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## Evolution of components distribution and its effect on low temperature properties of terminal blend rubberized asphalt binder



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### HIGHLIGHTS

- Evolution of molecular components was monitored in the degradation process.
- Degradation of crumb rubber affects the low temperature stiffness negatively.
- Relaxation property of is improved with the degradation.
- Relaxation property becomes more temperature sensitive with the degradation.

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### ABSTRACT

The addition of crumb rubber modifier (CRM) could enhance the low temperature properties of asphalt binder. However, in the production of terminal blend (TB) rubberized asphalt binder, the degradation of CRM results in the change of molecular components of TB rubberized asphalt binder. The effect of components evolution on low temperature rheological behaviour of TB rubberized asphalt binder was not clear yet. The present study aims to investigate the evolution of components distribution and its effect on low temperature properties of TB rubberized asphalt binders produced under various interaction temperatures and interaction times. The solubility test and the Gel Permeation Chromatography (GPC) test were performed to obtain the distribution of insoluble components, dissolved polymers, apparent asphaltenes and maltenes in various TB rubberized asphalt binder. Bending Beam Rheometer (BBR) was conducted to investigate the low temperature properties of TB rubberized asphalt binder before and after aging. Burgers model was used to retrieve the relaxation time at low temperature. The results indicated that interaction time plays a more important role in promoting the dissolution of CRM whereas the interaction temperature is more responsible for the breaking of backbone of the main chain of rubber. Low temperature stiffness of TB rubberized asphalt binder increased with the degradation of CRM, which has a negative effect on low temperature performance. Relaxation property of TB rubberized asphalt binder was improved with the increase in percentage of apparent asphaltenes. However, the loss of relaxation property of TB rubberized asphalt binder after aging is more remarkable with the gradual degradation of CRM. TB rubberized asphalt binder investigated in the present study exhibits better low temperature performance than styrene-butadienestyrene (SBS) modified asphalt binder and base binder. The parameter  $\Delta T_c$  increases linearly with the reduction of insoluble components and dissolved polymers, which indicates that the low temperature property is becoming less m-value controlled with the degradation of CRM.

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## 1. Introduction

The recycling use of waste tire rubber to enhance the pavement performance is a common practice in pavement construction and rehabilitation. The incorporation of crumb rubber modifier (CRM)

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into asphalt pavement can improve its fatigue performance, low temperature performance, vehicle riding comfort, aging resistance, and reduce traffic noise [1–3]. This technology has been successfully used in California, Arizona, Texas, Florida, et al. for many years [4]. There are mainly three technical routes in the incorporation of crumb rubber modifier (CRM) into asphalt pavement, namely dry process, wet process and Terminal Blend (TB). In the dry process, CRM is used as a substitute of 1%–3% of total aggregates in the asphalt mix and there is limited interaction between CRM and asphalt during blending in the mix plant [5]. In traditional wet-process, CRM acts as an asphalt binder modifier, generating asphalt rubber (AR) produced at 180–190 °C. This technique has been used for a long history and can significantly improve engineering performance compared to conventional paving grade bitumen. However, the construction of AR pavements is more challenging as temperature is more critical and the storage stability of AR also raises concern [6]. Although some warm-mix additives or bio-modifier were added to improve the workability of CRM binder [7–13], the storage stability properties were not clear.

In recent years, TB rubberized asphalt technology arose in America has shown its potential in greatly improving storage stability of rubber/asphalt composite because crumb rubber is fully digested into the asphalt, while maintaining considerable performance in pavement practice. TB rubberized asphalt binder is produced at the refinery (or terminal) like any other polymer modified asphalt and could be used in both dense and open graded asphalt mixtures [14]. It was reported that the production of TB rubberized asphalt binder can be achieved through digestion of crumb rubber into asphalt matrix using heat and shear [15]. However, the use of heat and shear to produce homogeneous rubberized binder will cause the loss of mechanical performance and elastic properties. Various studies confirmed that CRM will experience degradation and affect mechanical and elastic property significantly in the production of TB rubberized asphalt binders [16–19]. The effect of CRM degradation on molecular components and low temperature rheological behaviour of final modified product was not well understood. McGennis [20] found that an increase in rubber concentration results in lower stiffness and higher *m*-value of fine crumb rubber-modified asphalt binder and assumed that this phenomenon was due to the less aging of fine CRM asphalt or the softening effect caused by the released components from CRM. This phenomenon was also observed in CRM binders produced at high-cure conditions of 260 °C [17] and in CRM binders with wide range of CRM dissolution [21]. Zanzotto et al. [16] stated that the production of CRM binders by depolymerization and devulcanization of scrap tires in asphalt can improve low temperature performance grade of neat binder from –28 to –34. Billiter et al. [22] observed that the creep stiffness of asphalt binder decreases with the increase of CRM dissolution at interaction temperature of 191 °C, which can be explained by rubber devulcanization and depolymerization during the curing process according to the Gel Permeation Chromatography (GPC) test. Ghavibazoo and Abdelrahman [21] further investigated the CRM dissolution on low temperature properties of CRM binders before and after aging and concluded that at very low CRM dissolution levels (below 40%), the low stiffness of CRM binder matrix is attributed to the flexible behaviour of the elastic CRM particles; at higher CRM dissolution levels (above 40%), the liquid phase of CRM matrix contributed to the low stiffness of CRM binder due to the released rubbery components from CRM particle. Nevertheless, the relationship between chemical components and low temperature properties of asphalt binder was not discussed quantitatively.

The degradation of CRM during the preparation of TB rubberized asphalt binder will cause the transformation of chemical components in asphalt binder and reduce molecular size of released components, which will therefore affect the rheological properties

of asphalt binder. However, the evolution of released components from CRM and its effect on low temperature properties has not been investigated yet. This study is to investigate the evolution of components distribution and its effect on low temperature properties of TB rubberized asphalt binders produced under various interaction temperatures and times. The solubility test and the GPC test were performed to obtain the distribution of insoluble components, dissolved polymers, apparent asphaltenes and maltenes. Attempts were made to correlate each component in TB rubberized asphalt binder with low temperature creep stiffness, *m*-value and relaxation time. The terminology, apparent asphaltenes was defined as the amount of depolymerized material [17] and true asphaltenes with molecular weights between 3000 and 19,000 Da [23].

## 2. Materials and methods

### 2.1. Materials and preparation of TB rubberized asphalt binder

The base binder used in this study was PG 64–22. One minus 30 mesh crumb rubber that was processed from truck tires was utilized. The crumb rubber contains 54% natural rubber (NR) and synthesis rubber. The truck tire rubber contains more NR and can be dissolved faster than passenger tire rubber [24,25]. Based on the typical production conditions reported by Abdelrahman [18], Billiter [17], Zanzotto [16] and Flanigan [26], 20% crumb rubber by weight of asphalt binder were mixed with base binder at four temperatures of 220 °C, 240 °C, 260 °C and 280 °C for 2, 4, 6 and 8 h to produce TB rubberized asphalt binder. One mechanical blender was used to produce those binders using above interaction conditions and the blending speed was 400 rpm. As storage stability is very important for engineering use, the separation test was performed on each sample through holding an aluminium tube containing 50 ± 0.5 g asphalt binder at 163 °C for 48 h according to ASTM D7173. The top and bottom parts of the tube were collected for softening point test (ASTM D36) and the softening point difference between the top and bottom was recorded as an index to evaluate the storage stability of asphalt binder. Two replicates were used for each sample. In China, if the difference is less than 2.5 °C, the binder is considered to have good storage stability. The storage stability test results are summarized in Table 1. Generally, it can be seen that increasing interaction temperature and extending curing time could help to improve the storage stability of TB rubberized asphalt binder.

### 2.2. Solubility test and Gel Permeation Chromatography (GPC) test

About 20 mg virgin asphalt binder sample ( $W_{\text{binder}}$ ) was dissolved by tetrahydrofuran (THF) for 24 h before solubility test and GPC test. The solution was then filtered through a weighted 0.45 μm PTFE filter and was collected in a 0.5 mL centrifugal tube for GPC test. The remaining solution was filtrated through the weighted filter ( $W_{\text{filter before filtration}}$ ). After this, about 5 mL clean THF was filtrated through the filter until the filtrated solution was colourless. The filter was then kept in a vacuum oven at 35 °C for about 12 h until the retained THF was fully volatilized. Then, the filter was weighted again ( $W_{\text{filter after filtration}}$ ). The solubility was calculated using Eq. (1).

$$\text{Solubility}\% = \frac{W_{\text{binder}} - (W_{\text{filter after filtration}} - W_{\text{filter before filtration}})}{W_{\text{binder}}} \times 100\% \quad (1)$$

Waters 1515 High-Pressure Liquid Chromatography (HPLC) Pump and Waters 2414 Refractive Index (RI) detector were used to perform GPC test. THF (HPLC grade) was selected as the mobile

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