



Micro-heterogeneous modification of an asphalt binder using a dimethylphenol and high-impact polystyrene solution



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HIGHLIGHTS

- We verified the oxidative condensation from dimethylphenol to polyphenylene oxide.
- The modified asphalt exhibits an increase in $|G^*|/\sin(\delta)$ of 2.14 kPa at 82 °C.
- This indicates an improvement in its high-temperature rut resistance.

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ABSTRACT

This study proposes a modified asphalt binder using a solution of high-impact polystyrene (HIPS) and 2,6-dimethylphenol (DMP). The oxidative polymerization of 2,6-dimethylphenol synthesizes a thermoplastic polymer (polyphenylene oxide, PPO) in the presence of copper, amines, and vanadium in the asphalt binder. The polymerization and existence of PPO were verified using X-ray diffraction and Fourier transform infrared (FTIR) spectroscopic analyses. The maximum crystallinity of the solution occurred after a mixing time of 2 h with a homogenizer mixing energy of 3000 rpm. The rheological properties of the DMP- and HIPS-modified binder are also investigated using a dynamic shear rheometer (DSR) and a bending beam rheometer (BBR). The modified asphalt binder exhibits an increase in $|G^*|/\sin(\delta)$ of 2.14 kPa at 82 °C without aging, which improves the high-temperature rut resistance of the modified binder. The modification does not significantly affect the low-temperature crack resistance of the binder, considering that the m -value and stiffness at -12 °C are 0.31 and 237 MPa.

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1. Introduction

Asphalt binder is a black cementitious material in which the predominant constituent is bitumen, which either exists in nature or is a byproduct of petroleum processing. Similar to cement, asphalt binders have been widely applied to roadway pavement because of their strong cohesiveness, adhesion, waterproof nature, and durability. The material also provides flexibility to mixtures of aggregates and high chemical resistance to reactions with most acids, alkalis, and salts [1].

Since the development of modern petroleum refinement techniques in the early 1900s, asphalt binders have been widely utilized as a crucial ingredient of paving materials. Although asphalt binders are most commonly used as cementitious materials in paving, their chemical compositions are highly variable and depend on where and how the petroleum was formed; therefore, complete analysis and characterization of their chemical and mechanical properties would be almost impossible.

Thus, to set the mechanical grade of an asphalt binder as a useful paving material by engaging its viscoelastic nature, several methods were used, including the penetration grading, viscosity grading, and, most recently, performance grading systems. An example of which is the Superpave PG system, rutting and fatigue cracking could be correlated with rheological measurements, such as $|G^*|/\sin(\delta)$ in ranges greater than 1.0 kPa without aging or 2.2 kPa with short-term aging and $|G^*|/\sin(\delta)$ when less than 5000 kPa with long-term aging, to verify the performance specification of the appropriate binder. According to the temperature changes at a site, the PG specifications provided seven grades from PG46 to PG82 in 6 °C intervals between two adjacent temperatures [2].

For high temperature grades exceeding 70 °C, styrene-butadiene-styrene (SBS) has been widely used to modify bitumen because of its excellent engineering properties. However, the role and effect of SBS in bitumen are still ambiguous in the aging process. Although superior elastic recovery at low temperatures and enhanced viscosity at high temperatures are the main advantages of a SBS-modified binder, its drawbacks are phase separation and oxidation due to the carbon double bond (C=C) in the vinyl group

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of the styrene monomer, which expedites aging or oxidation by easily binding oxygen molecules in the air or water. After using a pressure aging vessel for approximately 500 h, the softening point of the SBS-modified binder is almost identical to that of a plain asphalt binder at 60–70 °C, meaning that the SBS was degraded after aging in the PAV, leading to the disappearance of the cross-linked network structure [3–5].

The copper-catalyzed oxidative coupling polymerization of 2,6-dimethylphenol (DMP) to polyphenylene oxide (PPO) was first proposed by Hay et al. to utilize the thermal, oxidative, and chemical stabilities of PPO [6,7].

To enhance the oxidative resistance and cohesiveness of asphalt binders at high temperatures through the oxidative polycondensation process, this study proposes an asphalt binder modified with a solution of high-impact polystyrene (HIPS) and DMP.

2. Selection of materials

2.1. Polymer-modified asphalt binder

An asphalt binder is a polar compound that consists of a variety of chemical components and has a solubility parameter between 16.5 and 19.3 (cal/cm³)^{1/2}; its specific gravity varies between 0.95 and 1.05 g/cm³ depending on the crude oil [8].

Asphalt binders can be separated into two major components, such as asphaltene and maltene, using differences in solubility between the various molecules. Asphaltenes, black amorphous solids, comprise 5–25% by weight of an asphalt binder and have relatively high molecular weights (10,000–100,000) that consist of various functional groups, such as carboxyl (–COOH), alcohol (–OH), ketone (–C=O), amine (–NH–), and sulfur (–S–). The remaining weight consists of the continuous phase (oils) and dispersant (resins) [2].

During the Strategic Highway Research Program, a complete review of colloidal systems of asphalt binders led to the conclusion that the colloidal model is not consistent with the rheological behavior of asphalt binders as a function of temperature, loading time, and aging. The chemical complexities of asphalt binders, compared to those of thermoplastic homopolymers, such as polyethylene and polypropylene, led to the development of physical property tests relevant to the performance of a given asphalt binder [2].

Although some asphalt binders satisfied conventional grading thresholds, such as viscosity or penetration, increases in heavy axle load above 80 kN, a high tire pressure above 600 kPa, and a great number of truck passes have recently resulted in severe failures, such as fatigue cracks and rutting in hot-mix asphalt pavement. Such failures may be related to some limitation in asphalt binder performance, the aggregate, or mix design. One alternative for enhancing the physical properties of asphalt binders is the use of polymer-modified asphalt binders in pavements that are exposed to heavy loading conditions or severe climatic environments.

Although several advantages of polymer-modified asphalt binders have been reported, the chemical and mechanical behaviors corresponding to physical benefits are still ambiguous. The recently developed SBS polymer-modified asphalt may be a good alternative to regular asphalt in situations requiring increased HMA strength at high temperatures and good resistance to rutting. However, the chemical and mechanical degradation of the binder and SBS modifier occur simultaneously due to aging and oxidation. Simultaneous aging and oxidation may contribute to the loss of molecular cross-linking in a polymer-modified binder. The degradation of SBS leads to the loss of the molecular network structure and results in a nearly identical softening point to that of a plain binder after aging in a pressure aging vessel for 500 h [4].

Even ultraviolet light has sufficient energy to break the carbon double bond (C=C) in a SBS backbone by splitting it into short chains; thus, a polymer containing C=C, such as styrene monomer, may be more brittle, causing the rubber to lose its flexibility [9].

The carbon double bond in the vinyl group of a styrene monomer can be replaced by a much stronger carbon single bond (C–C) through a polymerization process. Polystyrene is then very hard to depolymerize; however, in processing this modification, verification of the occurrence of the polymerization is difficult without X-ray diffraction (XRD) or Fourier transform infrared (FTIR) analysis. Some styrene monomer may exist after the modification process that can be easily bonded with oxygen molecules, expediting the aging process of the modified binder.

2.2. DMP and HIPS solution

To overcome these limitations in SBS-modified asphalt binders, HIPS, which is relatively inexpensive compared with SBS and is recycled from plastic waste, as shown in Fig. 1, was selected as an asphalt modifier in this study. HIPS is a thermoplastic polymer that can be recycled; it is also polymerized without a carbon double bond.

During the course of the polymerization reaction of the styrene monomer, polymerized polybutadiene is added to the reactor to obtain the grafted copolymer of HIPS. If polybutadiene is added during polymerization, it can become chemically bonded to polystyrene, which forms a graft copolymer that helps incorporate normal polybutadiene into the final mix and results in HIPS [10].

The conceptual diagram of the graft copolymer is shown in Fig. 2a. HIPS contains approximately 60–70% polystyrene and displays a phase of spherical polystyrene domains that extends through the elastomeric phase of approximately 30–40% polybutadiene, as shown in Fig. 2b.

The polybutadiene phase gives HIPS elastic features. The two phases are not compatible with each other. Thus, on the molecular scale, the two phases separate from each other, leading polybutadiene to take its own domain in the polymeric structures, as shown in Fig. 2b. The spherical polystyrene and elastomeric polybutadiene give the polymer elastic-recovery features against external loading. The polybutadiene domain gives rubber-like elasticity to the graft copolymer of HIPS, providing the HIPS-modified asphalt binder with enhanced elastomeric recovery features at temperatures above 40 °C [10].



Fig. 1. Recycled HIPS particles.

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