

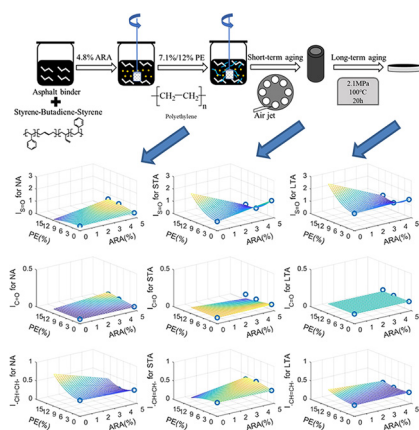


Full Length Article

Chemical, thermal and rheological characteristics of composite polymerized asphalts

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GRAPHICAL ABSTRACT



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ABSTRACT

Thermal oxidation has a significant effect on rheological and chemical properties of composite polymerized asphalt. In this study, polyethylenes (PE) and a multi-polymer (a type of anti-rutting agent) were incorporated into asphalt binders at four contents and the properties of polymer-asphalt composite systems were investigated through chemical functional group, molecular size, thermal behavior and rheological characteristics. Due to the formation of large conjugate structure by thermal oxidation, the carbonyl peak shifted to a lower wavenumber after a long-term aging procedure and the aging procedure increased the values of large molecular sizes. However, the degradation of PE played a dominant role during oxidation progress, which significantly decreased the LMS value of composite binders. Therefore the aging process changed the chemical compositions and element component distribution of binders and these changes were influenced by both binder source and polymer contents. In addition, with the increase of polymer modification, the penetration value and phase angle value at low frequency decreased; the endothermic peak area, softening point as well as complex shear modulus at low frequency increased. Therefore, the polymer degradation and polycyclic aromatic hydrocarbon formation should be considered during the aging process, which influenced the rheological performance of composite polymerized asphalts together. Furthermore, the correlation analysis showed that the carbonyl structure index $I_{C=O}$ has significant correlations with the difference between glassy modulus and equilibrium modulus, and phase angle master curve plateau δP , the LMS value has significant relationships with phase angle master curve factors δL and $\delta P + \delta L$ at a low frequency.

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1. Introduction

Asphalt material has been widely used in airfield and high-grade road pavement due to its numerous advantages such as smoothness, low-vibration, high-automated construction and easy-maintenance [1]. Asphalt binder is a composite material obtained from petroleum refinery, about 90 ~ 95% by weight of asphalt is composed of aliphatic, aromatic and naphthenic hydrocarbons and their chemical compositions. However, these chemical compositions cannot be defined exactly because they are extremely complex and variable, depending on the crude oil source and refining process [2,3]. In addition, due to the similarity between asphalt and polymer in nature, many test and analysis methodologies for asphalt have been employed [4]. With that in mind, the physical and chemical characteristics of asphalt binder are influenced by the interactions and cross links between organic molecules. Also, the organic molecules in asphalt binder can be oxidized by the environment air, which gradually changes the characteristics of the asphalt binder [5,6].

Following the rapid economic development, the increasing traffic load results in various severe distresses of asphalt pavement, especially high-temperature permanent deformation and ambient-temperature fatigue cracking [7]. The imperious demands of high quality asphalt binder and the limitation of refining process develop the research of polymer reinforced or modified asphalt binders [3]. Polymer modification is defined as the incorporation of polymers in asphalt by mechanical mixing or chemical reaction [8]. There are different polymer modifiers used to achieve the improved performance from original asphalt binder, although none of these are used for asphalt modification initially. The mainly used polymer modifiers include elastomers, plastomers, and resin [9].

Elastomers increase the elastic property of asphalt binder, and result in the increased complex modulus at high temperatures, the decreased complex modulus at low temperatures, the broadened relaxation range and the improved ductility [10]. These additives such as styrene-butadiene styrene triblock copolymer (SBS) reduce the fatigue and thermal cracking distresses of asphalt pavement [11], but these are quite expensive and easily subjected to degradation when exposed to mechanical stress and environment air [12,13]. The plastomers such as polyethylenes (PE) increase the consistency, resilient modulus, and rutting resistance of asphalt materials [14,15]. Based on the molecular weight and polymerization process, the PE polymer can be classified into several types and each has different rheological performances. Most importantly, the PE polymers can be recycled from waste plastics, which are relatively cheap and environmental friendly.

In addition, a synthetic modifier named Anti-Rutting Agent (ARA) has been used in some European countries and China to improve the rutting resistance of hot mix asphalt (HMA) [16–18]. This type of modifier is composed of synthetic rubber, polyethylenes, and lignin fiber. The main advantage of this modifier is to enhance the resistances of permanent deformations and fatigue cracking [19]. However, the content of polymer has a significant influence on binder modification. During the mixing progress, the hot asphalt binder penetrates the polymer particles, where long-chain molecules become solvated and/or swollen [20,21]. The two interlocked continuous phases of polymer modified binder in microstructure is an optimum polymer content [22,23]. For example, the optimal dosage of ARA is about 0.3% (by the weight of asphalt mixture) and the excess amount of ARA can lead to low fatigue life and poor low-temperature performance [24,25]. The reason for this trend is that the different properties such as density, molecular weight, solubility and polarity between polymers and asphalt binder result in poor compatibility and storage stability [26].

Previous studies have found that the oxidation procedure hardened the virgin asphalt binder and degraded polymer modifiers into smaller molecules [27]. Generally, the oxidation of asphalt binder increases its resistance to permanent deformation and reduces its flexibility, resulting in the formation of cracking under repeated traffic loading [18].

Gravari also found that the effect of polymer on increasing complex modulus and on reducing phase angle of aged asphalt was less pronounced compared with the unaged binders, indicating the degradation of polymer during the oxidation process [6]. Also, the molecules in asphalt binder can be affected by the oxidation level and molecular size changes during aging depend on temperature, modifier type and modification level. The oxidation also changes the structure and composition of asphalt binder [28].

Multiscale research of asphalt binder is of interest to fully understand the behavior of this composite material [29]. The molecular size and its distribution influence the chemical structure of the polymer modifier and finally changes the rheological properties of asphalt-polymer composite [23]. Also, the rheological properties of asphalt binder directly determine the performance of pavement [30]. Since a modified binder consists of two distinct modifiers have been studied by researchers, the modification mechanism and effect of three different materials should be considered [31]. Plastomers such as PE and ARA show great high-temperature performances but have some limitations in elasticity when incorporated into asphalt [3]. The elastomers such as SBS polymer have good elasticity but have a limitation to improve high-temperature performance [32]. Therefore, combining PE and ARA with SBS to form an asphalt-polymers composite system will be an effective method to enhance the properties of high-grade asphalt pavement, especially for heavy load pavement.

This study investigates the influence of variable contents of ARA and PE on the polymerized asphalts. The low and high temperature rheological properties of the SBS based asphalt binders are used as the reference to explore the properties of these PE and ARA asphalts. Several empirical indices such as penetration depths and softening points as well as master curves of complex shear modulus and phase angle value are conducted. In addition, the chemical characterizations are investigated by Fourier Transform Infrared Spectroscopy (FTIR) and Gel Permeation Chromatography (GPC). Moreover, the Thermogravimetry/ Differential Scanning Calorimetry (TG/DSC) tests are used to determine thermal behaviors of these modified asphalts.

2. Materials characteristics and testing methods

2.1. Material characterization

To prepare polymer reinforced asphalt binders, two PG 76-22 SBS modified asphalt binders were used as per AASHTO M320 [33], referred as A and B. The basic properties of original binders and two polymers are shown in Table 1.

2.2. Specimens preparation

The diagrams of specimen preparation and two aging processes are shown in Fig. 1. A high-speed shearing machine was used to produce laboratory blended binders from base asphalt binder and two polymers. First, base binders were heated up to 170 °C to become a sufficiently fluid state. Then, a certain additive was added slowly (5 g/min) to base binder in metal containers and blended for an hour at a shearing speed of 5500 ~ 6500 rpm and a temperature of 170 ~ 180 °C. After that, the blended binder was stored at 150 °C for another hour to achieve a homogeneous modified binder. The shearing speed and mixing temperature were determined by the properties of polymers. After mixed, the polymerized asphalt binder was poured into a small sealed can and then stored in a dark chamber thermostated at 25 °C to retain the obtained morphology.

In this study, three aging states including no aging (NA), short-term aging (STA) as well as long-term aging (LTA) were considered to evaluate the influence of oxidation on the properties of these composite asphalts. As shown in Fig. 1, according to ASTM D2872-04, the short-term aging was simulated by Rolling Thin-Film Oven Test (RTFOT). Asphalt binder was heated in an oven for 85 min at 163 °C under the air

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