



# Effect of Waste Nitrile Latex on Temperature Susceptibility of Bitumen

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**Abstract:** Deterioration of bitumen performance is generally due to inadequate viscoelastic response to loading and temperature changes. The deficiency of bitumen urges the need in developing high-quality bitumen through bitumen modification technology. In order to confirm the quality of the bitumen, laboratory tests were generally conducted through empirical physical testing. However, empirical physical bitumen testing (i.e. penetration, softening point and viscosity) is insufficient to predict binder performance at high temperature. Therefore, rheology testing was performed to compensate the empirical testing using Dynamic Shear Rheometer (DSR) test. Isochrones plots, rutting parameter and failure temperature obtained from DSR testing result were analysed to investigate the temperature susceptibility response of the bitumen. At commencement, conventional bitumen PEN 60/70 was modified with waste nitrile latex additive using mechanical high shear mixer in the laboratory. The modified binders were then undergone a short-term aging process through rolling thin oven film (RTFO) test. The results of modified binder indicated that added waste nitrile latex results in better temperature susceptibility response with the improvement of stiffness and elastic behaviour at high temperature. The result also indicated that, the positive improvement upgraded the PG level to PG82, hence, showed its higher capability to resist deformation.

**Keywords:** viscoelastic response, temperature susceptibility

## 1. Introduction

Bitumen is viscoelastic material; elastic solid at low temperature, and viscous liquid at high temperatures. An adequate viscoelastic response is fundamental to ensure a good performance of road pavement [1]. Pavement performance closely related to viscoelastic properties of the bitumen [2], and gradually deteriorates because of excessive heavy traffic loads and high daytime temperature. This situation demands the need of high-quality bitumen to improve rutting. The deficiencies of bitumen are compensated by improving viscoelastic characteristic of bitumen to lower temperature susceptibility, thus, minimize stress cracking at low temperature and plastic deformation at high temperature [3], [4].

Temperature susceptibility is more precisely, defined change in the consistency parameter as a function of temperature [5]. Bitumen with high temperature susceptibility will cause problem to asphalt mixture (i.e. permanent

deformation) during pavement service period. Generally, temperature susceptibility of modified bitumen is determined through empirical tests, in terms of penetration index (PI) and penetration viscosity number (PVN) [3,5-6]. In addition, researchers adopt rheological analysis as another criterion in expressing temperature susceptibility through rheological isochrones, master curve and black diagram [7]-[10].

In an effort to develop a new high-quality bitumen, the polymer is adopted in the modification process of conventional bitumen [3], [11], [12]. Polymer modified bitumen are purposely developed to prevent rutting, fatigue cracking and thermal cracking [13].

During the modification process, polymer particles absorb maltenes leaving higher asphaltenes in bitumen [14]. Higher asphaltenes resulting in changes of viscoelastic properties, which improves bitumen's stiffness and elastic behaviour. These improvements implying a good rutting resistance of the binder. Despite of its advantages, higher cost is one significant drawback of polymer modified binder [3], [15]. The binder cost may increase from 30% to 100%, lead to significant impact on hot mix asphalt (HMA) price hiking up to 40% [16].

Accepting this limitation and global trend toward sustainable industry, recycling waste material becoming a feasible option in bitumen modification technology. Recycling waste material is not only improving the bitumen properties, but also for safe method of waste disposal [17]. The most widespread waste material used as bitumen modifier includes scrap tires [9], [18], [19], waste nitrile rubber [20] and plastic waste [21], [22].

In this study, waste nitrile latex was incorporated to enhance bitumen's properties on high temperature performance. The effect of increasing content of waste nitrile latex on physical properties and rheological properties, therefore the reduction of temperature susceptibility has been investigated.

## 2. Methodology

### 2.1 Modified Bitumen Preparation

Bitumen penetration grade (PEN) 60/70 was used as base bitumen for modification purpose. 5%, 7% and 10% of waste nitrile latex by weight of bitumen were used during the modification process. At commencement of modification process, approximately 500g base bitumen (PEN 60/70) was heated at mixing temperature of  $145 \pm 5^\circ\text{C}$ . Upon reaching the desired temperature, waste nitrile latex was gradually added into the hot bitumen to mix at shearing speed of 1500 rpm for 45 minutes. Since waste nitrile latex is in liquid form, frothing and foaming could occur during the addition of latex into the hot bitumen due to the presence of water in latex. Fig. 1 illustrates bitumen modification process using high shear mixer. The term of rubberised bitumen will be used throughout this paper to represent waste nitrile latex modified binder.



Fig 1 - Bitumen modification process using mechanical high shear mixer

### 2.2 Aging Bitumen Preparation

Laboratory aged bitumen was prepared as representative of aging phenomenon in field. Established laboratory method, Rolling Thin Oven Film test (RTFOT) was adopted to simulate aging process of the bitumen in accordance to AASHTO T240. Simulation of aging process during application was performed by RTFOT on unaged bitumen. This process began with rotating glass container filled with 35g bitumen in the oven at temperature of  $163 \pm 0.5^\circ\text{C}$ . The rotation maintains at 15 rpm for 85 minutes with hot air flow horizontally into the opening of the glass container. RTFOT residue represents the binder condition during application stage. Afterwards, the unaged and aged bitumen were evaluated using Dynamic Shear Rheometer (DSR) at a frequency of 10 rad/s.

## 3. Results and Discussion

### 3.1 Physical properties

Empirical testing was conducted to measure physical properties of bitumen includes penetration at  $25^\circ\text{C}$  (AASHTO T49), softening point (AASHTO T53) and viscosity (AASHTO T316). The effect of waste nitrile latex

content on physical properties of conventional bitumen are presented in Fig. 1 Fig. 2. Generally, changes in physical properties of bitumen corresponds to waste nitrile latex content. As can be seen in Fig. 1, penetration dropped about 25% when 5% of waste nitrile latex was introduced to control bitumen. Penetration value continue to decrease to the lowest as waste nitrile latex content was increased to 10%. Contradict to penetration result, softening point displayed a noticeable increment with 5% of waste nitrile latex. However, incorporating waste nitrile latex more than 5% exhibited a reverse effect as softening point decreased gradually, yet, still showed remarkable improvement than control bitumen.

It is generally accepted that modification process resulted in bitumen hardening. This conclusion supported with the reduction of penetration and improvement in softening point due to the existence of waste nitrile latex in conventional bitumen.

Viscosity measures bitumen’s resistance to flow [23], which represents its workability during production and placement of asphaltic concrete mixture. Viscosity of each rubberised bitumen was tested using a Brookefield rotational viscometer. The effect of waste nitrile latex content on viscosity reading at temperature of 135°C is shown in Fig. 2. Strategic highway research program (SHRP) limits the bitumen viscosity at 3.0 Pa.s at temperature of 135°C. The experimental results indicated that as waste nitrile latex content increased, the viscosity reading steadily increased. The addition of 5% and 7% waste nitrile latex resulted in progressively increased in viscosity, though, they still recorded viscosity reading below 3 Pa.s. Viscosity soared to the highest with the addition on 10% waste nitrile latex content, however, exceeded the maximum limit. The increasing in viscosity can be explained through to the absorption of aromatic oil in bitumen by rubber particles [24,25], and it allows greater film thickness on aggregates in asphalt mixture [24,26-27]. However, higher viscosity of a binder could negatively affect its workability which later requires high mixing and compaction temperatures.

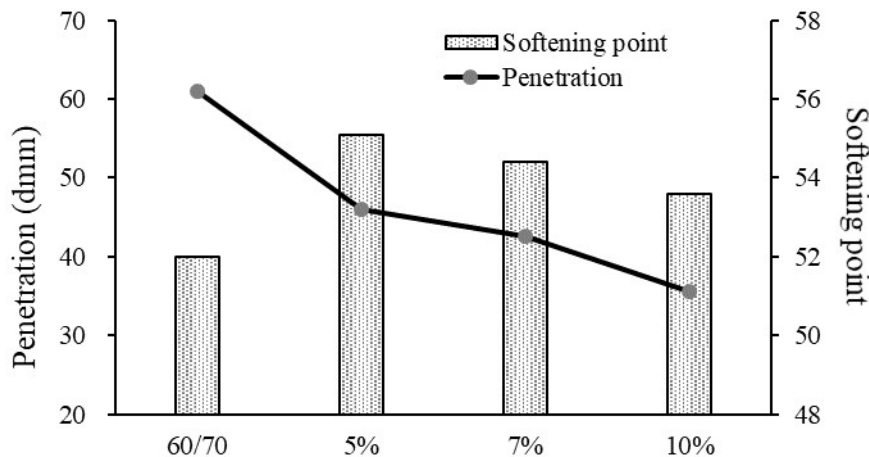


Fig. 1 - Penetration and softening point of rubberized bitumen

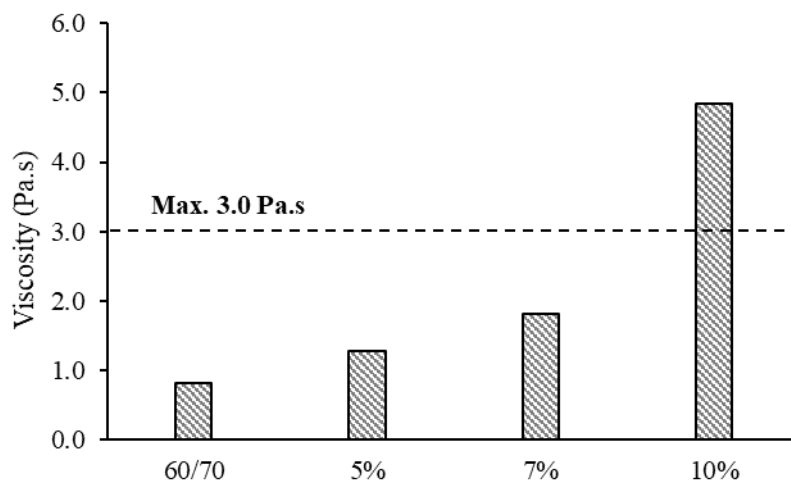


Fig. 2 - Viscosity reading at temperature of 135°C

### 3.2 Complex Modulus ( $G^*$ ) Isochronal Plot

It is acknowledged that bitumen shows elastic and viscous behaviour of a combination of both, which depends on the temperature and load frequency. Dynamic shear rheometer (DSR) test is used to determine the complex modulus and the phase angle [28]. Isochronal plot of complex modulus ( $G^*$ ) and phase angle ( $\delta$ ) at constant frequency of 10 rad/s for unaged (original state) and RTFO aged condition are shown in Fig. 3 and Fig. 4, respectively. Generally, isochronal plot illustrates rheological properties of bitumen in term of stiffness and viscoelastic behaviour at elevated temperature.

As can be seen in Fig. 3,  $G^*$  value of rubberised bitumen is more pronounced compare to control bitumen. Theoretically, higher  $G^*$  value reflect the binder's strength and show the tendency towards stiffer behaviour. In Fig. 3(a),  $G^*$  value shows a uniform increased over the entire testing temperature as waste nitrile latex content increased. In temperature range of 46°C to 58°C,  $G^*$  value of 5% waste nitrile latex showed a little improvement compared to control bitumen. The slope of  $G^*$  for 5% for waste nitrile latex gradually decreased presenting an improvement of  $G^*$  value within temperature range of 58°C to 76°C. The improvement of  $G^*$  is more evident as waste nitrile latex content increased up to 10%, indicating stiffness behaviour of unaged rubberised bitumen is significantly improved. High value of  $G^*$  in rubberised bitumen is mainly due to the presence of high percentage of elastic rubber particles in the binder [10].

During RTFO aged state, rubberised bitumen demonstrated an increasing in  $G^*$ . The changes showed the binder become stiffer due to oxidation. As can be seen in Fig. 3(b), all modified bitumen displayed a very similar value of  $G^*$ , regardless of the waste nitrile latex content. At high temperature (58°C to 76°C), control bitumen plotted a sharp increase in  $G^*$  slope indicating a rapid reduction in  $G^*$  value with the increasing of temperature. In comparison, rubberised bitumen displays continue decrease in  $G^*$  slope represents a better stiffness behaviour, and consequently reduces the temperature susceptibility.

### 3.3 Phase Angle ( $\delta$ ) Isochronal Plot

Isochronal plot of phase angle,  $\delta$  (Fig. 4) represents viscoelastic behaviour of bitumen during shear process. Generally, higher  $\delta$  value (approaching 90°) displays bitumen response towards viscous behaviour. Meanwhile, lower  $\delta$  value shows the bitumen behaves like elastic material [29].

As shown in Fig. 4(a),  $\delta$  value has significantly reduced with respect to waste nitrile latex content. The effect of waste nitrile latex on viscoelastic behaviour is shown through the reduction of  $\delta$  value, which indicated an improvement in elastic properties 10% of waste nitrile latex appears to have the lowest  $\delta$  value amongst all binders.

As the bitumen are exposed to high temperature (163°C) and oxidized during RTFO aged process, viscoelastic properties changed. RTFO aged bitumen display lower  $\delta$  values than unaged bitumen. In addition,  $\delta$  value of bitumen containing 5%, 7% and 10% waste nitrile latex resemble each other after undergone RTFO aged process.

It is easily seen that, in Fig. 4(b), there was remarkable different in  $\delta$  value between control and rubberised bitumen. As temperature was rising to 76°C,  $\delta$  value of control binder approaching 90° showing control bitumen losing its elasticity. In comparison, rubberised bitumen exhibited noticeable lower  $\delta$  value, which indicates a delay in elasticity response at high temperature [30]. In addition, aging did reduces the whole phase angle curve, demonstrating sol-like (viscous and less structured) is transformed to gel-like (elastic and more structured) behaviour has occurred [10].

### 3.4 High Temperature Performance

High temperature performance of rubberised bitumen is investigated by Dynamic Shear Rheometer (DSR) test according to AASHTO T315. The strategic highway research program (SHRP) recommended high service temperature (or failure temperature) of binder is determined at which  $G^*/\sin \delta$  value is greater than 1.0 kPa and 2.2 kPa for unaged and RTFO aged, respectively.

Fig. 5 displays failure temperature for control and rubberised bitumen. It was found that, rubberised bitumen showed higher failure temperature than control bitumen. High failure temperature gradually rose as waste nitrile latex content was increased. This increment directly contributed to the improvement of binder's performance grade (PG). PG system usually adopt to classify a binder based on its performance at the highest temperature it can sustain. Rubberised bitumen showed at least one level PG improvement. Addition of 5% and 7% waste nitrile latex maintains the PG level at PG76, before upgrade to PG82 at 10% waste nitrile latex.

Bitumen performance at high temperature is indicated by rutting resistance ( $G^*/\sin \delta$ ), where higher  $G^*$  with lower  $\delta$  is preferred. In order to resist rutting, bitumen should be stiff enough and sufficiently elastic, so that it will be able to return to its original shape after load deformation [30]. The effect of waste nitrile latex on  $G^*/\sin \delta$  values is illustrated in Fig. 6 with special reference to temperature of 76°C. Obviously, incorporating waste nitrile latex exhibited higher  $G^*/\sin \delta$  value compared to control bitumen for both conditions.

During unaged condition, the increasing waste nitrile latex content revealed a consistent improvement of  $G^*/\sin \delta$ . As waste nitrile latex content increase,  $G^*/\sin \delta$  value gradually increased presenting an improvement in capability to resist rutting. For instance, addition of 10% waste nitrile latex displayed highest  $G^*/\sin \delta$  at unaged condition amongst

all modified bitumen. On contrary, control bitumen recorded  $G^*/\sin \delta$  value lower than minimum requirement at 76°C. This because control bitumen used in this study is equal to PG70, which remarks the highest temperature performance at maximum of 70°C.

As oxidation occurs during RTFO process,  $G^*/\sin \delta$  values exhibit noticeable changes compare to  $G^*/\sin \delta$  during unaged condition.

It was also found that, regardless of different waste nitrile latex content,  $G^*/\sin \delta$  demonstrated small changes between each content during RTFO aged condition. It is well known that aging increases  $G^*$  value of the binder, caused by oxidation and volatilization of light molecular component from bitumen. As the result, stiffness increase and improving the deformation resistance at high temperature [10].

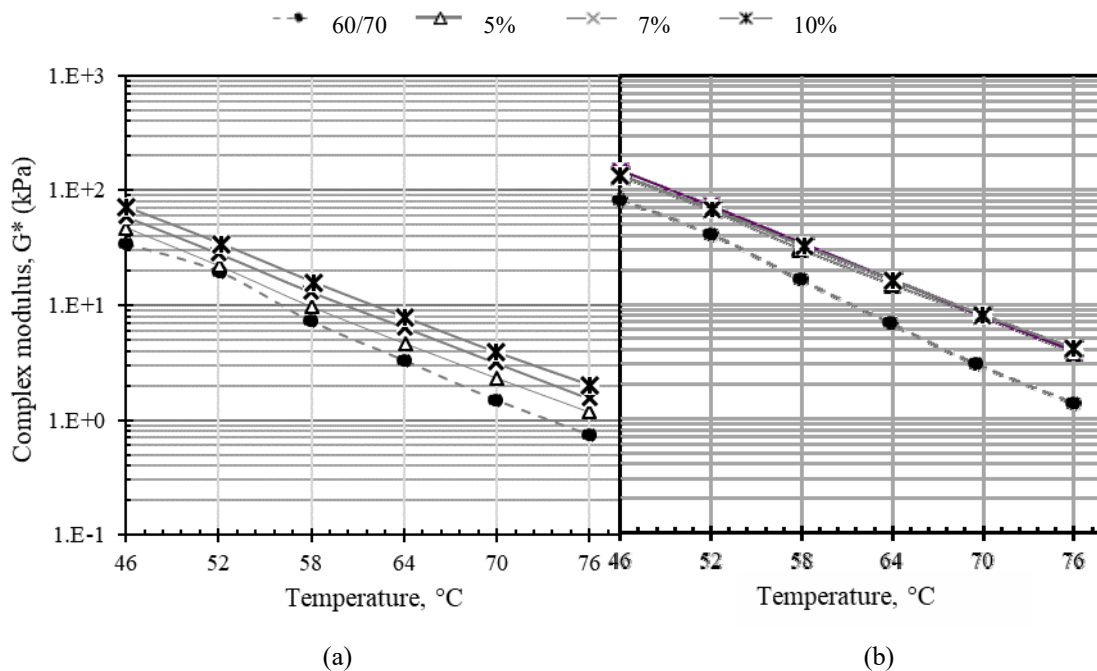


Fig. 3 - Isochronal plot of complex modulus at 10 rad/s for (a) unaged, and (b) RTFO aged conditions

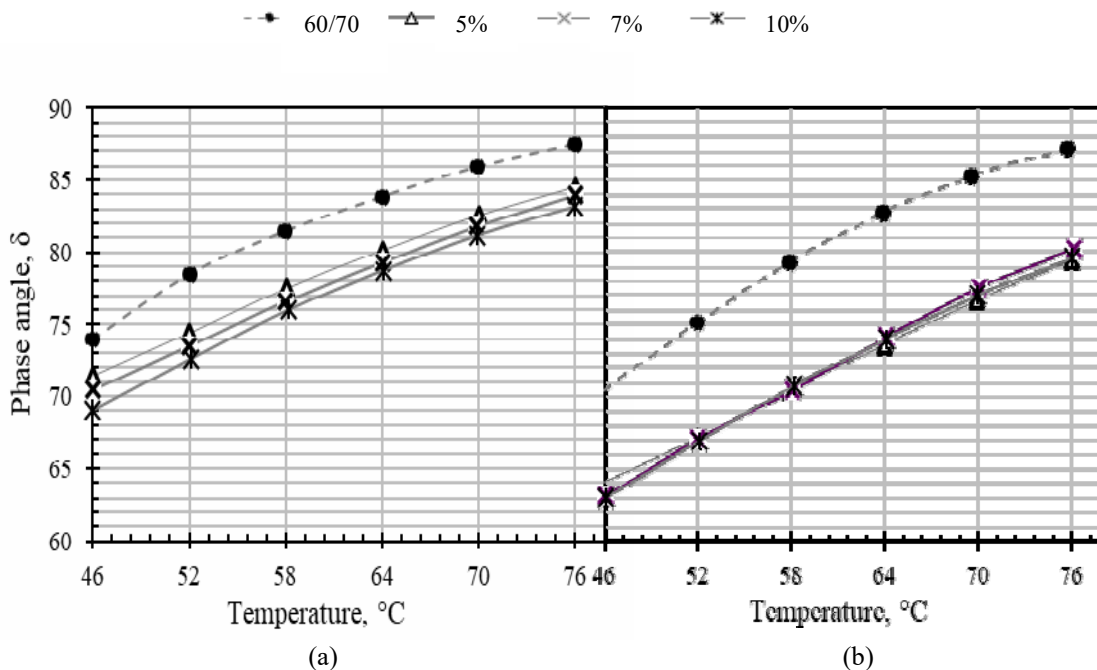


Fig. 4 - Isochronal plot of phase angle at 10 rad/s for (a) unaged, and (b) RTFO aged conditions

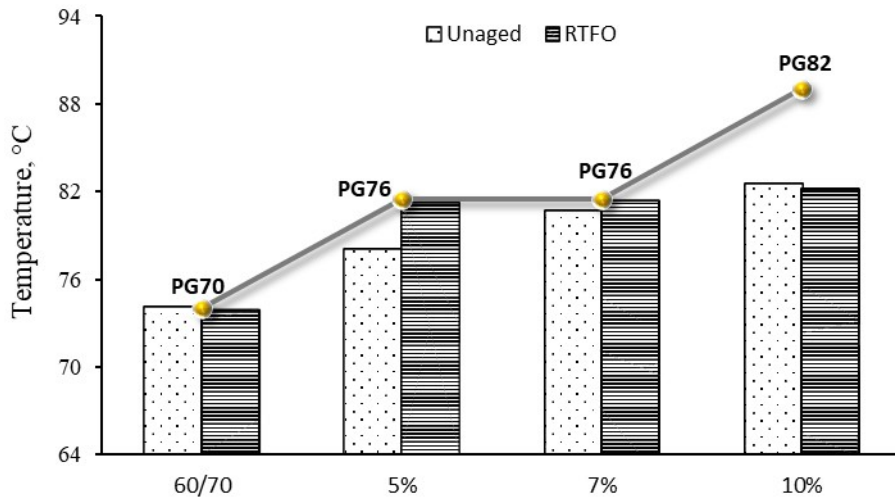


Fig. 5 - Failure temperature for rubberised bitumen

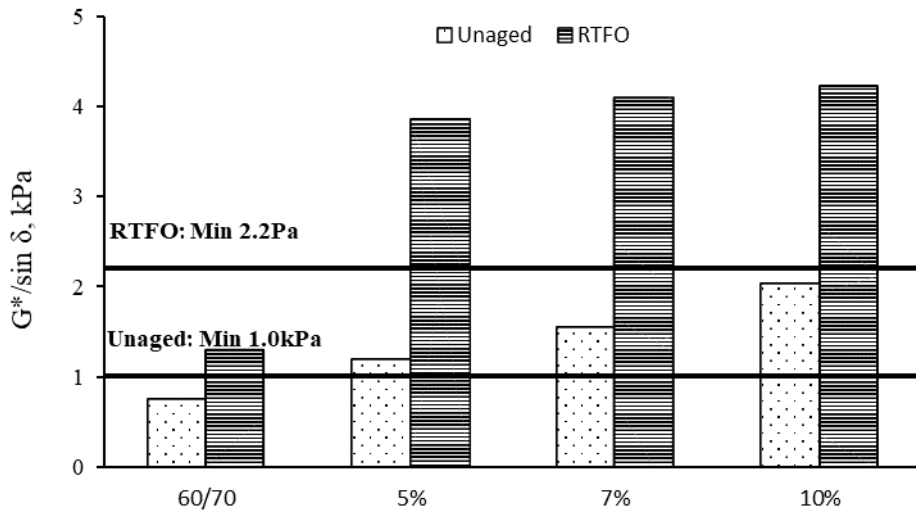


Fig. 6 - Rutting resistance of rubberised bitumen for unaged and RTFO condition

#### 4. Summary

In this study, bitumen PEN 60/70 was modified with three different concentration of waste nitrile latex. Empirical testing included penetration, softening point and viscosity were performed to study the changes in physical properties of modified bitumen. It was found that, modifying conventional bitumen with waste nitrile latex results in improvement of physical properties. For instance, results showed decreased penetration, increased softening point and viscosity, indicates that modified bitumen become harder and stiffer.

Considering the result obtained from DSR, isochrones plots confirmed that modifying conventional bitumen with waste nitrile latex resulting in the increasing complex modulus and decreasing phase angle value. The positive changes of these rheology parameters demonstrated a better stiffness and elasticity behaviour, which directly contribute to higher capability to resist deformation, hence, upgrading the binder’s performance grade from PG70 to PG82. It was also found that, the addition of waste nitrile latex between 5% to 7% is suitable in modifying bitumen. The significant enhancement of binder’s properties suggesting an improvement in temperature susceptibility response of the studied binders. The improved temperature susceptibility of rubberised bitumen corresponds to the improvement of rutting resistance. It is predicted that these improvements contribute to the high permanent deformation resistance of asphalt mixture due to binder’s capability to sustain at high temperature.

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## References

- [1] Fu, H., Xie, L., Dou, D., Li, L., Yu, M. and Yao, S. (2007). Storage stability and compatibility of asphalt binder modified by SBS graft copolymer. *Construction Building Material*, Volume 21, pp. 1528–1533.
- [2] Liang, M., Xin, X., Fan, W., Luo, H., Wang, X. and Xing, B. (2015). Investigation of the rheological properties and storage stability of CR/SBS modified asphalt. *Construction Building Material*, Volume 74, pp. 235–240.
- [3] Firoozifar, S. H. and Foroutan, S. S. (2011). The effect of asphaltene on thermal properties of bitumen. *Chemical Engineering Research and Design*, Volume 89, pp. 2044–2048.
- [4] Kalantar, Z. N., Karim, M. R. and Mahrez, A. (2012). A review of using waste and virgin polymer in pavement. *Construction Building Material*, Volume 33, pp. 55–62.
- [5] Sengoz, B., Topal, A. and Isikyakar, G. (2009). Morphology and image analysis of polymer modified bitumens, *Construction Building Material*, Volume 23, pp. 1986–1992.
- [6] Si Bachir, D., Dekhli, S. and Ait Mokhtar, K. (2016). Rheological evaluation of ageing properties of SEBS polymer modified bitumens. *Periodica Polytechnica Civil Engineering*. Volume 60, pp. 397–404.
- [7] Airey G. D. (1997). Rheological characteristics of polymer modified and aged bitumen. University of Nottingham, PhD Thesis.
- [8] Da Silva L. S., De Camargo Forte M. M., De Alencastro Vignol L. D. and Cardozo N. S. M. (2004). Study of rheological properties of pure and polymer-modified Brazilian asphalt binders. *Journal of Material and Science*. Volume 39 pp. 539-546.
- [9] Asgharzadeh S. M. (2015). Modified Asphalt Binders Evaluation Using Rheological Master Curves and Black Diagrams Mixing Conditions. *Proceedings of 5th IASTEM International Conference*, pp. 5-11.
- [10] Wang Q., Li S., Wu X., Wang S., and Ouyang C. (2016). Weather aging resistance of different rubber modified asphalts. *Construction Building Material*, Volume 106, pp. 443-448.
- [11] Grabowski W. and Kuczma, M. S. (2002). Mathematical Modelling of Rheological Properties of Polymer Modified Bitumens, in *Foundations of Civil and Environmental Engineering*. House of Poznan University of Technology, Poznan..
- [12] Al-mansob, R. A., Ismail, A., Izzi, N. and Ismael, S. (2016). Rheological characteristics of unaged and aged epoxidised natural rubber modified asphalt. *Construction Building Material*, Volume 102, pp. 190–199.
- [13] Fuentes-Audén C., et al. (2008). Evaluation of thermal and mechanical properties of recycled polyethylene modified bitumen. *Polymer Testing*. Volume 27, pp. 1005–1012.
- [14] Ramprasad, D.S., Umesha, T.S., Dinesh, S.V. and Dattatreya, J.K. (2013). Comparison of physical and rheological properties of plain and crumb rubber modified bitumen. *International Journal Research Engineering Technology*. pp. 216–220.
- [15] Wang, Y., Chong, D. and Wen, Y. (2017). Quality verification of polymer-modified asphalt binder used in hot-mix asphalt pavement construction. *Construction Building Material*, Volume 150, pp. 157–166.
- [16] Angelo, J.D. (2004). Modified Binders and Superpave Plus Specifications, in *Superpave Technical Issues*, Asphalt Institute.
- [17] Ghuzlan, B.K.A. (1997). Rheological properties of polyethylene-modified asphalt binder. *Athens Journal Technology and Engineering*, pp. 1–14.
- [18] Sarathy, R.V., Ravindraraj, B.J. and Kumar, S.D. (2015). Analysis of Properties in Bitumen and Asphalt with Partial Replacement of Rubber Tyres. *International Journal of Innovative Research & Development*. Volume 4, pp. 172–176.
- [19] Lei, Z., Chao, X., Fei, G., Tian-shuai, L. and Yi-qiu, T. (2016). Using DSR and MSCR tests to characterize high temperature performance of different rubber modified asphalt. *Construction Building Material*, Volume 127, pp. 466–474.
- [20] Soudani, K., Cerezo, V. and Haddadi, S. (2016). Rheological characterization of bitumen modified with waste nitrile rubber (NBR). *Construction Building Material*, Volume 104, pp. 126–133.
- [21] Hınıslioglu S. and Agar, E. (2016). Use of waste high density polyethylene as bitumen modifier in asphalt concrete mix. *Material Letters*, Volume. 58, pp. 267–271.
- [22] Kwabena, J., Berko-boateng, V.N. and Ama, T. (2017). Case Studies in Construction Materials Use of waste plastic materials for road construction in Ghana. *Case Studies in Construction Materials*, Volume 6, pp. 1–7.
- [23] Yero S. and Hainin, M. (2011). Viscosity Characteristics of Modified Bitumen. *ARPN Journal Science and Technology*, Volume 2, pp. 500–503.
- [24] Kim H., Geiger A., Park T., and Kim K. (2008). Effects of Asphalt Ratios on Properties of Crumb Rubber Modified Asphalts, 6th ICPT.
- [25] Hassan N. A. (2012). Microstructural characterisation of rubber modified asphalt mixtures. University of Nottingham, PhD Thesis.
- [26] Kim H. H. and Lee S. (2015). Effect of crumb rubber on viscosity of rubberized asphalt binders containing wax additives. *Construction Building Material*, Volume 95, pp. 65–73
- [27] Papagiannakis, A.T. (1995). A review of crumb rubber modified asphalt concrete technology. Washington.

- [28] Luo, W., Zhang, Y. and Cong, P. (2017). Investigation on physical and high temperature rheology properties of asphalt binder adding waste oil and polymers. *Construction Building Material*, Volume 144, pp. 13-24.
- [29] Gama, DA, Rosa, JM, DeMelo, T.JA and Rodrigues, JK.G. (2016). Rheological studies of asphalt modified with elastomeric polymer. *Construction Building Material*, Volume 106, pp. 290-295.
- [30] Mahrez A and Karim, MR (2010). Rheological evaluation of bituminous binder modified with waste plastic material. *5th International Symposium on Hydrocarbons & Chemistry*.