

Enhancement of storage stability and rheological properties of polyethylene (PE) modified asphalt using cross linking and reactive polymer based additives

Rabindra Kumar Padhan^{a,*}, Anand Sreeram^b

^a Indian Oil R&D Centre, Sector-13, Faridabad, Haryana, India

^b Department of Civil and Environmental Engineering, Hong Kong Polytechnic University, Hong Kong, China

HIGHLIGHTS

- Improved storage stability and rheological performance of PE modified binders.
- Cross linking agents significantly enhance elastic properties of modified binders.
- Modified binders exhibit homogenous polymeric distribution of PE in bitumen matrix.

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ABSTRACT

The incorporation of waste polymeric materials such as polyethylene (PE) has shown to improve the rheological properties of asphalt pavement but it is often beleaguered with other concerns such as storage stability and phase separation. This study looks that the incorporation of specific reactive polymers and cross-linking additives to resolve the practical difficulties associated with the application of PE modified asphalt. Binders with waste PE were firstly prepared followed by chemical modification using trans-polyoctenamer (TPOR) and cross-linking additives at various percentages. The samples were then evaluated by the means of various tests such as conventional binder tests, storage stability, superpave performance tests and fluorescence microscopy. The results indicated that such binder modification significantly aids to reduce the storage stability problems, greatly increase elastomeric properties and improve overall rheological properties. From the microscopic analysis, it was also observed that a network of PE linkages in the modified binders is well dispersed, contributing to a more balanced and stable binder performance.

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1. Introduction

Every year, millions of metric tons of polyethylene (PE) is produced and predominantly utilised as a packaging material making it one of the most popularly used plastic materials in the world. Chemically, PE is highly robust and degrades very slowly as compared to other traditional plastic materials. Hence, it remains in the environment for a long time causing widespread pollution. The use of waste materials for road construction has been promoted due to conservational and economic benefits. In view of this, an effective way to reuse waste PE in pavement would be welcomed by practitioners around the world [1,2]. In terms of binder modification, it is now common that polymer modified binders are

used in asphalt pavement applications as they offer considerable advantages over unmodified binders in relation to rheological and ageing resistance properties [3–6]. The addition of polymers such as styrene–butadiene–styrene (SBS), ethyl vinyl acetate (EVA), polyvinyl chloride (PVC), polyethylene (PE), polyoctenamer, polyethylene terephthalate (PET) materials in asphalt has been well researched and its benefits widely recognised [5]. PE in particular has been the focus of many researchers and can be mainly divided into two categories; low density polyethylene (LDPE) and high-density polyethylene (HDPE). LDPE entails of carbon atoms attached to a long chain of PE and has density between 0.91 and 0.94 while HDPE is comparable but does not have as much branching and has a density of more than 0.94. LDPE is traditionally synthesised through polymerisation at very high pressures and temperatures while HDPE is obtained by polymerisation at relatively low temperatures and pressures. Published literature has

* Corresponding author.

E-mail address: rabinpadhan@gmail.com (R.K. Padhan).

indicated that LDPE with low molecular weight and broader molecular distribution to be more effective and compatible than HDPE in terms of asphalt binder modification [7]. In the early studies, positive rheological effects were observed on the linear viscoelastic properties when a combination of recycled ethylene vinyl acetate (EVA) and LDPE was used as a modifier for asphalt binder [7]. In another study, various rheological tests were conducted on PE modified asphalt binder to investigate the impact of temperature, mixing time and other parameters on binder performance [8]. Additionally, the ageing mechanism of recycled PE-modified asphalt binder with rubber powder has also been investigated [9]. When LDPE modified asphalt was used in conjunction with maleic acid, an improvement in viscosity, softening point and penetration was observed [10]. Overall, from these studies it has been well established that PE modified asphalt is more rut and fatigue resistant than conventional asphalt binder. However, one of the major drawbacks of PE modified asphalt is its poor mixability and storage stability in asphalt. It has been noted that partial phase separation might occur during the application of these binders in pavement [11]. Therefore, in order to overcome these issues; it would be meaningful to look for suitable additives to incorporate in PE modified asphalt to improve its mixability, storage stability and overall performance. Studies have already reported the possibility of the improvement in homogeneity and mixability of PE modified binder by the addition of various additives [12]. Generally, asphalt binder is modified with two types of polymeric materials: non-reactive polymers such as thermoplastics, elastomers, plastomers and reactive polymers (eg-anhydrides, isocyanates and epoxide) which contain reactive groups capable of reacting with specific functional groups. An example of such a reactive polymer is trans-polyoctenamer (TPOR) which has been formerly used to improve the performance properties of various types of bituminous binders [13,14]. TPOR, due to its unique double bond chemical structure has also been used in conjunction with cross linking agents such as sulphur, now widely available in pelletised form; and shown to further enhance the elastomeric character and rheological properties of the binders [14]. Some studies have also indicated that asphalt binders modified through sulphur vulcanisation of trans-polyoctenamers led to improved rheological performance especially in terms of storage stability and elasticity [15]. Hence it is anticipated that similar modification with selected reactive polymer-based additives can have a notable positive effect in improving the performance properties of PE modified asphalt. The overall objective of the present work is to investigate the combined effect of polyoctenamer polymer and a cross-linking agent to overcome the traditional deficiencies such as low elastic recovery and high-temperature storage stability in PE modified asphalt. A binary mixture of PE and TPOR was firstly prepared followed by the addition of sulphur at three different proportions to characterise the rheological properties of the mixtures. Microscopical characterisation was also subsequently conducted on the various binders to shed light on the interaction between the additives and the asphalt binder.

2. Material preparation and research methodology

2.1. Materials

The asphalt binder used in the present work was of commercial grade VG 10 binder obtained from Indian Oil refinery in Mathura, India. Typical properties of the binder used are presented in Table 1. The Trans-polyoctenamer (TPOR) used was supplied by Micu Traders, Vadodara, India. The TPOR had a molecular weight of 90,000, its basic properties and chemical structure are listed in Table 2 and Fig. 1 respectively. The sulphur used was of industrial grade and of 99.9% purity.

The PE low density plastic bags were procured locally and cut into small pieces of around 5 cm by 5 cm for convenience in mixing as illustrated in Figs. 2 and 3. Homogeneity between PE samples are important in the context of the asphalt modification hence the bags were firstly characterised by IR analysis using an ISSO FTIR

Table 1
Properties of Asphalt Binder.

Properties	Asphalt Binder
Ductility at 27 °C (ASTM D113-17)	100+
Flash Point Pen Cup, °C	350

Table 2
Properties of Trans-Polyoctenamer.

Tensile impact strength kJ/m ² , 23 °C	165
Molecular weight (MW)	90,000
Tensile Stress Yield	7.5 MPa
Glass transition temperature	−65 °C

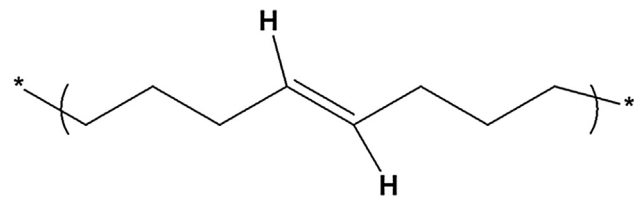


Fig. 1. Chemical structure of trans-polyoctenamer.



Fig. 2. Waste PE bags.

manufactured by the Thermo Fisher Scientific Inc to ascertain its functional groups which showed the strong bands at 2919 cm^{−1} and 2851 cm^{−1} attributable to CH₂ stretching and bands at 1366 cm^{−1} and 1350 cm^{−1} attributable to CH₂ and CH₃ stretching respectively as show in Fig. 4. The bags were also studied under a SEM microscope which showed the clear layered polymeric structure characteristic of low density PE as indicated in Fig. 5.

2.2. Preparation of modified asphalt

Asphalt binder per 100 units, in a free-flowing form was blended with a 2:1 ratio of PE and TPOR at around 165 °C for 1 h at a mixing rate of 4000 r/min with the help of a high shear mixer to synthesize PE-TPOR. The dosages of the TPOR and PE chosen was based on the prior work conducted by the authors and preliminary mixing studies which showed that a higher PE content (greater than 2%) may not be suitable for producing storage binders irrespective of other additive content [16]. Afterwards, three percentages of cross linking agent i.e. sulphur at 0.1%, 0.5% and 1% was mixed with PE-TPOR for a further 30 min to produce PE-TPOR-2, PE-

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