copolymer, . . . etc. [11–13]. General Poly methacrylates are polymers of the esters of methacrylic acids. The most commonly used among them is poly (methyl methacrylate) (PMMA) which has high mechanical strength, high Young's modulus and low elongation at break. It does not shatter on rupture and it is one of the hardest thermoplastics i.e. highly scratch resistant. Also, it exhibits low moisture and water absorbing capacity, and accordingly it

has good dimensional stability. Both of these characteristics increase as the temperature rises. PMMA is one of the polymers that are most resistant to direct sunshine exposure. Its strength characteristics exhibit fairly small variations under the effect of UV radiation, as well as in the presence of ozone. These properties of PMMA make it suitable for products intended for long open air operation [14,15].

The use of polymer modified asphalt in different applications has generated a lot of interests in its rheological properties, because of their importance in the manufacture and quality of bituminous applications. As a matter of fact, the development of the early colloidal model of PMA samples was based on the rheological observations. Long before that, ancient users of bitumen observed the strong effect of temperature on its consistency but due to its highly viscous character at room temperature, giving rise to a confusing and somewhat imprecise description such as pasty or semi-solid [3], bitumen rheological behavior remained hard to quantify. In the present paper, the rheological and thermal behavior of blends of bitumen with different concentrations of a PMMA/EGDMA was investigated as effective properties for asphalt to be used in coating applications. In this manner, bitumen/PMMA/EGDMA blends, in contents ranging as 5, 10 and 15 w/w, were prepared and evaluated. The rheological tests, thermal and microstructure analysis revealed that, the addition of reactive polymers, enabled to chemically interact with certain bitumen compounds, and caused some advantages in the resulting binder, regarding both improved storage stability and enhanced in-service performance.

2. Experimental

2.1. Materials used

- *Asphalt Cement*: Local virgin asphalt cement of penetration grade (AC 60/70) produced by El-Nasr Petroleum Company in Suez, Egypt.
- *Chemicals*: Methylmethacrylate MMA, ethylene glycol dimethacrylate (EGDM), Benzoyl peroxide. All the chemicals were supplied from the Fluka chemical company.

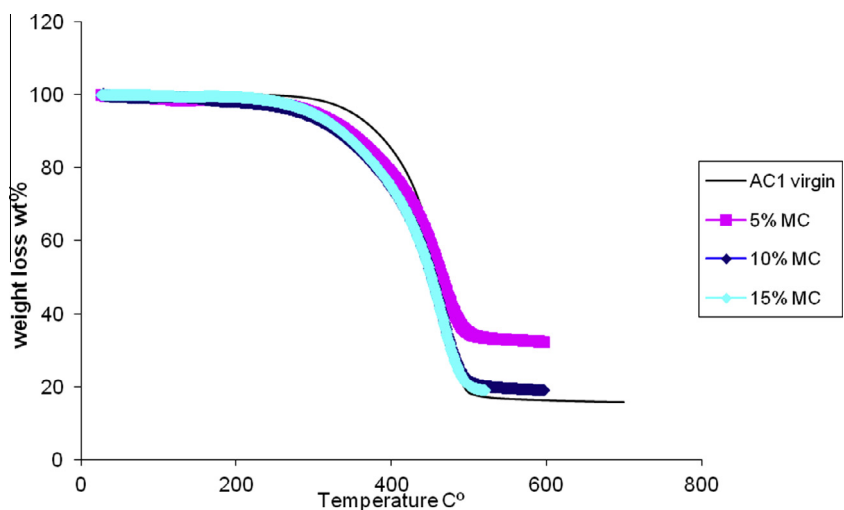
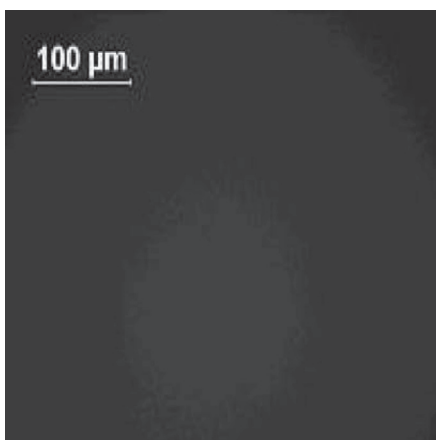
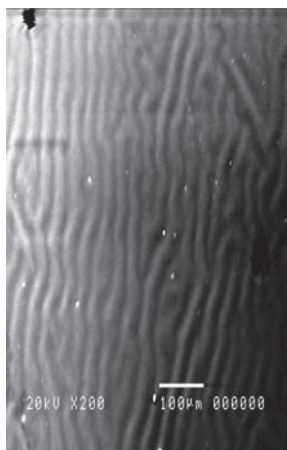


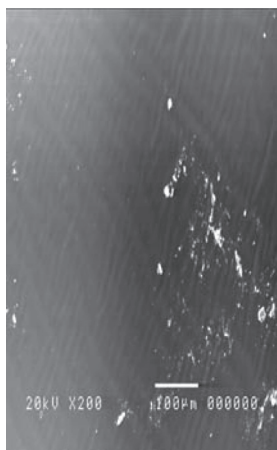
Figure 1 TGA curves of virgin and PMAs with MC.



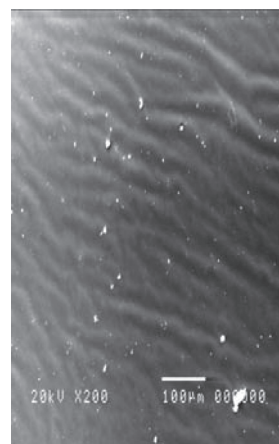
(a) virgin AC



(b):5% MC



(b)10% MC



(2b):15 % MC

Figure 2 SEM photos of virgin and PMA samples.

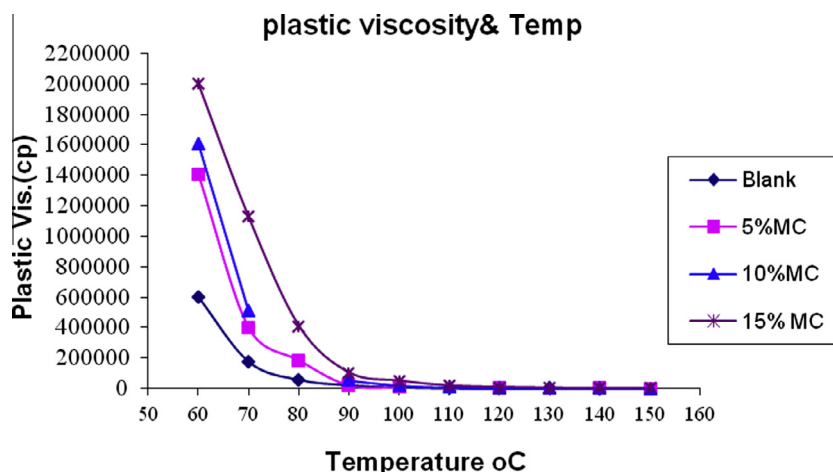


Figure 3 Plastic viscosity vs temperature for virgin and PMA samples.

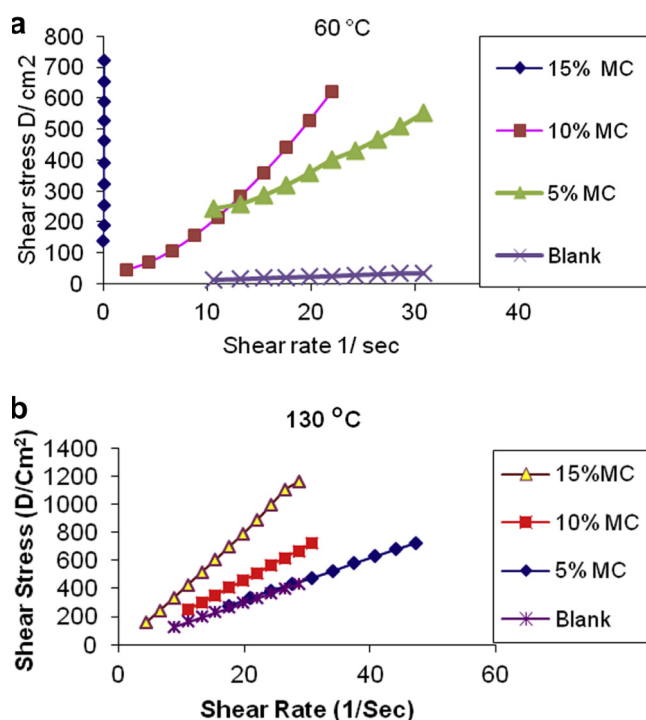


Figure 4 (a) Shear stress shear rate curves of PMA using MC at 60 °C. (b) Shear stress shear rate curves of PMA using MC at 130 °C.

2.2. Experimental procedure

The testing program included the following steps.

2.2.1. Characterization of virgin asphalt 60/70

- The virgin asphalt sample was tested as illustrated in Table 1 for penetration (ASTM D5), softening point (ASTM D36), specific gravity (ASTM D70), Brookfield viscosity (ASTM D4402) and n-heptan insoluble (ASTM D3297). The results are illustrated in Table 1.

- The temperature susceptibility of virgin asphalt sample was expressed in term of penetration index (P.I) using the penetration (@ 25 °C) and softening point values. P.I can be calculated using the following equation [16]:

$$P.I = \frac{1952 - 500 \times \log(\text{pen } 25) - 20 \times SP^*}{50 \log(\text{pen } 25) - SP - 120}$$

where Pen 25 = penetration value at 25 °C, 0.1 mm & SP softening point, °C

Also, asphalt sample was tested for TGA (Fig. 1), SEM (Fig. 2), rheology test (Figs. 3 and 4), DMA analysis (Fig. 5a and b) and bending test (Fig. 9).

2.2.2. In situ polymerization of MMA/EGDM asphalt samples [16]

In-situ polymerization of virgin asphalt and three levels of MMA (5, 10 and 15 percent by weight of asphalt) were prepared in suitable cans. Then a high shear mixer was dipped into the can and set to 3000 rpm. The polymer was added gradually (5 g/min). Temperature was kept within 180 ± 1 °C during the polymer addition and subsequent mixing. Then, stirring was performed for 2 h after complete addition of polymer, initiator and cross linking agent with fixed amount 2 ml into asphalt [17]. Under high temperature, partial carbon-carbon double bonds of MMA molecules were opened to form free radicals. EGDM were then added to crosslink those free radicals where a network structure would be formed when reaching high polymer concentration [18]. Moreover, asphaltene molecule, or micelle, contains more than one carboxylic group so; a chemical network theoretically will form. Unfortunately, due to the extremely complex chemical nature and composition of asphalts, it is difficult to detect the real nature of chemical bonds formed during cross linking. Scheme 1 represents schematic reaction of in-situ polymerization of MMA and asphalt in the presence and absence of EGDMA crosslinker.

2.2.3. Characterization and evaluation of Polymer Modified Asphalt Samples (PMAs) prepared

The PMAs were tested for physical characteristics (Table 2), TGA (Fig. 1), SEM (Fig. 2), rheology test (Figs. 3 and 4), DMA analysis (Figs. 5–8) and bending test (Fig. 9).

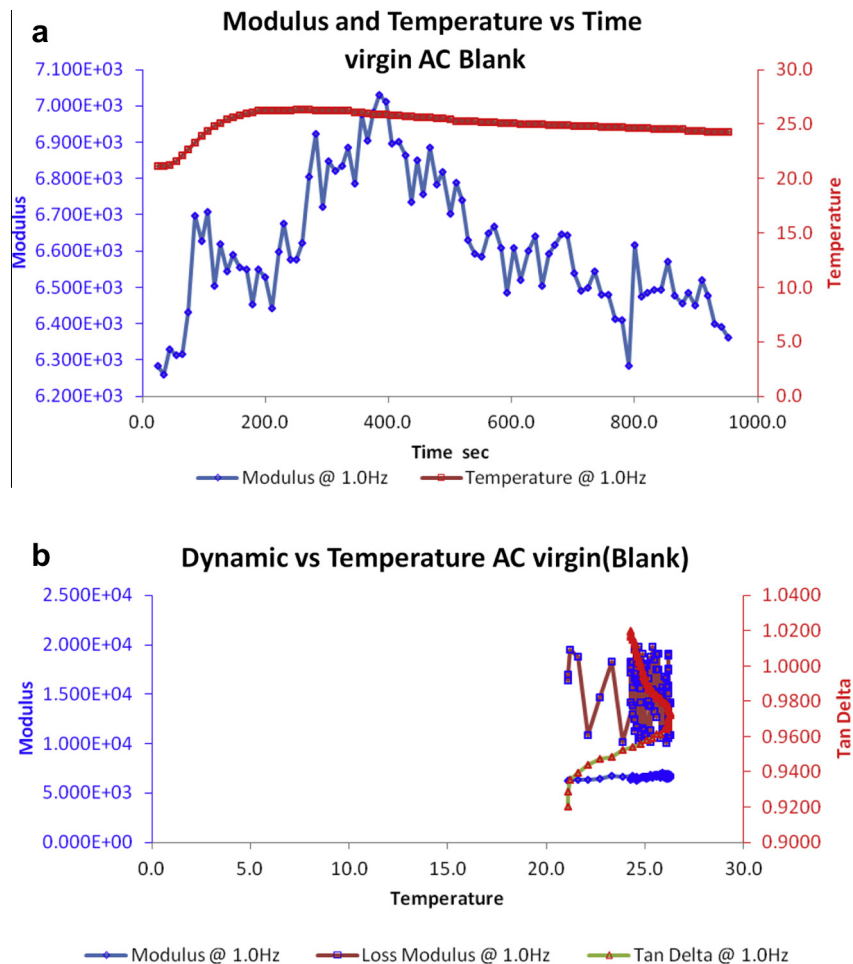
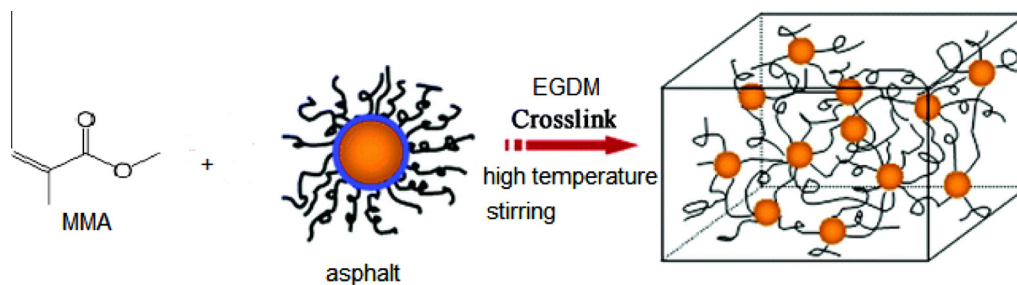


Figure 5 (a) Modulus and temperature Vs time for Blank AC. (b) Dynamic and Tan Dltta Vs temperature for Blank AC.



Scheme 1 Schematic reaction of in-situ polymerization of MMA and asphalt in the presence of EGDMA crosslinker.

2.2.3.1. Physical characteristic. Physical characteristics for PMAs are illustrated in Table 2.

2.2.3.2. TGA analyses. Thermal) as shown gravimetric analysis (TGA in Fig. 1 was carried out using SDTQ 600 thermo-gravimetric analyzer (TA-USA) to test the thermal stability of the virgin asphalt sample in the temperature range of 25–800 °C with a heating rate of 10 °C/min under dynamic nitrogen gas.

2.2.3.3. SEM photographs. The SEM photographs of asphalt virgin and its modified blends have been determined using

Scanning electron microscopy (SEM; Philips) as shown in Fig. 2.

2.2.3.4. Rheology test. The Brookfield DV-III Ultra Programmable Rheometer was used to measure fluid parameters as shear stress and viscosity at given shear rates and temperature. Viscosity is a measure of a fluid's resistance to flow. The principle of operation of the DV-III Ultra is to drive a spindle (which is immersed in the test fluid) through a calibrated spring. The viscous drag of the fluid against the spindle is measured by the spring deflection. Spring deflection is measured with a rotary transducer. The rheology test was run in 60 °C and 130 °C.

Table 2 Physical characteristics of PMAs.

Characteristics	AC	MMA/EGDM		
		5%	10%	15%
Penetration (at 25 °C, 100 g, 5 s) 0.1 mm	62	50	41	20
Softening point (ring and ball) °C	50.6	54	57	70
Specific gravity (at 25 °C)	1.02	1.025	1.030	1.039
Dynamic viscosity (at 60 °C) c.p	599186	1,279,384	1,959,582	2,243,543
Dynamic viscosity (at 130 °C) c.p	1499.68	1570.22	2298.18	3877.57
Penetration Index	-0.51	-0.21	0.38	0.85

2.2.3.5. *Dynamic mechanical analysis (DMA)*. DMA measures the dynamic mechanical behavior of a material as it is deformed under a sinusoidal strain deformation as a function of frequency and temperature. There are several modes in DMA such as three points bending, tension and shear. Actually, the selection of test mode depends upon the specimen nature and its stiffness. Dynamic mechanical analysis was performed by Triton Technology-TTDMa which determines

storage modulus, loss modulus and $\tan \delta$. Disk-formed specimens with 10 mm dia. and 3 mm thickness were tested at room temperature and 1 Hz frequency. Dynamic mechanical properties refer to the response of a material as it is subjected to a periodic force. These properties may be expressed in terms of a dynamic modulus, a dynamic loss modulus, and a mechanical damping term. Typical values of dynamic moduli for polymers range from 106 to 1012 dyne/cm² depending upon the type of polymer, temperature, and frequency. For an applied stress varying sinusoidally with time, a viscoelastic material will also respond with a sinusoidal strain for low amplitudes of stress. The sinusoidal variation in time is usually described as a rate specified by the frequency ($f = \text{Hz}$; $\omega = \text{rad/s}$). The strain of a viscoelastic body is out of phase with the stress applied, by the phase angle, δ . This phase lag is due to the excess time necessary for molecular motions and relaxations to occur. Dynamic stress, σ , and strain, ϵ , given as:

$$\sigma = \sigma_o \sin(\omega t + \delta) \tag{1}$$

$$\epsilon = \epsilon_o \sin(t\bar{\omega}) \tag{2}$$

where ω is the angular frequency. Using this notation, stress can be divided into an “inphase” component ($\sigma_o \cos\delta$) and an “out-of-phase” component ($\sigma_o \sin\delta$) and rewritten as,

$$\sigma = \sigma_o \sin(\omega t) \cos \delta + \sigma_o \cos(\omega t) \sin \delta. \tag{3}$$

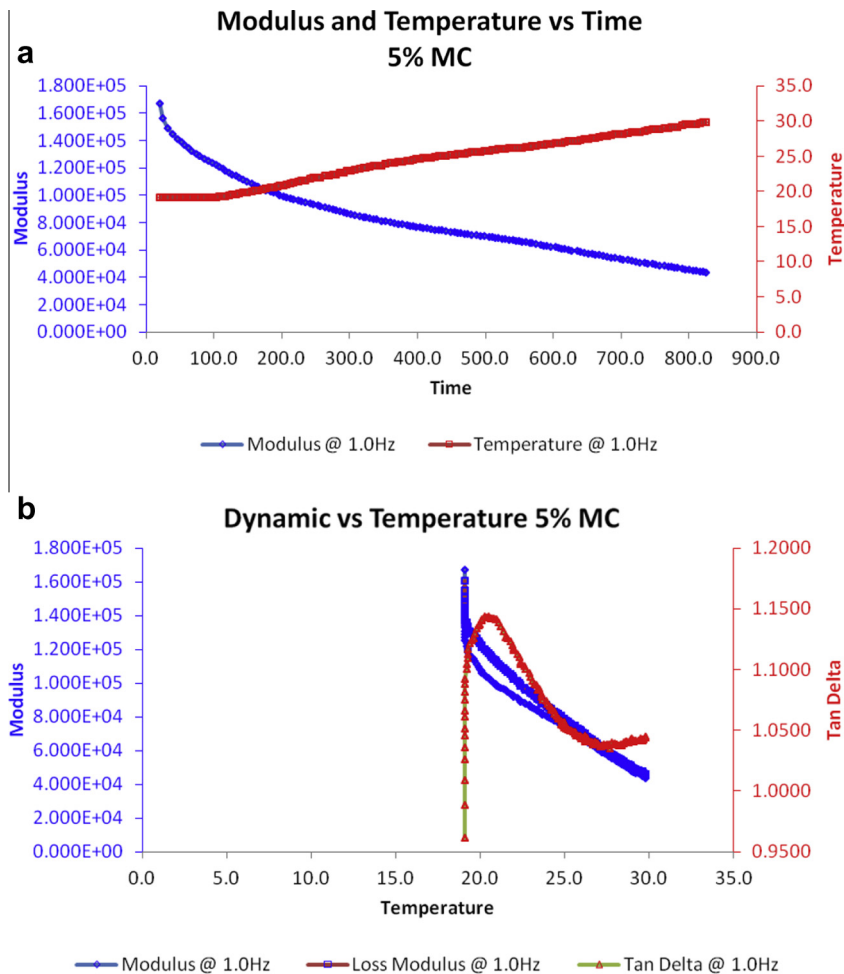


Figure 6 (a) Modulus and temperature Vs time for 5% MC. (b) Dynamic and Tan Dlta Vs temperature for 5% MC.

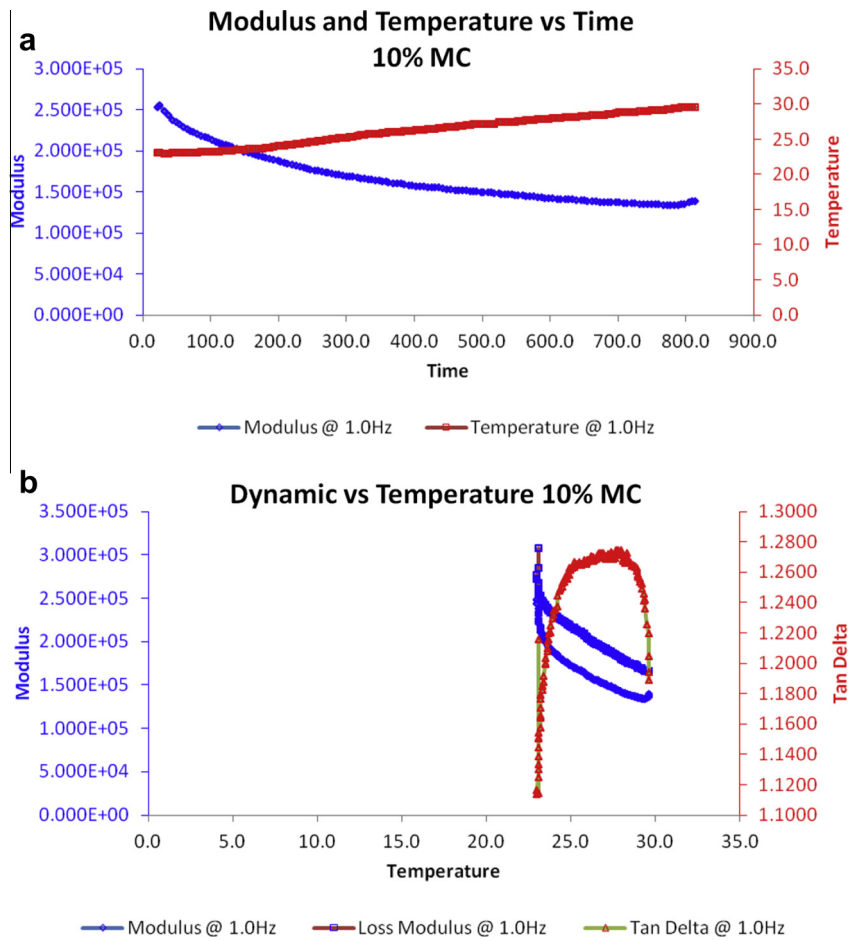


Figure 7 (a) Modulus and temperature Vs time for 10% MC. (b) Dynamic and Tan Dlta Vs temperature for 10% MC.

Dividing stress by strain to yield a modulus and using the symbols E' and E'' for the inphase (real) and out-of-phase (imaginary) moduli yields:

$$\sigma' = \varepsilon_0 E' \sin(\omega t) + \varepsilon_0 E'' \cos(\omega t) \quad (4)$$

$$E' = \sigma_o / \varepsilon_o (\cos \delta) \quad E'' = \sigma_o / \varepsilon_o \sin \delta \quad (5)$$

$$\varepsilon = \varepsilon_o \exp(i\omega t) \quad \sigma = \sigma_o \exp(\omega t + \delta)i \quad (6)$$

$$E^* = \varepsilon / \sigma = \varepsilon_o / \sigma_o e^{i\delta} = \varepsilon_o / \sigma_o (\cos \delta + i \sin \delta) = E' + iE'' \quad (7)$$

Eq. (7) shows that the complex modulus obtained from a dynamic mechanical test consists of “real” and “imaginary” parts. The real (storage) part describes the ability of the material to store potential energy and release it upon deformation. The imaginary (loss) portion is associated with energy dissipation in the form of heat upon deformation. The above equation is rewritten for shear modulus G^* as,

$$G^* = G' + iG'' \quad (8)$$

where G' is the storage modulus and G'' is the loss modulus. The phase angle δ is given by:

$$\tan \delta = G'' / G' \quad (9)$$

The storage modulus is often times associated with “stiffness” of a material and is related to the Young’s modulus, E . The dynamic loss modulus is often associated with “internal friction” and is sensitive to different kinds of molecular motions, relaxation processes, transitions, morphology and other structural heterogeneities. Thus, the dynamic properties provide information at the molecular level to understanding the polymer mechanical behavior.

2.2.3.6. Bending test. Mandrel test (ASTM E 522) was used to evaluate the flexibility and the resistance to cracking for the prepared modified asphalt samples (organic coatings) on the substrates of sheet metal. In this way, the coating materials were applied to a uniform thickness using a brush in one direction for one time to panels of sheet metal. After complete curing, the coated panels were bent over a mandrel and the resistance to cracking of the coating was determined. Coatings attached to substrates are elongated when the substrates are bent during the manufacture of articles or when the articles are abused in service. Conical mandrel bend tester is applicable also to determine the extensibility of asphalt coatings on metal panels which are clamped in position and formed round the conical mandrel by rotating the roller frame. The panels were examined visually to evaluate crack resistance detachment

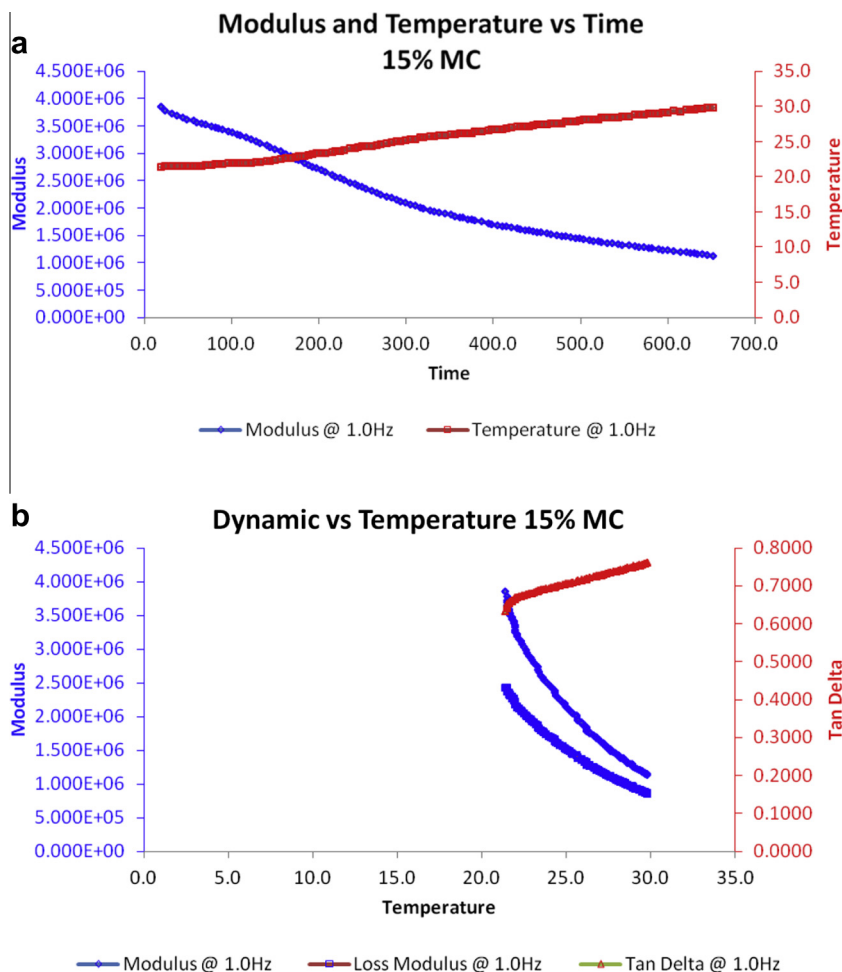


Figure 8 (a) Modulus and temperature Vs time for 15% MC. (b) Dynamic and Tan Delta Vs temperature for 15% MC.

from the metal substrate of coated surface which was coated with PMA samples under the standard condition.

3. Results and discussion

3.1. Characteristics of virgin and modified asphalt samples

From Tables 1 and 2 and Figs. 1–9 the following results were detected:

3.1.1. Physical characteristics of samples

Generally, data in Tables 1 and 2 indicated that the modified samples are more hardening than the virgin one as there was an increase in softening point, specific gravity and dynamic viscosities and decrease in penetration value with the addition of MC to asphalt.

Increasing the MC content from 5 to 15 wt% produced more hardness asphalt samples compared to properties of virgin asphalt AC. Penetration of PMAs decreased in percentages of 19.35%, 33.87% and 67.74%, while softening point increased in percentages of 6.7%, 12.65% and 38.34% respectively with addition of MC as 5%, 10% and 15%. This is attributed to the nature and accordingly the chemical molecular composition of the PMMA which react with asphalt to form chemical bond. It is a known fact that, PMMA has high

mechanical strength, high Young's modulus and low elongation at break. It does not shatter on rupture. It is one of the hardest thermoplastics and is also highly scratch resistant. It exhibits low moisture and water absorbing capacity, due to which products made have good dimensional stability. Both of these characteristics increase as the temperature rises [19]. The formation of more hardness modified samples than the virgin one may be explained by spreading PMMA (which is a macromolecule with long chain and three-dimensional network) within asphalt which is composed of hard, and large molecules polar aromatic material dispersed in saturated paraffin materials, will cause difficulty to soften the mixture. As a result of asphalt modification, its cohesion and elasticity are both enhanced. At higher service temperatures, the stiffness modulus of polymer phase is higher than that of matrix. These reinforcing properties of the polymer phase contribute to the increase in viscosity. At low temperatures, the stiffness modulus of the dispersed phase is lower than that of the matrix, which reduces its brittleness. Consequently, the dispersed polymer phase enhances the engineering properties of asphalt in terms of viscosity, softening point and toughness [20].

3.1.2. Temperature susceptibility of all asphalt samples

The temperature susceptibility of the modified bitumen was investigated in the term of P.I. The results showed the more



(a) of virgin AC



(b) Oxidized asphalt (OA).

Figure 9 Mandrel bend testing panel of virgin AC and Oxidized asphalt (OA).

P.I which indicated that there was an increase in asphaltene contents therefore, the less temperature susceptibility was obtained [21].

From Table 2 it is obviously that, the modification of asphalt reduces the temperature susceptibility of the produced samples. Comparing to virgin AC the P.I values increased in values as – 0.21%, 0.38% and 0.85% for 5%, 10%, 15% of MC content respectively. Also, the increase in P.I will increase the resistance of asphalt samples to cracking at low temperature [22].

3.1.3. TGA analysis

Fig. 1 shows TGA curves for virgin and prepared PMA samples. Virgin AC shows decomposition stage from 290 to 490 °C with a mass loss of 84.17% suggesting the decomposition of asphaltenes to produce coke. On the other hand, the decomposition of PMMA proceeds in three general stages, as shown by the TGA output. From the TGA data, the first stage begins at about 120 °C, although the small detectable mass loss (2%) and its corresponding thermal effect occur only from 150 to 200 °C in this stage. The second stage destroys about 40% of the sample, most rapidly at 270 °C, with an apparent change of mechanism at about 290 °C. Finally, the sample is completely decomposed by 410 °C, reaching a maximum in rate of mass loss and heat uptake at 370 °C [23].

From Fig. 1, PMA samples showed a very similar thermal behavior when compared to virgin AC. the complete weight loss was 67.75%, 81.08% and 79.32% in case of using MC modifier in percentages of 5%, 10% and 15% (w/w) respectively. This may be attributed to a decrease in the thermal stability as a result of an increase in P.I. values for the prepared PMAs. In the other word, the increase in asphaltene content causes the decomposition temperatures to decrease and also char yield of bitumen. However, the rate of decomposition after initial decomposition temperature (IDT) has an opposite

Table 3 Plastic viscosity vs temperature for virgin and PMA samples.

Temperature, °C	Plastic viscosity (cp)			
	Blank	5% MC	10% MC	15% MC
60	599,186	1,408,093	1,608,093	2,003,440
70	177,824	400,000	509,855	1,126,908
80	59,757	186,757	250,645	410,094
90	24,943	19,037	54,929	103,198
100	10,386	9146	21,680	49,767
110	5087	6542	9297	18,133
120	2486	2322	4245	8471
130	1546	1853	2386	4075
140	929.3	1491	1421	2289
150	559.3	884.5	884.5	1408

effect. Bitumen with higher P.I decomposes slower after IDT than bitumen with a lower P.I. [24].

3.1.4. SEM of PMAs

The SEM photos of the virgin and polymer modified blends are displayed in Fig. 2. It is shown that; all PMAs are compatible and homogeneously formed mono phase picture and this is due to the addition of the cross-linking agent during preparation of the PMAs [25]. The modification process allows the polymer rich phase to slightly cross-link and prevents polymer rich phase droplets to coalesce. High cross-link density is not sought for, because it would result in a lower swelling extent. Another proposed stabilization mechanism, although of little industrial relevance, makes use of low molecular weight polymers thought to act as emulsifiers and finally this leads to a decrease in the phase separation [26].

3.1.5. Rheology test

3.1.5.1. Viscosity and temperature. In this test the effect of temperature variation on the viscosity for all samples is detected. The test is also used to develop temperature–viscosity charts.

As shown in Fig. 3 and Table 3, it can be observed that the viscosity values reduce as the test temperature increases regardless of polymer content. Fig. 3 indicates that the base binder has the lowest viscosity values at all the chosen test temperatures as compared to the modified asphalt binders. The blend viscosity increased with the increase in polymer content at the chosen temperatures from 60 to 150 °C, and this increase was observed in both the viscous and elastic moduli. For example and comparing to virgin sample the percent increase in viscosity at 60 °C was 213%, 360% and 371% in case of the content of MC is 5%, 10% and 15% respectively. At higher temperatures, the viscosity also increases but in percentages lower than that recorded at lower temperatures. At a temperature of 130 °C viscosity increased in percentages of 4.7%, 105% and 158.5% for the same level of addition. This indicated that the added polymer react with virgin asphalt and formed cross linking between asphalt and MC forming a chemical network caused the formed PMAs to be more rigid and difficult to flow [27].

3.1.5.2. Shear rate and shear stress. Fig. 4 shows that the virgin asphalt and PMA samples do not undergo shear rates proportional to the applied shear stress that means these undergo as A non-Newtonian matter.

A non-Newtonian fluid is broadly defined as one for which the relationship is not a constant. In other words, when the shear rate is varied, the shear stress doesn't vary in the same proportion (or even necessarily in the same direction). The viscosity of such fluids will therefore change as the shear rate is varied. Thus, the experimental parameters of Viscometer model, spindle and speed all have an effect on the measured viscosity of a non-Newtonian fluid. Non-Newtonian flow can be envisioned by thinking of any fluid as a mixture of molecules with different shapes and sizes. As they pass by each other, as happens during flow, their size, shape, and cohesiveness will determine how much force is required to move them. At each specific rate of shear, the alignment may be different and more or less force may be required to maintain motion. There are several types of non-Newtonian flow behavior, characterized by the way a fluid's viscosity changes in response to variations in shear rate.

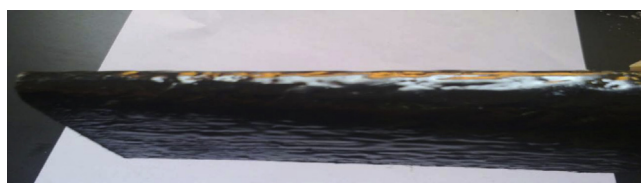
3.1.5.3. Dynamic mechanical analysis. Figs.5(a and b)–8(a and b) shows that: The modulus increased with polymer added percent increased at the same time Tan Delta increased which means that loss modulus decreases with increasing polymer added percent this is due to reaction between MC and asphalt forming internal network caused high plasticity in PMAs than



(a) 5% MC



(b) 10% MC



(c) 15% MC



(d) MC

Figure 10 Mandrel bend testing panel of asphalt modified using MC.

the AC virgin. Also, figures show the modulus decrease in a small amount with time which means that the PMA samples became more stable and more resistant for deformation conditions and has high storage stability modulus. PMAs show the higher storage modulus due to stronger intermolecular interactions this can refer to occurrence of crosslinking between MC and virgin asphalt. The gap as distance between the G' and G'' curves (whatever be: amplitude, time or frequency scans) has not much relevance. Instead, the ratio G''/G' which is $\tan(\delta)$, is a measure of the internal friction of the material in that condition. When loss modulus is higher than modulus $\tan(\delta)$ is >1 and one can say that the sample is more viscous than elastic, and vice versa when modulus is higher and $\tan(\delta)$ is <1 . At $\tan(\delta) = 1$ is the crossover or gel point as shown in figures in PMAs with 15% $\tan(\delta)$ is <1 which mean that the sample more elastic than others. When the sample starts to cure, modulus grows faster because of the cross linking reaction. When modulus is equal to loss modulus ($\tan \delta = 1$, loss tangent), that this is the “gel point” but at that frequency and temperature. In addition, when both curves are flat in time, then reaction has finished.

3.1.5.4. Mandrel bending test. From Figs. 9(a and b) and 10(a–d) it is shown that AC and OA samples failed to elongate with Mandrel bending tester and some cracks appear). From the corresponding elongation to such added values of the polymer, it is clear that when MC content increased from 5 to 15%, the sample became completely flexible and no cracks appeared as shown in Fig. 10(a–d). Mandrel test clearly showed that incorporating 15 w% of MC enhances the flexibility of the modified asphalt samples to the required value.

4. Conclusion

This research aims to prepare polymer modified asphalt based on AC 60/70 which is used only in paving applications with different weight ratios of methylmeth acrylate in presence of ethylene glycol dimethacrylate (EGDM) as a cross linker for use in infrastructure applications. The produced materials have low cost, high storage stability and superior characteristics as compared to the commercially traditional material (oxidized asphalt).

The results showed that:

- The mixes comply with the standards have reduced temperature susceptibility.
- It is not well known how the polymer interacts with asphalt molecules; however, it is believed that the partial carbon–carbon double bonds of MMA molecules were opened to form free radicals and then react with functional groups prevalently present in asphaltene micelles. However, the presence of an interpolymer cross-linking reaction cannot be excluded.

But the curves obtained from dynamic data clearly show that during the evaluation the material tends to the behavior of a cross-linked network.

- The prepared PMAs samples have desired engineering properties, such as increased shear modulus and reduced plastic flow at high temperatures and increased resistance to thermal fracture at different temperatures.

- Polymer modified asphalts have better mechanical properties and higher durability than the non-modified ones.
- The addition of reactive polymers, able to chemically interact with bitumen compounds, may yield some improvements in the resulting binder, regarding both improved storage stability and enhanced in-service performance.
- Polar groups present in polymer molecules can react with polar constituents of bitumen. Subsequently, phase separation is prevented, which in turn enhances the material's consistency, and decreases oxidative aging.
- 15 w% of MC enhances the flexibility of the modified asphalt samples to the required value.

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