Effect of Short Term Aging on Unmodified and Local Crumb Rubber (LCR) Modified Bitumen

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Abstract

Two grades of bitumen, (60/70 and 80/100), were modified by addition of Local Crumb Rubber (LCR) by weight of base bitumen. To investigate the effects of short-term aging on modified and LCR modified bitumen, Rolling Thin Film Oven (RTFO) test was used to simulate the short term aging. Dynamic shear rheometer (DSR) was used to assess the rheological properties of bitumen, both before and after aging. It was observed that at 65°C on aging, the phase angle (δ) of unmodified bitumen decreased by 3 and 4% for 60/70 and 80/100 bitumen respectively; whereas, for 60/70 LCR modified bitumen, the phase angle (δ) on RTFO aging decreased by 7% for all LCR content (10, 15 and 20%) by weight of base bitumen. However, on RTFO aging (δ), values for LCR modified 80/100 bitumen varied across different LCR content and could not be generalized. The complex modulus |G*| of LCR modified 60/70 bitumen increased on aging for both modified and unmodified bitumen at 65°C, but with the increase in LCR content, the difference between bitumen aged and un-aged values of |G*| was considerably lowered. An interesting finding was for 80/100 bitumen modified with 20% LCR content by weight, which showed a reduction in |G*| values and an increase in (δ) values on aging. This shows that to some extent, LCR compensates for the stiffening effects of aging.

Keywords—Rolling Thin Film Oven (RTFO), Dynamic Shear Rheometer (DSR), Local Crumb Rubber (LCR), Aging, Bitumen

1 Introduction

Crumb Rubber (CR) from end of life tyres is used to improve the engineering properties of bitumen. Two types of processes, namely, wet process and dry process are currently being used to modify the properties of bitumen. Wet process involves mixing bitumen with CR at elevated temperature prior to its mixing with aggregates, whereas, in dry process the CR is mixed with heated aggregates before addition of bitumen. [1][2]. Bitumen consists of two broad fractions: the lighter fractions called the maltenes, and heavier fractions called the asphaltenes. During bitumen-CR interaction, the CR particles absorb the lighter fractions of bitumen and swell [3]. Thus CR modified bitumen consists of a liquid phase and swollen particle phase. The modification of bitumen generally occurs due to migration of lighter fractions from bitumen into CR particles. This leads to a reduction of lighter fractions of bitumen and results in an increase in complex modulus |G*| values and a reduction in phase angle (δ) values [4][5]. Factors affecting the properties of CR modified binder include time and temperature of bitumen – CR interaction, CR particle size, shape and content [6][7]. The reaction between CR and bitumen is highly dependent on time and temperature. If any of these two parameters are in excess, the rubber particles will go into a de-polymerization phase and beneficial effects of modification will be lost [4]. According to Mashan et. al., the properties of CR modified bitumen are directly affected by the quantity of CR modifier [7]. With respect to size and shape of CR particles, various researchers have shown that increasing the size of crumb rubber particles results in decrease in (δ) values and an increase in |G*| values. Thus, CR modified bitumen produced from coarser particles tend to have better elastic properties due to
Fig. 1: The methodology adopted in the research

lower value of ($\delta$) and high rutting resistance due to higher $|G'^*|$ values [8].

In general, bitumen undergoes two types of aging: (i) short-term aging that occurs during the initial asphalt production and laying, (ii) and a long term aging that continues throughout the service life of the pavement and is the result of environmental factors [9][10]. Main types of aging processes that are considered when studying the aging of bitumen include oxidation, evaporation of lighter fractions, absorption of bitumen fractions by aggregates, and physical hardening, also known as steric hardening. Oxidation is considered to be the main cause of aging and results in the changes in chemical structure of bitumen [9]. The changes in chemical structure result from increase in asphaltenes contents and their molecular weight, and is accompanied by decrease in the maltenes (aromatic oils) [11][12]. The chemical changes are mostly the result of combination of processes that involve oxidation, polymerization and evaporation of lighter fractions. This leads to hardening of the binder, and makes it more susceptible to cracking failures [12].

Aging of bitumen modified with crumb rubber is a complex phenomenon, since aging of crumb rubber modified bitumen is affected by both oxidation process and polymer degradation [10]. According to Lesueur [12], short term aging that occurs at the temperature of 160°C, shows good correlation with aging using the rolling thin film oven test. This study focuses on short-term aging of bitumen and its effects on bitumen properties, since before the asphalt mix reaches the site, considerable aging has taken place during mixing process that affects the in situ performance of bitumen. The CR that is being used for modification of bitumen is obtained from two distinct industrial processes: ambient and cryogenetic. The local crumb rubber (LCR) that is used in this research is obtained from neither of the two processes, but is a waste product generated from local shoe industry during grinding of shoe soles which are made up of waste rubber products such as end of life tyres, conveyor belts, etc. In this research, two grades of bitumen 60/70 and 80/100 are modified with LCR using the wet process. This study demonstrates the usefulness of LCR as a modifier for bitumen.

2 Material & Methods

Local Crumb Rubber (LCR) used for this research was obtained from local shoe makers, where it is generated as a result of utilization of waste rubber products such as end of life tyres, conveyor belts, etc. in shoe making and is dumped as a waste product. LCR passing sieve# 30 having following gradation (Table 1) was used in this research as bitumen modifier. Two grades of bitumen (pen grade 60/70 and 80/100) were used as base bitumen. LCR was mixed with based bitumen at a temperature of 180°C. Mixing was carried out in a laboratory asphalt mixer for 30 minutes at a speed of 1400 rpm. Glass jars filled with binder were placed in RTFO apparatus to simulate short-term aging. The temperature was kept at 163°C and test duration was set to 80 minutes in accordance with AASHTO - T240 [13]. On un-aged and the residue from RTFO test, frequency sweep was run from (0.1 to 100) rad/sec using DSR for temperature range 15°C, 25°C, 35°C, 45°C, 55°C, 65°C and 75°C. Dynamic Shear Rheometer (DSR) test assembly used was 25 mm plate with 1 mm plate gap.

| Sieve Number | #30 | #50 | #100 | #200 |
|--------------|-----|-----|------|------|
| Percent Passing | 100 | 49.1 | 12.87 | 4.19 |

TABLE 1: LCR gradation

3 Results & Discussion

Following sections describe the results of this study.

3.1 Rolling Thin Film Oven (RTFO) Test

The result of mass loss percentage after RTFO aging is given below. The results indicate that the LCR
modified bitumen has higher mass loss percentage than the unmodified bitumen. As the LCR content is increased, the mass loss percentage increases up to a maximum value of 0.08% at 15% LCR content by weight of base bitumen for 60/70 bitumen. On further increase in the LCR content, the mass loss percentage reduces to 0.07% which is still comparatively higher than the unmodified bitumen. For 80/100 bitumen, the mass loss percentage increases to a maximum value of 0.19% at 10% LCR content after which it begins to reduce on further increase in the LCR content.

3.2 Isochronal Plot

The phase angle ($\delta$) isochronal plots for 60/70 and 80/100 bitumen are given below. It can be observed that addition of LCR results in decrease in the ($\delta$) values at high temperature. The ($\delta$) continues to decrease with the increase in LCR content. This finding is similar to that of conventional crumb rubber modified bitumen [7][4][8]. This indicates that the increase in LCR content results in a decrease in the viscous component of bitumen. From Figure 3 and 4, it can be seen that with the increase in temperature, the ($\delta$) value approaches 90°C which indicates that the material is approaching a viscous state [14]. It can be observed that on aging, the phase angle values reduces, indicating a loss of viscous component of bitumen. The difference between ($\delta$) values is more evident at temperature greater than 45°C. Furthermore, at temperature less than 20°C, the LCR modifier bitumen has higher $\delta$ values. As the temperature increases, the graph plummets and then steadily rises. This behaviour is similar for both RTFO-aged and un-aged samples, and can be attributed to the particle effect of LCR and DSR plate geometry [15][16]. Both grades of bitumen 60/70 and 80/100 show the same behaviour for phase angle parameter at 10 rad/sec.

Comparison of aged and un-aged bitumen for modified and unmodified bitumen is given below in the bar charts to further quantify the effects of aging on bitumen. It can be observed that for 60/70 bitumen, aging increases the value of ($\delta$) at low temperature of 15°C, whereas at a high temperature of 65°C, it results in the decreases in ($\delta$) values. At temperature of 15°C, the value of ($\delta$) decreases by 3% on aging unmodified bitumen, and increased by 10%, 9% and 27% for 10, 15 and 20% LCR content respectively by weight of base bitumen. At temperature of 65°C, the value decreased by 3% for unmodified bitumen and decreased by 7% for all quantities of LCR content. 80/100 bitumen showed varying response. At low temperature, the ($\delta$) increased by 15% on aging for unmodified bitumen, and increased by 10% and 20% for 10% and 15% LCR content respectively by weight of base bitumen. The smallest increase of 5% in ($\delta$) value was observed for 20% LCR content. At high temperature of 65°C, ($\delta$) decreased by 4% and 1% for unmodified bitumen and for 15% LCR content, respectively. Whereas, for 10% and 20% LCR content, there is an increase in the ($\delta$) value.

From figure 9 and 10, after aging using RTFO, the values of complex modulus ($|G^*|$) increased. The increase in the values of ($|G^*|$) was evident both at high and low temperature, except for the RTFO aged 60/70 bitumen as the temperature is reduced. Its ($|G^*|$) value also reduces and from the temperature range of 25°C begins to fall behind un-aged 60/70 bitumen. The trend of increase in ($|G^*|$) values is similar for all LCR modified samples.

To further quantify the response of bitumen, at the high and low temperature range, the bar charts of ($|G^*|$) of bitumen sample at 65°C and 15°C are given below in Figure 11-14. It can be observed that for 60/70 bitumen RTFO, aging results in an increase in the ($|G^*|$) value for both 65°C and 15°C except for unmodified bitumen at 15°C, where there is a slight reduction in the ($|G^*|$) values from 257KPa to 249 KPa. On RTFO aging, both modified and unmodified bitumen showed an increase in the ($|G^*|$) at 65°C, however, the extent of increase was limited to the increase in LCR content, as value of ($|G^*|$) for unmodified bitumen increases by 102% on RTFO aging. Whereas, for 10%, 15% and 20% LCR content, it only increases by 35%, 29% and 17% respectively. This shows that, to some extent, LCR reduces the stiffness of bitumen. For 80/100 bitumen, the effects of RTFO aging vary across different LCR content at both 65°C and 15°C. At 15°C, an increase in the value of ($|G^*|$) is observed with the increase in LCR content. The RTFO aged 80/100 bitumen attain the maximum value of 350 KPa at 15% LCR content.
Fig. 3: Phase angle ($\delta$) isochronal plot 60/70 bitumen

Fig. 4: Phase angle ($\delta$) isochronal plot 80/100 bitumen

Fig. 5: Phase angle at 15°C

Fig. 6: Phase angle at 65°C
at 15°C. At 65°C, 80/100 bitumen show unique behaviour for each LCR content. The value of $|G^*|$ increase from 0.85 KPa to 2.09 KPa, and from 7.82 KPa to 8.68 KPa for 0% and 15% LCR content respectively. It reduces from 5.79 KPa to 3.63 KPa and 8.27 KPa to 4.87 KPa for 10% and 20% LCR content. According to Haopeng et al. [17], during aging, oxidation and polymer degradation for polymer modified bitumen occur simultaneously. Due to this reason, the stiffening effect of oxidation to some extent are compensated by the softening effect of polymer degradation. However, inconsistencies do exist in the values of $|G^*|$ and $(\delta)$, therefore, it is not possible to completely characterize the aging phenomenon based on the selected values. Furthermore, the lowering of $(\delta)$ values and increase in $|G^*|$ is visible at high temperature than at low temperature. Studies attribute this phenomena to the increased CR contribution to the bitumen rheology at high temperature as compared to low temperature [18].

3.3 Master Curves

For 60/70 bitumen, the $(\delta)$ master curves fall to lower $(\delta)$ values on aging, which is consistent for both modified and unmodified bitumen; however, at low temperature, considerable overlapping is observed between RTFO aged, modified and unmodified bitumen. Some scattering of the graph is also observed for RTFO-aged 20% LCR modified bitumen. A plateau is observed in the low frequency region (high temperature) for LCR modified bitumen, from where the value of the $(\delta)$ begins to fall below the curve in the $(\delta)$ master curve. The curvature of the curve at this low frequency region increases on both RTFO aging and on increasing LCR content. For 80/100 bitumen, some scattering of $(\delta)$ values is observed in the high frequency region, but it was lower than the 60/70 bitumen. Interestingly, RTFO-aged 80/100 bitumen modified with 15% LCR gives the lowest values of $(\delta)$ in the low frequency region. Its curvature is also visibly the highest. Another interesting thing is the behaviour of RTFO-aged 20% LCR modified bitumen, which is characterised by overlapping with RTFO-aged 10% LCR modified bitumen both at high and low frequency region.

For 60/70 bitumen, the values of $|G^*|$ increase both on aging and with the increase in LCR content. The effect of LCR addition and aging are more distinguishable in the frequency range 1.E-06 rad/sec-1.E-01 rad/sec. The un-aged samples show increase in the $|G^*|$ values on addition of LCR. The stiffening effect of LCR continue to increase with the increase in LCR content, as evident from the increase in $|G^*|$ value. For un-aged sample, the general behaviour remains the same with the LCR modified samples showing higher $|G^*|$ values. For the case of 80/100 bitumen, RTFO-aged LCR 20% values overlap with RTFO-aged LCR 10%. The values of RTFO-aged LCR 20% was also lower than the un-aged LCR 20% values, which shows that some form of softening effect of LCR modification is present to balance the stiffening effect of aging. According to Haopeng et al. [17], bitumen modified with higher percentage of crumb rubber shows better resistance against stiffening effects of aging. The difference in response between 60/70 and 80/100 bitumen can be attributed to the difference in grades of bitumen, as softer grade bitumen are known to enhance the swelling of crumb rubber particles due to the presence of greater amount of light-weight fractions. This results in better high and low temperature performance of modified binder [19].

4 Conclusion

From the results it can be concluded that:

- Addition of LCR reduces $(\delta)$ value of bitumen. The effect of $(\delta)$ modification is more visible at high temperature than at low temperature.
reduction in $(\delta)$ value of bitumen indicates a shift towards a more elastic response. This observation indicates that LCR can be used to improve the high temperature performance of bitumen. The modification of $(\delta)$ on addition of LCR is similar to that of conventional crumb rubber as observed.
by other researchers.

- RTFO aging results show an increase in $|G^*|$ and a decrease in $(\delta)$ values for both grades (60/70 and 80/100) of unmodified bitumen. Similar result is obtained for 60/70 LCR modified bitumen. This shows that aging results in a more stiff and less viscous bitumen.

- For LCR modified 80/100 bitumen, the effects of RTFO aging vary for different LCR content. It is observed that for 20% LCR modified bitumen, there is a decrease in stiffness and $|G^*|$ values, but an increase in viscous component and $(\delta)$ values. This shows that the characteristics of base bitumen have a considerable effect on the properties of RTFO-aged LCR-modified bitumen. The increased LCR content for softer grade bitumen results in better aging resistance.

- To generalize these findings, further studies are needed with different bitumen sources and grades.

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Fig. 15: $|G^*|$ and $(\delta)$ master curve 60/70 aged and un-aged bitumen

Fig. 16: $|G^*|$ and $(\delta)$ master curve 80/100 aged and un-aged bitumen

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