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Rheological characteristics of unaged and aged epoxidised natural rubber modified asphalt

Ramez A. Al-Mansob [a,b], Amiruddin Ismail [a,b,*], Nur Izzi Md. Yusoff [a,b], Shaban Ismael Albrka [a,b], Che Husna Azhari [c], Mohamed Rehan Karim [d]

[a] Sustainable Urban Transport Research Centre (SUTRA), Universiti Kebangsaan Malaysia, 43600 UKM Bangi, Selangor, Malaysia
[b] Dept. of Civil and Structural Engineering, Universiti Kebangsaan Malaysia, 43600 UKM Bangi, Selangor, Malaysia
[c] Dept. of Mechanical and Materials Engineering, Universiti Kebangsaan Malaysia, 43600 UKM Bangi, Selangor, Malaysia
[d] Dept. of Civil Engineering, Universiti Malaya, Jalan Lembah Pantai, 59100 Kuala Lumpur, Malaysia

HIGHLIGHTS

- ENR increased asphalt stiffness and elasticity behaviour before and after ageing.
- ENR modified asphalt was affected by the ageing.
- ENR decreased effect of ageing on the rheological properties of binders.

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ABSTRACT

Durable pavement with long service life and low maintenance and rehabilitation costs are preferred; hence, the demand for high-quality asphalt is growing. This situation has led to the research and development of new materials with increased performance, such as polymer-modified asphalt (PMA). This study was conducted to investigate the use of epoxidised natural rubber (ENR) mixed with asphalt, both unaged and aged. The physical and rheological properties of base asphalt and ENR-modified asphalt (ENRMAs) were measured. Fundamental parameters were used to describe the significant benefits of ENR as a modifier. However, it was observed that the rheological properties differ considerably between the base asphalt and ENRMAs. The rheological parameters of complex modulus and phase angle indicate that the presence of ENR increases the stiffness and elasticity behaviour of the binder, particularly at high temperatures. For the aged binders, the dynamic mechanical analysis of the effect of ageing on the rheological performance of the binders indicated that there is a considerable difference in behaviour between the base asphalt and PMAs. Based on the dynamic mechanical analysis of the binders, it can be concluded that the asphalt component of the modified binders ages in a manner similar to that of the base asphalt. However, the rheological changes that occur after ageing differ from the base asphalt in regions where the polymer is the dominant component. Finally based on the physical and rheological tests, it can be concluded that ENR can increase the durability of asphalt pavements.

1. Introduction

The use of polymers as asphalt modifiers is not new. As early as 1823, an English cork manufacturer was granted a patent for a binder containing natural rubber [1]. After the Second World War, synthetic polymer began to compete with natural rubber as an additive in asphalt pavements. Over the years, an increased interest in asphalt modification using different types of synthetic polymers has been observed in many countries [2–15].

It was reported that the incremental life cycle cost analysis showed that polymer-modified asphalt (PMA) is cost-effective, providing that the cost of the PMA does not exceed the cost of the base asphalt by more than 100% [16]. PMAs are selected, therefore, to reduce life cycle costs in many cases. Note that binders often require modifications to achieve high temperature resistance to rutting and low temperature resistance to thermal cracking. An

* Corresponding author at: Sustainable Urban Transport Research Centre (SUTRA), Universiti Kebangsaan Malaysia, 43600 UKM Bangi, Selangor, Malaysia

E-mail addresses: Ramizizzi@gmail.com (R.A. Al-Mansob), aismail@ukm.edu.my (A. Ismail), izzi.alkutani@gmail.com (Nur Izzi Md. Yusoff), shabarofking10@gmail.com (S.I. Albrka), husna.azhari@gmail.com (C.H. Azhari), rehan@um.edu.my (M.R. Karim).

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increase in the use of PMAs for specific applications in the near future is predicted because of their outstanding performance. However, ideal asphalt modifiers would contribute to the followings properties [17,18]:

- Higher stiffness at high temperatures to reduce rutting and shoving.
- Lower stiffness at cold temperatures to reduce cracking.
- Improved adhesion of asphalts to aggregates in the presence of moisture to reduce stripping.

The ideal behaviour of the binder is only theoretical and cannot be achieved in practice [19]. The major real advantage in using PMA is its ability to change the rheological properties of asphalts. Polymer modification leads to significant changes in the stress–strain behaviour, as well as in the creep response. The ability of some polymers to elastically recover provides added durability to asphalts and expands the range of service temperatures.

Asphalt technologists have recognised that while the physical properties of some asphalt binders may be the same initially, the oxidation and volatilisation that occurs during use in a hot mix facility and subsequent placement on the road may lead to a difference in service performance. Stated more simply, not all asphalt binders age the same, and this difference in ageing may result in a difference in actual performance [18,20,21]. Ageing procedures were developed to subject an asphalt binder sample to hardening conditions that approximate the conditions that occur in normal, hot mix facility operations. The thin film oven tests (TFOT) and rolling thin film oven test (RTFOT) are two common short-term ageing procedures, and the pressure-ageing vessel (PAV) is used as a long-term ageing procedure. Comparing with TFOT, RTFOT more accurately simulates the asphalt mixing conditions [17]. However, the performance graded (PG) asphalt binder specifications now requires the use of the RTFOT [22–24].

RTFOT and TFOT ageing causes oxidation of the asphalt and degradation of the polymer, and consequently change the microstructure and rheological properties of the modified binders. These changes largely depend on the characteristics of the polymer used and its content. For the modified binders with styrene–butadiene–styrene (SBS), ageing increases the complex modulus and elastic response (decreased phase angle) and reduces the temperature susceptibility. On the other hand, for the modified binders with a sufficiently high content of SBS, the influence of ageing on those parameters is strongly dependent on the temperature and frequency [25,26]. According to a previous study, styrene butadiene rubber- (SBR-) modified asphalt improved the elasticity, adhesion, and cohesion and reduced the age-hardening [27].

Epoxidised natural rubber (ENR) is a chemical modification product of natural rubber (NR). Reacting natural rubber with peroxy formic acid can produce ENR as shown in Fig. 1. ENR-50 is the integer designate 50-mol% of epoxide incorporated into the natural rubber chain. However, the potential application of ENR as a modifier was realised in the 1980s [1].

In general, ENR has 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 [2,3]. As a modifier, ENR is able to increase the complex viscosity, the storage and the loss modulus of the blends [4]. In addition, the epoxide groups bonded to rubber polymer chains influence rubber properties such as improve resistance to swelling in oils, modify adhesion properties to other materials, improve their resistance against penetration of gases and vapours [5]. On the other hand, it was found that epoxidised natural rubber- (ENR)- modified asphalt (ENRMA) improved the temperature susceptibility and increased the viscosity, stiffness, and elastic behaviour [28–30]. Moreover, based on the storage stability and the rheological properties, it was found that the optimum content of ENR as a modifier is 6% by weight [28].

2. Experimental program

2.1. Asphalt modification (blending)

The blending of ENR and asphalt was carried out using a high-shear mixer. Table 1 summarises the mixing parameters used in the blending procedure. All of the samples, differing in terms of mixing time, were tested to determine the viscosity of the binder. This protocol was set up to attempt to obtain a dispersive mixing of asphalt and the ENR by applying a constant, high-shear strain (4000 rpm) for two hours. In order to understand the modification process, every 20 min, a sample of approximately 100 g of binder was taken and stored for the viscosity test. ENR particles react with the asphalt improving its stiffness and elastic properties until a peak performance is reached. In practical applications of binders, this peak is considered to be achieved when the apparent viscosity, after a constant increasing trend, reaches a plateau level [31].

2.2. Penetration test

The penetration test is a physical test that measures binder consistency at intermediate service temperatures. The penetration is defined as the distance in tenths of a millimetre that a standard needle penetrates into the sample under given conditions of loading, time, and temperature at 100 g, 5 s, and 25 °C, respectively. The value of the penetration depends on the hardness or softness of the binders. The higher the penetration value, the softer the binder is. The test was conducted in accordance with ASTM D5 [32].

2.3. Ring and Ball Softening Point test

The Ring and Ball Softening Point test ($T_{R\&B}$) is also a physical test that determines the temperature at which a phase change occurs in the binder. The test is used to measure the temperature at which two horizontal disks of asphalt, cast in shouldered brass rings, are heated at a controlled rate in a liquid bath while each disk supports a steel ball. The softening point is reported as the mean of the temperatures at which the two disks soften enough to allow each ball, enveloped in asphalt, to fall a distance of 25 mm. The test was conducted in accordance to ASTM D36 [33].

2.4. Ductility

The ductility of asphalt binders is considered as an indirect measure of the tensile properties of the material. This test measures the distance in centimetres to which the sample can be elongated before breaking when two ends of a briquette sample of the material are pulled apart at a specified temperature and speed. In this test, the sample was melted and poured into a brass mould and allowed to cool to room temperature, and then the mould was transferred to a water bath at a specified temperature. In this study, two different temperatures were used to perform this test, 25 °C and 10 °C. Samples were maintained at the specific temperature by adding very cold water to the ductility bath before and during testing. The two ends of the test sample were then pulled apart at a speed of 5 cm/min. The test was conducted in accordance with ASTM D113 [34].

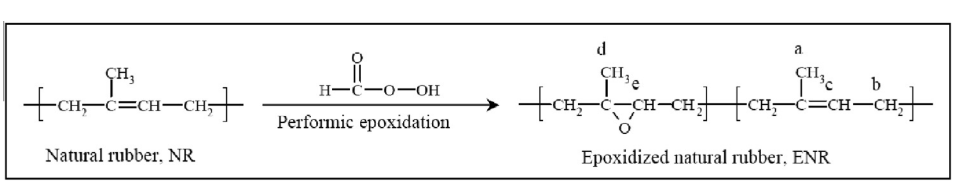

Fig. 1. Performic epoxidation of NR.