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Physical and rheological properties of epoxidized natural rubber modified bitumens

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HIGHLIGHTS

- Adding ENR to bitumen decreased bitumen temperature susceptibility.
- ENR modified bitumen increased in the complex modulus and decreased in the phase angle.
- ENR modified bitumen decreased the rutting for the binder and improved the fatigue behavior at low temperatures.

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ABSTRACT

The use of polymers as bitumen modifiers in pavements has been growing rapidly in the last decades because of the distresses on asphalt pavements. However, using polymers as a modifier mostly changes the properties of the asphalt pavement to be more stable and stiffer at high temperatures and more flexible at low temperatures. Therefore, this study was conducted to investigate the empirical and rheological properties of bitumen modified with epoxidized natural rubber (ENR). Four percentages (3%, 6%, 9% and 12%) of epoxidized natural rubber were used as modifiers. The effects of the modifier on the conventional properties, storage stability and rheological properties were investigated. Requirement tests (e.g., penetration, softening point, ductility and viscosity) and rheological analysis (e.g., isochronal plot, master curves, Black diagram and SHRP parameters) using a dynamic shear rheometer (DSR) were conducted to characterize the ENR-modified bitumen. The fundamental parameters were used to describe significant benefits of ENR as a modifier. The results indicated that the storage stability of epoxidized natural rubber modified bitumens (ENRMBs) depended mainly on the ENR content. Based on the results obtained from the DSR test, ENR reduces the temperature susceptibility and facilitated polymeric modification using a highly elastic network within the bitumen. This elastic network increases the viscosity, stiffness and elastic behavior of the ENRMBs. ENR improves rutting resistance and fatigue behavior at high temperatures and low temperatures respectively. The best results were recorded for polymer-modified bitumen containing 6% of ENR.

1. Introduction

Over the past decades, research on polymer-modified bitumens (PMBs) and its benefits has dramatically increased and has been reported in many studies [1–3]. The use of PMBs can improve the performance of asphalt mixtures and substantially increase the service life of highway surfaces. However, the addition of polymers significantly improves various bitumen properties, such as elasticity, cohesion, stiffness and adhesion, thus resulting in an overall performance improvement of asphalt pavements, which become more stable and stiffer at high temperatures and more flexible at low temperatures. In addition to rutting resistance, a good polymer can supply a degree of flexibility or elasticity to an asphalt mixture, thereby improving the fatigue and thermal cracking properties of the asphalt mixture [4–6].

One of the most well-known, widely used category of polymers is thermoplastic elastomers (TE). Thermoplastic elastomers (TE) are polymers with thermoplastic and elastomeric properties [7]. TE polymers derive their strength and elasticity from a physical

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cross-linking of molecules within a three-dimensional network. Styrene–butadiene–styrene (SBS) is the most commonly known TE that can improve the rheological properties of bitumen [8]. Furthermore, SBS increases binder elasticity at high temperatures and improves flexibility at low temperatures. This improvement led to increase the resistance to asphalt rutting at high temperatures, and decrease cracking at low temperatures [9]. Styrene–butadiene–rubber (SBR) is another example of an elastomer polymer and it increases the ductility of asphalt pavement and consequently becomes more flexible and crack-resistant at low temperatures [10]. In addition, other types of additives have also been used as modifiers, i.e., as TE, such as styrene isoprene styrene (SIS), styrene ethylene butadiene styrene (SEBS), ethylene–propylene–diene-terpolymer (EPDM), isobutene–isoprene-copolymer (IIP), crumb-rubber, polybutadiene (PBD), polyisoprene and natural rubber (NR) [11,12]. However, the major types of styrenic block copolymers such as SBS, SIS and hydrogenated styrenic block copolymers (HSBC) have the best modifying potential when blended with bitumen [13–15].

In some cases, the rubbers have been used in a vulcanized state, e.g., crumb-rubber, but are difficult to disperse throughout the bitumen. Ineffective dispersion requires high temperatures and long mixing times and can yield a heterogeneous binder, with the rubber acting mainly as flexible filler [11]. However, crumb-rubber is a combination of mainly natural rubber (improves the elasticity of the compound), carbon black and synthetic rubber (both of which materials improve the thermal stability properties). In addition, crumb-rubber has been found to improve rutting resistance and to decrease reflective cracking [16]. However, it is found that natural rubber shows better reactivity compared to crumb-rubber [17]. The reacted particles become tacky and improve adhesion properties. The cohesion between aggregates, thus holding the aggregate together, is one advantage of using natural rubber to modify bitumen [18]. Natural rubber increases the stiffness of the binder, thereby enhancing the latter's performance at high temperatures but rendering it brittleness at low temperatures [19]. Natural rubber displays high mechanical strength, outstanding resilience and excellent elasticity. However, natural rubber is known to exhibit poor wet grip properties and poor weather resistance [20].

The potential application of epoxidized natural rubber (ENR) as a modifier was realized in the 1980s [20]. ENR is a chemically modified natural rubber created by reacting natural rubber with proxy formic acid, as shown in Fig. 1 [20]. This material has a good properties, offering high strength because of its ability to bear strain crystallization, along with increased glass transition temperature. These properties facilitate increased oil resistance, enhanced adhesion properties, damping and reduced gas permeation [21,22]. As a modifier, ENR is able to increase the complex viscosity, the storage and the loss modulus of the blends [23]. However, the details of ENR preparation, are reported in [24–26] and not discussed in this paper for the sake of brevity.

This study was conducted to present a laboratory evaluation of base bitumen and ENR modified bitumen (ENRMB) in terms of conventional properties, temperature susceptibility, storage stability and rheological properties. The ENRMBs were produced by laboratory mixing using four polymer contents. The rheological properties (viscoelasticity) of the ENRMBs were determined by means of an oscillatory type testing of dynamic mechanical analysis (DMA), generally conducted within the region of linear viscoelastic response. DMA allows the viscoelastic nature of bitumen to be determined over a wide range of temperatures and loading times (or frequencies) [27–32].

2. Experimental works

2.1. Materials

The bitumen used in this study is 80/100 penetration grade, supplied by the bitumen factory at Port Klang, Malaysia. The ENR was obtained from the Malaysian Rubber Board under the trade name of ENR 50, with 53% epoxidization and passed through 2.36 mm mesh sieve (before shearing). The physical properties of the bitumen and the ENR used are shown in Table 1.

2.2. Preparation of binders

Five ENRMBs were produced by mixing ENR-50 (0%, 3%, 6%, 9% and 12% content by weight of bitumen) with base bitumen. All ENRMBs were prepared using a Silverson high shear mixer at 160 °C (±1 °C) under 4000 rpm of speed for one hour. ENR was added to the bitumen after the temperature stabilized at 160 °C. The different ENRMBs were coded as ENR (polymer), MB (modified bitumen) and (ENR content), e.g., ENRMB3 corresponds to the bitumen modified with 3% epoxidized natural rubber content (by weight).

2.3. Conventional binder tests

According to ASTM standards, many conventional tests were conducted on the base bitumen and the modified bitumen, such as penetration, softening point, ductility, viscosity (using the Brookfield Model DV-III) and storage stability test [33–36]. The penetration test is used as a measure of consistency. Higher values of penetration indicate softer consistency. The sample is melted and then cooled to 25 ± 0.5 °C. The penetration is measured with a penetrometer by means of which a standard needle is applied to the sample at a temperature of 25 ± 0.5 °C [33]. In the softening point test, two horizontal disks of bitumen, cast in shouldered brass rings, are heated at a controlled rate in a liquid bath while each supports a steel ball. The softening point is reported as the mean temperatures at which the two disks soften sufficiently to allow each ball, enveloped in bitumen, to fall 25 mm [34]. Ductility testing provides one measure of tensile properties of bituminous materials. In a ductility test, bitumen is stretched in a water bath lower than 10 °C and at a stretching velocity of 5 cm/min. Ductility length is measured after the sample is cut [36].

2.4. Temperature susceptibility

Changing the temperature susceptibility with ENR was investigated with a calculating penetration index (PI) and a penetration viscosity number (PVN). The higher the PI and PVN values of bitumen, the lower is its temperature susceptibility. For PI, the temperature susceptibility for the binders is measured by calculating the PI using the penetration at 25 °C and softening point results.

Penetration index is calculated using an equation, shows as follows [11]:

$$\mathrm{PI} = \frac{1952 - 500\log \mathrm{Pen} - 20\mathrm{S.P}}{50\log \mathrm{Pen} - \mathrm{S.P} - 120} \quad (1)$$

where Pen is the penetration test at 25 °C and S.P is the softening point.

The penetration viscosity number (PVN) is calculated based on penetration at 25 °C and viscosity at 135 °C, such that

$$\mathrm{PVN} = \frac{\log L - \log X}{\log L - \log M} \times 1.5 \quad (2)$$

where PVN is the penetration viscosity number, L is the logarithm of viscosity at 135 °C for PVN of 0.0, X is the logarithm of viscosity at 135 °C, and M is the logarithm of viscosity at 135 °C for a PVN of −1.5.

The viscosity values of L and M can be determined using the following equations. The equation for the line representing a PVN of 0.0 is:

$$L = 4.25800 - 0.79670\log \mathrm{Pen} \quad (3)$$

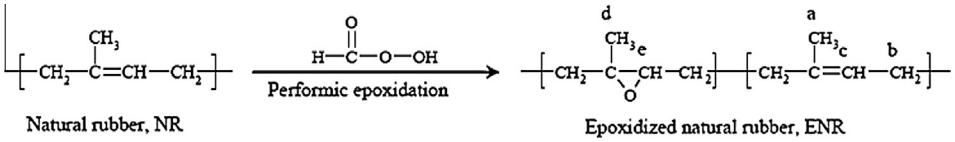

Fig. 1. Performic epoxidation of NR.