RHEOLOGICAL TESTING OF CRUMB RUBBER MODIFIED BITUMEN

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ABSTRACT

The development of improved test protocols and procedures for asphalt materials is currently been pursued by the Council for Scientific and Industrial Research (CSIR) to support the revision of the existing South Africa flexible pavement design method.

The determination of binder rheological properties using the dynamic shear rheometer (DSR) is specified in AASHTO (American Association of State Highway and Transportation Officials) Test Method T 315-05. A major draw-back of this test method when used to analyse crumb rubber modified (CRM) bitumen is that the specified gap setting in the configuration of the DSR between the upper and lower test platens is too small to accommodate crumb rubber particles. DSR testing of CRM bitumen therefore requires a plate gap adjustment to avoid any influence by the rubber particles. This paper covers key aspects of experimental work currently being undertaken at the CSIR Transport Infrastructure Research Group laboratories to address some of the issues related to rheological characterisation of CRM bitumen. The paper illustrates the effect of the DSR plate gap setting on the measured linear visco-elastic properties of the sample. The optimum gap setting is defined as that range where repeatable values are obtained for the complex shear modulus within the linear visco-elastic limit.

KEY WORDS

Crumb Rubber Modified Bitumen, Rheological Testing, Dynamic Shear Rheometer

INTRODUCTION

In South Africa, CRM bitumen is manufactured through blending penetration grade bitumen, rubber crumbs and extender oil, all complying to the requirements set by TG1: The Use of Modified Bituminous Binders in Road Construction (2nd Edition, November 2007). The blending is done at elevated temperatures of between 190 - 210ºC (Potgieter, 2003) by a high speed stirring device; the process taking between 1 to 4 hours.

Rubber crumbs are obtained by the shredding of vulcanized tyres at ambient temperature. Rubber crumbs consist primarily of a three dimensional network of poly-isoprene and poly-butadiene polymers, linked by sulphur-sulphur bonds. Upon blending, the rubber particles initially swell as the oil and/or lighter components of the bitumen diffuse into the rubber increasing particle dimensions (Airey *et al.*, 2002). This results in an initial increase in viscosity. Subsequently, thermal dissociation of the sulphur-sulphur linkages occurs resulting in a further incorporation of the aromatically ‘peptized’ rubber particles into the binder. This causes an additional viscosity increase in a process referred to as the ‘digestion of the rubber particles’. This process continues until the CRM bitumen blend reaches a maximum viscosity. Beyond this point, the viscosity decreases with digestion time due to the gradual loss of the three dimensional network as a result of thermal disassociation of the sulphur links. Hence CRM bitumen properties are not constant, they change with temperature, digestion time and energy consumed during the digestion process.

Rheological testing practices available in the literature for testing CRM bitumen differ substantially from each other due to the varying compositions of these binders in terms of rubber crumb particle size, type and percentage in a given blend. Additionally, the actual testing of the binder has its challenges such as the settling of rubber particles during measurements. Fortunately, the type, amount and size of rubber crumbs in South African CRM bitumen are fairly consistent.

DSR testing of CRM bitumen requires a plate gap adjustment to avoid the influence of the rubber particle size (Shen and Amirkhanian, 2005). It has been shown by Tayebali *et al.* (1997) that linear visco-elastic properties can be compared at different gap sizes. Researchers have attempted to adjust the plate gap (Lee *et al.*, 2008 and Jeong *et al.*, 2010) to counter the particle size effect, but only Souza *et al.* (2005) have proposed a ratio between rubber particle size to DSR gap size. But they do not indicate how they arrived at such a ratio; whether the particle size referred to is the mean or maximum and/or whether the size used was before or after the swelling of the rubber particles. This paper investigates the effect that changing the DSR gap setting has on the measured linear visco-elastic properties of South African CRM bitumen.

CRM BITUMEN CHARACTERISATION

An 80/100pen grade bitumen was used as the base bitumen (as typically done in South Africa) complying with South African National Standard (SANS) 307 (2005) specification requirements.

Rubber Crumbs of the grading depicted in Figure 1 were used. The rubber particles essentially pass the 1.18mm sieve and the majority retained on the 0.6mm sieve. The rubber crumb surface texture/morphology is shown in SEM photographs of Figures 2-4. The porous microstructures are consistent with comminution via an ambient process (Shen and Amirkhanian, 2005) as opposed to cryogenic crushing. The white spots seen in Figures 3 and 4 are most likely filler (Xiao *et al.*, 2006) and not metal fragments (Shen and Amirkhanian, 2005) since South Africa only imports non-steel reinforced tyres for rubber crumb manufacture.

The identification of the elemental composition of the rubber crumbs shown in the SEM micrographs was done using an Energy Dispersive System (EDS). It uses an energy dispersive X-ray spectrum to determine the elemental composition of specific points on an SEM image. The results of the elemental analysis of the rubber crumbs are shown in Table 1, typically (Xiao *et al.*, 2006) containing Carbon, Oxygen, Silicon, Sulphur and traces of the metals: Calcium, Sodium, Aluminium and Zinc from the vulcanization component, Zinc Oxide (ZnO) (Mark *et al.*, 2005).

The resultant CRM bitumen blend consisting of an 80/100pen grade bitumen (72-82%), rubber crumbs (18-24%) and extender oil (0-4%) (TG1, 2007) was blended by Much Asphalt (Pty) Ltd and when tested conformed to national TG1 (2007) specification requirements for all tests except resilience, for which it was slightly above specification.

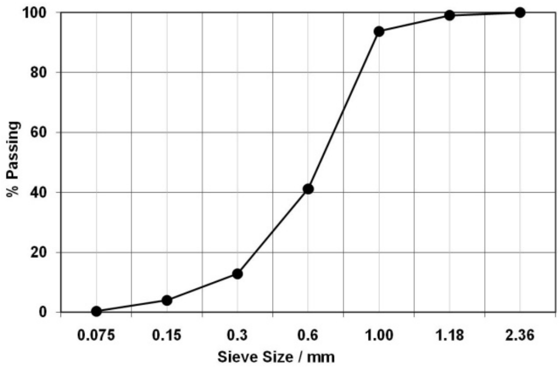


Figure 1: Grading of Rubber Crumbs.

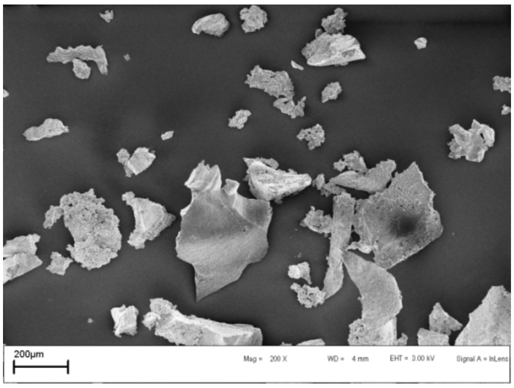


Figure 2: SEM analysis of the rubber crumbs.

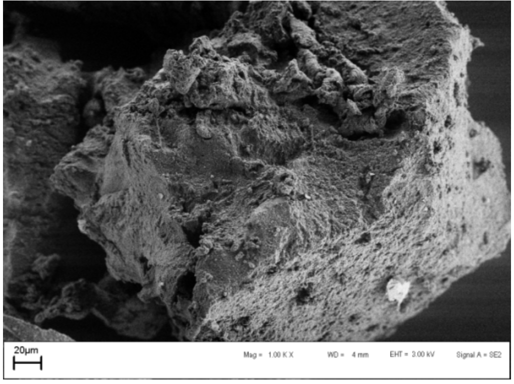


Figure 3: SEM analysis of the rubber crumbs.

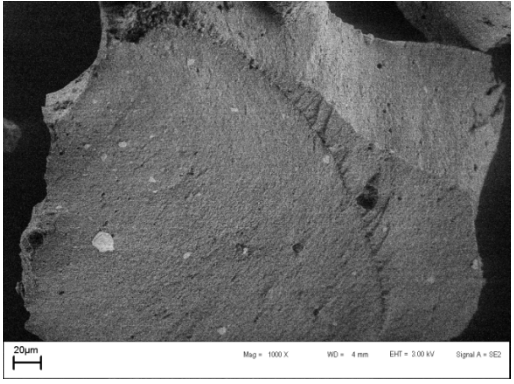


Figure 4: SEM analysis of the rubber crumbs showing surface texture.

Table 1: Elemental Composition (by weight) of the rubber crumbs

Spectrum	**C**	**O**	**Si**	**S**	**Ca**	**Na**	**Al**	**Zn**	**Total**
Sample 1	81.93	16.60		0.91		0.55			100.00
Sample 1 Repeat 1	76.54	23.46				<0.005			100.00
Sample 1 Repeat 2	86.42	11.19	0.58	1.36	0.44			<0.005	100.00
Sample 1 Repeat 3	85.91	11.52		2.08			0.50	<0.005	100.00
Sample 1 Repeat 4	88.71	8.02	0.75	2.03			0.51	<0.005	100.00
Max. Detected	**88.71**	**23.46**	**0.75**	**2.08**	**0.44**	**0.55**	**0.51**	**<0.005**	
Min. Detected	**76.54**	**8.02**	**0.58**	**0.91**	**0.44**	**<0.005**	**0.50**	**<0.005**	

DSR GAP INVESTIGATIONS

According to AASHTO Test Method T 315-05, the limits of the test temperature and frequency ranges are a function of the binder stiffness. The DSR model employed in this investigation was an Anton Paar Physica Smartpave Plus that uses a Peltier system with a parallel plate measuring configuration. SHRP-A-370 (1994) guidelines were used for selecting plate diameters and the initial sample thickness (gap), which was subsequently varied.

The effect of rubber crumb particle size in relation to the gap size was investigated by monitoring the effect of gap size on the CRM bitumen stiffness. A 60/70 penetration grade bitumen was used as a control instead of the base binder, since it is similar in stiffness to the CRM bitumen characterized in this investigation. It is also the most commonly used unmodified binder in South Africa. Frequency sweeps of unmodified 60/70pen grade bitumen were run at various gap settings using the 25mm DSR plate. A strain of 1% was used at a testing temperature of 55°C where the amplitude was still within the linear viscoelastic limit. Figure 5 shows good reproducibility for repeated frequency sweeps at various gap heights (between 1-2mm) for the control unmodified 60/70pen grade bitumen. This implied the sample successfully adhered to both plates without slippage, and deformed homogenously throughout the shear gap.

The CRM bitumen was similarly tested at various gap settings. The same strain of 1% was selected at the testing temperature of 55°C. Figures 6-8 show how at the lower gap settings, frequency sweeps were not reproducible whether repeated at the same gap or at different gaps. Since the calculation of the rheological parameters of the 60/70pen grade bitumen was not affected by the shear conditions, such variations must be due to interference by the rubber crumbs. Upon increasing the gap, a range is reached at approximately 1.6-2.6 mm where the frequency sweeps were once more reproducible (see Figure 9). This signifies that crumb rubber particles no longer affect the measurements. Testing specimens at progressively bigger gap settings setting also resulted in a loss of test reproducibility (see Figure 10). This signifies that the binder sample did not deform homogenously throughout the shear gap.

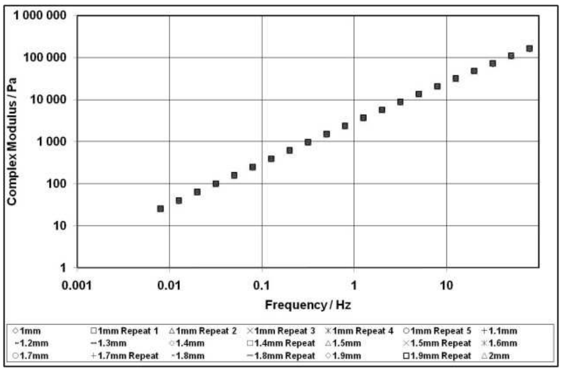


Figure 5: Frequency Sweeps of 60/70pen grade bitumen at various gap settings.

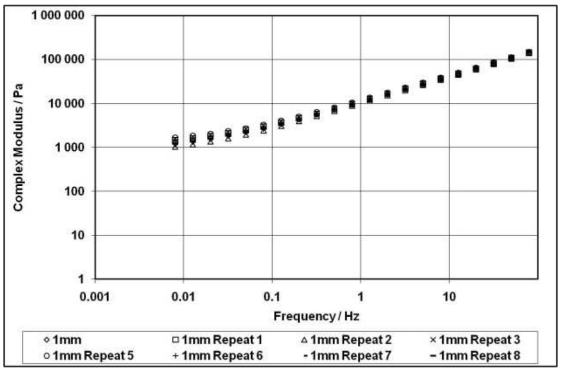


Figure 6: Frequency sweeps of CRM bitumen at 1mm gap setting.

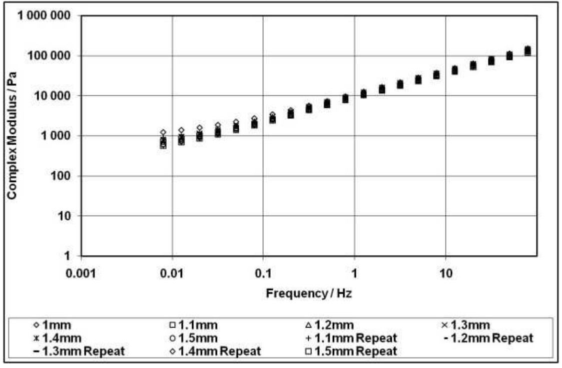


Figure 7: Frequency sweeps of CRM bitumen at gap settings between 1.0 – 1.5mm.

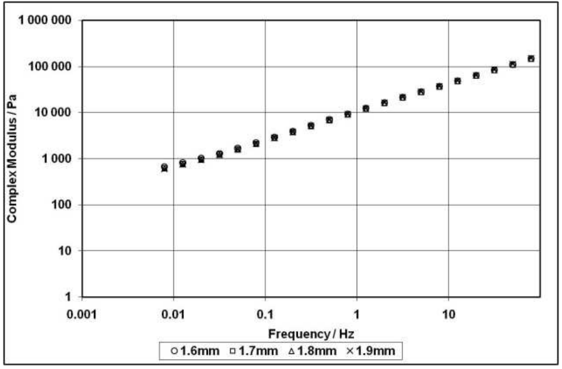


Figure 8: Frequency sweeps of CRM bitumen at gap settings between 1.6 – 1.9mm.

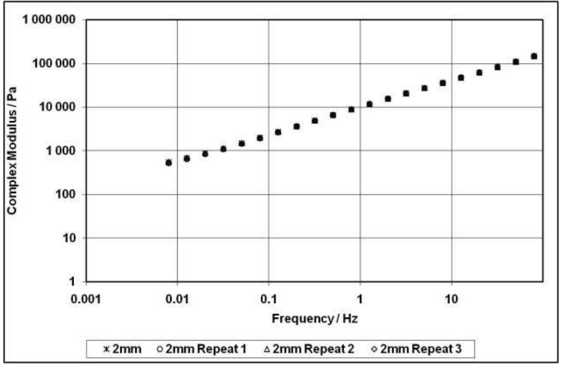


Figure 9: Repeated frequency sweeps of CRM bitumen at 2mm gap.

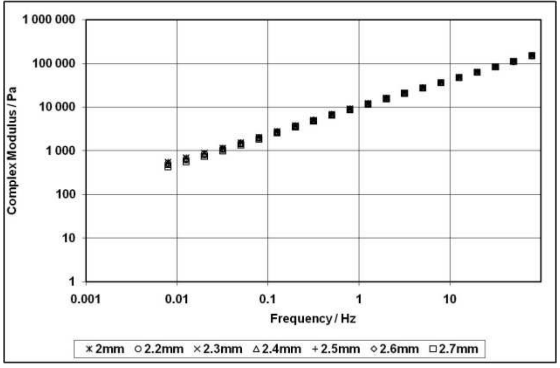


Figure 10: Frequency sweeps of CRM bitumen at gap settings between 2.0 - 2.7mm.

It is important to note that the optimum gap setting determined above applies to the particular CRM bitumen under test. Such an optimum gap setting would be dependent on the maximum rubber particle size. The maximum rubber particle size is time dependant, and it may well be that the CRM modified bitumen under test may not be at its greatest maximum particle size. The determination of the maximum particle size over time is the subject of a separate investigation.

Figure 11 shows a combined plot of a CRM bitumen frequency sweep compared to those of standard 20/30, 40/50, 60/70 and 80/100pen grade bitumens all at 55°C. The unmodified binders exhibit characteristic curves with G* values increasing in line with increased test frequency. In order of decreasing stiffness, the pen grade binder lines can therefore be ranked as follows: 20/30 > 40/50 > 60/70 > 80/100.

The effect of crumb rubber modification on the base 80/100pen curve is clearly observed in Figure 11. At the lower frequencies, the CRM bitumen tends towards G* values above those of the 20/30pen grade bitumen. At higher frequencies, the curve approaches that of the 60/70pen grade bitumen. In this way, the modified binder exhibits enhanced elasticity at lower frequencies (i.e. longer loading times where unmodified bitumens normally behave as Newtonian fluids and readily creep in response to stress) accompanied by reduced stiffness at high frequencies (where creep becomes a less important factor). The graph thus illustrates improved stability of asphalt mixes provided by CRM bitumen for slow moving traffic.

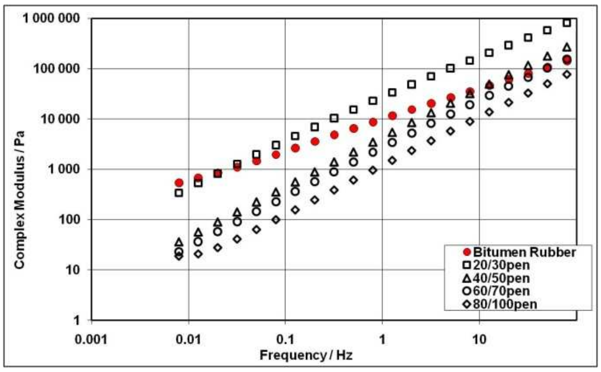


Figure 11: Frequency sweeps for CRM bitumen, 20/30pen, 40/50pen, 60/70pen and 80/100pen grade bitumen at 55°C.

The relationship between the optimum gap distance and the rubber crumb particle size was calculated. The gap distance was found to be between 1.4-2.2 times the respective maximum size of the rubber crumb particles prior to blending. The swollen rubber crumb particle size could not be determined through extraction as suggested by Attia and Abdelrahman (2009). The extraction process changes the size of the rubber particles through solvent loss and agglomeration (see Figure 12). Using SEM micrographs of the CRM bitumen blend proved too difficult for size determination (see Figure 13).

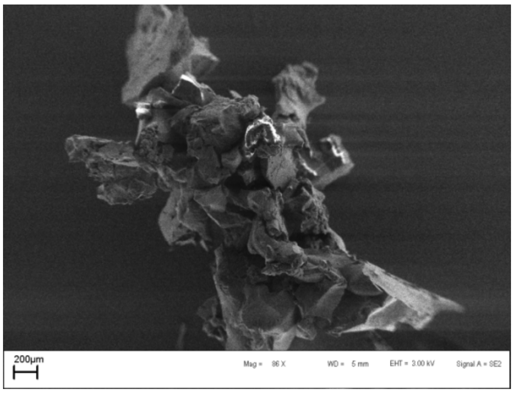


Figure 12: Recovered crumb particles.

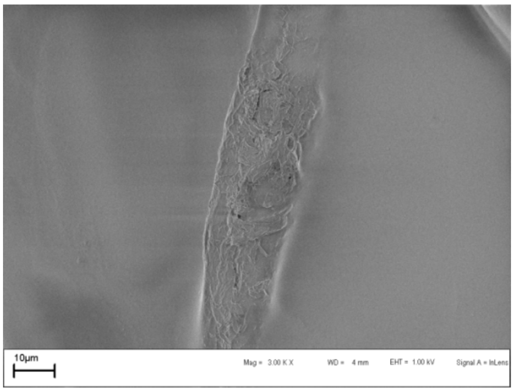


Figure 13: Rubber crumb particles in the CRM bitumen.

STRAIN SWEEP TESTS ON CRM BITUMEN

The 2mm plate gap setting was used to conduct strain sweeps on the CRM bitumen sample at 20, 40, 55 and 70°C, and the linear viscoelastic range (LVE range) was determined accordingly. Although gap settings were not investigated at temperatures other than 55˚C, it was the authors' assumption that the 2 mm gap setting would fall within the ideal gap range for the other temperatures. This assumption, however, remains to be proven. Figure 14 shows strain sweep results of the CRM bitumen compared to the base 80/100pen grade bitumen. Similar to the effect observed in previous investigations of other South African binders (Mturi *et al.*, 2010), LVE limit values for CRM bitumen increases with an increase in test temperature. The effect of rubber/oil addition on the complex modulus values is clearly demonstrated. At 20°C, the base binder has higher G* values and an almost identical LVE range to the CRM bitumen, consistent with the trend that polymer modified bitumen have lower stiffness and improved crack resistance at lower temperatures compared to unmodified bitumen. At 70°C, enhanced elastic behaviour of the CRM bitumen sample is evident from the higher G* values and enhanced resistance to deformation would be expected. Surprisingly the sample exhibited a lower LVE range to that of the base binder. Generally, polymer modification extends the LVE range and the observed reduction is most likely a function of the heterogeneous nature of the CRM binder.

The effect of rubber crumb at various temperatures may be better explained in a plot of damping factor (tan δ = G"/G') versus test temperature for a high and a low frequency in Figure 15. If the binder displays ideal-elastic behaviour, the phase angle δ = 0° or tan δ = 0, since G' fully dominates G". On the other hand, ideal-viscous behaviour occurs at δ = 90° or tan δ = ∞ since G" fully dominates G'. At the crossover frequency, the phase angle δ = 45°, which is the frequency at which G'=G", i.e. when tan δ = 1, also referred to as the sol/gel transition point. Figure 15 shows that crumb rubber modification reduces the damping factor of straight run bitumen to a level close to the sol/gel transition point, irrespective of temperature or frequency. This suggests a shift from a more sol-like viscous behaviour to a more gel-like elastic character occurs. At the higher test temperatures of 70°C and the lower frequency of 0.03Hz, the sol/gel transition point is surpassed as tan δ < 1 (since G'>G") implying the CRM bitumen blend exhibits a gel-like structural behaviour.

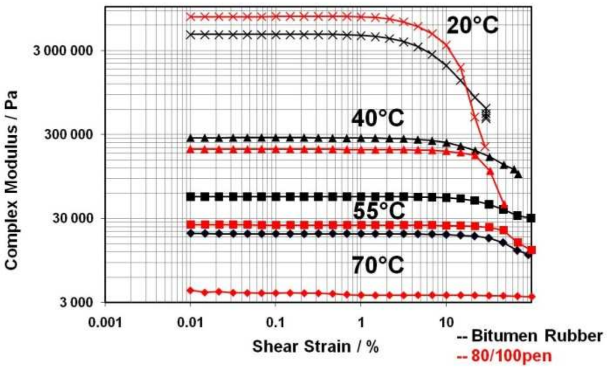


Figure 14: Strain sweeps at 20Hz

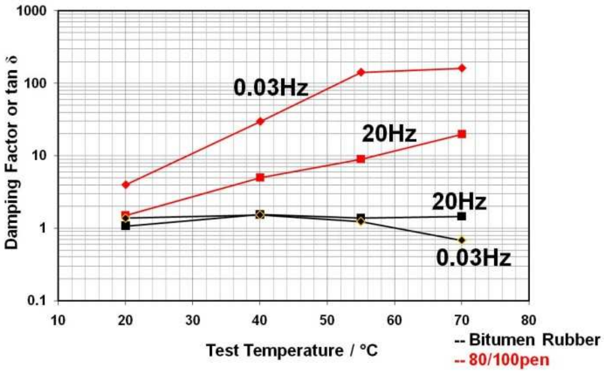


Figure 15: Damping factor at 0.03Hz and 20Hz

BLACK DIAGRAM REPRESENTATIONS

Having identified the gap distance and LVE range required for this particular CRM bitumen sample, a plot of G* (complex modulus) vs. δ (phase angle) commonly referred to as a 'black diagram' was determined. Black diagrams are often used as a 'fingerprint' of bituminous binders for quality control purposes. A combined plot of the black diagrams for CRM bitumen, SBS-modified binder and the unmodified 80/100pen grade bitumen (used as the base bitumen for the CRM bitumen blend in this investigation) are shown in Figure 16. The black diagram of CRM bitumen shows the addition of crumb rubber changes the curve of conventional bitumen into a "3" like profile. This plot indicates that crumb rubber as a modifier improved the elastic response (reduced phase angles) of the binder compared to the base bitumen. Comparing the CRM bitumen blends with the SBS sample shows that at all temperatures the CRM bitumen blends have lower δ values (higher elasticity) primarily due to the much higher elastomer content.

The rheology of the elastomer modified binders differs from that of unmodified bitumen. As the temperature increases, the complex modulus (G*) of the modifier-rich phase of modified binders decreases more slowly than the G* of the bitumen-rich phase. As a result, the overall decrease in G* of the modified binder is less than that of the unmodified bitumen. This can also be achieved through the introduction of any inert filler to an unmodified bitumen. Figure 17 shows examples of how a commercial very fine (< 0.8μm) calcium carbonate inert filler (Kulu products) high shear blended at 10% and 20% by volume of bitumen can easily be used to increase the G* of a 60/70pen grade bitumen. Due to the fineness of the filler such blends are storage stable at high temperatures. The figure compares the stiffening effect of filler loading with G* frequency sweeps of 2 other bitumen grades (namely a 40/50pen and a 20/30pen).

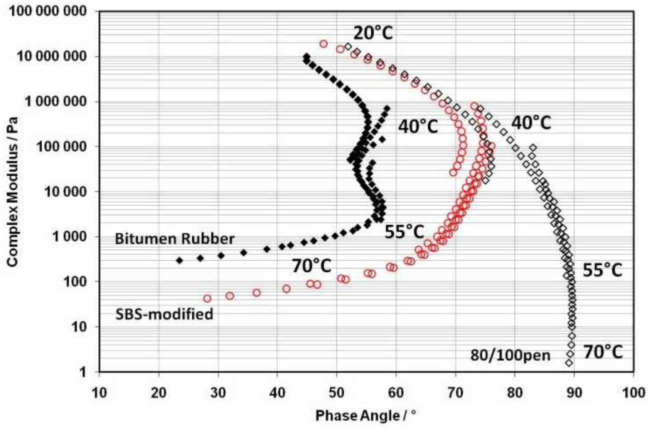
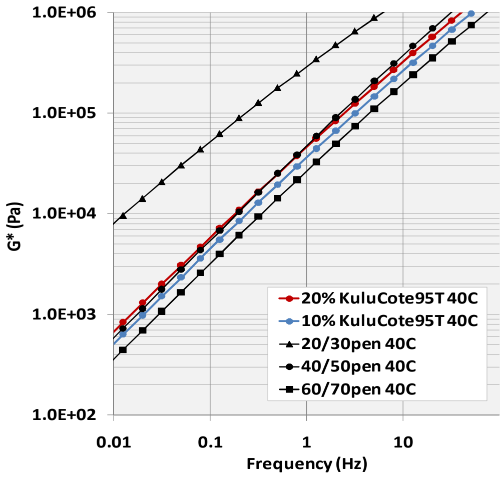


Figure 16: Black diagrams for CRM bitumen, SBS-modified and 80/100pen grade bitumen, data obtained from frequency sweeps.



Figures 17: Stiffness ranking of 10% and 20% filler-bitumen (KuluCote95T very fine Limestone) blends in comparison to other penetration grade bitumens at 40°C.

The key advantage of adding an elastomer as opposed to an inert volume filling powder is the dramatic effects on the phase angles at higher temperatures. As the temperature increases, the phase angle of the bitumen-rich phase approaches 90° signifying viscous flow or Newtonian behaviour, whereas the modifier-rich phase tends towards 0°, signifying a more elastic behaviour. Consequently, the effect of these modifiers on the unmodified bitumen is that of increasing the complex modulus whilst simultaneously decreasing the phase angle. This enhanced elastic response means that these binders would have higher resistance to deformation than pure binders. The CRM bitumen in turn displays higher G* at higher temperatures with much lower phase angles than the SBS-modified binder, it is thus expected to be more superior in this regard.

The CRM bitumen also provides enhanced resistance to low temperature cracking. At low temperatures, Figure 16 shows the addition of crumb rubber reduces both the G* and phase angle values of the bitumen. This results in a more flexible yet elastic binder.

CONCLUSION

The DSR rheological characterisation of CRM bitumen requires gap adjustment depending on the maximum size of the rubber crumbs. Since the type and size of rubber crumbs used in South African CRM bitumen blends are fairly consistent, their swollen particle size when blended with the standard 80/100 penetration grade bitumen could also be assumed as constant. DSR gap adjustment can therefore be done directly from the maximum size of rubber crumbs prior to blending. The black diagram of the CRM bitumen blend showed higher G* and reduced phase angles at the higher temperatures relative to the SBS-modified binder and base bitumen. This binder should therefore exhibit improved resistance to rutting and deformation. At the lower temperatures, its lower G* values at a higher elastic response indicates superior resistance to low temperature cracking than the other two binders.

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