Influence of Ageing Process on Rheological Characteristic of Waste Natural Rubber Latex Modified Bitumen

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Abstract. Bitumen is widely used in highway pavement construction. Changes of bitumen properties occurs during the modification and ageing process. The ageing process does consequences in road deterioration; however, it is also benefits in determination of optimum properties during bitumen modification process. Thus, this study was conducted to evaluate the changes of bitumen (unmodified and modified) properties as it was exposed to artificial ageing process. In order to achieve the objective, base bitumen (PEN 60/70) was modified with waste natural rubber (NR) latex. Subsequently, the modified bitumen was subjected to a laboratory ageing process by the aided of rolling thin oven film (RTFOT) and pressure ageing vessel (PAV). Its rheological characteristic was evaluate using dynamic shear rheometer (DSR). It was found that, associating waste NR latex with bitumen improved its properties during short term and long term ageing, indicated by G*/sinδ and G*.sinδ. The performance grade (PG) also successfully improved to PG76 with the addition of 5% waste NR latex.

1. Introduction

Bitumen ageing behaviour has a crucial impact on durability of pavement, subsequently resulting to the deterioration of asphalt pavements [1]. Basically, bitumen ageing is induced by chemical and physical changes that occurs during storage, mixing, transport and construction process as well as during its service life [2,3]. During production and construction process (short term ageing), bitumen ageing or hardening primarily associated with loss of volatile components. Meanwhile, progressive oxidation occurs during asphalt pavement service life (long term ageing) [4-7]. Both short term and long term ageing resulting in an increase of binder’s viscosity, therefore, stiffen the mixture. Subsequently, the mixture become hard and brittle, and susceptible to disintegration and cracking failures. Moreover, ageing may render the mixture’s durability, in term of wear resistance and moisture susceptibility. However, ageing is not necessarily negative phenomenon, since some ageing may help a mixture to achieve its optimum properties [7].
To simulate bitumen ageing condition, laboratory ageing method has been conducted to quantitatively determine the ageing of bitumen. Simulation of field ageing entails increasing the temperature, decreasing bitumen film thickness, increasing oxygen pressure, or using combinations of these factors [8-10]. The most utilized test to simulate the age hardening occurring during plant mixing and laydown is Rolling Thin Film Oven Test (RTFOT). Meanwhile, Pressure Ageing Vessel test (PAV) is used to simulate long term ageing during in-service after 5 to 10 years [11-13].

In this paper, changes of NR latex modified bitumen properties due to short term and long term aging was performed using Dynamic Shear Rheometer (DSR).

2. Material and method
All the modified bitumens were prepared using a high shear mixer a shearing speed of 1500 rpm. The mixing procedure began with heating the approximately 500g base bitumen (PEN 60/70) to 145 ± 5°C. Subsequently waste NR latex (3-7% by weight) was gradually added into the hot bitumen under stirring condition for 45 minutes. Caution should be taken as the addition of latex into hot bitumen could produce frothing and foaming.

As specified in Superpave binder specification, short term and long term laboratory ageing of both unmodified and modified bitumen were performed using Rolling Thin Oven Film test (RTFOT, AASHTO T240) and Pressure Ageing Vessel test (PAV, AASHTO R28), respectively. During the ageing simulation, bitumen was exposed to high temperature of 163°C for 85 minutes during RTFOT; and afterwards, RTFOT residue was subjected to high pressure of 2.1 MPa for 20 hours at 100°C during PAV test. The unaged and aged bitumen were evaluated using Dynamic Shear Rheometer (DSR) at a frequency of 10 rad/s.

3. Result and discussion

3.1. Isochronal plot
Isochrones plot of complex modulus, G* and phase angle, δ at constant frequency of 10 rad/s in original state (unaged) and RTFO aged state (short term aged) were shown in Figure 1. Isochronal plot of the rheological properties of bitumen illustrates bitumen’s stiffness and viscoelastic behaviour with the changes of temperature [5]. With increasing of NR latex content, G* value of modified bitumen in original state was promoted. Moreover, G* values of modified bitumen were more pronounced compared to base bitumen within temperature ranges of 46 to 58°C. An obvious increment of G* was clearly seen when NR latex content was increased up to 7% resulting in improvement in binder’s stiffness. However, 7% NR latex showed a sharp increase in slope of G* isochrones with the increasing of temperature indicating a rapid reduction of G*.

During RTFO aged state, within temperature 46 and 52°C, base bitumen and 3% NR latex modified binder exhibit greater G* amongst all modified bitumen. However, both base bitumen and 3% NR latex modified bitumen recorded a reduction in G* values, therefore, reducing its stiffness and subsequently increased the temperature susceptibility. At high temperature (58 to 76°C), 5% and 7% NR latex modified binder resulted in greater G* indicating better temperature susceptibility compared to base bitumen.5% and 7% NR latex modified binder resulted in greater G* indicating better temperature susceptibility compared to base bitumen.

Meanwhile, isochronal plot for phase angle, δ illustrated that as temperature increased, δ for base bitumen approaching 90°, therefore, predominantly showed viscous behaviour. When rapid reduction of stiffness occurred, bitumen lost its elastic component and becoming a viscous liquid at high temperature [5]. With the increasing of NR latex content, δ value reduced towards elastic behaviour. 7% NR latex modified bitumen exhibited the lowest δ amongst all modified bitumen. As temperature changes from medium to high temperature, both unmodified and modified bitumen slowly behave toward viscous behaviour in response to the increasing of δ value and loses of elastic component.
Figure 1. Rheological properties of (i) unaged and (ii) RTFO aged bitumen versus changing temperature

3.2. $G^*/\sin \delta$

In order to evaluate rutting resistance characteristic ($G^*/\sin \delta$), bitumen with higher $G^*$ and lower $\delta$ are preferred. Rutting resistance at high temperature was determined by limiting the minimum value of $G^*/\sin \delta$ are 1.0 kPa for original and 2.2 kPa for RTFOT residue, in accordance with Superpave binder specification. Figure 2 presented $G^*/\sin \delta$ values for original and RTFO aged state at 76°C. Obviously, bitumen associated with NR latex exhibited higher capability to resist rutting compared to base bitumen. As NR latex content was increased, $G^*/\sin \delta$ value was also gradually increased. It was recorded that 7% NR latex has the highest $G^*/\sin \delta$ value amongst all modified bitumen. Highest $G^*/\sin \delta$ value revealed that rutting resistance of bitumen was improved by the addition of NR latex. The changes in $G^*/\sin \delta$ of RTFO aged with response to NR latex content showed similar trend with original state bitumen. The increase of $G^*/\sin \delta$ could be attribute to the bitumen-rubber interaction during wet process. The interaction occurs as the light component of bitumen absorb into rubber particles causing binder hardening.

Moreover, the increasing in $G^*/\sin \delta$ value contributed to the improvement of bitumen’s performance grade (PG), as shown in Table 1. Despite of the improvement in $G^*/\sin \delta$, the addition of 3% NR latex remained the same performance with control bitumen (PG70). However, incorporating more than 5% NR latex successfully improved PG by at least one level (PG76). According to JKR/SPJ/2008-S4 requirement, polymer modified binder should achieve PG 76. Thus, 5 and 7% concentration satisfy the high service temperature and fulfil the JKR/SPJ/S4-2008 requirement.
Table 1. Rutting resistance properties of unmodified and modified bitumen before and after RTFO aged

| Binder type | Temperature (°C) | Unaged (G*/sin δ > 1.0 kPa) | Short term aged (G*/sin δ > 2.2 kPa) | Performance Grade (PG) |
|-------------|-----------------|-----------------------------|-------------------------------------|-----------------------|
| 60/70       | 70              | 1.53                        | 3.02                                | PG 70                 |
|             | 76              | 0.74                        | 1.37                                |                       |
| 3NRL        | 70              | 1.81                        | 3.58                                | PG 76                 |
|             | 76              | 0.89                        | 1.71                                |                       |
| 5NRL        | 70              | 2.24                        | 5.44                                | PG 76                 |
|             | 76              | 1.19                        | 2.70                                |                       |
|             | 82              | 0.68                        | 1.41                                |                       |
| 7NRL        | 70              | 6.89                        | 7.68                                | PG 82                 |
|             | 76              | 3.80                        | 4.16                                |                       |
|             | 82              | 2.24                        | 2.39                                |                       |

3.3. G*.sin δ

G*.sin δ value is measured in order to determine the characteristics of binders in terms of fatigue ability resistance. According to Superpave binder specification, G*.sin δ should have a maximum value of 5000 kPa. Low G* and δ are considered desirable attributes from the standpoint of resistance to fatigue cracking. In other word, the lower value of G*.sin δ at 10 rad/s are generally considered as good indicator for fatigue cracking resistance [12 (Ali, Mashaan, & Karim, 2013) (Abdullah, 2014).

Fatigue life of NR latex modified bitumen were tested on RTFO+PAV residue at 25°C and the results were depicted in Figure 3. In general, the higher content of NR latex contributed to lower G*.sin δ values of modified bitumen. The reduction in G*.sin δ was about 10%, 23% and 48% for NR latex content of 3%, 5% and 7%, respectively. From Table 2, both unmodified and modified bitumen recorded G*.sin δ values lower than 5000 kPa at 25°C. As the testing temperature was lowered to 22°C, G*.sin δ values exceeded the maximum value except for 7% NR latex which still recorded lower than 5000 kPa. It meant that, the increasing of NR latex content has a positive effect on modified bitumen during long...
term aging condition. Also, incorporating more than 5% NR latex could improve the resistance to fatigue cracking.

![Figure 3. G*.sin δ of PAV residue at 25°C](image)

**Table 2. Fatigue cracking properties of modified bitumen after RTFO+PAV aged**

| Binder | G*.sin δ (kPa) |
|--------|----------------|
|        | 28°C | 25°C | 22°C |
| 60/70  | 3002 | 4420 | 6739 |
| 3NRL   | 2769 | 3993 | 5949 |
| 5NRL   | 2488 | 3425 | 5109 |
| 7NRL   | 1706 | 2309 | 3425 |

**4. Conclusion**

Based on the result obtained, associating bitumen with NR latex has successfully improved its rheological properties during ageing condition (short term and long term ageing). It was also found that, an increasing of NR latex content enhances the bitumen’s stiffness and elasticity properties, which indicated an improvement in temperature susceptibility and bitumen’s performance at high temperature. In term of long term ageing, the higher NR latex content has positively affected on fatigue cracking resistance as lower G*.sin δ was recorded. Taken together, results from this study showed that the use of NR latex as bitumen modifier successfully improved at least one level of PG. It is also can benefits in alleviating permanent deformation and fatigue cracking problems that later affected the asphalt pavement performance during its service life.

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