The European Proceedings of
Multidisciplinary Sciences
EpMS

FUTURE ACADEMY

ISSN: 2421-826X

https://doi.org/10.15405/epms.2019.12.56

ICRP 2019

4th International Conference on Rebuilding Place

EFFECTS OF LABORATORY SHORT-TERM AGEING ON BITUMEN PROPERTIES

Noor Halizah Abdullah (a)* Meor Othman Hamzah (b), Nur Izzi Md. Yusoff (c), Ahmad Shukri Yahaya (d)
*Corresponding author

(a) School of Housing, Building and Planning, Universiti Sains Malaysia, 11800 Penang, Malaysia, nhalizah@usm.my
(b) School of Civil Engineering, Universiti Sains Malaysia, 14300 Nibong Tebal, Penang, Malaysia, cemeor@usm.my
(c) Faculty of Engineering and Built Environment, Universiti Kebangsaan Malaysia, 43600 UKM Bangi, Malaysia, izzi@ukm.edu.my
(d) School of Civil Engineering, Universiti Sains Malaysia, 14300 Nibong Tebal, Penang, Malaysia, ceshukri@usm.my

Abstract

The fundamental parameters affecting e.g. durability of asphalt mixtures are their properties after ageing. Generally, ageing is quantified into two stages namely; short-term and long-term ageing. The aim of this paper is to understand the rheological and chemical properties of different bitumen subjected to various combinations of extended laboratory short-term ageing conditions. Two bitumen with the same grade (80/100 pen) from two sources, and one 60/70 pen bitumen were used in this study. The short-term ageing procedure was performed using the standard procedure for rolling thin film oven (RTFO) test, but with varying combinations of temperature and duration. The rheological properties of short-term aged bitumen were evaluated using the dynamic shear rheometer, viscometer, and Fourier transform infrared spectroscopy. Viscosity result indicated that the increment of ageing duration and time linearly increases bitumen viscosity. However, as the temperature rises, the viscosity difference is smaller.Based on the temperature sweep test, further laboratory ageing than the standard RTFO will not be severe enough to alter the performance grade of stiffer bitumen, except for the higher part of the ageing condition. The G* ageing indices indicate that the effects of oxidative ageing becomes more severe as the ageing duration and temperature progresses but there is no obvious trend found between different bitumen. While the carbonyl and sulfoxide indices increases eveidently with ageing severity.

Keywords: Short-term ageing, ageing, DSR, viscosity, ATR-FTIR, RTFOT.

https://doi.org/10.15405/epms.2019.12.56
Corresponding Author: Noor Halizah Abdullah
Selection and peer-review under responsibility of the Organizing Committee of the conference
eISSN: 2421-826X

1. Introduction

In Malaysia, the most prevalent form of surface distress in asphalt pavement is top down cracking due to repeated loading accelerated by bitumen ageing. Top down cracking normally forms over the service life of a pavement (long-term ageing). However, this occurrence may take place more rapidly due to a more severely aged asphalt mixture during its production process (short-term ageing). Asphalt pavement begins to age as soon as the bitumen is heated for mixing with the aggregates prior to lay-down. Ageing of bitumen and asphalt mixtures is a natural occurrence. When mixes aged, the bitumen becomes stiff and makes it more susceptible to cracking. A stiffening of the mixture and a decrease in the pavement's flexibility may lead to cracks formation with the possible consequence of complete failure (Vallerga, 1981). However, ageing does not necessarily imply a negative outcome, as some materials require a certain amount of ageing to achieve improved characteristics.

1.1. Short-term ageing

According to Shalaby (2002), bitumen short-term ageing is simulated in the laboratory using different methods, namely, the thin film oven (TFO) and the rolling thin film oven (RTFO) tests. Lewis and Welborn (1941) first introduced the TFO test to distinguish between bitumen with different hardening properties and volatility. The test apparatus involved a 50 ml bitumen sample placed on a 140 mm diameter dish with a 3.2 mm film thickness. The dishes containing samples were then placed in a rotating shelf, with 5 to 6 rpm rotation, in the oven for 5 hours at 163°C. In 1959, AASHTO adopted the TFO test method and ASTM in 1969 (ASTM, 1995) for simulating bitumen hardening during plant mixing. It was observed that the major issue with the TFO test was the thick bitumen layer that caused a large volume of exposed surface area for aged bitumen. Since the bitumen was not rotated during the test duration, there was concern that ageing may be limited to the upper layer of the sample. Due to this reason, the modified thin film oven (MTFO) test was developed (Edler, Hattingh, Servas, & Marais, 1985). The bitumen film thickness was reduced to 0.1 mm with additional increase in exposure time (24 hours). This minor modification was done to enhance the ageing process severity and taking account hardening and volatile loss of bitumen. According to Hintz, Velasquez, Li, and Bahia, (2011), the first stage of ageing occurs during transport, storing and handling, followed by asphalt mixture production and construction. The second stage, named as long-term ageing occurs over an extended time during pavement's service. The characteristics of bitumen change during mixing, transporting, construction and its service life (Lee, Amirkhanian, Shatanawi, & Kim, 2008). According to the Asphalt Institute (1996), the two mechanisms that change the property and cause bitumen ageing are volatilization of bitumen and its reaction with oxygen in the environment (oxidation). During asphalt mixture production, bitumen aged due to increased temperatures and constant exposure to air flow (oxygen). Over time, bitumen ageing in the asphalt mixture becomes more severe due to oxidation and greater variation in air temperature (Chen & Huang, 2000; McGennis, Shuler, & Bahia, 1994). According to Basu, Marasteanu, and Hesp, (2003), the properties of bitumen are reliant on loading time and temperature, thus temperature and time may be used interchangeably. It is also concluded that the behaviour of bitumen at high temperature in short loading times is equivalent to lower temperatures and longer loading times.

Bitumen ageing rate has been known as one of the primary factors influencing the durability of asphalt pavement in service (Apeagyei, 2011; Collop, McDowell, & Lee 2007). However, it is complicated to investigate bitumen ageing in the field since such studies are particularly costly and time consuming. It is also quite challenging to investigate due to the various variables such as precipitation, temperature, sunlight (UV), wind and others (Mouillet et al., 2009; Garrick, 1994). Therefore, many researchers have diverted their attention and proposed substantial evaluation tests and parameters to assess the ageing behaviour and ageing resistance of bitumen (Akbarnejad, Houben, & Molenaar, 2014; Mollenhauer, Pierard, Tusar, Mouillet, & Gabet, 2010; Li, Zhang, & Wang, 2007; Zhang, Liang, & Li, 2005; Airey, 2003; Shenoy, 2002). Lu, and Isacsson (2002) observed that bitumen ageing may be affected concurrently by numerous factors, such as bitumen content and properties, aggregate nature and particle size distribution, air voids, factors related to production, and external conditions such as time and temperature. Information on bitumen properties is imperative in designing pavement to provide optimal durability and service performance. A study was carried out by Mallick and Brown (2004) to evaluate the RTFO and pressurised ageing vessel (PAV) ageing methods. Bitumen from loose asphalt mixtures obtained during construction were recovered and from cores obtained after first, second and third year of service. The recovered, RTFO and PAV samples were tested using the bending beam rheometer (BBR) and dynamic shear rheometer (DSR). The results indicated that in the plant mix process, bitumen were aged more compared to the RTFO test. However, it was indicated through statistical analysis that RTFO method seemed capable of simulating short-term aged bitumen to a reasonable degree. Li, Zofka, Marasteanu, and Clyne, (2006) investigated the rheological properties of recovered bitumen and base bitumen aged in the laboratory using standard RTFO and PAV test procedures. The results were compared and significant differences were found between laboratory aged and recovered bitumen. Lee et al. (2008) evaluated the short-term ageing effects of nine bitumen aged in the RTFO and short-term oven ageing (STOA) methods using the GPC. It was concluded that the ageing effects among laboratory short-term aged and field aged bitumen varied widely and difficult to conclude. Good correlation was found between RTFO and STOA. However, no proper correlation existed between laboratory short-term ageing and field samples. According to Superpave binder specifications (McGennis, Shuler, & Bahia, 1994), rutting potential of asphalt mixture is currently evaluated by testing short-term aged bitumen, aged using the RTFO test method which was originally suggested by Hveem, Zube, and Skog (1963). In this method, the RTFO test procedure (ASTM D2872) was assumed to be capable of simulating short-term ageing of bitumen (Hicks & Finn, 1994). This method can be evaluated by comparing the DSR test results from bitumen extracted from asphalt mixture plant with the test results from unaged bitumen subjected to RTFO test conditions..

2. Problem Statement

Many established laboratory test methods and ageing models have been developed to simulate field ageing during the execution of the Strategic Highway Research Program (SHRP) and several other studies in the USA. Production of asphalt mixture practices in the USA and Malaysia do differ in terms of environmental conditions and asphalt mixture production practices. These differences may influence the extent of short-term aged bitumen and asphalt mixtures.

Ageing is a physico-chemical phenomenon that affects performance of asphalt mixtures. It stiffens bitumen over time. Ageing occurs rapidly during asphalt mixture production and it ages at a slower rate during its service life. It can potentially cause structural and functional failures in asphalt pavements. Therefore, prediction of formation patterns of short-term ageing or extent of ageing can be a useful tool for pavement management sectors to plan short-term and long-term strategies for pavement maintenance and rehabilitation. One of the methodologies to evaluate the ageing patterns is simulation of field ageing in the laboratory. In this regards, field ageing can be correlated with laboratory ageing via some indices indicating bitumen properties, aggregate characteristics, environment temperatures and number of years in which a pavement has been in service. Since traffic volume, climatic conditions, mode of manoeuvre of transportation fleet, incident ray of light and latitude could adversely affect the cumulative effects of ageing, it is necessary to investigate short-term ageing in detail on the basis of production and quarry conditions.

3. Research Questions

Since environmental conditions, ambient temperature, exposure to ultra-violet light, humidity levels, construction materials and methods are different; the field condition can differ country by country. Therefore, it is necessary to develop laboratory ageing methods that truly simulates field conditions given the materials used and environmental conditions (humidity and temperature) in Malaysia. This study is carried out to answer the following research questions:

- What is the ageing severity or indices when the bitumen is aged at various ageing conditions?
- How does extended laboratory short-term ageing affects the rheological and chemical properties of bitumen?

4. Purpose of the Study

This study is a part of a research carried out by Abdullah, Hamzah, Golchin, and Hasan, (2018) to evaluate laboratory short-term ageing. The objective of this paper is to present the supplementary results of that study to better comprehend the effects of laboratory ageing on bitumen. The virgin and short-term aged sample are assessed by rheological and chemical tests. This is to characterize and understand the impacts of extended short-term ageing, by varying ageing duration and temperature, to the advancement of bitumen properties.

5. Research Methods

5.1. Bitumen

Two types of bitumen were used, namely 80/100 and 60/70 penetration grade bitumen. The bitumen were obtained from Kuad Kuari Sdn. Bhd. (Quarry A) and Kamunting Premix Plant Sdn. Bhd. (Quarry B). All bitumen obtained from Quarry A originated from PETRONAS Penapisan (Melaka) Sdn. Bhd. and bitumen from quarry B originated from Kemaman Bitumen Company Sdn. Bhd. Bitumen were collected and sealed in containers to minimise premature ageing and oxidation. A total of three bitumen were collected; one 60/70 pen bitumen (from Quarry B) and two 80/100 pen bitumen from Quarry A and Quarry B.

5.2. Laboratory ageing of bitumen

A design of experiment was utilised for developing the laboratory short-term ageing criteria based on Malaysian asphalt mixture production practices. This experiment design is also used to characterize the effects of various short-term ageing condition on the rheological properties of bitumen. The Central Composite Design (CCD) method was applied to design experimental programs for three ageing temperatures and ageing durations. This procedure was explained in detail by Abdullah at al. (2018).

The ageing duration and temperature levels were determined based on the maximum duration and temperature that can safely handle the RTFO bottles. Nine duration and temperature combination conditions were considered for the design of experiment as shown in Table 1. All unaged bitumen were aged in the RTFO at different temperature and duration combinations to simulate short-term ageing.

Table 01. Levels of investigated parameters

Parameter	Min.	Central	Max.
Time (minutes)	85	130	175
Temperature (°C)	148	163	178

5.3. Rheological and chemical tests

To ensure sufficient fluidity of bitumen when pumped and mixed in the mixing plants, the mixing and compaction temperatures were usually established using the Brookfield Rotational Viscometer (RV). Unaged and laboratory aged bitumen from three sources (A80, B80, and B60) after subjected to different ageing conditions were tested. A sample chamber was filled with 10g of bitumen. The viscosities were measured at 20 rpm with a number 21 spindle in accordance to ASTM D4402 (ASTM, 2015) standard procedures. Temperatures from 110 to 170°C were applied at 10°C increments. The temperature sweep test was conducted at test temperatures ranging from 46 to 82°C at 6°C inntervals for unaged and laboratory aged bitumen following the Superpave specifications (SP-1, 2003). Samples were tested with the 25 mm diameter plate with 1 mm gap. Following the Superpave requirements, 1.59 Hz loading frequency was applied to simulate the shear stress which is equivalent to an approximately 90 km/hr traffic speed. According to Superpave, the failure temperatures were determined at 1.0 kPa for unaged and 2.2 kPa for short-term aged bitumen (McGennis et al., 1994). The frequency sweep test was carried out according to the test procedure described by Airey (1997). This test was carried out under strain controlled conditions at temperatures between 5 and 65°C (with 10°C increment) and frequencies between 0.1 and 10 Hz. The parallel plates with 8 mm diameter and a 2 mm gap and parallel plates with 25 mm diameter and a 1 mm gap were used for testing temperatures below and above 35°C, respectively. The use of different testing geometry is shown in Table 2. The bitumen stiffness characteristics were interpreted in the form of shifted master curves at a 25°C reference temperature based on time-temperature superposition principle (TTSP). The G^* and δ at 25°C test temperature and 1 Hz loading frequency were used to determine the ageing indices of the tested bitumen. The attenuated total reflectance Fourier transform infrared spectroscopy (ATR-FTIR) was used to describe the ageing effects on bitumen. Infrared spectroscopy measures the absorbed infrared light by covalent bonds in molecules. A diamond crystal was used in the ATR-FTIR machine, permitting spectra between 4000 and 600 $cm^{-}1$. Each sample was scanned in the Mid Infrared

Region (600 – 4000 cm^{-1}). The IR Solution software was used to convert (by Fourier transformation) the interferogram into an absorption spectrum for further analysis.

6. Findings

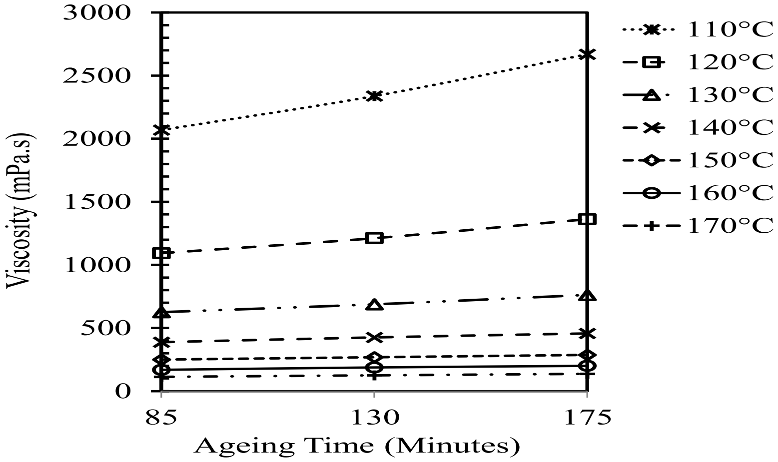
The rheological properties of laboratory aged bitumen subjected to various shear loading, test temperatures and ageing conditions are characterised. For easy identification of bitumen and ageing conditions, the designation system in Table 2 was adopted throughout the texts and graphs.

Table 02. Bitumen and ageing condition designation

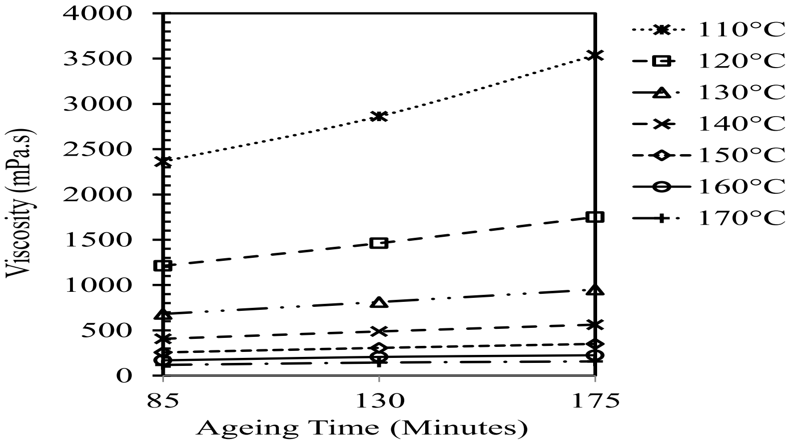
Bitumen Type	**Ageing Condition**		**Designation**
	Temperature (°C)	Time (Minutes)	
A80D	Unaged		A80
B80D			B80
B60D			B60
A80D B80D B60D	148	85	148-85
		130	148-130
		175	148-175
	163	85	163-85
		130	163-130
		175	163-175
	178	85	178-85
		130	178-130
		175	178-175

6.1. Rheological properties

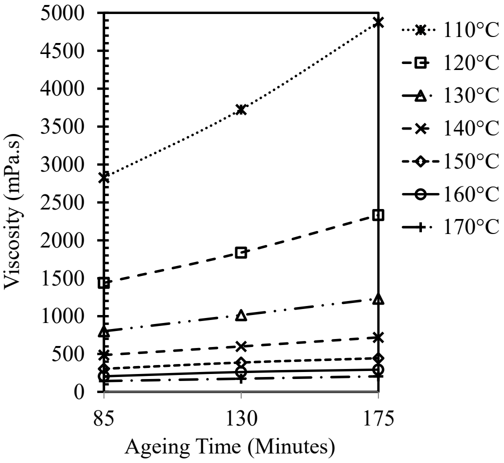
Viscosity is one of the fundamental characteristics that refers to the resistance of fluids to flow. It is crucial to ensure that bitumen has sufficient viscosity for easy pumping and adequately coat every aggregate particle during the mixing process (Behl, Chandra, Aggarwal, & Gangopadhyay, (2014). The relationships between ageing extent and viscosity for Bitumen A80, B80 and B60 subjected to different ageing conditions and temperatures are shown in Figures 1 through 3. In general, the results show that the increment of ageing duration linearly increases the viscosity of Bitumen A80, B80, and B60. Similar trend is shown for bitumen subjected to higher ageing temperature. The rise in ageing temperature also increases the viscosities of bitumen. The increase in viscosities is also higher for higher ageing temperature. This is indicated by the slope of the graph which is steeper as the ageing temperature rises. The increment in viscosity is more pronounced when the bitumen was aged at 178°C and tested at 110 to 130°C regardless of bitumen type. The difference in viscosity rises with ageing temperature. For example, for Bitumen A80, the viscosity increases about 24% with ageing duration when aged at 148°C. At 178°C ageing temperature, the viscosity increases up to 62% with ageing duration. This indicates that at high ageing temperature, the more severe the bitumen aged. However, as the test temperature rises, the viscosity difference is smaller. This is apparent for all bitumen types. For example, the viscosity of Bitumen B60 enhanced up to 58% with ageing duration at 120°C test temperature whereby 160°C only registers 33% increment.



a) [Ageing Temperature: 148°C]

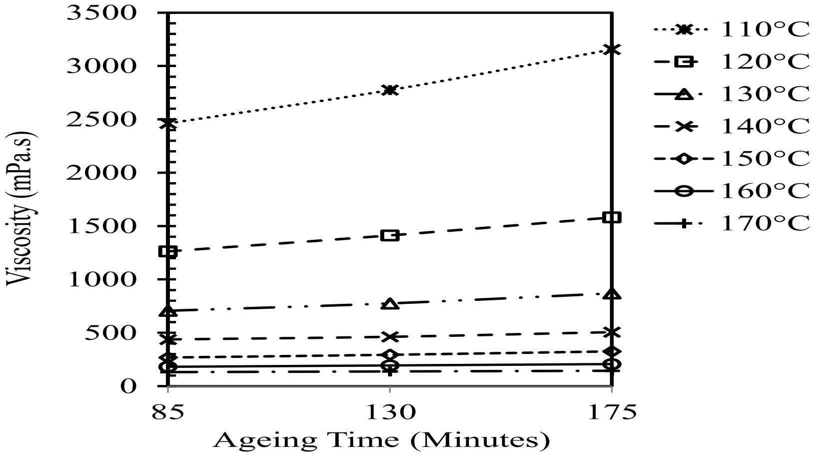


b) Ageing Temperature: 163°C

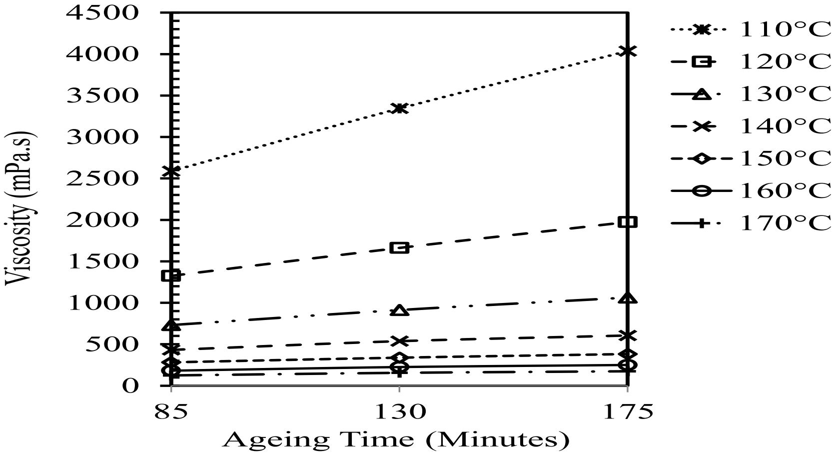


c) Ageing Temperature: 178°C

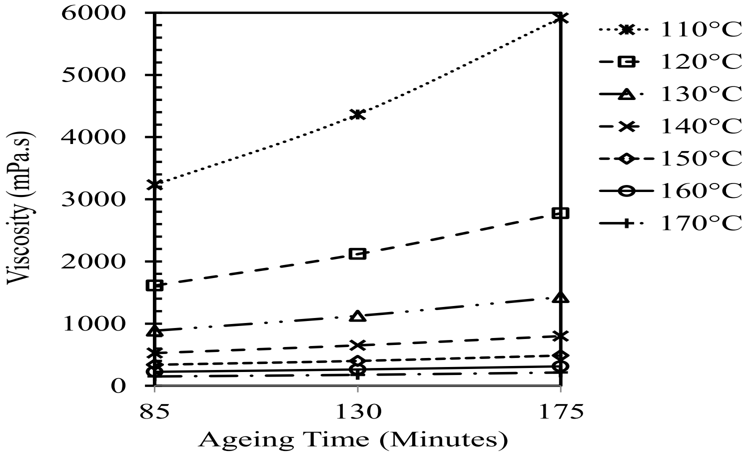
Figure 01. Viscosity ageing condition dependency for Bitumen A80



a) Ageing Temperature: 148°C

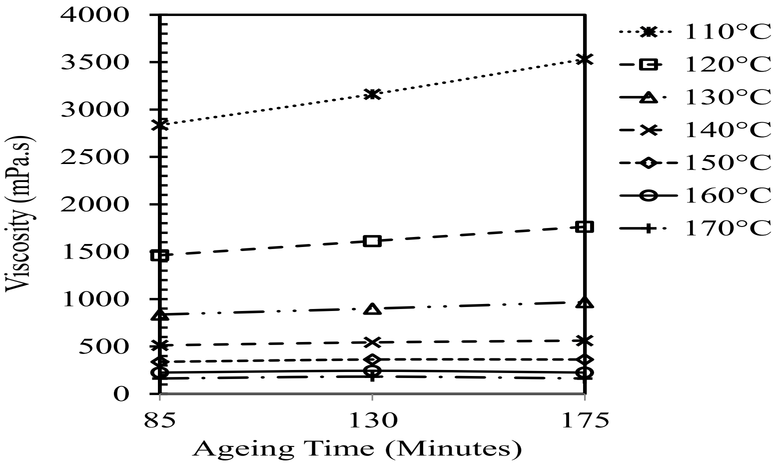


b) Ageing Temperature: 163°C

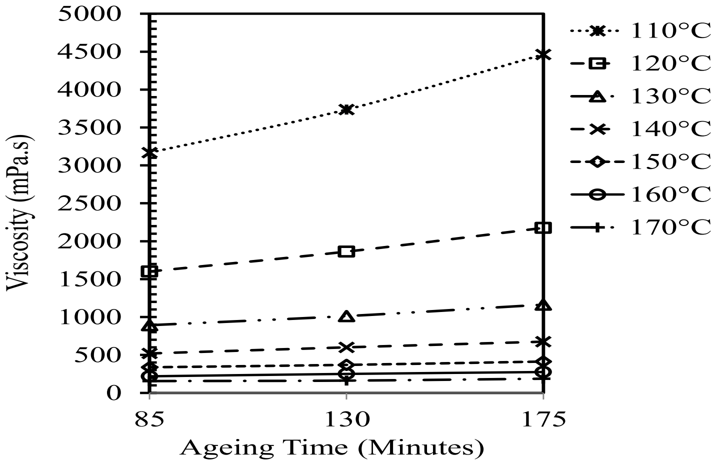


c) Ageing Temperature: 178°C

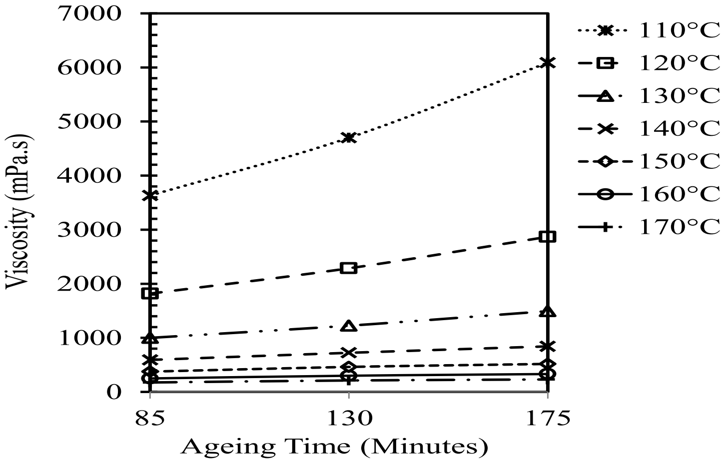
Figure 02. Viscosity ageing condition dependency for Bitumen B80



a) Ageing Temperature: 148°C



b) Ageing Temperature: 163°C



c) Ageing Temperature: 178°

Figure 03. Viscosity ageing condition dependency for Bitumen B60

Temperature sweep test was carried out between 46 and 82°C to determine the G* and δ of laboratory aged samples. According to Superpave, rutting is controlled by restricting the G*/sin δ at a test temperature that corresponds to values greater than 1.0 and 2.2 kPa for unaged and short-term aged bitumen, respectively (SP-1, 2003). Table 3 exhibits the effects of laboratory ageing on bitumen rutting parameter. The G*/sin δ increases with ageing temperature and duration. Table 4 shows the effects of laboratory ageing on failure temperature of bitumen.

The effects of ageing on bitumen performance grade can be evaluated using this parameter. The results show that an increase in ageing duration does not noticeably effect the performance grading of stiffer bitumen. However, as the ageing duration and temperature progress, the bitumen performance grade increases. For instance, for Bitumen A80, the performance grade changes from PG 58 to PG 64 when subjected to 148-175 and higher than 163-85 ageing conditions. Since the rutting parameter of Bitumen A80 lies at the border of PG 58 and PG 64, any further ageing than the standard RTFO ageing will shift the performance grade to PG 64. As for Bitumen B80 and B60, further laboratory ageing will only transition the performance grade if the ageing extent is severe. In this case, 178-175 for Bitumen B80 and 163-175, 178-130 and 178-175 ageing conditions for Bitumen B60. This suggests that depending on the stiffness of the unaged bitumen, further laboratory ageing than the standard RTFO will not be severe enough to alter the performance grade of Bitumen B80 and B60, except for the higher part of the ageing condition. The actual failure temperatures and performance grades are shown in Table 4.

Table 04. Effects of laboratory ageing on G*/sin δ (kPa)

Test Temperature (°C)	**148**			**163**			**178**		
	85	**130**	**175**	**85**	**130**	**175**	**85**	**130**	**175**
A80									
46	27.70	32.03	47.43	36.68	49.11	75.85	59.14	79.07	119.49
52	10.28	12.91	18.22	13.40	18.06	26.72	18.70	30.25	44.81
58	4.38	4.89	6.64	5.02	6.60	10.30	6.86	11.32	17.70
64	1.75	1.91	2.81	2.02	2.76	4.01	2.66	4.37	6.18
70	0.72	0.84	1.33	0.88	1.12	1.59	1.20	1.75	2.76
76	0.35	0.39	0.58	0.43	0.53	0.74	0.57	0.83	1.29
82	0.18	0.19	0.35	0.21	0.24	0.36	0.28	0.41	0.55
B80									
46	33.02	43.66	60.20	38.30	55.94	72.23	56.07	77.14	125.23
52	13.21	17.30	22.88	15.04	22.62	30.32	21.60	31.42	46.79
58	5.17	6.41	8.54	5.86	8.34	11.05	8.25	11.62	16.48
64	2.07	2.64	3.38	2.28	3.32	4.42	3.35	4.73	6.53
70	0.90	1.14	1.45	1.06	1.41	1.87	1.49	1.96	3.02
76	0.43	0.55	0.69	0.54	0.69	0.87	0.71	0.97	1.45
82	0.21	0.27	0.34	0.27	0.36	0.43	0.34	0.48	0.61
B60									
46	45.06	58.30	65.62	49.23	70.11	125.72	51.70	111.60	141.89
52	16.62	23.22	26.49	20.41	27.81	44.37	23.53	44.36	59.39
58	6.10	8.33	9.76	7.47	10.37	15.53	9.47	16.21	21.91
64	2.65	3.13	3.80	2.83	4.03	5.55	3.78	5.80	8.41
70	1.17	1.32	1.66	1.27	1.75	2.40	1.70	2.40	3.43
76	0.56	0.62	0.77	0.60	0.81	1.14	0.80	1.10	1.57
82	0.24	0.31	0.37	0.30	0.39	0.58	0.35	0.53	0.74

Table 05. High-temperature performance grading changes due to ageing

Ageing Temperature (°C)		**A80**			**B80**			**B60**		
		85	**130**	**175**	**85**	**130**	**175**	**85**	**130**	**175**
148	FT	63	63	66	64	65	67	65	66	68
	PG	58	58	64	58	64	64	64	64	64
163	FT	64	66	68	64	67	69	66	68	71
	PG	58	64	64	64	64	64	64	64	70
178	FT	65	69	72	67	69	73	68	71	73
	PG	64	64	70	64	64	70	64	70	70

The G* and δ at 25°C test temperature and 1 Hz loading frequency are presented in Table 5. Generally, G* increases with higher frequency and also rises as the ageing duration progresses implying that the bitumen rheological properties are influenced by ageing duration. As the ageing duration continues, G* increases, while δ decreases at a given frequency. The G* for bitumen aged at 148°C show little difference between ageing duration. However, the differences are more evident as the ageing temperature increases. The δ master curves show a reduction in δ indicating an elastic behaviour improvement after ageing. The results provide similar findings, that is, increase in stiffness and elastic behaviour as the bitumen aged. The G* ageing indices indicate that the effects of oxidative ageing becomes more severe as the ageing duration and temperature progresses. However, there is no obvious trend found between different bitumen.

Table 06. Changes in G* and δ due to laboratory ageing

Bitumen	**Ageing Condition**		**Properties**		**Ageing Index**
	Ageing Temperature (°C)	Ageing Timen (Minutes)	G* (Pa)	δ (°)	G* Aged/ G* Unaged
A80	Unaged		1,309	62	-
	148	85	1,442	64	1.10
		130	1,537	58	1.17
		175	1,760	57	1.34
	163	85	1,526	63	1.17
		130	1,553	57	1.19
		175	2,047	55	1.56
	178	85	1,586	56	1.21
		130	2,299	54	1.76
		175	3,894	53	2.97
B60	Unaged		1,563	63	-
	148	85	1,695	59	1.08
		130	1,720	57	1.10
		175	2,580	53	1.65
	163	85	2,103	57	1.35
		130	3,053	56	1.95
		175	3,484	56	2.23
	178	85	2,721	52	1.74
		130	4,156	53	2.66
		175	3,622	57	2.32

6.2. Chemical property

The infrared spectroscopy has been utilised to signify the level of bitumen oxidation by characterising the chemical functional group (Lau, Lunsford, Glover, Davison, & Bullin, 1992; Petersen et al., 1993). Ageing effects evaluation is determined by evaluating the spectra changes, particularly the S=O (1030 cm$^-$1) and C=O (1700 cm$^-$1) stretching. Figure 4 demonstrates the typical FTIR spectra plot and its absorbance at 1030 cm^{-1} and 1700 cm^{-1}.

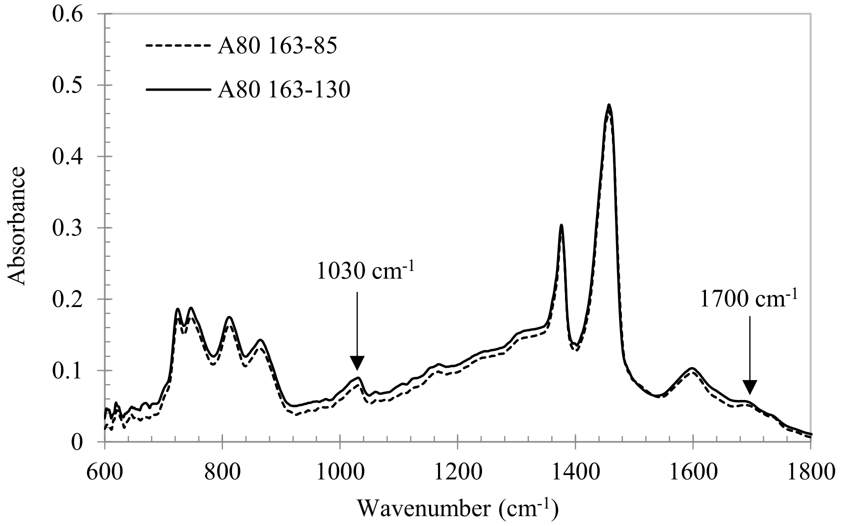


Figure 04. Typical plot of FTIR spectra for laboratory aged bitumen

Figures 5 and 6 show the oxidation indices ICO and ISO for Bitumen A80 and B60. For both bitumen, the carbonyl and sulfoxide indices increase evidently with ageing severity. This is a consequence from the content increment of the most polar component in bitumen. The relative increment in C=O and S=O functional groups are also associated with bitumen ageing, accompanied by hardening (Farcas, 2010). Lopes et al. (2014) and Zhang, Yu, and Han (2011) also observed similar findings where carbonyl functions increase with ageing intensity.

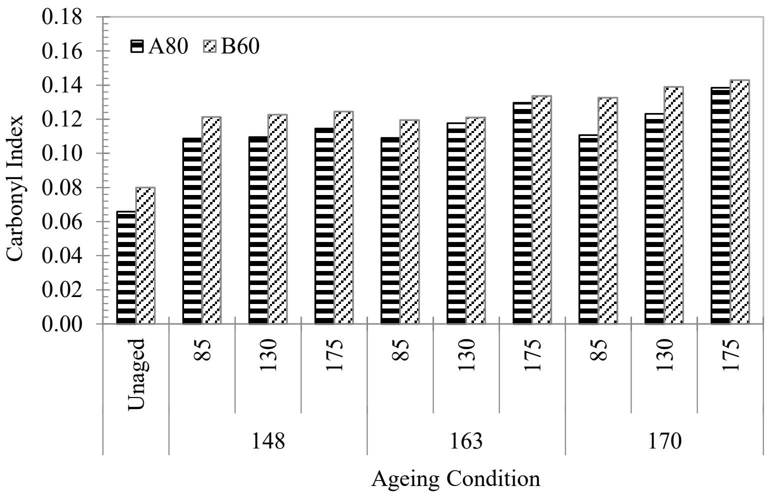


Figure 05. Carbonyl indices for laboratory aged bitumen

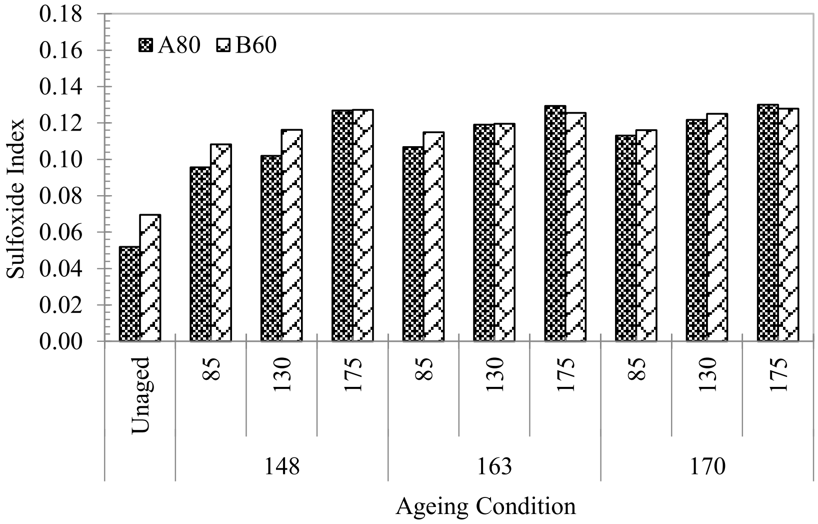


Figure 06. Sulfoxide indices for laboratory aged bitumen

7. Conclusion

In general, the viscosity results show that the increment of ageing duration linearly increases the viscosity of Bitumen A80, B80, and B60. The rise in ageing temperature also increases the viscosities of bitumen. The increase in viscosities is also higher for higher ageing temperature. The increment in viscosity is more pronounced when the bitumen was aged at 178°C and tested at 110 to 130°C regardless of bitumen type. This indicates that at high ageing temperature, the more severe the bitumen aged. However, as the test temperature rises, the viscosity difference is smaller. The results show that an increase in ageing duration does not noticeably effect the performance grading of stiffer bitumen. However, as the ageing duration and temperature progress, the bitumen performance grade increases. This suggests that depending on the stiffness of the unaged bitumen, further laboratory ageing than the standard RTFO will not be severe enough to alter the performance grade of Bitumen B80 and B60, except for the higher part of the ageing condition. Generally, the G* increases with higher frequency and also rises as the ageing duration progresses. The G* ageing indices indicate that the effects of oxidative ageing becomes more severe as the ageing duration and temperature progresses. However, there is no obvious trend found between different bitumen. As for the oxidation indices of ICO and ISO for Bitumen A80 and B60, the carbonyl and sulfoxide indices increase evidently with ageing severity. This is a consequence from the content increment of the most polar component in bitumen.

Acknowledgments

This study was funded through the Exploratory Research Grant Scheme (Grant No. 203/PAWAM/6730111) by the Malaysian Ministry of Higher Education. The authors would also like to acknowledgement Kuad Sdn. Bhd. and Kamunting Premix Plant Sdn. Bhd. for supplying bitumen and asphalt mixtures used in this study. Many thanks to the technicians of the Highway Engineering Laboratory at the Universiti Sains Malaysia for their support and assistance. The opinions, findings and conclusions conveyed in this study are those of the authors and do not necessarily represent the views of the Malaysian Ministry of Higher Education.

References

Abdullah, N. H., Hamzah, M. O., Golchin, B., & Hasan, M. R. M. (2018). An Alternative Protocol to Artificially Simulate Short-term Ageing of Binders for Selected Regional Condition. *Construction and Building Materials*, *161*, 654-664.

Airey, G. (1997). Rheological Characteristics of Polymer Modified and Aged Bitumens (Doctoral dissertation), University of Nottingham.

Airey, G. D. (2003). State of The Art Report on Ageing Test Methods for Bituminous Pavement Materials. *International Journal of Pavement Engineering*, *4*(3), 165-176.

Akbarnejad, S., Houben, L. J., & Molenaar, A. A. (2014). Application of Aging Methods to Evaluate The Long-Term Performance Of Road Bases Containing Blast Furnace Slag Materials. *Road Materials and Pavement Design, 15*(3), 488-506.

Apeagyei, A. K. (2011). Laboratory Evaluation of Antioxidants for Asphalt Binders. *Construction and Building Materials, 25*(1), 47-53.

Asphalt Institute. (1996). *Superpave mix design,* Asphalt Institute.

ASTM. (1995). *Standard Test Method for Effect of Heat and Air on Asphaltic Materials* (Thin Film Oven Test) (ASTM, Philadelphia, USA), D1754–94.

ASTM. (2015). ASTM D4402 / D4402M-15. *Standard Test Method for Viscosity Determination of Asphalt at Elevated Temperatures Using a Rotational Viscometer,* ASTM International, West Conshohocken, PA.

Basu, A., Marasteanu, M., & Hesp, S. (2003). Time-Temperature Superposition and Physical Hardening Effects in Low-Temperature Asphalt Binder Grading. *Transportation Research Record: Journal of the Transportation Research Board,* (1829), 1-7.

Behl, A., Chandra, S., Aggarwal, V. K., & Gangopadhyay, S. (2014). Zero Shear Viscosity of Bitumen-Filler Mastics of Warm-Mix Binders. *Journal of Materials in Civil Engineering, 27*(10), 04014260.

Chen, J. S., & Huang, L. S. (2000). Developing an Aging Model to Evaluate Engineering Properties of Asphalt Paving Binders. *Materials and Structures*, *33*(9), 559-565.

Collop, A. C., McDowell, G. R., & Lee, Y. (2007). On The Use of Discrete Element Modelling to Simulate The Viscoelastic Deformation Behaviour of An Idealized Asphalt Mixture. *Geomechanics and Geoengineering: An International Journal, 2*(2), 77-86.

Edler, A. C., Hattingh, M. M., Servas, V. P., & Marais, C. P. (1985). Use of Aging Tests to Determine The Efficacy of Hydrated Lime Additions to Asphalt in Retarding Its Oxidative Hardening. In *Proceedings, Association of Asphalt Paving Technologists*, *54*, 118-139.

Farcas, F. (2010). The Characterization of Organic Materials Used In Civil Engineering by Chemical and Physico-Chemical Methods. In: *Specific organic materials for construction, Chapter 13*, HERMES-LAVOISIER, 271-300.

Garrick, N. W. (1994). Use of Gel-Permeation Chromatography in Predicting Properties of Asphalt. *Journal of materials in civil engineering, 6*(3), 376-389.

Hicks, R. G., & Finn, F. N. (1994). *Stage 1 Validation of The Relationship Between Asphalt Properties and Asphalt-Aggregate Mix Performance*. Report, SHRP-A-398, SHRP, National Research Council, Washington, DC, 91.

Hintz, C., Velasquez, R., Li, Z., & Bahia, H. (2011). Effect of Oxidative Aging on Binder Fatigue Performance. *Journal of the Association of Asphalt Paving Technologists, 80.*

Hveem, F. N., Zube, E., & Skog, J. (1963). Proposed New Tests and Specifications for Paving Grade Asphalts. In *Association of Asphalt Paving Technologists Proceedings*, *32*.

Lau, C. K., Lunsford, K. M., Glover, C. J., Davison, R. R., & Bullin, J. A. (1992). Reaction Rates and Hardening Susceptibilities as Determined From Pressure Oxygen Vessel Aging of Asphalts. *Transportation Research Record,* (1342).

Lee, S. J., Amirkhanian, S. N., Shatanawi, K., & Kim, K. W. (2008). Short-term Aging Characterization of Asphalt Binders Using Gel Permeation Chromatography and Selected Superpave Binder Tests. *Construction and Building Materials, 22*(11), 2220-2227.

Lewis, R. H., & Welborn, J. Y. (1941). *The Properties of the Residues of 50-60 and 85-100 Penetration Asphalts from Oven Tests and Exposure*. Public Roads.

Li, X. M., Zhang, X. N., & Wang, S. H. (2007). Study of High Temperature Performance of Asphalt Mastic Based on Dynamic Viscoelastic Mechanics. *Journal of Highway and Transportation Research and Development (English Edition), 2*(2), 16-20.

Li, X., Zofka, A., Marasteanu, M., & Clyne, T. R. (2006). Evaluation of Field Aging Effects on Asphalt Binder Properties. *Road Materials and Pavement Design, 7*(sup1), 57-73.

Lopes, M., Zhao, D., Chailleux, E., Kane, M., Gabet, T., Petiteau, C., & Soares, J. (2014). Characterisation of Ageing Processes on The Asphalt Mixture Surface. *Road Materials and Pavement Design, 15*(3), 477-487.

Lu, X., & Isacsson, U. (2002). Effect of Ageing on Bitumen Chemistry and Rheology. *Construction and Building materials, 16*(1), 15-22.

Mallick, R. B., & Brown, E. R. (2004). An Evaluation of Superpave Binder Aging Methods. *International Journal of Pavement Engineering, 5*(1), 9-18.

McGennis, R. B., Shuler, S., & Bahia, H. U. (1994). *Background of Superpave asphalt binder test methods*. Federal Highway Administration, Office of Technology Applications.

Mollenhauer, K., Pierard, N., Tusar, M., Mouillet, V., & Gabet, T. (2010). Development and Validation of a Laboratory Aging Method for The Accelerated Simulation of Reclaimed Asphalt. *Journal of Wuhan University of Technology--Materials Science Edition, 25*(4), 631-636.

Mouillet, V., Farcas, F., Battaglia, V., Besson, S., Petiteau, C., & Le Cunff, F. (2009). *Identification and Quantification of Bituminous Binder's Oxygenated Species – Analysis by Fourier Transform InfraRed spectroscopy, LCPC testing method 69*. Technical Report, ME 69, ISSN 1167-489X.

Petersen, J. C., Branthaver, J. F., Robertson, R. E., Harnsberger, P. M., Duvall, J. J., & Ensley, E. K. (1993). Effects of Physicochemical Factors on Asphalt Oxidation Kinetics. *Transportation Research Record,* (1391).

Shalaby, A. (2002). Modelling Short-Term Aging of Asphalt Binders Using The Rolling Thin Film Oven Test. *Canadian Journal of Civil Engineering, 29*(1), 135-144.

Shenoy, A. (2002). A Method to Estimate The Rheological Properties of Aged Asphalt Binders Without Actually Aging Them. *Road Materials and Pavement Design, 3*(3), 331-343.

SP-1. (2003*). Superpave Performance Graded Asphalt Binder Specifications and Testing*. Asphalt Institute, KY, USA.

Vallerga, B. A. (1981). Pavement Deficiencies Related to Asphalt Durability. In *Association of Asphalt Paving Technologists Proceedings, 50*.

Zhang, F., Yu, J., & Han, J. (2011). Effects of Thermal Oxidative Ageing on Dynamic Viscosity, TG/DTG, DTA and FTIR of SBS-and SBS/sulfur-modified asphalts. *Construction and Building Materials, 25*(1), 129-137.

Zhang, Z. Q., Liang, X. L., & Li, P. (2005). Evaluation Method of Asphalt Aging Properties. *Journal of Traffic and Transportation Engineering, 1*.