



# Optimizing the use of reactive terpolymer, polyphosphoric acid and high-density polyethylene to achieve asphalt binders with superior performance

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## HIGHLIGHTS

- Individual use of PPA and HDPE stiffen the base binder but induce minor effect on binder elasticity.
- EMA-GMA improves binder stiffness and elasticity however, its allowable content is not sufficient.
- PPA is more effective than HDPE in enhancing the chemical reaction between EMA-GMA and base binder.
- EMA-GMA with HDPE and/or PPA result in equivalent or better binder performance than a SBS polymer.

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## ABSTRACT

Styrene-butadiene-styrene (SBS) is the most widely used polymer in asphalt binders because of its success in mitigating pavement rutting and cracking distresses. However, the asphalt industry has been searching for alternatives that provide equivalent or possibly better performance. This study investigated the individual and combined effects of reactive terpolymer (ethylene/methylacrylate/glycidyl methacrylate (EMA-GMA)), polyphosphoric acid (PPA) and high-density polyethylene (HDPE) on binder rheological properties with the goal of formulating new binders to replace a PG 76–22 SBS binder. The PPA and HDPE were found to only increase binder stiffness (i.e., reduced penetration number, non-recoverable compliance and increased high temperature performance grades) whereas the EMA-GMA also improved the binder elasticity (i.e., increased elastic recovery and strain recovery values). Binders with single modifier failed to meet the strain recovery requirement indicating insufficient elasticity, which is known to be positively correlated with binder cracking performance at intermediate temperatures. Conversely, the combined use of EMA-GMA with PPA or with PPA plus HDPE not only increased binder stiffness but also induced sufficient elasticity. Moreover, PPA as a catalyst, promoted the chemical reaction between EMA-GMA and base binder, better than HDPE did. Finally, comparisons were made in abovementioned properties between the PG 76–22 SBS binder and selected new binders. It was concluded that both PPA and HDPE enhanced the efficiency of EMA-GMA to achieve binders with equivalent or potentially better performance than a standard PG 76–22 SBS binder.

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## 1. Introduction

Polymer modification has been extensively adopted by the asphalt industry to enhance binder properties that strongly affect pavement resistance to rutting, fatigue and thermal cracking. In addition, polymer modification is an environmentally sustainable solution that can also result in significant economic savings by

extending the service life of asphalt pavements. Following sections provide detailed descriptions of reported experiences of the main classes of polymers currently used in asphalt modification.

Polymers can be generally classified into three main classes: elastomer, plastomer and reactive polymer (RET) [29]. Styrene-butadiene-styrene (SBS) is the most widely used elastomer in the asphalt industry because of its success in mitigating rutting and cracking in asphalt pavement. Over 40 states in U.S.A. have adopted elastomers (e.g., SBS) as modifiers and some states like Florida require exclusive use of SBS for binder modification.

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However, SBS binders are much more expensive than unmodified binders, mainly due to the high cost of the SBS polymer. The typical content of SBS used in a binder ranges from 2 to 5 wt% (% by weight of final product) and the use of higher percentages is an economic disadvantage [21]. Moreover, the use of a crosslink agent is necessary to avoid phase separation and ensure properties [22].

Plastomeric polymers, which are known to be less expensive than elastomers, increases binder stiffness resulting in better resistance to permanent deformation [23,30]. The two most common plastomers are Polyethylene (PE) and Polypropylene (PP), which are categorized as polyolefin. Polyolefin can be used either alone or blended with elastomers as co-modifiers. The primary disadvantage of using plastomers as modifiers is phase separation. Pérez-Lepe et al. [24] investigated asphalt binders modified with high-density polyethylene (HDPE) at contents of 1–5 wt%. Their test results indicated that although HDPE enhanced high temperature performance, severe storage instability occurred which makes this type of modification inefficient for pavement application. Also, the use of plastomeric polymer to bump binder performance grade (PG) can yield binders that are more susceptible to oxidative aging and, consequently, it degrades cracking performance.

RET polymers are typically based on ethylene, glycidyl-methacrylate (GMA) and an ester group, either methyl, ethyl or butyl acrylate. One unique feature of RET is that it chemically bonds to a base binder by establishing covalent linkages between molecules [21]. Certain functional groups in RET are able to react with asphalt molecules and form a 3-dimensional network, resulting in improved binder properties and performance. Typically, RET polymers are used in low percentages (between 1.5–2.5 wt%) because higher dosage leads to formation of an insoluble and infusible asphalt gel [25]. Bulatovic et al. [14] evaluated binders modified with RET polymers that contain difference percentages of reactive functional groups (i.e., GMA's). The use of 1.9 wt% RET clearly caused binder gelification although improved binder elasticity and rutting resistance were also reported. Conversely, Polacco et al. [26] highlighted that low RET content had a minor effect on base binder properties which are quite poor comparing to a SBS modified binder. They suggested that the efficiency of RET can be improved when combined with polyolefins; however, this suggestion has not been investigated.

Berkley and Romagosa [13] reported that in 2009, the SBS market was hit by a shortage of butadiene which affected the supply and cost of the most commonly used binder (i.e., PG 76-22 SBS) throughout the U.S.A. High cost of SBS polymer has driven the asphalt industry to search for economic alternatives, and RET polymer is a promising option [31]. RET polymer chemically reacts with the base binder providing acceptable storage stability as well as improvements in performance to some extent. However, the amount of RET that can be added without causing binder gelification is not sufficient to ensure satisfactory performance. Therefore, further research is needed to optimize the use of RET polymer by

adding co-modifiers and/or catalysts to achieve enhanced binder properties in an effective way.

## 2. Objectives and scope

The objectives of this study were: 1) to investigate the individual and combined effects of RET terpolymer, polyphosphoric acid (PPA) and high-density polyethylene (HDPE) on the properties and performance of a conventional asphalt binder and 2) to optimize combination of these three modifiers to formulate high performance binders that can be used as alternatives to a standard PG 76-22 SBS PMA binder.

## 3. Materials and specimen preparation

Three modifiers used in this study were: 1) reactive terpolymer of ethylene/metil-acrilate/glycidyl metacrylate (EMA-GMA) (metil-acrilate 20% by weight, glycidyl metacrylate 6% by weight); 2) HDPE, a polyolefin, and 3) polyphosphoric acid (PPA 116%), as catalyst. Fig. 1 shows the modifiers used in this study.

PG 67-22 unmodified and PG 76-22 SBS PMA binders that are routinely used in the state of Florida were utilized as the base and reference binders, respectively. Eleven binders named from B to L were formulated and produced. The amount of EMA-GMA was fixed at 1.8 wt.% to achieve improvement in binder elasticity without causing gelification. Details regarding the formulations are presented in Table 1 and described as follows:

- A: PG 67-22, base binder for modifications;
- B and C: various contents of PPA;
- D, E and F: fixed EMA-GMA with various contents of PPA;
- G, H and I: fixed HDPE and with various contents of EMA-GMA and PPA;
- J, K and L: fixed EMA-GMA and PPA, with various contents of HDPE;
- M: PG 76-22 SBS PMA, a reference binder.

Binders were modified in the laboratory using a low shear mixer at a rate of 2500 rpm. A temperature control heating mantle was used to maintain a temperature of 165 °C during mixing. Modifiers were added in the preheated base binder as necessary following the sequence of priority: 1) EMA-GMA for 60 minutes, 2) HDPE for 30 minutes, and 3) PPA for 30 minutes. After mixing, samples were transferred to an oven at 160 °C for 12 hours for curing.

## 4. Research approach

Penetration and softening point tests, which are known as traditional empirical methods, were applied for binder classification. In addition, dynamic shear rheometer (DSR) and bending beam

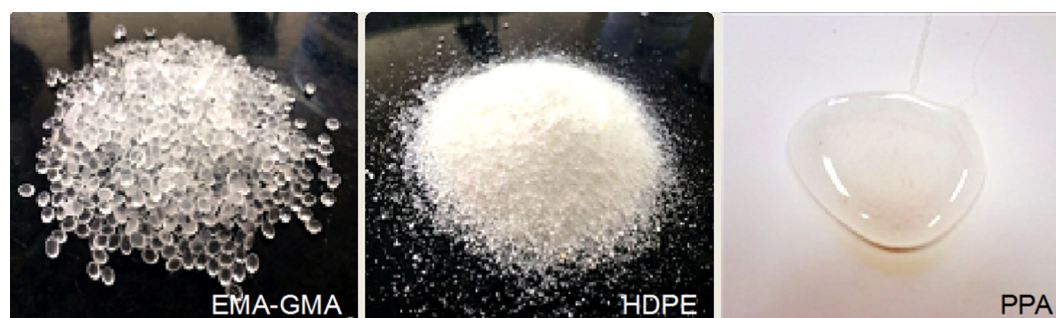


Fig. 1. Illustration of EMA-GMA, HDPE and PPA.

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