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Thermal analysis on the interactions among asphalt modified with SBS and different degraded tire rubber

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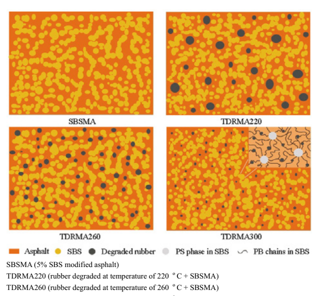
HIGHLIGHTS

- T_g and elastic modulus of the hybrid modified asphalts varied with degraded rubber.
- Increased degradation leads to the sub-millimeter, micro and nano-sized dispersion of CR in SBSMA.
- Increased degradation of crumb rubber improves its compatibility with SBS modified asphalt.
- The schematic of degraded rubber and SBSMA is presented by thermal and morphological studies.

GRAPHICAL ABSTRACT



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ABSTRACT

The cross-linked network present in crumb rubber (CR) limits its uniform dispersion in styrene-butadienestyrene modified asphalt (SBSMA). For this purpose, CR was degraded using a twin screw extruder to improve its dispersion properties in SBSMA. Different degraded rubbers (DR) prepared at different degradation temperatures were blended with SBSMA. Interaction in asphalt, SBS and degraded rubber were analyzed and compared using thermo-gravimetric analysis (TGA), differential scanning calorimetry (DSC) and dynamic mechanical analysis (DMA). The degradation behavior, glass transition temperature and the low temperature rheological properties of the modified asphalt were evaluated. The results indicate that the degree of degradation in the CR affects its dispersion and interactions with SBSMA, which further influences the performance of the modified asphalts. The morphological analysis explains the mechanism of interactions within the degraded rubber, asphalt and SBS, specifies that the modification of the asphalt occurs *via* the exchange of materials among these three components.

1. Introduction

Asphalt modifiers have been used to improve roadway performance and reduce different types of pavement stresses such as low temperature cracking, rutting, stripping and fatigue cracking associated with load [1]. Extensive research in most recent years has paid attention on improving the usage properties of the asphalt. The introduction of synthetic polymers *via* physical or chemical blending has been shown to significantly improve the performance of base asphalts. The conditions for selecting a polymer additive are: (i) the asphalt's viscosity should not be considerably changed. (ii) compatible with the asphalt so as to prevent phase separation in the course of application at different temperatures). (iii) mechanical properties of the asphalt must be improved such as resistance to high temperature, fracture and permanent deformation. The polymer additive should be capable to improve the low temperature plasticity or flexibility and lower the glass transition temperature (T_g) of the asphalt in low temperature [2]. Many polymers such as styrene butadiene rubber (SBR),

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styrene-butadienestyrene block copolymers (SBS) and ethylene vinyl acetate copolymers (EVA) are increasingly being applied in asphalt modification. The exact quantity of polymer's addition depends on the desired properties. For example, 6% of SBS is enough to produce a continuous phase in the asphalt which acts as an elastic element and intensely limits crack propagation in the asphalt mix [3].

Generally, the base asphalt possesses a glass transition temperature (T_g) around −20 °C. It can be detected that the transition of the asphalt from a viscoelastic nature to a Newtonian fluid is above 60 °C. During the service life, pavement temperatures are between −30 °C and 60 °C (apart from extreme weathers, where the temperatures can be as low as −40 °C and as high as 80 °C), and paving asphalt is typically characterized by its dynamic functions [4]. As a result of the addition of some polymers, an obvious increase in complex modulus (G^*) and decrease in phase angle is attained at high temperature and lower angular frequency [5]. Together, the storage and loss modulus of the asphalt increase by increasing the polymer content.

SBS is a special polymer exhibiting elastomer and plastic type properties and is widely used as an asphalt modifier all over the world [6,7]. It is composed of hard polystyrene (PS) domains linked by poly-butadiene (PB) segments, which shows a diphase morphology [8,9]. PS domains has T_g around 95 °C while that of PB is around −80 °C [10]. Several investigators tried to examine the microstructure and road performance of SBSMA [11–13]. The results indicate that SBSMA significantly enhances the fatigue cracking as well as low temperature performance [14,15]. So SBS is most favorable modifier for the asphalt. Aging resistance and high cost also presents challenge for SBSMA. Extensive research has been done on using waste materials as asphalt modifiers, such as waste plastic, waste tire rubber, waste fibers and tall oil pitch [16,17]. Hence, use of waste materials is an economical method for the production of modified asphalt.

Crumb rubber (CR) obtained from waste tire has gained more attention for use in the asphalt modification. The performance of CR in asphalt is influenced by numerous factors, such as the properties of the base asphalt, rubber size, rubber content and mixing conditions. CR modified asphalt shows improved mechanical properties by decreasing fatigue cracking, reflective cracking and by enhancing the durability of the pavement [18,19]. However, characteristic properties and poor processing ability due to inherent chemical crosslinking prevent it from large scale use [20]. When CR is degraded or reclaimed; it can be well dispersed into the asphalt and can even be used as a modifier of asphalt [21].

The degradation process involves the use of mechanical, thermal or chemical energy. When CR is degraded extremely by thermo-mechanical extrusion method, the three dimensional network of the cross-linked macromolecule rubber shortens which decreases the rubber's viscosity. The micro-sized rubber particles can be easily compatible with the asphalt compounds [22]. For high performance roadways, modified asphalt has been prepared *via* the addition of modifier composites including CR and SBS, which has better low and high temperature performance [23]. Though substantial development has been attained in asphalt-rubber application in the last four decades, the interaction relating to CR, SBS and base asphalt is not quite clear. This might be, due to the crosslinking characteristic of CR and the complicated composition of SBS modified asphalt [24].

Thermal analysis is a technique that can offer some understanding as to why some asphalts and modified asphalts do not perform properly. The application of thermal analysis to asphalt relies on the fact that they suffer physiochemical changes upon heating [25]. The interaction between different materials can be detected using this tool. The thermal stability of asphalt is a key property to consider for its proper application. The thermal strength of various blends is highly dependent on their compatibility [26]. Thermo-gravimeter (TG) can be used to monitor the process of degradation at high temperature and to study the kinetics. It is proposed that the outcome properties and parameters related with thermal degradation should help to reinforce more studies in this area of research. Differential scanning calorimetry (DSC) and dynamic mechanical analysis (DMA) can be used consistently to determine the glass transition temperatures (T_g). Collectively, both TG and DSC can provide a degree of the degradative features which can lead to propose an interaction mechanism between CR and SBSMA.

Several studies that investigate the effects of CR on the physical and rheological properties of SBS modified asphalt have been reported. Studies concerning the effect of highly reclaimed rubber or degraded rubber (DR) on the physical properties of SBS modified asphalt have also been carried out previously [27]. However, very few studies have investigated the mechanism of interaction and dispersion state of different degraded rubbers with SBS modified asphalt. To address this problem, the aim of our present research is to investigate the dispersion state of different degraded rubbers (that has been reclaimed at high temperature) on the thermal behavior of DR/SBS blend modified asphalt. The DR/SBS modified asphalt composites are envisaged to be able to improve the workability and physical properties of asphalt. For this purpose, CR from waste tires was degraded at high temperature and then used as a future asphalt alternative or modifier and the cost of high viscosity asphalt can be reduced.

To gain more understanding of the interactions between DR, SBS and asphalt, DR/SBS modified asphalt (DRMAs) has been thermally analyzed in this study. Thermo gravimetric analysis (TGA/DTG) and differential scanning calorimetric analysis (DSC) were carried out to study the thermal behavior of DRMAs. Dynamic mechanical analysis (DMA) was conducted to evaluate the compatibility between DR and SBSMA by the rheological compatibility criteria. In the meantime, the microstructure of different DRs in SBSMA was analyzed with the help of fluorescent microscope in order to report a detail about interaction of DR and SBS with asphaltic components. Finally, this research is focused on discovering an inexpensive and environment friendly preparation of high performance modified asphalt.

2. Experimental and methodology

2.1. Materials

Asphalt was provided by China Ocean Oil Corp, and its conventional physical properties are presented in Table1. The recycled engine oil having viscosity of 46 centi-poises (25 °C) was used as processing oil. The SBS selected in this research is a linear structure polymer, with 30 wt% styrene content and average molecular weight of 6×10^4 g/mol, was supplied by Petrochina, Co., Ltd. Crumb rubber (40 mesh) was ground rubber derived from whole truck tire (65% natural rubber, 35% synthetic rubber) and car tire (35% natural rubber, 65% synthetic rubber) prepared at room temperature.

Table 1
Conventional performance index of original asphalt (OA).

Property (units)	Value	Standard test used
Penetration (dmm)	71	ASTM D 5 (25 °C, 100 g, 5 s)
Softening point (°C)	45.5	ASTM 36
Viscosity (Pa. s)	0.201	ASTM D 4402-87 (135 °C)
Ductility (cm)	Over 100	ASTM D113 (25 °C)