Int. J. Chem. Sci.: 10(4), 2012, 2048-2056
ISSN 0972-768X
www.sadgurupublications.com

STUDIES ON THE MODIFICATIONS OF ASPHALT BINDER USING SOME SELECTED POLYMERS

U. M. IBEAGI[a], S. O. E. OKEREKE[b], O. C. OKPAREKE[*] and P. O. UKOHA[c]

Department of Pure and Industrial Chemistry University of Nigeria, NSUKKA
[a]Department of Polymer and Textile Engineering Federal University of Technology, OWERRI
[b]Department of Industrial Chemistry Abia State University, UTURU
[c]Department of Pure and Industrial Chemistry, University of Nigeria, NSUKKA

ABSTRACT

Increase in pavement distress has prompted the study and investigation of the use of polymers as asphalt cement modifiers. It was observed that rubber-related polymers are most suitable and they improved on the resilience, toughness, and reduced temperature susceptibility of the asphalt. Development of cracks, rutting and potholes were reduced, and water-proofing effect on the pavement was improved. Compatibility of asphalt mix, a major factor in the determination of extent of modification was enhanced by incorporation of catalysts, and application of mechanical method.

Key words: Polymers, Asphalt, Toughness, Resilience.

INTRODUCTION

Asphalt, a constituent of petroleum is obtained as a residual product after crude oil has been separated into its constituents or fractions in the refinery[1]. It is black in colour and is 92-97% soluble in carbon (II) sulphide. It is composed of about 83% carbon, 11% hydrocarbon and heterocyclic compounds containing, 6% sulphur, 1% nitrogen and oxygen; its components vary in molecular weight from 400-5000. The exact molecular structure of asphalt varies from source to source[2].

A laquinillas petroleum asphaltene fraction is shown as :

Three main types of asphalt have been recognized:

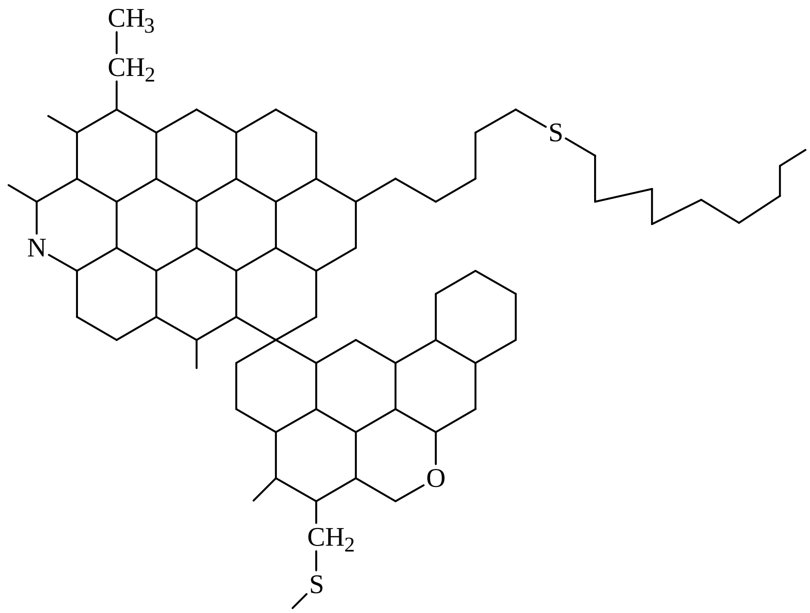
(a) Residual or straight–run asphalt used for road constructions, underbody coatings, and hydraulic works.

[*]Author for correspondence; E-mail: obinna.okpareke@unn.edu.ng

(b) Air-blown asphalt used in refining pipe-coating, paints, underbody coatings, paper laminates, and

(c) Cracked asphalt used as insulant, dust laying, etc.

Other categories include solid, semi-solid, and liquid asphalts whose divisions are based on viscosity, plasticity and their behaviours with reference to temperature[3].



The viscous nature of asphalt allows for permanent deformation or rutting of asphalt pavements to occur at service temperatures. During dry seasons or summer months, and temperature of pavements surfaces in Nigeria reach 60°C or even higher. The asphalt loses its resilience and a single heavy truck does as much rutting damage as 7000 automobiles. Its resilience and temperature susceptibility can be improved by adding rubber and some other polymers[4]. In the ambient temperature range of 5°C to 35°C, fatigue cracking occurs due to repeated cyclic stresses of values less than the tensile strength of the pavement. Fatigue resistance of the pavement can be achieved by using electrometric polymers and plastomers. It has been reported that styrene-butadiene polymer and neoprene extend fatigue life of asphalt pavement 30 to 10 times[5,6].

At low extreme temperatures between 4°C and 7°C as in early winter months when large temperatures changes can occur overnight. Normal asphalt becomes too brittle to withstand significant thermal shrinkage. Brittleness occurs when the asphalt used in the mixture has insufficient ductility at low temperatures. It may also occur because of a combination of factors: the shrinkage which takes place during a volume change because of lowering of temperature and the low tensile strength of the binder[7]. Brittleness may also develop due to steric hardening of the asphalt binder or the oxidation or polymerization of the asphalt film from exposure to air and sunlight. The brittle asphalt binder has no toughness, and small cracks initiated by naturally occurring flaws in the pavement can form large transverse cracks in a short time due to recurring freeze-thaw cycle[8].

The addition of rubbers, such as natural rubber, reclaimed rubber wastes, lower the brittle point, and increases the toughness thereby lowering the rate of cracking of the asphalt pavement. A number of polymers and their physical properties have been considered.

(a) Natural rubber in form of liquid suspension is called latex. It is processed into ribbed smoke sheets, air-dried sheets, and processed crumbs[9]. It has a unique combination of low hysteresis, and therefore high resilience at low strains. It has the ability to crystallize when stretched, thereby inducing strength and high elastic modulus. The strain-induced crystallization is responsible to natural rubber's resistance to fatigue by a crack growth mechanism.[10-11]

The ability of natural rubber to crystallize at low temperature is a desirable feature for asphalt at relatively low temperatures. The rubber fraction in the natural rubber-asphalt mixture will help to increase its modulus at temperatures less than 0^oC thereby preventing brittleness. The ability of natural rubber to crystallize at high strains helps the asphalt pavement to resist resilience of natural rubber and enhance the elastic response of the asphalt at high ambient temperature, with improved resistance to flow[12].

(b) **Low Density Polyethylene (LDPE)**: It is obtained in form of pellets by high pressure vapour-phase polymerization of ethylene monomer, is characterized by low melting point and low opacity. LDPE has reduced molecular weight, reduced strain rate, decreased tensile strength, decreased brittle point and high ductility.[13-14]

It is a wax-like thermoplastic, with softening range of 80^oC - 130^oC, and density of 0.910-0.925 g/cm^3. Ductility and low brittleness of LDPE are advantages to asphalt modification and use. LPDE is susceptible to thermal degradation, oxidation and hence anti-oxidants and carbon black are incorporated for protection[15].

(c) **Recycled Rubber**: It is a rubber waste, which has been converted into an economically used form such as reclaimed rubber, ground tyre, and reprocessed synthetic rubber.

Incompatibility in asphalt-polymer blends and solution

Polymer-modified asphalts lack phase homogeneity. Due to high molecular weight and rigid nature of the asphaltene fraction in the asphalt binder, high molecular weight synthetic polymers are not usually miscible in these systems. For polyethylene-modified binder, it has been reported that an initially homogenous system easily phase-separates to form a thick congealed layer on top of the hot asphalt mixture. This phenomenon tends to inhibit the large-scale acceptance of polyethylene-modified asphalt binders[16,17].

However, steric stabilizers which prevent the particles in a colloidal suspension from precipitating have been developed to permanently prevent coalescence at typical storage temperatures of the binders[18].

Styrene-butadiene rubber and natural rubber were added in conjunction with small amounts of sulphur. The sulphur both grafts onto the rubber back-bone, thus making it soluble and cross-links the polymer to form a higher molecular weight with better rheological properties[19].

Compatibility of polymers and asphalt can also be enhanced by addition of polymers to hot asphalt in conjunction with small amounts of Friedel-Crafts catalyst (anhydrous aluminum chloride, $AlCl_3$). This catalyst facilitates the reaction between the polymer and asphalt phase. It rearranges and disproportionates the alkyl aromatic asphaltene fraction. It is observed that intramolecular migration of chloride atoms occurs on the aromatic structure.[20]

A large number of aromatic alkyl substitute units and allylic hydrogen in the asphalt molecule make the material generally amenable to both radical-induced and electrophilic substitution reactions[21]. The influence of the different polymers on asphalt binders was investigated based on a number of parameters, namely penetration and viscosity. These parameters were used to determine the performance of asphalt in paving applications and in addition, make comparisms between the polymer-modified asphalt binders and the unmodified binders.

EXPERIMENTAL

Materials and methods

1. 60-70 penetration grade of asphalt obtained from National Oil Nigeria Plc. Port-Harcourt.
2. (a) Natural rubber latex from the Imo Rubbers Estate Ltd, Owerri.

(b) Ground Rubber Tyre (GRT) produced by Dunlop Nigeria Plc. and processed to powder of average particle size of 65 nm by Ferdinand Rubber processing Company Urualla.

(c) High Density Polyethylene and Linear Low Density Ethylene (Elpene FIU), and polypropylene (M130ST) resins from Eleme Petrochemical Company Limited.

3. The catalyst and stabilizer, anhydrous aluminum chloride and sulphur, respectively, obtained from Port-Harcourt and Owerri.

Method

Different samples of 60-70 penetration-grade asphalt binders were mixed with various polymers as shown in Table 1

Table 1: Blends of 60-70 penetration asphalt binder and polymers

S. No.	% by weight polymer	% by weight of asphalt
1.	5% Natural Rubber Latex	95%
2.	5% LDPE Film	95%
3.	5% Ground Rubber Tyre (GRT)	95%

The prepared samples were subjected to penetration and viscosity measurement according to ASTM standard. The hardness of the unmodified and the various polymer - modified asphalt binders were evaluated by determining the depth of penetration, using penetrometer at 15^{o}C and 25^{o}C.

Viscosity measurements were taken with unmodified and polymer-modified asphalt at 15^{o}C and 25^{o}C using Engler viscometer; for each evaluation 100 g of the binder sample was used.

RESULTS AND DISCUSSION

Values of penetration at 15^{o}C and 25^{o}C and the penetration index (PI) for different Polymer-modified asphalt are as shown (Table 2). This was compared to the values for unmodified asphalt binder at 15% and 25^{o}C.

Table 2: Penetration index (PI) for polymer-modified and asphalt binders

Asphalt (Penetration Grade)	Polymer	Penetration (mm) at 15°C				Average (mm)	Penetration (mm) at 25°C				Average (mm)
60-70	-	92.5	96.5	94.0	95.0	94.5	186	192	188	190	189
60-70	GRT	45.0	57.0	56.0	55.0	53.0	92	98	96	95	95.30
60-70	LATEX	43.0	39.0	42.0	43.0	41.80	70	71	68	71	70
60-70	LDPE	32.5	28.0	27.0	36.0	30.80	60	59	60	60	60

Penetration Index is a widely used empirical parameter in asphalt to indicate the temperature sensitivity of a particular asphalt binder as shown, for the various samples (Table 3).

Table 3: Penetration index (PI) for unmodified and polymer-modified asphalt binders

Asphalt	Penetration index (PI)
60-70 Unmodified	1.98
GRT – Modified	3.198
Latex –modified	4.136
LDPE – modified	2.325

The slope of plots of logarithm of penetration against temperature, for each binder sample, was indicative of the temperature susceptibility of the asphalt binder sample as shown in (Table 4) and Fig. 1.

Table 4: Logarithm of penetration versus temperature for each binder sample

Asphalt sample	Penetration (mm)	Penetration (mm)	Log P	Log P
	At 15°C	25°C	15°C	25°C
60-70 asphalt (unmodified)	94.5	189	1.9756	2.2765
GRT-modified	53.0	95.3	1.7243	1.9786
Latex-modified	41.8	70	1.6212	1.8451
LDPE-modified	30.9	60	1.4900	1.7782

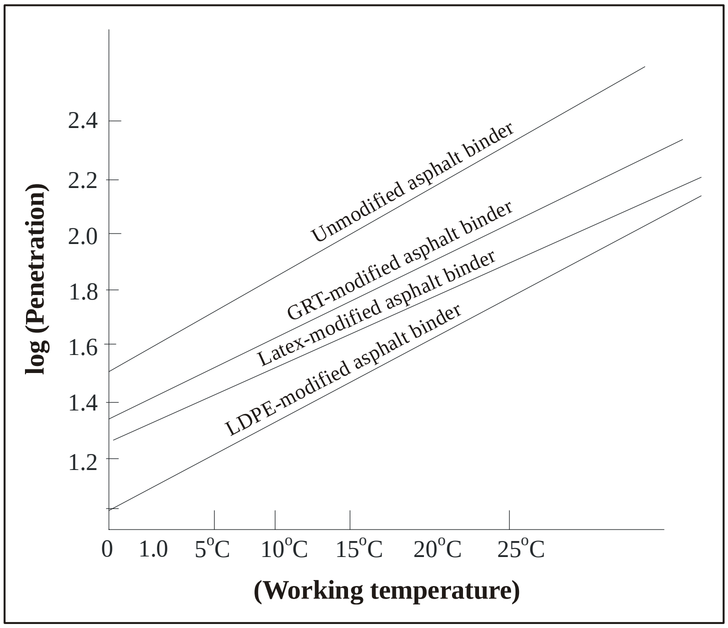


Fig. 1

Viscosity data using the Engler Specific Viscosity are as shown in Table 5.

Table 5: Viscosity data using engler specific viscosity

Asphalt penetration grade	Polymer additive	Efflux time for 50 mL (sec)	Engler specific viscosity
60-70	-	63	1.235
60-70	GRT	1609	31.54
60-70	Latex	2081	40.80
60-70	LDPE	546	10.71

Both the penetration index[6,22] and the viscosity indicate the temperature sensitivity of a particular asphalt binder. A low temperature susceptible asphalt binder will have a reasonable flexibility at low temperatures and a reasonable high viscosity at high service temperature and often fail at both temperature extremes; that is, they were too brittle at low temperatures and too soft at high temperature[23]. From Table 3, it appeared that latex-modified asphalt with PI of 4.136 and GRT modified asphalt with PI of 3.198 were highly acceptable[8]. They can withstand higher temperatures. Their viscosities of 40.80 and 31.54 (Table 5) show that they improved flow[24-25].

CONCLUSION

Three asphalt additives namely Natural Latex, Ground recycled tyre (GRT), and Low density polythylene (LDPE) were investigated for their effect on high temperature rheological behaviour and low temperature fraction behaviour of asphalt binders. The polymers used showed improved compatibility with the asphalt binders after reactive processing at elevated temperatures, which resulted in a significant increase in both the penetration index (PI) and the viscosity over the unmodified binder.

At 5% GRT, 5% Natural Rubber Latex, and 5% LDPE blends the Penetration Index (PI) increased from 1.98 to 3.198, 4.136 and 2.325, respectively therefore the most desirable additives recommended for asphalt binder are natural rubber latex and ground rubber (GRT).

ACKNOWLEDGEMENT

The authors are gratefull to National Oil Nigeria Plc Port-Hacourt, Imo Rubbers Plc Owerri, Dunlop Nigeria Plc, Ferdinand Rubber processing Company and Eleme Petrochemical Company Limited for their assistance during the course of the research.

REFERENCES

1. R. Steven, Highway Design Reference, The Mc-Graw Hill Engineering Reference Guide Series, New York, USA (1989) p. 7.
2. P. C. Painter and M. M. Coleman, Fundamentals of Polymer Science, An Introductory Text, Technomic Publishing Co. Inc. Lan. Custer P. A. (1997).
3. C. V. Horie, J. Appl. Poly. Sci., **54**, John Wiley and Sons Inc., London, UK (1994) pp. 231-240.
4. M. N. Partl and L. Francken, Introduction to Bituminous Binders and Mixes RILEM Report, London, **17** (1998) pp. 2-10.
5. X. U. Lu, Issacson and J. Ekblad, Construction and Building Materials, 12405, 414 (1998).
6. X. Lu and U. Issacson, J. FRB, 1661, 83 (1999).
7. H. Green, An Acceptance Test for Bitumen for Rolled Asphalt Wearing Course, Transport and Road Research Laboratory Report, Griffen, George and Sons Ltd., London (1998) pp. 5-11.

8. I. K. Pellinen, M. W. Witezak and R. F. Bonaquist, Asphalt Mix Mater Currc. Construction Using Sigmoidal Kitting Function Wilt Non Linear Least Squares Optimization 15th ASCC Engineering Mechanics Conferences June 2-5, Columbia University, New York (2002).

9. J. M. Krishann and K. O. R. Rajagopal, Mechanicals of Materials, **37**, 1085 (2005).

10. A. Sainton, Transportation Research Record, 1265 (1990).

11. J. R. Stastna, L. Zanzotto and O. J. Vacin, J. Colloid and Interface Sci., 200 (2003).

12. A. Whelan and K. S. Lee, Developments in Rubber Technology, Holloway, London (1993) pp. 20-22.

13. A. Yields, Construction and Building Materials, **2**, 66 (2007).

14. I. M. Ward and D. W. Hadley, An Introduction to the Mechanical Properties of Solid Polymers, Wiley, New York (1993).

15. Waleed Abd El-Aziz. Mohamed, Crumb Rubber in Improving Asphalt Mixes, Ph.D. Civil Eng., El-Mandona University (2004).

16. B. Noor Zainab, K. Ibrahim, N. Madzlan and M. I. Isa, Proceeding of Malaysian Universities Transportation Research Forum and Conference (2010).

17. C. Wekumbra, J. Stastna and L. Zanzottoet, J. Mater. Civil Engg., 227 (2007).

18. I. S. Chem, M. C. Lia and M. S. Shiah, J. Merter Civil Engg., **14(3)**, 1224 (2002).

19. A. A. Youseki, Iran Polym. J., **18**, 207-215 (2009).

20. Garcia-Moralesm, P. Partal, P. J. Navarro, F. G. Marti-nez Boza and C. Gallegor, Rheo Acta, **45**, 1329 (2007).

21. American Society for Testing and Materials (ASTM) Standard Specification, Section 4, **Vol. 04-03** (2000).

22. A. A. Youseki, J. Color Sec. Tech. (Persian), **2**, 10 (2008).

23. A. E. Langrouidi and A. A. Youseki, J. Polym. Sci. Tech. (Persian), **16**, 293 (2003).

24. J. Stastna, L. Zanzotto and O. J. Vacin, J. Colloid Interface Sci., **259**, 220 (2002).

25. D. Drozdor Aleksey Yuan, Int. J. Solid Structure, **40**, 2221 (2003).

Revised : 18.07.2012 *Accepted : 20.07.2012*