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RESEARCH PAPER

Ageing Resistance of Bituminous Binder Modified with Propitious Antioxidant

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ABSTRACT: Bituminous mixes are prepared with aggregates, filler and bitumen as binder. Ageing of bitumen due to oxidation reduces the durability of such mixes. Various research efforts are therefore put in to decrease the ageing potential of binders and use of anti-oxidants has shown substantial success. In this research, Irganox 1010 has been identified as an anti-oxidant which has been found to be economical and yet of limited use in the field of pavement engineering. Various physical and rheological tests were conducted to determine the influence of Irganox 1010 on short-termed as well as long termed aged binders and results were analyzed. Softening Point Index (SPI) and Viscosity Ageing Index (VAI) were determined to understand the influence of Irganox 1010 on bituminous mixes. It was observed that Irganox 1010 significantly reduce ageing potential with usage of 0.6% by weight of bitumen. It has been found to be cost-effective due requirement of small quantities and being of comparatively lesser cost. This material has the potential of large scale usage to increase durability of bituminous mixes.

Keywords: Ageing, Anti-Oxidant, Bitumen, Irganox 1010.

1. Introduction

The top layers of flexible pavements are made up of aggregates bonded by bitumen as a binding material and are termed as bituminous mixes. The properties of bitumen greatly influence the performance of these mixes and depend upon exposure to environmental conditions such as presence of moisture and oxidation (Chakravarty and Sinha, 2020). Ageing of bitumen is considered to be one of the important factors for deterioration of bituminous mixes. Due to simultaneous action of external factors such as temperature, moisture, oxygen, solar radiation and internal factors such as thermal conductivity, reactive oxygen species, volumetric properties etc., ageing of bitumen occurs over its useful service life which further results in various distresses leading to decrease in serviceability of pavements (Aley et al., 2021; Omairey et al., 2021; Omaimey et al., 2022). Due to

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ageing, the stiffness and brittleness of the binder increases, thereby reducing the flexibility of the binder as well as the mix. Under repeated traffic loading, cracks are developed due to reduced flexibility of the bituminous mixes (Apeagyei, 2011). Ageing of bitumen occurs due to the exposure of heat, UV rays and atmospheric conditions due to thermal oxidation and photo-oxidation (Dickinson, 1980). The ageing process can be broadly subdivided into short term ageing and long-term ageing for hot mix asphalt (Singh and Kumar, 2015). Short term ageing is caused due to loss of volatile components and oxidation of bitumen due to high temperatures during storage, mixing, laying and compacting stages which happen within a span of hours. Environmental oxidation and UV radiations lead to long term ageing during service life of pavements (Hofko et al., 2017; Feng et al., 2021). Volatization of light components, physical changes of polar substances also results in micro-transformation and intricate chemical changes in the process of ageing (Wang et al., 2020). Moreover, the increase in share of asphaltene and decrease in aromatic compounds due to ageing of the bitumen matrix leads to greater stiffness (Hofko et al., 2017; Read and Whiteoak, 2003). The short-term ageing process is simulated in laboratory using Rolling Thin Film Oven (RTFO) in accordance with ASTM D2872 whereas, long-term ageing is simulated by Pressure Ageing Vessel (PAV) in accordance with ASTM D6521. The recently developed Viennese Binder Ageing (VBA) gives expected to result in realistic long term field ageing simulation by use of highly oxidative gases such as ozone and nitrogen oxides, and can be performed for both bitumen and bituminous mixes (Hofko et al., 2020; Sreeram et al., 2021; Steiner et al., 2020; Mirwald et al., 2020). Various advanced testing techniques have been used to qualitatively and quantitatively analyze chemical components, characteristic functional groups and molecular weights before and after ageing by Thin-Layer Chromatography-Flame Ionization Detector (TLC-FID) (Margaritis et al., 2020; Yao et al., 2015), Fourier Transform Infrared spectrometer (FTIR) (Yao et al., 2015) and Gel Permeation Chromatography (GPC) (Lee et al., 2008), respectively. For assessing microstructural changes that occur due to ageing, florescent microscopy (Zhang et al., 2017), Scanning Electron Microscopy (SEM) (Ji et al., 2020) and Atomic Force Microscopy (AFM) (Xu et al., 2017) have been used for assessing ageing related microstructural changes.

Delaying ageing process can lead to enhanced serviceability life for bituminous pavements. Moreover, acceptance of hot, warm as well as cold Reclaimed Aasphalt Pavement (RAP) materials which could significantly reduce environmental loads is greatly dependent upon proper understanding of the aged binder and influence of various anti-ageing additives to virgin binders (Luo et al., 2018; Wang et al., 2018; Gu et al., 2019; Qian et al., 2020). Contemporary literatures mention that various modifiers enhance the binder performance by delaying ageing caused due to oxidation (Apeagyei, 2011; Perez and Maicelo, 2021; Sirin et al., 2018). Researches on anti-oxidants to resist ageing have increased in recent times. Some of the compounds such as Vitamin E, Irganox 1010, carbon black, Hydrated Lime (HL), Di-Lauryl-Thio-Di-Propionate (DLTDP), wood lignin, Styrene-Butadine-Styrene (SBS) etc. can work as anti-oxidants (Apeagyei, 2011; Feng et al., 2011; Cong et al., 2012; Cong et al., 2013; Feng et al., 2016). Bitumen modified with a combination of furfural and DLTDP (2.0% and 1.5%) was observed to have a lower ageing tendency. A comparison was done for Vitamin E, Irganox 1010, Irgafos P-EPQ, Carbon black, hydrated lime and the combination of furfural and DLTDP (Apeagyei, 2011). The combination of Irganox 1010 and UV326, which is a UV absorber, showed good effect in resisting the thermal and photo-oxidative ageing of bituminous binder, which results in outstanding ageing resistance behavior of modified bituminous binder (Feng et al., 2011). Usage of grape pomace antioxidant exhibited optimal behavior in all tests performed at 10% content and for all aging conditions with respect to the controls (Floody and Thenoux, 2012). Experimental results indicated that the improvement of the anti-aging resistance of asphalt binder is noticeable when anti-aging agents containing 1% antioxidant Zinc dialkyldithio phosphate (ZDDP) and UV absorbers (0.5% UV531) were used (Cong et al., 2012; Cong et al., 2013). Wood lignin at dosages of 5% and 10% were used as an anti-ageing asphalt binder substitute (Wang and Derewecki, 2013). It was found that the 2% furfural modification of bitumen had the lowest ageing index after ageing, indicating an improvement in ageing properties of binder (Fini et al., 2015). The combination of layered double hydroxide (LDHs) and Irganox 1010 (3 wt% and 0.6 wt%) modified bitumen shows outstanding anti-ageing behaviour to both thermo-oxidative ageing and UV ageing (Zhao et al., 2015). The aged epoxy natural rubber (ENR) modified binder show better physical and rheological properties than the base binder (Al-Mansob et al., 2016). The thermal- and photo-oxidative aging resistance of bitumen was improved significantly by the pyrolysis carbon black (PCBs) (Feng et al., 2016). Usage of 0.5% poly phosphoric acid (PPA) as polymer replacement indicates better anti-ageing properties of binders (Liu et al., 2018). Replacing petroleum-based binder (bitumen) with up to 6% of lignin precipitated from black liquor can be used for hot mix asphalt, and may even be feasible for lower temperature warm mix asphalt process (Arafat et al., 2019). The alternative use of waste vegetable cooking oil (WVCO) as anti-ageing agent for bitumen shows noteworthy benefits to the environment, human health, and economy (Gokalp and Uz, 2019).

As can be observed from literature, few studies have been conducted using Irganox 1010 as anti-oxidant in binders with promising results (Apeagyei 2011; Feng et al., 2011; Zhao et al., 2015). The quantity requirement for optimized performance is also observed to be quite low. Moreover, market study showed that Irganox 1010 has low cost, good availability and better compatibility with organic products. Further researches conducted on ageing resistance with anti-oxidants with viscosity grade binders are limited. These factors served as motivation for adopting Irganox 1010 in this study and determine the influence of the modifier on performance of the bituminous binder of viscosity grade (VG) 30 which is widely used in India.

2. Materials

2.1. Bitumen

Viscosity Grade (VG) 30 bituminous binder was obtained from Indian Oil, Barauni refinery, Bihar, India. Basic physical properties were determined to ascertain the quality of binder.

2.2. Anti-oxidant

Irganox 1010 was selected as the anti-oxidant as depicted in Figure 1a and the molecular structure is shown in Figure 1b. It is a high-grade antioxidant for natural and synthetic oil. Irganox 1010 was procured from Eklingjee Polymers Pvt. Ltd., Rajasthan, India. The physical properties of Irganox 1010 are presented under:

2.3. Methodology

The modified asphalt binders with antioxidant Irganox 1010 were prepared by melt blending. The contents of antioxidant are 0.2, 0.4, 0.6, 0.8 and 1.0 % by weight of bitumen had been taken as shown in Figure 2. In this way, five modified binders were prepared with different compositions. Firstly, base binder was melted by heating it at 150 ℃ in a steel container and then antioxidant Irganox 1010 added into the container of bitumen while continuously stirring it. After the complete addition of antioxidant, mixture was mechanically blended at mixing speed of 1000 rpm for 30 min at the temperature of 150 ℃ for obtaining the modified bituminous binder. Short-term ageing and long-term ageing were done using RTFO and PAV testing facilities. The RTFO ageing was done as per ASTM D2872 at 163 ^{o}C for 85 minutes. The PAV ageing was performed on the RTFO aged samples at 305 psi for 20 hours in accordance to ASTM D6521. Thereafter, physical parameters such as penetration and softening point for modified and aged binders were determined. Viscosity measurement was performed using Brookfield rotational viscometer at 135 ^{o}C and 20 rpm in accordance to ASTM D4402. Complex modulus and phase angle were determined using Dynamic Shear Modulus (DSR) test with oscillation rate of 10 rad/sec and stain level of 12% in accordance to ASTM D7175-15. Comparison between these parameters were drawn to determine the influence of Irganox 1010 modifications on unaged, RTFO aged and PAV aged binders. The temperature range was in the range of 46-76 ^{o}C as this is the range of operational temperature within India. Thereafter, Softening Point Increment (SPI) and Viscosity Ageing Index (VAI) were determined. SPI is given by the difference of softening points of aged and unaged binders and VAI is given by the ratio of difference of viscosity of aged and unaged binders to the viscosity of unaged binders. Smaller values of SPI and VAI indicate better ageing resistance and are used to estimate the ageing characteristics of binder.

3. Results and Discussions

3.1. Physical Tests

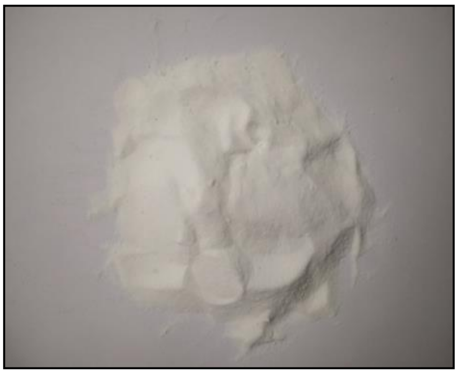
3.1.1. Penetration Value

The penetration value was determined in accordance with ASTM D6. Tests were performed at a temperature of 25 ^{o}C.

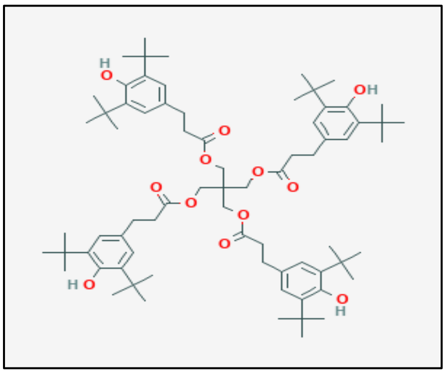
Penetration test result of modified binder is plotted in Figure 3. Addition of Irganox 1010 makes binder softer, so their penetration value increases gradually for unaged and RTFO aged binders while increasing Irganox 1010 content. It is lesser in case of PAV aged binders but for PAV aged binder penetration values increase up to 0.6% Irganox 1010 and then after decreases up to some extent on further addition of antioxidant. As observed, after ageing the binder becomes stiff and thereby a reduction in penetration value is observed. It is observed that penetration value decreases upon increase in oxidative ageing. Thus, PAV aged binders have lesser penetration values as compared to RTFOT aged binders.

Table 1. Physical properties of bitumen

Physical properties	Test method	Observed value
Penetration value	ASTM D5	61
Softening point value	ASTM D36	49 ^{0}C
Specific gravity value	ASTM D70-03	1.02



(a) Irganox 1010



(b) Molecular Structure of Irganox1010 (Pubchem, National Centre for Biotechnology Information, 2021)

Fig. 1. Physical and chemical form for Irganox 1010

Table 2. Physical properties of Irganox 1010

Material	Appearance	Melting range	Specific gravity (g/ml, 20 ºC)	Molecular formula
Irganox 1010	White powder	110 ~ 125 ºC	1.116	$C_{73}H_{108}O_{12}$

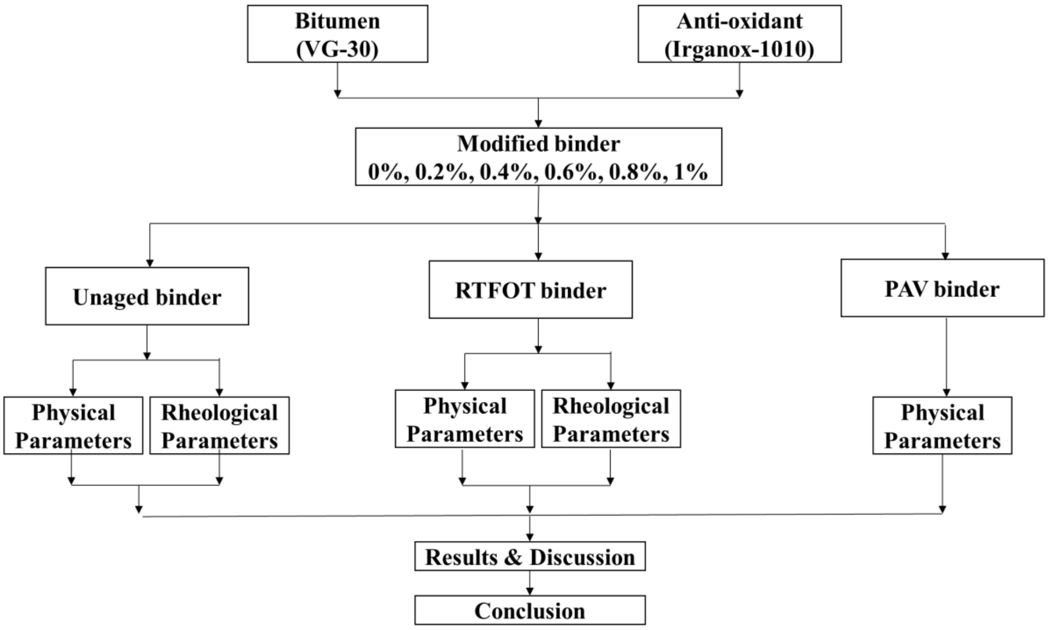


Fig. 2. Methodology for evaluation of Irganox 1010 as anti-oxidant

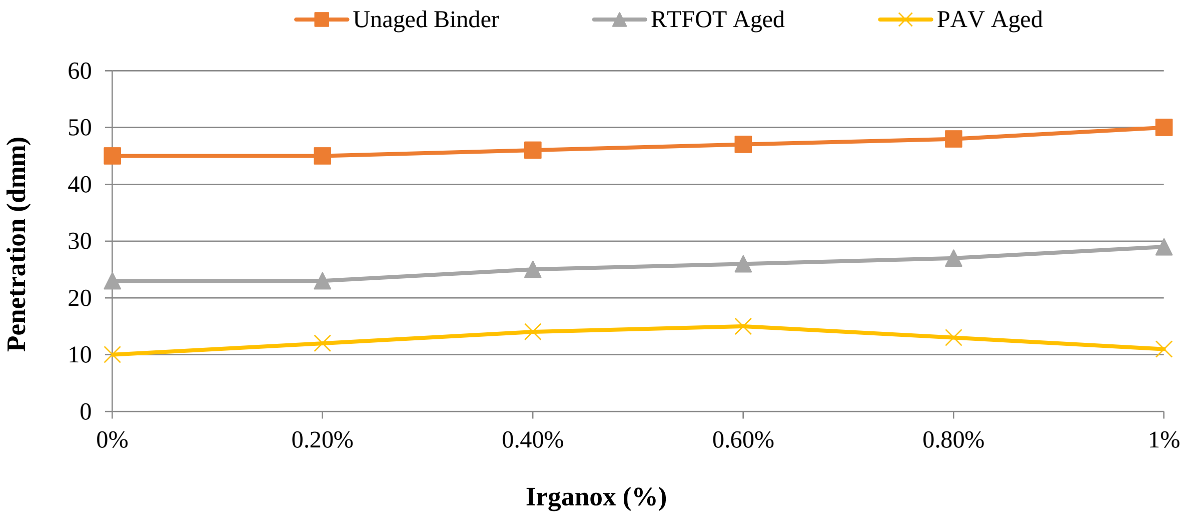


Fig. 3. Effect of Irganox 1010 on penetration values of unaged, RTFO aged and PAV aged binders

3.1.2. Softening Value

The softening point was determined in accordance with ASTM D36. The results obtained are provided in Figure 4.

After ageing of binders, they become harder in nature and their softening point increases. It can be observed from Figure 4 that softening point of base binder after RTFO ageing is more than un-aged binder. After PAV ageing, softening point is further increased. Addition of Irganox 1010 makes binder softer, so their softening point decreases gradually for un-aged and RTFO aged binders while increasing Irganox 1010 content. But for PAV aged binders softening point values decrease up to 0.6% Irganox 1010 and then after increases up to some extent on further addition of antioxidant.

Thus, observing the softening point and the penetration results, it can be concluded that after ageing, the binder becomes harder. This can be attributed to oxidative ageing due to the short-term as well as long-term ageing simulation process.

3.2. Rheological Parameters
3.2.1. Rotational Viscosity

Bolin's viscometer was used for Rotational Viscosity (RV) tests to determine the dynamic viscosity of binder in accordance with ASTM D4402. RV test result of different binders at 135 °C is provided in Figure 5.

After ageing of binders, these become more viscous. It can be observed from Figure 5 that PAV aged binders are highly viscous followed by RTFOT aged binders and the unaged binders have the least viscosity. With the addition of Irganox 1010, it is observed that the viscosity decreases for all conditions. For PAV aged binder, however, viscosity increases after modification by 0.6% Irganox 1010 thereby setting an optimum value of modification at 0.6%.

3.2.2. Complex Modulus (G*)

Dynamic Shear Rheometer (DSR) was used for determination of complex modulus (in kPa) in accordance with ASTM D7175-15. The G* values were obtained for unaged as well as RTFO and PAV aged binders. The values obtained are plotted in Figure 6-8, representing the values in graphical format.

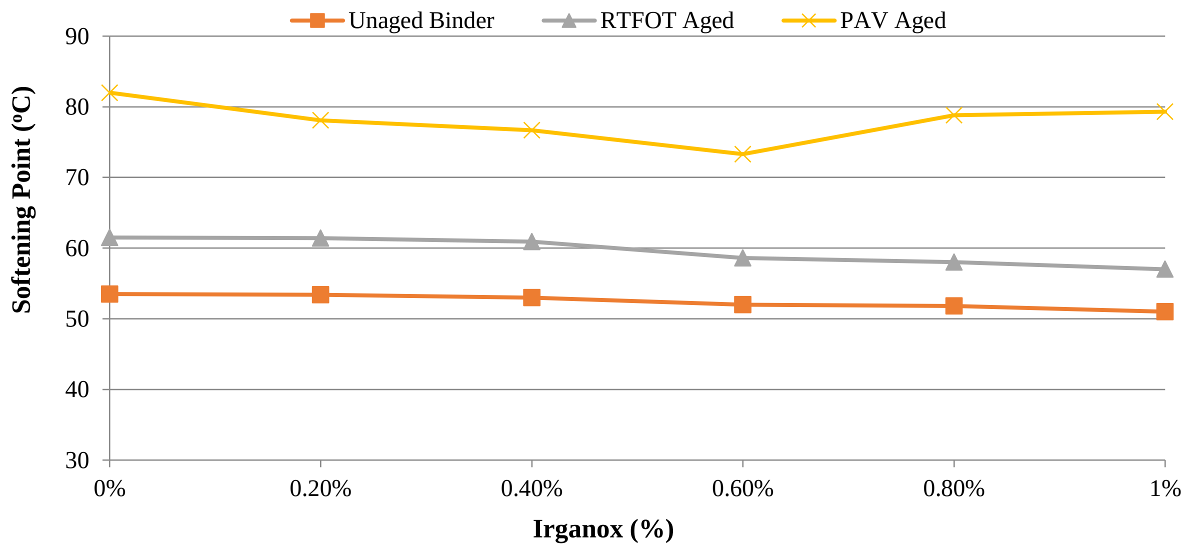


Fig. 4. Effect of Irganox 1010 on softening point of un-aged, RTFO aged and PAV aged binders at 135 ºC

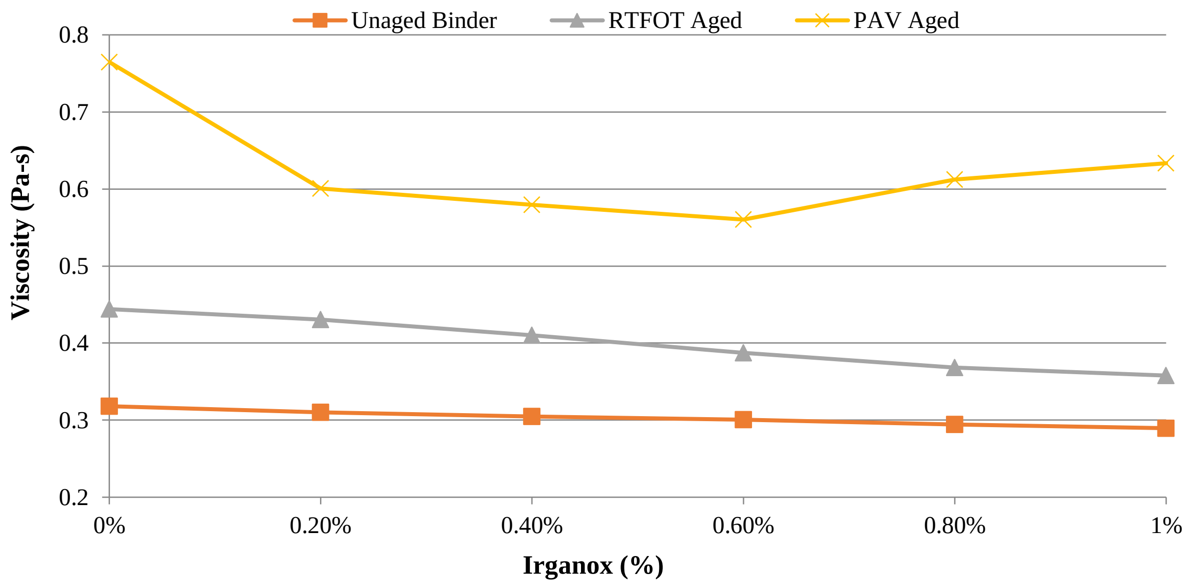


Fig. 5. Effect of Irganox 1010 contents on viscosity of un-aged, RTFO aged and PAV aged binders at 135 ºC

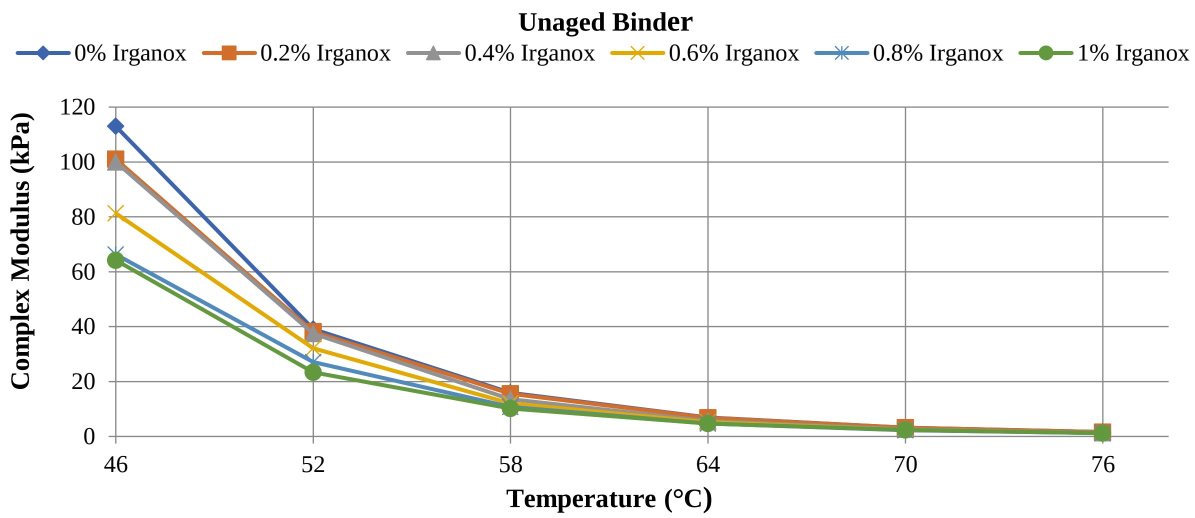


Fig. 6. Complex modulus variation of binders of unaged binders

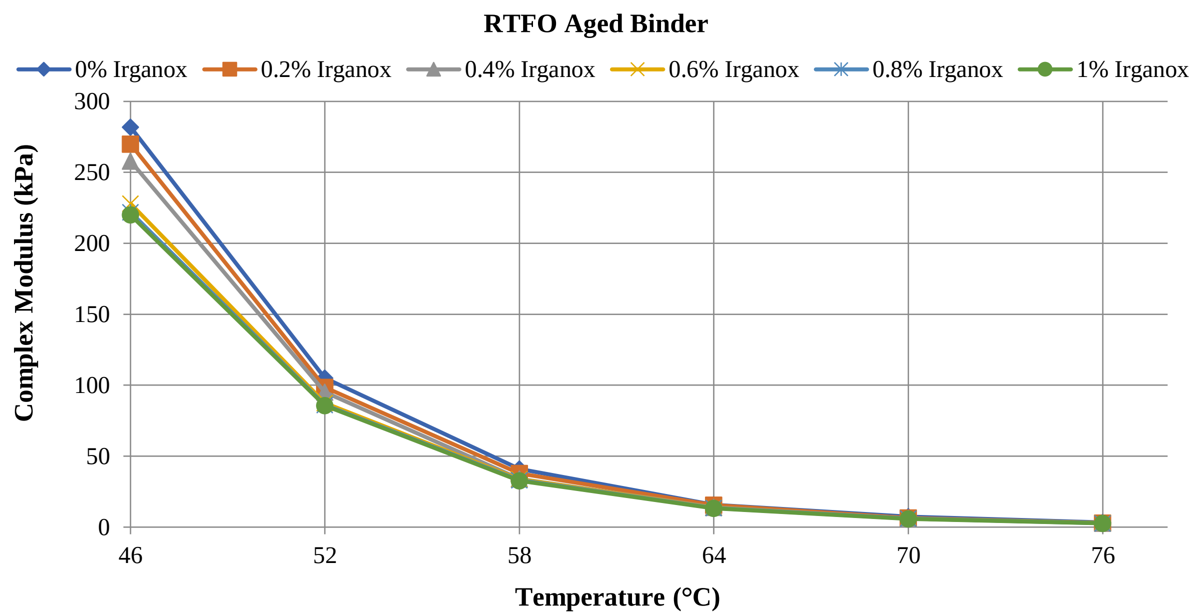


Fig. 7. Complex modulus variation of binders after RTFO ageing

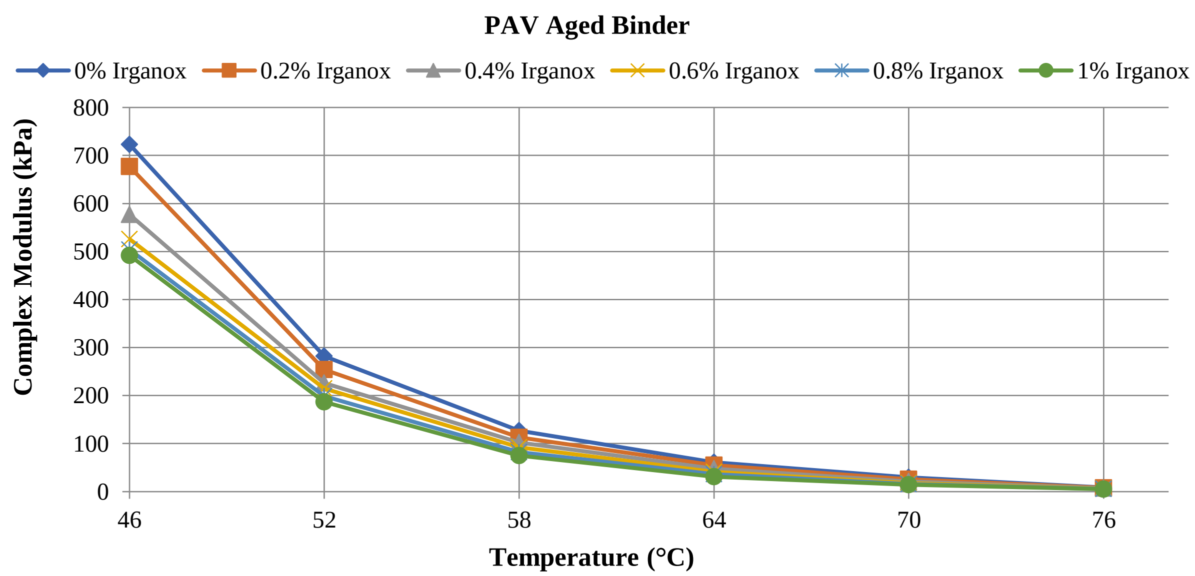


Fig. 8. Complex modulus variation of binders after PAV ageing

It can be observed that from the Figures 6-8 that for both unaged and aged binders complex modulus values decrease with subsequent introduction of Irganox 1010 signifying decrease in stiffness with Irganox 1010. The complex modulus values for PAV aged binders were higher than RTFO aged binders and both were subsequently higher as compared to unaged binders corresponding to individual temperatures meaning thereby that the binders became stiffer with ageing. The results are in line with observations made for penetration and softening point values and can be attributed to loss of volatile materials and increasing the asphaltene content leading to rise to stiffness (Hofko et al., 2017; Read and Whiteoak, 2003).

3.2.3. Phase Angle (δ)

Phase angle was also obtained from DSR results performed in accordance with ASTM 7175-15. Phase angle provides an overview about the viscous and elastic nature of binders. Higher phase angle signifies an elastic binder. The results found have been plotted in Figures 9-11. It was observed from the phase angle values that with the ageing there is a loss of binder elasticity due to decreased phase angle. The phase angles were compared for individual temperatures at different ageing conditions. This loss of elasticity or increase in brittleness subsequently promotes fatigue cracking. However, when Irganox 1010 was introduced, it was found that phase angle values increases thereby signifying a more elastic binder.

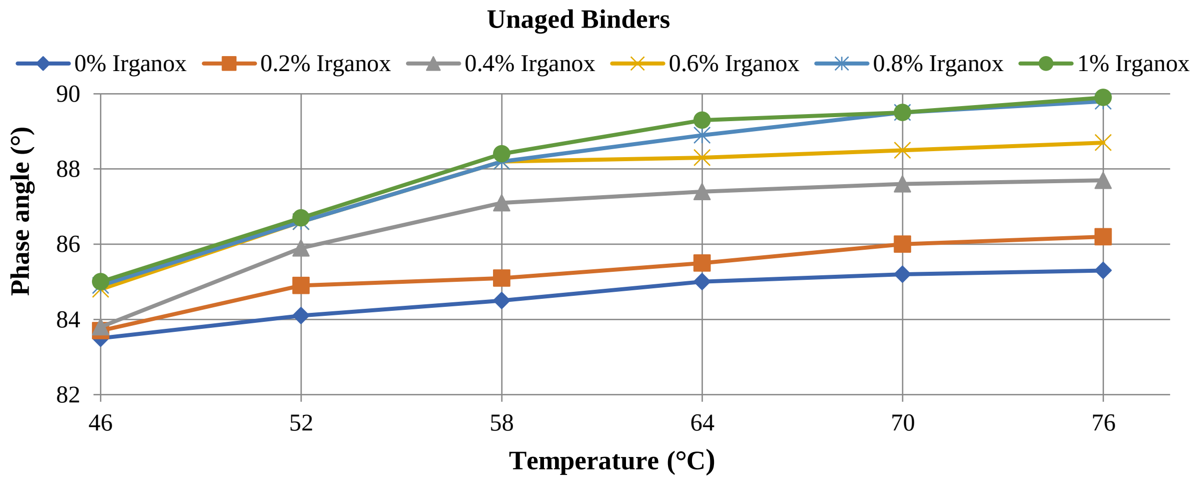


Fig. 9. Phase angle variation of unaged binders

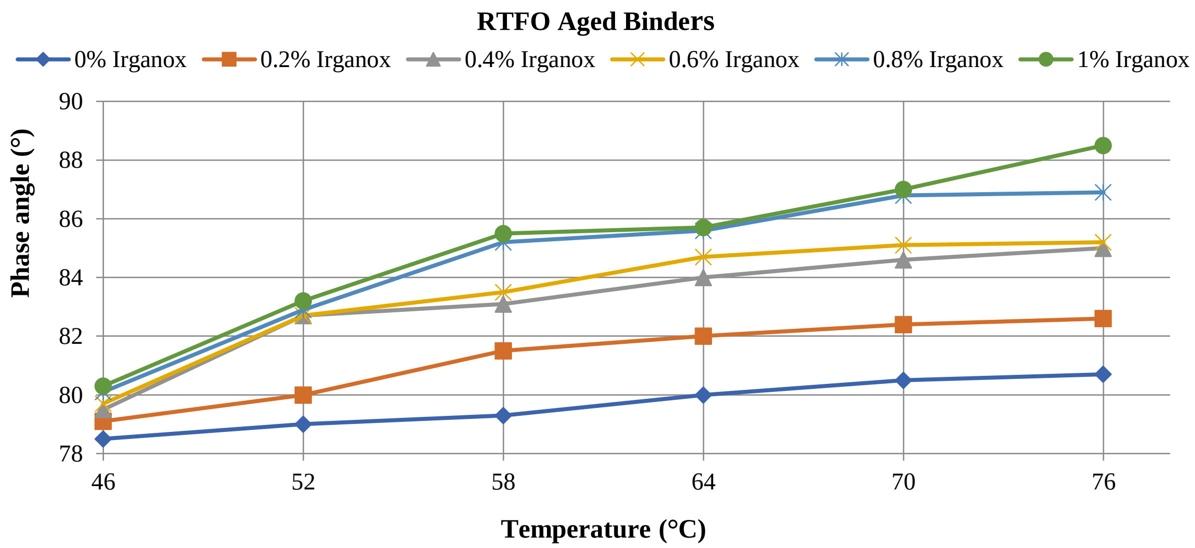


Fig. 10. Phase angle variation of binders after RTFO ageing

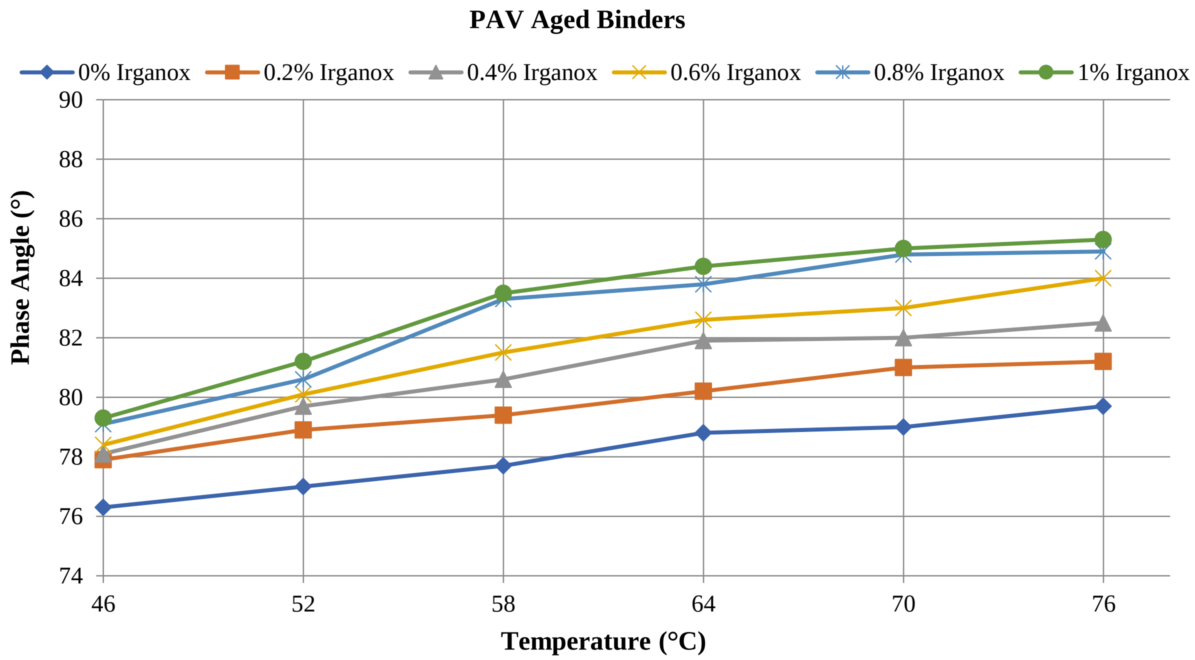


Fig. 11. Phase angle variation of binders after PAV ageing

3.2.4. Softening Point Increment (SPI) and Viscosity Ageing Index (VAI)

The ageing characteristics of bituminous binder have been estimated with the help of two indexes in this study namely the Softening Point Increment (SPI) and Viscosity Ageing Index (VAI) (Feng et al., 2011; Cong et al., 2012; Cong et al., 2013; Zhao et al., 2015). These are as can be observed in Eqs. (1) and (2).

$$SPI = SP_2 - SP_1 \quad (1)$$

$$VAI = \frac{V_2 - V_1}{V_1} \quad (2)$$

where, SP_1 and SP_2: indicate softening point of un-aged and aged bitumen, respectively; V_2 and V_1: indicate viscosity of un-aged and aged bitumen, respectively in Pa-s. Lesser values of SPI and VAI signify enhanced anti-ageing performance of bitumen (Cong et al., 2013).

The ageing characteristics of binder with different amount of Irganox 1010 are provided in Figures 12 and 13. After RTFO ageing, binders modified using Irganox 1010 show continuous decrement in SPI and VAI values when the amount of Irganox 1010 is increased gradually. It indicates the improvement of thermal-oxidative anti-ageing behaviour of bitumen.

The consequence of Irganox 1010 on PAV aged binder can be observed to have decreasing trend initially. However, after 0.6% addition, a rise in values can be seen for both SPI and VAI results. The Irganox 1010 shows supporting results on the anti-ageing behaviour of PAV aged binder while the amount of Irganox 1010 is not greater than 0.6 wt%, but opposing influence on the anti-ageing behaviour is shown when the amount of Irganox 1010 is more than 0.6% weight of bitumen. This behavior is observed for PAV aged binder and not for RTFO aged binder. It can be attributed to the fact that long term ageing simulated by PAV gives a wholistic overview of the thermal oxidative ageing of binder as compared to short term aged RTFO binders.

The reason and mechanism of action of Irganox 1010 on the aged bitumen has been explained in few contemporary literatures (Feng et al., 2011; Zhao et al., 2015). It may be concluded that Irganox 1010 deflocculates the asphaltene which is responsible for imparting hardness to the aged bitumen by the process of peptization (Zhao et al., 2015; Lian et al., 1994). This action thus improves the resistance of the bitumen for both short- and long-term aging.

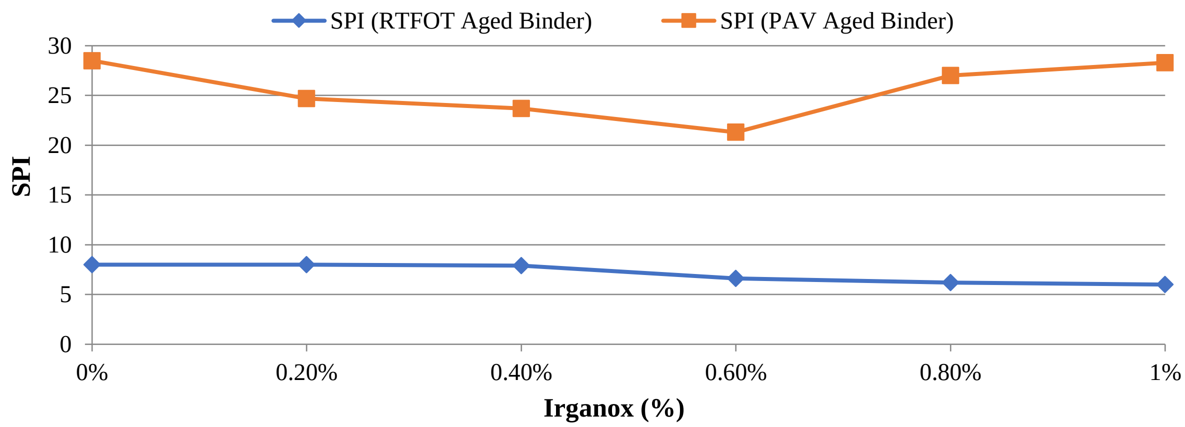


Fig. 12. Effect of Irganox 1010 contents on SPI (°C) of bitumen

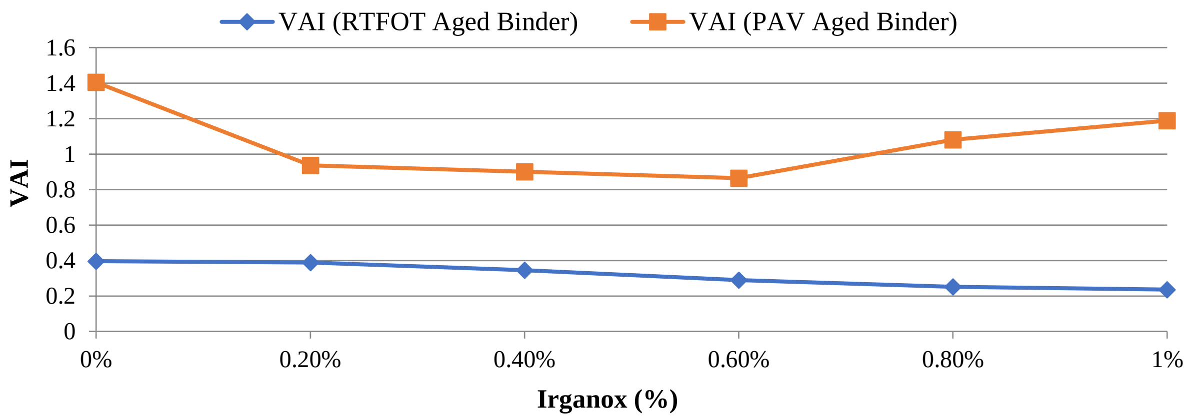


Fig. 13. Effect of Irganox 1010 contents on VAI of bitumen

4. Conclusions

The process of ageing of bitumen leads to stiffer bitumen which can be observed from respective physical and rheological tests. As observed from literature, this change is attributed to loss of volatile aromatic compounds and increase in asphaltene contents due to the process of ageing.

Various anti-ageing materials are used to mitigate the problem of ageing and Irganox1010 is one of them. The use of this anti-oxidant was found to be limited in literature. However, due to lower cost and wide scale availability this material was selected for this study. While increasing the antioxidant Irganox 1010 content in bitumen, the softening point, viscosity, complex modulus decreases gradually whereas penetration and phase angle increases gradually. After RTFO ageing, SPI and VAI values of binders gradually decrease with increase in antioxidant content, which indicates the improvement in ageing resistance of binders. SPI and VAI values of binders after PAV ageing are observed to be having slightly different nature. Firstly, both indexes decrease up to 0.6% of Irganox 1010 and then after both of them increases. It can thus be concluded that Irganox 1010 improves resistance of bituminous binders to ageing and 0.6 wt% of Irganox 1010 is the appropriate content of antioxidant for VG30 binder. Literature explains that Irganox 1010 deflocculates the asphaltene by the process of peptization and inhibits gelatinization of the binders subjected to ageing. This however, needs to be ascertained by carrying out detailed chemical analysis and studying of the microstructures of the bituminous binders and the mixes.

Thus, it can be concluded that Irganox 1010 can be considered as potential anti-ageing bitumen modifier considering smaller quantity, lower cost and wide scale availability to enhance durability of flexible pavements. However, other performance evaluation for Irganox 1010 modified binders would further help for confirmation of appropriate content of antioxidant.

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