Comparison of the Zero Shear Viscosity of Bitumen-Filler Mastics Using Different Measurement Techniques

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ABSTRACT: Zero shear viscosity (ZSV) or the viscosity at zero shear rate is an intrinsic property of a bituminous binder that has been proved to have a good correlation to the rutting potential of most binders, particularly modified binders. Various techniques and procedures exist to calculate or estimate ZSV. These include creep flow with or without creep recovery, viscosity measurements at low shear rates, sinusoidal oscillation at low frequencies and repeated creep (pulsed creep). Most of these techniques are performed on a dynamic shear rheometer with the ZSV being determined directly from the plotted data or, particularly in the case of the oscillation test, extrapolated to zero frequency using an appropriate mathematical model. This paper investigates the effect of modifying a bituminous binder through the addition of filler to produce bitumen-filler mastic similar to that found within an actual asphalt mixture. The ZSVs of bitumen-filler mastics with different filler types and bitumen-filler ratios have been tested. Three types of filler have been included in the testing programme including inert fillers (limestone and gritstone) as well as an active filler (cement) at various filler-bitumen ratios ranging from 15% to 65%. The ZSV results show varying degrees of modification (increases) of ZSV as a function not only of filler content but also of filler type. In addition, the ZSVs determined by means of the different procedures gave statistically different values, again as a function of both filler content and filler type.

KEY WORDS: Zero shear viscosity, dynamic shear rheometer, creep, bitumen, filler.

1 INTRODUCTION

Bitumen rheology is a major factor influencing the permanent deformation of an asphalt mixture. Rheological parameters such as the Superpave bitumen rutting parameter, $G^*/\sin\delta$, have been used as a performance indicator for rutting (Anderson et al. 2002). For conventional, penetration grade bitumens this parameter provides a good indication and correlation with the rutting behaviour of asphalt mixtures. However, this correlation is not particular good for specialist binders such as polymer modified bitumens (PMBs) (Phillips and Robertus 1996) due to the inability of the Superpave parameter to account for the effect of delayed elasticity.

The use the concept of zero shear viscosity (ZSV) has therefore been suggested as more appropriate rutting parameter (Phillips and Robertus 1996, Sybilski, 1996a). The ZSV of a binder is an intrinsic property that has been shown to correlate with the rutting performance of the binder. However within an asphalt mixture it is probably more appropriate to consider the performance of the bitumen-filler mastic (bitumen plus filler material passing 75 μm) rather than simply considering the pure bitumen. This paper looks at the suitability of different laboratory testing techniques at measuring ZSV of bitumen-filler mastics and the relative effect of filler type and concentration on the measured ZSV.

2 ZERO SHEAR VISCOSITY CONCEPT

Bitumen viscosity, as a rational, physical material property, has commonly been accepted as an indicator of asphalt mixture permanent deformation resistance (Sybilski, 1996b). However, as bitumen and bitumen-filler mastics tend to show non-Newtonian behaviour at 40°C and 60°C, the viscosity needs to be determined in the form of ZSV, which is independent of shear rate and testing condition. Within this linear regime, the ZSV reflects dissipated motions in a negligibly perturbed, equilibrium "no-flow" structure (Phillips and Robertus 1995). Various techniques can be used to measure ZSV, including small amplitude, low frequency oscillations, low stress creep tests and low strain rate viscometry measurements.

2.1 Oscillatory Testing

The ZSV of bitumen or bitumen-filler mastics can be determined by means of dynamic, oscillatory testing using a dynamic shear rheometer (DSR). Using this testing technique, measurements of complex viscosity approach a value equal to ZSV, as loading frequency tends to zero (Phillips and Robertus 1996, De Visscher and Vanelstraete 2003) or at least at very low oscillation frequencies (De Visscher et al. 2004) as calculated using Equation 1.

$$\eta_0 = \eta' = \frac{G''}{\omega} \qquad (1)$$

where η_0 is the ZSV (Pa.s), η' is dynamic viscosity (Pa.s), G'' is the loss (viscous) modulus (Pa) and ω is loading frequency (rad/s). Figure 1 shows the concept of dynamic viscosity approaching an upper limiting viscosity, ZSV, at low testing frequencies.

2.2 Viscometry Testing

Strain rate tests using the DSR can also be undertaken to determine the ZSV using viscometry testing and the power law relationship between stress and strain rate:

$$\sigma = \eta \dot{\gamma}^{n} \qquad (2)$$

where σ is the applied shear stress (Pa), η is the apparent viscosity (Pa.s), $\dot{\gamma}$ is the shear rate (1/s) and n is the power exponent. Using this testing technique the ZSV can be determined as strain rate tends to zero and the viscosity becomes Newtonian in nature. The concept of viscosity approaching Newtonian ZSV at low strain rates is shown in Figure 2.

2.3 Creep Testing

At low stress levels and with increasing time of loading, ZSV can be measured in a creep test as a limiting value (steady state viscosity) based on Equation 3 (Desmazes et al. 2000, De Visscher et al. 2004):

$$\eta_0 = \left(\frac{dJ(t)}{dt}\right)^{-1} \tag{3}$$

where $J(t)$ is the Creep Compliance (1/Pa) and t is the creep time (s).

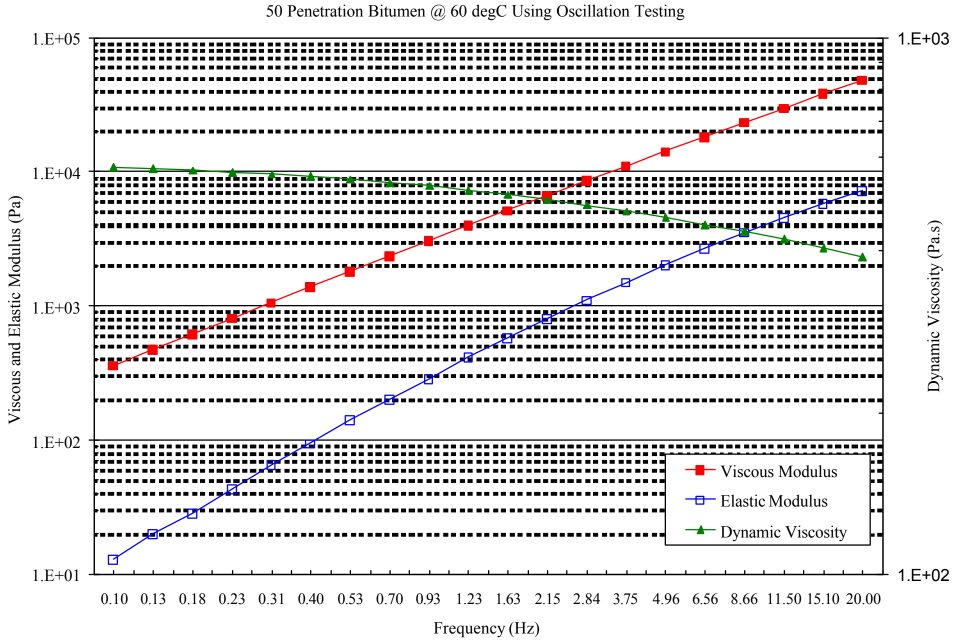

Figure 1: The Concept of Zero Shear Viscosity from Oscillation Testing

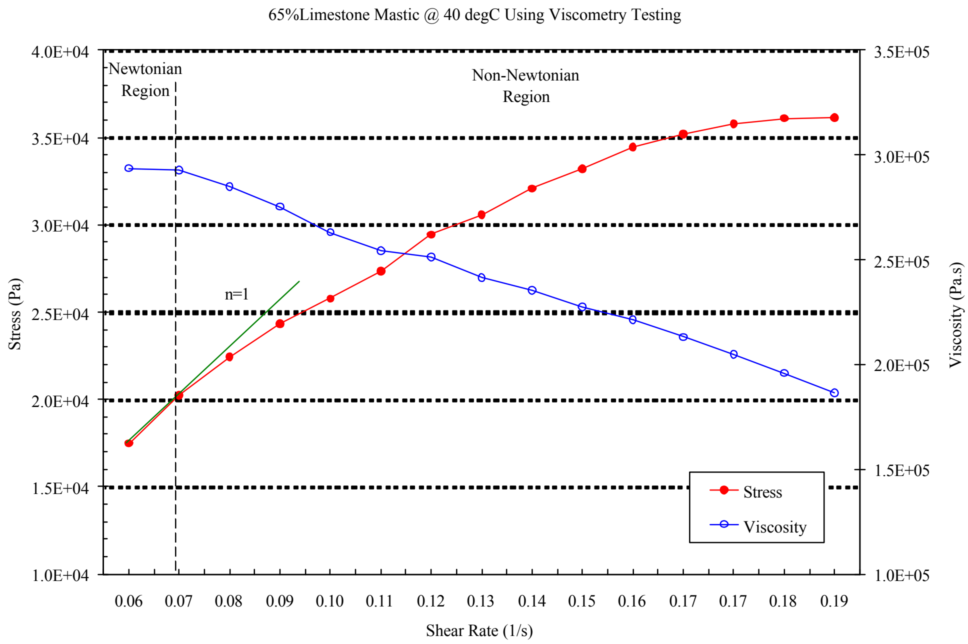

Figure 2: The Concept of Zero Shear Viscosity from Viscometry Testing

In a long time creep test, the effect of delayed elasticity decreases with time and after a sufficiently long time period, the rheological behaviour of the bitumen is dominated by viscous flow. Under low stress creep, structures within the bitumen or bitumen-filler mastic deform so slowly that they can continuously adapt thereby maintaining a situation close to equlibrium without building up any significant structural change in the material. An example of using the creep test to calculate ZSV is shown in Figure 3.

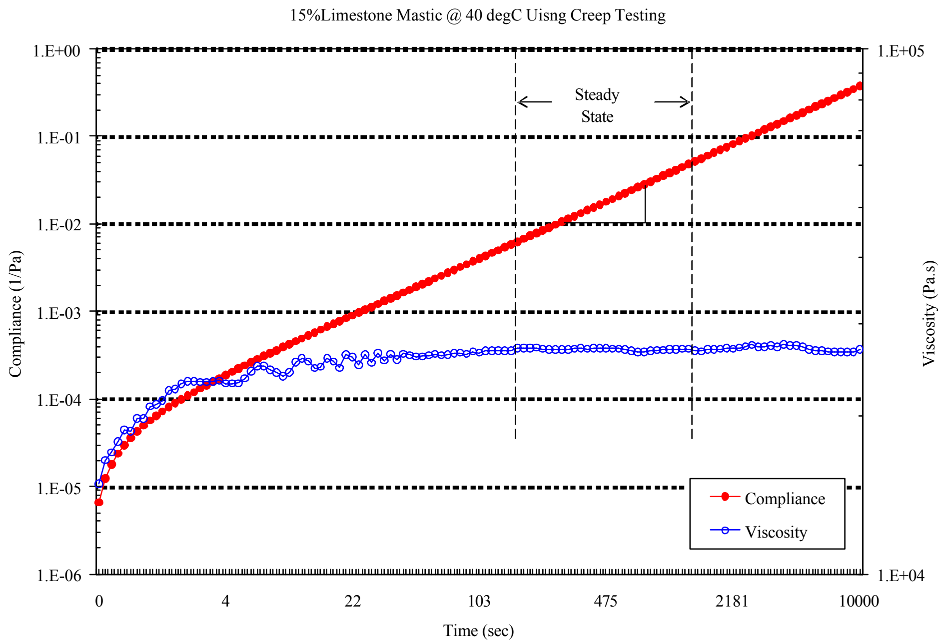

Figure 3: The Concept of Zero Shear Viscosity from Creep Testing

3 MATERIALS

A combination of one penetration grade bitumen, three filler types and three filler contents (by mass) were included in the testing programme. The matrix of materials that were tested is listed in Table 1. The base bitumen consisted of a 50 penetration grade bitumen with the fillers being limestone, cement and gritstone.

Table 1: Base Bitumen and Bitumen-Filler Mastics

Materials / Filler Contents	Base Bitumen	Bitumen-Filler Mastics		
	50 Pen Bitumen	Limestone	Cement	Gritstone
	x			
15%		x		
35%		x	x	x
65%		x	x	x

4 TESTING PROCEDURES

4.1 Oscillation Testing

The oscillatory tests were undertaken with a Bohlin controlled stress rheometer using the testing parameters given in Table 2.

Table 2: The Test Programme of Oscillation Testing

Temperature	Geometry	Gap	Frequency	Model
40°C	8 mm PP	2 mm	0.1-20 Hz	Cross Model
60°C	25 mm PP	1 mm	0.1-20 Hz	Cross Model

As measurements were not taken down to "zero" frequency, the 4-parameter Cross model was used to extrapolate measurements of complex viscosity to zero frequency (Cross, 1965).

$$\eta' = \eta_\infty + \frac{(\eta_0 - \eta_\infty)}{(1+(k\omega)^n)} \tag{4}$$

where η' is dynamic viscosity (Pa.s), η_0 is ZSV (Pa.s), η_∞ is infinite viscosity (Pa.s), ω is frequency (Hz) and k and n are material constants. The Cross model fitted to DSR experimental data can be seen in Figure 4.

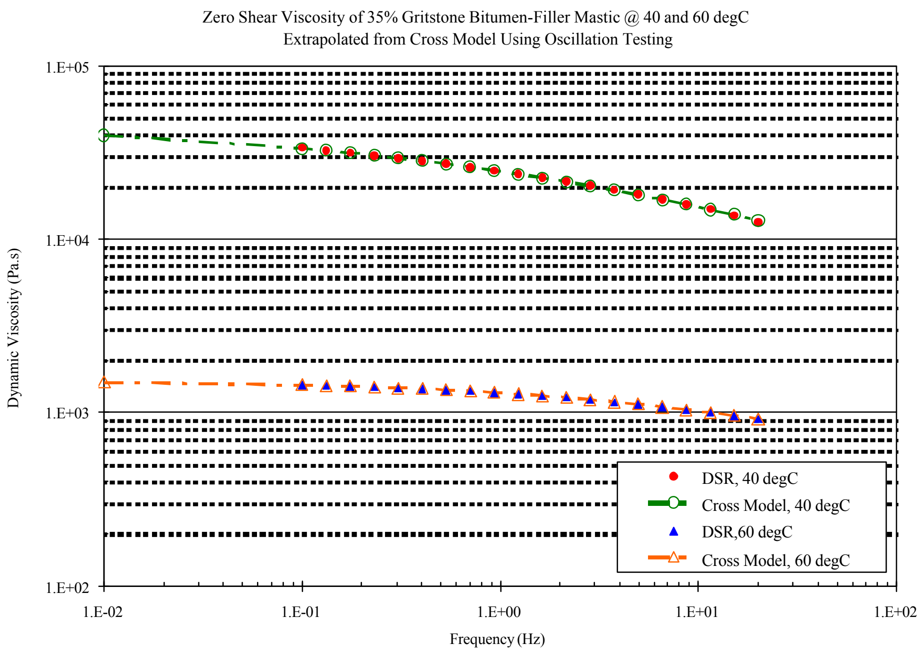

Figure 4: The Zero Shear Viscosity Extrapolated from Cross Model Using Oscillation Testing

4.2 Viscometry Testing

The viscometry testing was also undertaken with a Bohlin controlled stress rheometer using the viscometry mode and the testing variables detailed in Table 3.

Table 3: The Test Programme of Viscometry Testing

Temperature	Geometry	Gap	Shear Rate	Model
40	8 mm PP	2 mm	0.05-1 1/sec	Cross Model
60	25 mm PP	1 mm	0.05-1 1/sec	Cross Model

Identical to the oscillatory tests, a 4-parameter Cross Model was used to determine the ZSV from the low strain rate viscometry data:

$$\eta = \eta_\infty + \frac{(\eta_0 - \eta_\infty)}{(1 + (k\dot{\gamma})^n)} \tag{5}$$

where η is apparent viscosity (Pa.s), η_0 is zero shear viscosity (Pa.s), η_∞ is infinite viscosity (Pa.s), $\dot{\gamma}$ is the shear rate (1/s), and k and n are material constants. The Cross model fitted to low shear rate viscosity is shown in Figure 5.

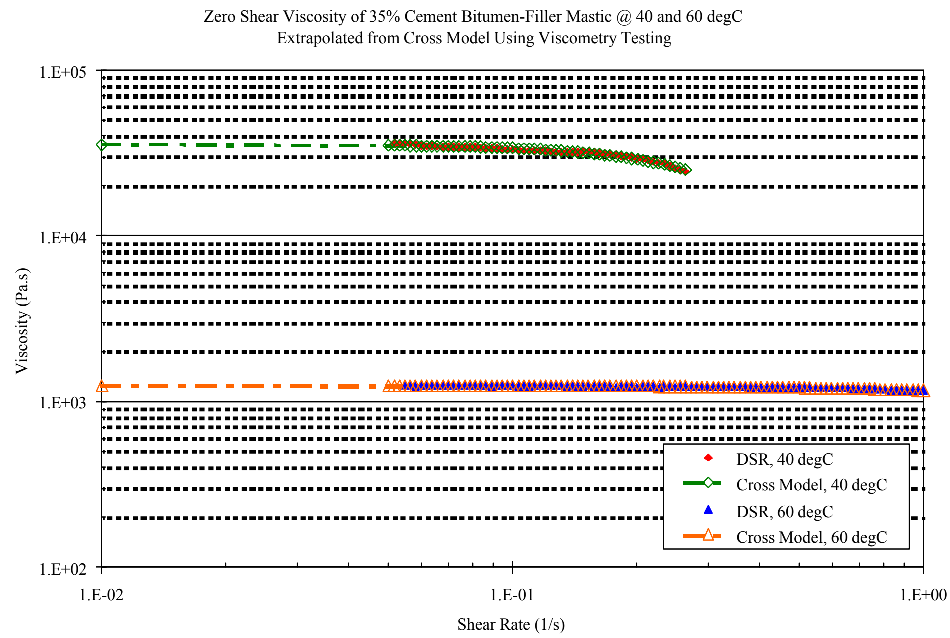

Figure 5: Zero Shear Viscosity Extrapolated from Cross Model Using Viscometry Testing

4.3 Creep Testing

The creep tests were carried out using a Bohlin constant stress dynamic shear rheometer with 8 mm and 25 mm parallel plates. The creep testing programme is detailed in Table 4.

Table 4: The Test Programme of Creep Testing

Temperature	Geometry	Gap	Stress	Creep Time	Model
40	8 mm PP	2 mm	300 Pa 1000 Pa	5000 sec 20000 sec	Multiple Creep
60	25 mm PP	1 mm	300 Pa	5000 sec	Multiple Creep

The ZSVs for the bitumen and bitumen-filler mastics were determined by fitting the experimental data to Equation 6.

$$J(t) = J_g + t/\eta_0 + \sum_{i=1}^{n} J_i(1 - e^{-t/t_i}) \tag{6}$$

where J_g is instantaneous compliance (1/Pa), η_0 is ZSV (Pa.s), t is the creep time (s), J_i is creep compliance (1/Pa) and $\sum_{i=1}^{n} J_i(1 - e^{-t/t_i})$ is the retardation spectrum. An example of the creep data used in the study is shown in Figure 6.

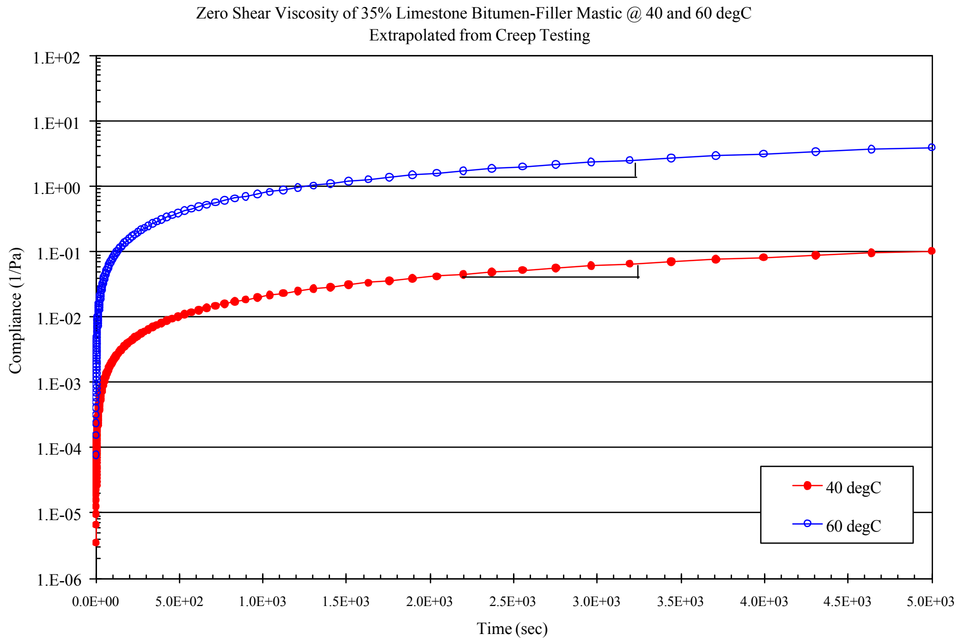

Figure 6: The Zero Shear Viscosity Extrapolated from Multiple Creep Model Using Creep Testing

5 RESULTS

Table 5 compares the ZSV results obtained for the base bitumen and bitumen-filler mastics as measured using the three techniques of oscillatory, viscometry and creep testing at both 40°C and 60°C. In addition to the values of ZSV, the relative increase in viscosity compared to the base bitumen is included as a ratio.

5.1 Influence of Filler Type and Concentration

The influence of filler type and concentration is shown in Figure 7, where the ZSVs for the 35% and 65% bitumen-filler mastics as determined using low frequency oscillatory testing are presented. The results show that at 35% filler by mass, the influence of filler type is marginal with all three mastics having a similar ZSV at both 40°C and 60°C. However, at 65% filler by mass the results show a significantly larger value of ZSV for the gritstone filler mastic compared to the limestone and cement fillers.

Table 5: Zero Shear Viscosities of Materials

Materials	ZSV @ 40°C (Pa.s)			ZSV @ 60°C (Pa.s)		
	Oscillation	Viscometry	Creep	Oscillation	Viscometry	Creep
50 pen	25,800	18,900	22,000	597	672	753
15% limestone	29,700 (1.15)	26,100 (1.38)	26,900 (1.22)	768 (1.29)	837 (1.25)	838 (1.11)
35% limestone	42,500 (1.65)	33,600 (1.78)	49,800 (2.26)	1,581 (2.65)	1,114 (1.66)	1,296 (1.72)
65% limestone	364,000 (14.11)	321,000 (16.98)	639,000 (29.05)	7,093 (11.88)	14,320 (21.31)	15,790 (20.97)
35% cement	39,900 (1.55)	35,500 (1.88)	65,400 (2.97)	1,363 (2.28)	1,251 (1.86)	1,281 (1.70)
65% cement	629,000 (24.38)	568,000 (30.05)	1,220,000 (55.55)	7,572 (12.68)	35,710 (53.14)	35,770 (45.50)
35% gritstone	46,300 (1.79)	47,700 (2.52)	80,400 (3.65)	1,518 (2.54)	1,542 (2.29)	1,386 (1.84)
65% gritstone	55,300,000 (2143.4)	41,200,000 (2179.9)	6,340,000 (288.2)	16,740 (28.04)	100,900 (150.15)	146,900 (195.09)

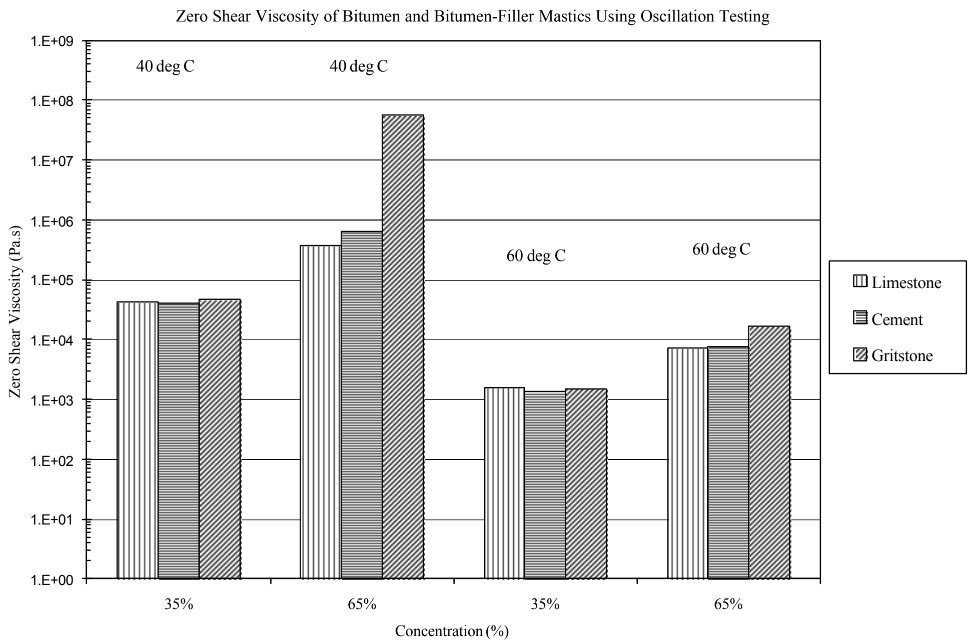

Figure 7: The Influence of Filler Type

In terms of the limestone filler mastics, the results for the 50 pen bitumen together with the three limestone filler mastics are presented in Figure 8. The results only show a marginal increase in ZSV for the 15% and 35% filler mastics but a considerable increase for the 65% filler content mastic. The sharp increase in ZSV is caused by the filler contributing such a large volume to the binder that it becomes the predominant component in the binder-filler mastic and therefore has a dramatic effect on viscosity. In addition this volume and stiffening effect is greater for the cement and gritstone mastics compared to the limestone as shown in Table 5.

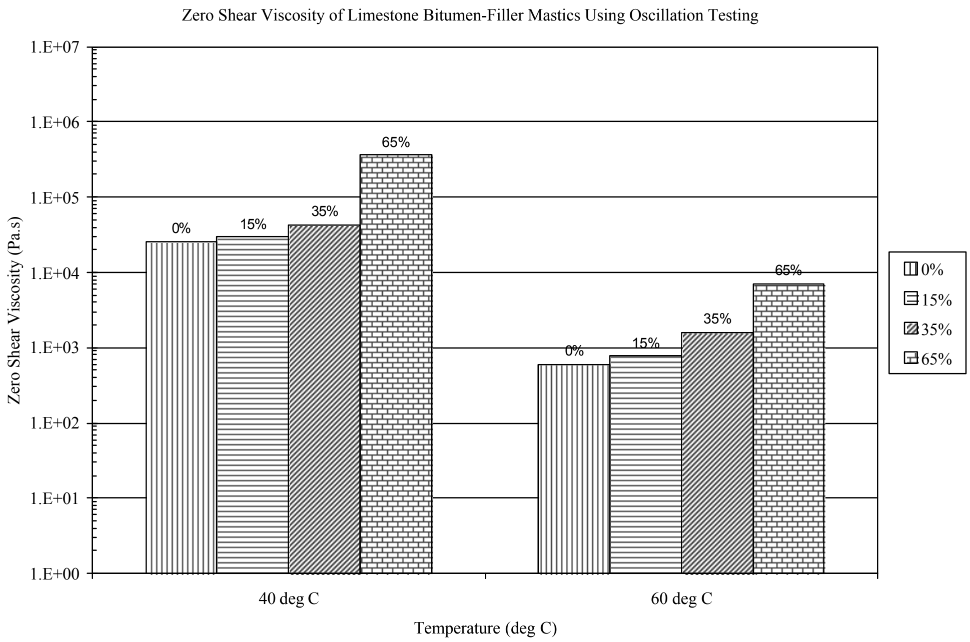

Figure 8: The Influence of Filler Concentration

5.2 Influence of Testing Methods

The influence of test method on the ZSVs determined for the 50 pen bitumen and various bitumen-filler mastics is shown in Figure 9. In general all three methods; oscillatory, viscometry and creep, produce basically the same results for the 50 pen bitumen and the low filler content (15%) mastic. However, there are significant differences among the methods for the intermediate and high filler content (35% and 65%) mastics, with the oscillatory and viscometry methods producing different values of ZSV than the creep method.

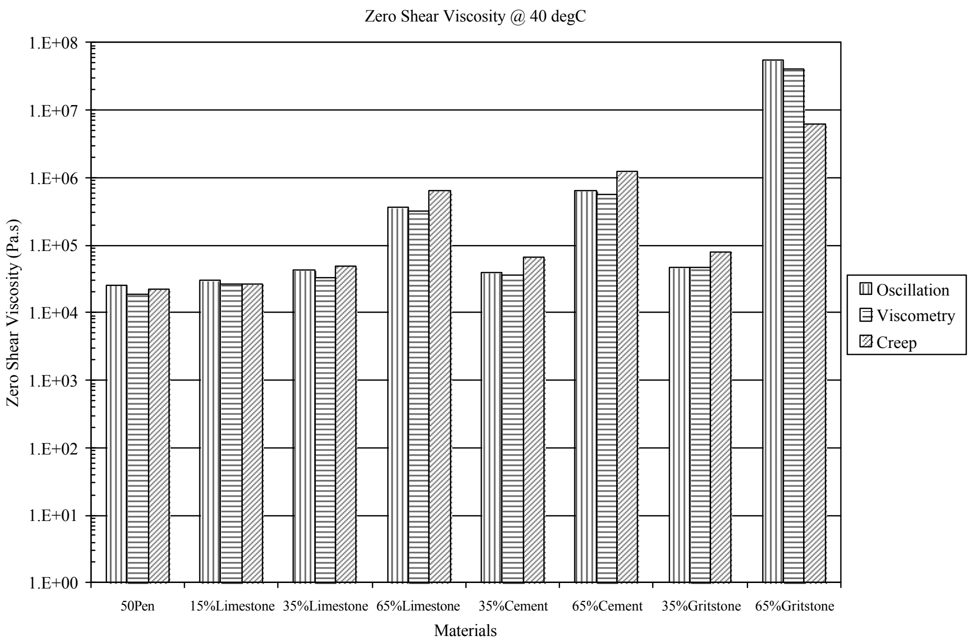

Figure 9: The Influence of Test Methods @ 40°C

6 DISCUSSION

The results presented in this paper have shown that ZSV tends to be independent of the measurement mode (oscillation, viscometry or creep) for conventional, penetration grade bitumens and low filler content mastics but significantly influenced by testing method for intermediate and high filler content mastics. In the creep test, the steady state may not be attained within the testing time for the 65% filler content mastics and therefore the ZSV, although approximated by determining the slope of the compliance curve after a long time period, will be less than the actual ZSV determined under steady state conditions (Binard et al. 2004). In theory the test method that produces the highest ZSV will generally be closer to the exact ZSV as approximations systematically underestimate ZSV.

However, for highly modified systems, such as bitumen-filler mastics with high filler contents, there is a danger that the gradient of dynamic viscosity may become too large at low frequencies (oscillation) and/or low strain rates (viscometry) leading to unrealistically high and unreliable values of ZSV from extrapolation or model fitting. Under these circumstances it may be better to determine the viscosity at very low frequencies (10^{-3} Hz) as an approximation of ZSV (De Visscher et al. 2004).

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