

## Characterization of Viscoelastic Properties of Bitumen-Filler Mastics

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**Abstract:** The linear viscoelastic (LVE) limits and rheological properties of bitumen-filler mastics were determined using a dynamic shear rheometer (DSR). Strain and frequency sweeps were performed on mastic specimens over a wide range of test temperature and frequency conditions. The use of an empirical algebraic equation of the Christensen and Anderson model (CA model) was used to characterize the LVE rheological behavior of the mastics in this study. Test results showed that the LVE limits for the concentrated suspension mastics were more restrictive than those for the dilute suspension mastics. Filler particles were likely to cause a hydrodynamic interaction in the concentrated suspension system. A significant difference in stiffening effect for the concentrated suspension mastics was observed due to the filler effective volume as well as the inter-particle interaction. For the dilute suspension mastics, the difference in filler stiffening effect was minimal irrespective of filler type in this study.

*Keywords:* Bitumen, Filler, Mastic, Linear viscoelastic, Master curve

### 1. INTRODUCTION

The role of mineral fillers has been known to considerably influence the mechanical properties of asphalt paving mixtures. The addition of fillers are generally shown to affect moisture damage, stiffness, oxidation, rutting, cracking behavior, workability and compaction characteristics in asphalt pavements (Anderson, 1987). The mastic (combination of mineral filler and bitumen) effectively coats all aggregates in asphalt concrete.

Researchers have reported that the physical properties and chemical constituents of mineral fillers affect the rheological and mechanical properties of bitumen-filler mastics (Heukelom, 1965; Huschek and Angst, 1980; Ishai and Craus, 1977; Tunnicliff, 1967). Kavussi and Hicks (1997) evaluated the effects of filler type, grading and shape on the physical and rheological properties of mastics. They found that at equal filler volume concentration, mastics containing kaolin and fly ash had higher viscosities compared to mastics prepared with quartz and limestone due to their fineness characteristics. The authors concluded that the higher the Rigden voids, the more stiffening the effect of the filler. The Rigden voids are defined as voids of dry compacted filler by means of Rigden apparatus (Rigden, 1947). Chen and Peng (1998) identified a considerable effect of filler particle size on tensile strength of mastics containing inert silica fillers. The results showed that smaller particles with higher surface areas carried more tensile loads than bigger ones. However, for the larger particles, the distance between particles was smaller than the stress concentration

area, thereby leading to an overlap area between particles. The strength of the particulate-filled composite was deteriorated. Soenen and Teugels (1999) examined bitumen-filler interaction by means of dynamic shear rheology. Mastics were prepared using four penetration grade bitumens and three types of mineral fillers having similar physical properties of bulk density and specific surface. The results showed little effect of a possible chemical interaction between bitumen and filler on the linear viscoelastic behavior of the mastics. The relative stiffening effect was only dependent on the filler volume concentration, but independent on the type of filler as well as on the origin of the bitumen. Little and Petersen (2005) investigated the effect of filler particles and bitumen interaction on the rheological properties of mastics in terms of physical and chemical interactions. Unaged and aged bitumens with two types of fillers (limestone filler and hydrated lime filler) at one filler content of 20% by volume were tested. The two types of mineral fillers had similar particle size distributions. The rheological results showed that the hydrated lime filler had considerably more impact on the loss tangent ( $\tan \delta$ ) than the limestone filler, but the impact was bitumen dependent. Mastic with lower loss tangent value may improve rutting resistance of asphalt pavements (Lesueur and Little, 2003; Petersen, 1984).

In addition, bitumen-filler mastic is regarded as a suspension system where mineral filler particles are suspended in a bitumen. For a dilute suspension in a mastic, when the filler content is low, the filler inter-particle distance is larger compared to the mean particle size. There exists a complete absence of any hydrodynamic interaction between the filler particles. The filler particles practically disperse in the bituminous matrix flow. For a concentrated suspension, when the content of suspended filler particles is increased, the inter-particle distance gets closer. Each particle begins to be dependent on the presence of other neighboring particles. These particles cause a hydrodynamic interaction in this suspension system, thereby affecting the mastic rheological properties (Shenoy, 1999; Vinogradov and Malkin, 1980). Researchers (Faheem *et al.*, 2008; Faheem and Bahia, 2009) investigated filler inter-particles and bitumen-filler interaction to interpret the stiffening effect using a tackiness technique. The tackiness was a test to obtain cohesion of bitumen. The normal tension force using a DSR was measured as the mastic sample deformed. The influenced of bitumen volume and filler volume were quantified by means of the conceptual model to illustrate the effect of filler on the mastic stiffness. The filler concentration could be divided into two regions of dilute suspension and concentrated suspension. Other researchers have utilized rheology-based approaches and micromechanical composite models to quantify the stiffening potential (Kim and Little, 2004; Shashidhar and Romeo, 1998).

Conventional tests such as penetration, softening point and absolute viscosity for bituminous binders have been used to measure their physical properties and link these to the performance of asphalt pavements. In reality, these conventional tests and specifications are only used to determine the characteristics of bituminous binders at particular temperature and loading rate conditions. Bitumen-filler mastics might not be properly characterized by the traditional parameters obtained from these empirical tests. Fundamental rheological testing using a dynamic shear rheometer (DSR) enables the linear viscoelastic behavior of mastics to be determined over a range of temperature and time conditions associated with asphalt mixtures. Considerable research in the linear viscoelastic analysis of bituminous binders using a rheometer has been studied (Airey *et al.*, 2004; Delaporte *et al.*, 2007; Delaporte *et al.*, 2008; Kim and Little, 2004; Yusoff *et al.*, 2011). However, bitumen-filler mastic has still not been comprehensively characterized due to the complex nature and interaction of the filler particle in the bitumen-filler system. Concern has been raised with interpreting the effect of fillers on dilute suspension and concentrated suspension mastics according to the linear rheological behavior. In this study, fundamental rheological testing by means of strain and frequency

sweeps was conducted on the bitumen-filler mastics. In addition to the DSR testing, a mathematical model was applied to the test results. The statistical techniques of curve fitting and nonlinear regression were also employed for rheological data interpretation. Thus, the objectives of this study are (1) to determine the linear viscoelastic (LVE) limits of the bitumen-filler mastics, (2) to evaluate the viscoelastic properties of the mastics, and (3) to characterize the rheological properties of the bitumen-filler mastics using the CA model.

## 2. THEORETICAL BACKGROUND

In order to characterize the rheological mechanism of mastics, the following theoretical concepts need to be addressed.

### 2.1 Dynamic Mechanical Analysis

Dynamic mechanical analysis (DMA) is used to help understand the viscoelastic properties of mastics by means of oscillatory-type testing. This testing is to measure the viscous and the elastic nature of materials over a range of temperatures and loading rates. Although the dynamic test can be conducted under either stress-controlled or strain-controlled conditions, strain-controlled testing is more common for bituminous binders and mastics. During the dynamic strain-controlled testing, the shear stress and shear strain vary sinusoidally with time. The resulting shear stress response is measured as a function of frequency ( $\omega$ ). The principle responses of interest are complex modulus,  $G^*$ , and phase angle,  $\delta$ . Since the  $G^*$  depends on the angular frequency, the magnitude of  $G^*$  is calculated from the peak-to-peak stress ( $\tau$ ) and strain ( $\gamma$ ) values.

The phase angle ( $\delta$ ) is the lag in the resulting shear stress compared to the applied shear strain in oscillatory testing, and varies between  $0^\circ$  and  $90^\circ$ . When the resulting shear stress is in-phase ( $\delta=0^\circ$ ) with the applied shear strain, the binder behaves as a purely elastic solid. If the resulting shear stress is out-of-phase ( $\delta=90^\circ$ ), the binder can be considered to be purely viscous in nature. Between the two extreme conditions, the binder behavior is to be viscoelastic in nature with a combination of elastic and viscous responses.

The other two viscoelastic parameters represented as in- phase and out-of-phase are the storage modulus (elastic component),  $G'$ , and the loss modulus (viscous component),  $G''$ , respectively. These two components are directly related to the  $G^*$  and to each other through the  $\delta$ . In addition, the loss tangent,  $\tan \delta$ , is calculated as the ratio of the loss modulus to the storage modulus. The viscoelastic parameters related to the  $G^*$  and  $\delta$  can be computed through a series of relatively simple equations:

$$G' = G^*(\omega)\cos\delta \quad (1)$$

$$G'' = G^*(\omega)\sin\delta \quad (2)$$

$$\tan\delta = \frac{G''(\omega)}{G'(\omega)} \quad (3)$$

$$G^*(\omega) = \sqrt{[G'(\omega)]^2 + [G''(\omega)]^2} \quad (4)$$

### 2.2 Linear Viscoelastic Limits

It is usual to confine the bitumen's characteristics to its linear viscoelastic response in order to simplify the mathematical modeling of the material, as nonlinear response is difficult to characterize and model in practical problems. As viscoelastic parameters are all ensured under linear viscoelastic (LVE) conditions, the relationship between stress and strain is influenced only by temperature and loading time. Based on the linearity of the rheological data, the master curves of bitumen-filler mastics can therefore be generated. To determine the LVE limits, strain or stress sweep testing is usually conducted at selected temperatures and frequencies. The LVE strain and stress limits were determined as the point beyond which the measured shear complex modulus reduces to 95% of its initial value. In the Strategic Highway Research Program (SHRP), the LVE shear stress and strain limits for forty aged and unaged penetration grade bitumens were found to be functions of  $G^*$  as defined by the following equations (Petersen et al., 1994):

$$\gamma = 12.0/(G^*)^{0.29} \quad (5)$$

$$\tau = 0.12(G^*)^{0.71} \quad (6)$$

where,  $\gamma$  is the shear strain (%),  $\tau$  is the shear stress (kPa), and  $G^*$  is the complex modulus (kPa).

### 2.3 Modeling of Linear Viscoelastic Properties

The use of an empirical algebraic equation of the Christensen and Anderson model (CA model) is used to describe the LVE rheological behavior of the mastics in this study. The model is able to predict bitumens' LVE properties and followed the time-temperature superposition principle (TTSP). In the CA model, there are three primary rheological parameters of glassy modulus ( $G_g$ ), crossover frequency ( $\omega_0$ ) and rheological index (R), as shown in Figure 1. For the  $G^*$  viscoelastic function, the following mathematical equation is shown as follows (Christensen and Anderson, 1992):

$$G^*(\omega) = G_g \left[ 1 + \left( \frac{\omega_0}{\omega} \right)^{\log 2 / R} \right]^{-R / \log 2} \quad (7)$$

where,  $G_g$  is the glassy modulus,  $\omega_0$  is the crossover frequency (rad/s), and R is the rheological index.

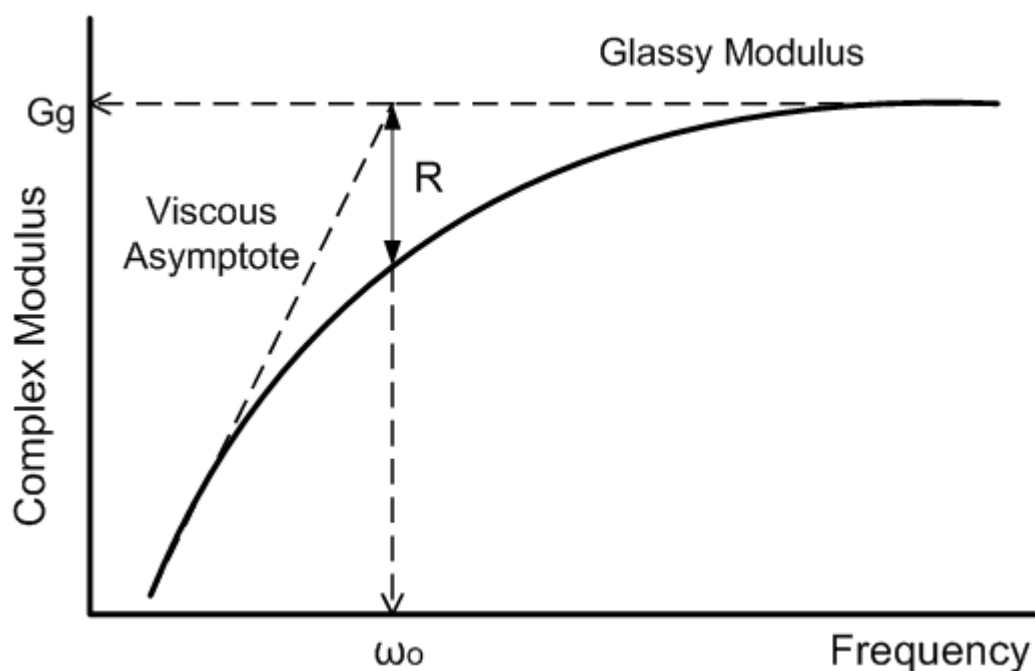


Figure 1. Illustration of three parameters in the CA model

The glassy modulus is the value that the complex modulus or storage modulus approaches at low temperatures and high frequencies. The value is typically close to 1 GPa in shear loading for most bituminous binders. The crossover frequency is defined as the frequency where the storage and loss moduli are equal. The rheological index is distance between the glassy modulus and the complex shear modulus at the crossover frequency.

### 3. MATERIALS AND TEST PROGRAM

#### 3.1 Materials

The following one straight-run bitumen, three types of mineral fillers and three filler concentrations were incorporated in this linear viscoelastic analysis program:

- Base bitumen: 40/60 penetration grade bitumen,
- Type of mineral filler: limestone, cement and gritstone, and
- Filler concentration (by mass): 15%, 35% and 65%.

The three fillers used in this study included limestone, cement and gritstone. The cement is considered to be more active with bitumen, while the limestone and gritstone are relatively inert to bituminous binder. Figure 2 shows the mineral filler gradations which were measured by performing a laser diffraction technique. The limestone filler undoubtedly has the finest particles. Although the particle size distribution of the gritstone filler is similar to that of the cement filler, the gritstone filler particles are slightly finer below the particle size fraction of 10  $\mu\text{m}$ .

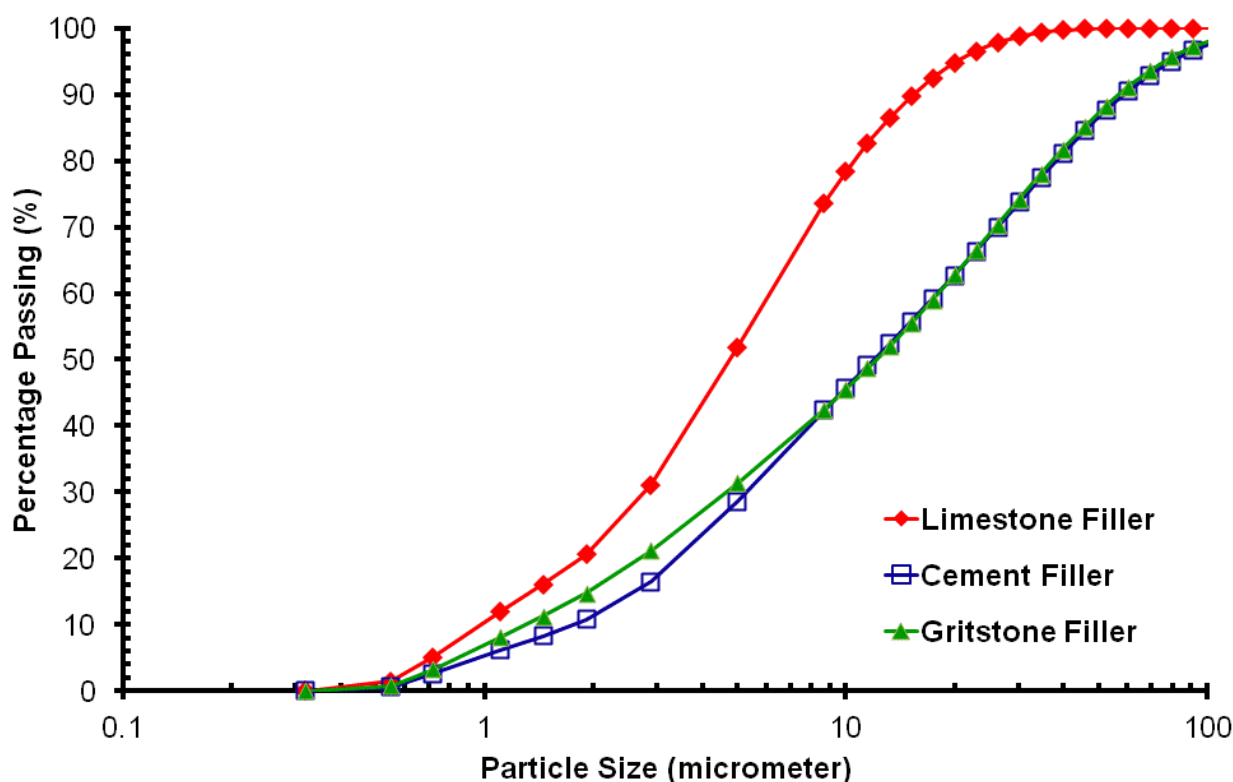


Figure 2. Particle size distribution of three mineral fillers

A total of seven bitumen-filler mastics with the base bitumen were produced using a Silverson high shear mixer at 160°C. The silicon mould method was used for the DSR sample preparation in order to conquer the filler settlement in the mastics. To simply compare with the filler proportion in practical asphalt mix design, the filler concentration levels were therefore decided in terms of filler weight in this test program. It is noted that although the weight ratios of filler to base bitumen were maintained at 0.18(15/85), 0.54(35/65) and 1.86(65/35), the percentages of filler volumetric fraction in the mastics should be taken into account. The filler physical properties, compositional volume ( $V_f$ ) and effective volumes ( $V_e$ ) in the mastics are shown in Table 1. The filler compositional volume is given by:

$$V_f = \frac{\frac{M_f}{S_f}}{\frac{M_f}{S_f} + \frac{M_b}{S_b}} V_f = \frac{\frac{M_f}{S_f}}{\frac{M_f}{S_f} + \frac{M_b}{S_b}} V_f = \frac{M_f / S_f}{M_f / S_f + M_b / S_b} \quad (8)$$

where  $M_f$  is the mass of filler;  $S_f$  is the specific gravity of filler;  $M_b$  is the mass of bitumen and  $S_b$  is the specific gravity of bitumen.

As bitumen was added to the filler, the bitumen first filled the voids. Any bitumen within these voids was called fixed bitumen since it was fixed within the minimum void structure. Bitumen in excess of the fixed bitumen was called free bitumen because it was free of the voids and was free to lubricate the filler particles (Anderson, 1973). The filler effective volume consists of the solid filler particles together with the presence of an adsorption layer around the particles. Different types of mineral fillers show dissimilar degrees of filler

dispersion in bitumen-filler systems due to the difference in maximum packing fraction in bitumen. The formula of the effective volume is given by Heukelom (1965):

$$V_{fe} = (100/V_{FR})V_f V_e = \left[ \frac{100}{(1-\varepsilon)} \right] V_f \tag{9}$$

where  $\varepsilon$  is the Rigden voids of mineral filler.

In addition to the filler volume fractions, the properties of specific gravity as well as the specific surface are also shown in Table 1.

Table 1. Mineral filler concentration levels of bitumen-filler mastic

Filler type	Specific gravity	Specific surface (m <sup>2</sup> /g)	Rigden void (%)	Mass content (%)	V <sub>f</sub> (%)	V <sub>e</sub> (%)
Limestone	2.74	1.3	24.9	15	6.2	8.3
				35	16.7	22.2
				65	40.9	54.4
Cement	3.18	1.4	28.4	35	14.7	20.6
				65	37.3	52.1
Gritstone	2.65	6.0	32.9	35	17.2	25.6
				65	41.7	62.1

### 3.2 Strain Sweep Testing

Strain sweep test was carried out using a DSR at selected temperatures of 30°C, 45°C and 60°C, and frequencies of 1.6, 10 and 25 Hz. DSR parallel plates (PP) of 8 mm diameter with 2 mm gap were used for low temperature (30°C) testing, while 25 mm diameter PP with 1 mm gap were used for high temperatures (45°C and 60°C) testing. The strain sweeps at each temperature/frequency combination were replicated on different specimens. The strain and stress LVE limits were determined as the point where G\* reduced to 95% of its initial value.

### 3.3 Frequency Sweep Testing

Once the LVE limits for the base bitumen and mastics were established, rheological parameters were generated by performing frequency sweeps under relatively small strain levels to ensure responses within a linear viscoelastic range. The frequency sweeps were carried out at seventeen temperatures between 5 and 85°C, and twenty frequencies between 0.1 and 20 Hz. The viscoelastic characteristics of the bitumen and mastics based on time-temperature superposition principle (TTSP) were interpreted in the form of shifted master curves at a reference temperature of 25°C.

## 4. RESULTS AND DISCUSSION

### 4.1 Linearity of Bitumen-Filler Mastics

Figure 3 shows the LVE strain limit as a function of complex modulus,  $G^*$ . The SHRP LVE strain criteria are also included to compare with the rheological properties of the base bitumen and bitumen-filler mastics in this study. There is a general decrease in the LVE strain limit with an increase in  $G^*$ . The SHRP LVE strain criteria are relatively conservative compared to the linearity limits of the 40/60 pen bitumen, 15% and 35% mastics. The 15% and 35% bitumen-filler mastics have very similar linearity trends compared to the base bitumen, and thus they are regarded as dilute suspension mastics. However, for the 65% bitumen-filler mastics, the LVE strain limit tends to be below the SHRP LVE criteria, reflecting considerably narrower linearity limit. This behavior is indicative of the presence of a filler-filler interaction which becomes increasingly more dominant on the linearity behavior for the 65% mastics. The 65% mastics are therefore regarded as concentrated suspension mastics and have more conservative linearity limits. The LVE strain limit as a function of  $G^*$  provides a clear indication of the difference in the linear performance between the two filler suspension systems. The LVE strain limits for the dilute suspension mastics are approximately 10 times greater than that of the concentrated suspension mastics.

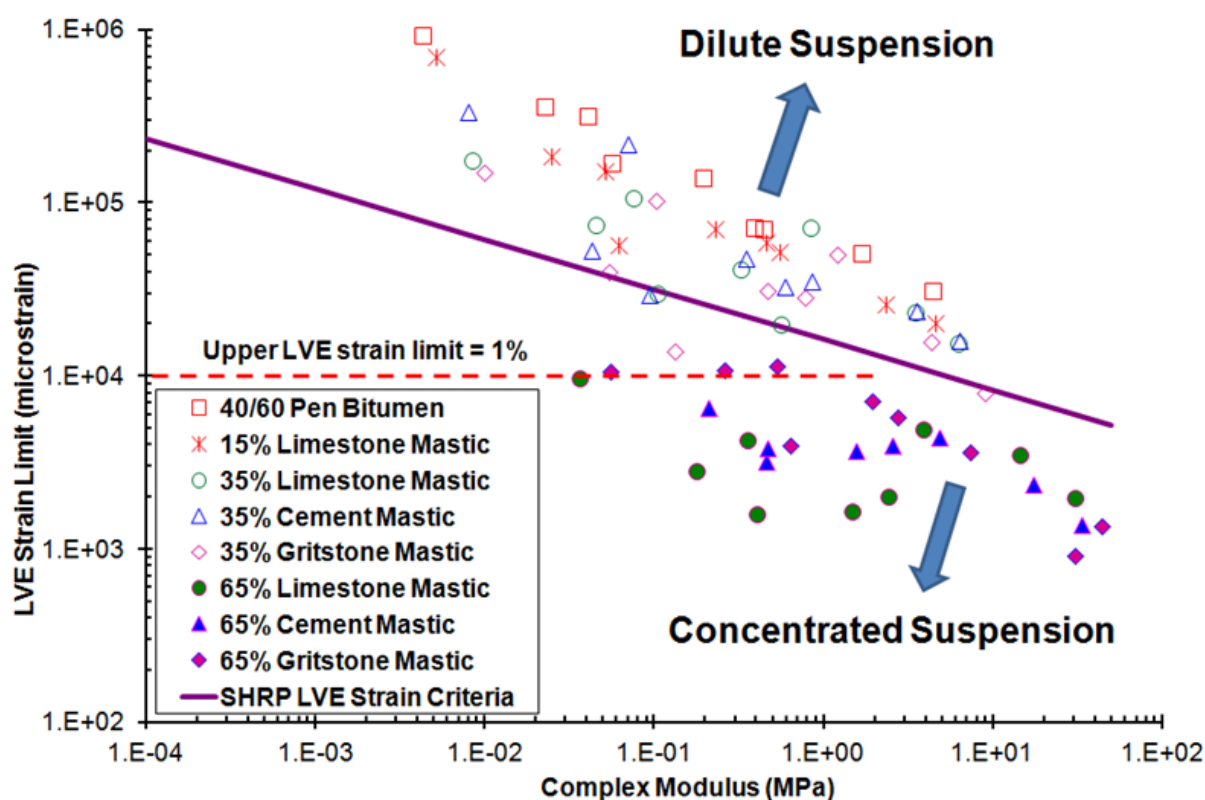


Figure 3. LVE strain limits for the dilute and concentrated suspension mastics

In addition, the results show the LVE strain limit decreases with an increase in  $G^*$  for the dilute suspension mastics. However, it seems that there is no strain dependent LVE criteria for the concentrated suspension mastics at lower  $G^*$  values (higher temperatures). The results display a upper LVE limit at 10,000 microstrain (1%) at  $G^*$  values between 0.04 and 1 MPa for the concentrated suspension mastics. It infers that the difference in the modulus between bitumen and filler particle becomes larger at high temperatures. The presence of the dominant filler particulate matrix may lead to more conservative strain limits.



Figure 4 shows the LVE stress limit as a function of complex modulus,  $G^*$ . The base bitumen and all mastics show an increase in the LVE stress limit with increasing  $G^*$ . Two distinct regions are found with the concentrated suspension mastics appearing to have more conservative stress limits compared to the dilute suspension mastics in terms of the linearity limit results. It is noted that the concentrated suspension mastics do not show a plateau of the stress limit at low  $G^*$  values, whereas an upper strain limit of 1% is displayed as seen in Figure 3. It infers that the presence of the dominant filler particles has little effect on the LVE stress limit at high temperatures.

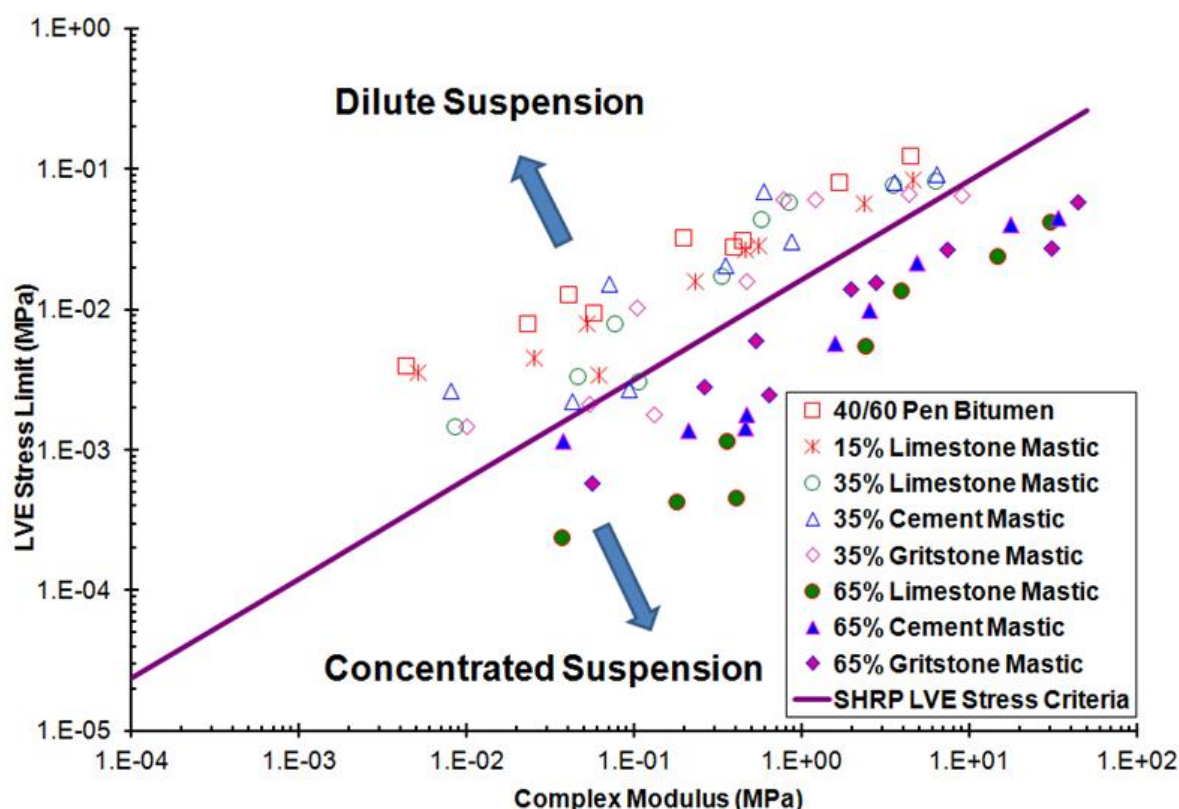


Figure 4. LVE stress limits for the dilute and concentrated suspension mastics

The indication implies that the LVE strain and stress limits for the concentrated suspension mastics are lower than those for the dilute suspension mastics due to the increased elastic response. The reduction in the linear limit appears to result primarily from the filler particle interaction. In addition, the linearity results provide no indication that the active filler (cement) mastic has lower LVE strain and stress limits than the inert fillers (limestone and gritstone) mastics.. The LVE limits therefore seem to be insensitive to the filler type.

#### 4.2 Master Curves for Bitumen-Filler Mastics

Based on the time- temperature superposition principle (TTSP), the master curves of  $G^*$  at a reference temperature of 25°C for the base bitumen and the limestone bitumen-filler mastics have been shown in Fig. 5. The smooth and continuous  $G^*$  master curves are produced because the base bitumen and bitumen-filler mastics exhibit simple rheological behavior. The limestone mineral filler shifts the master curves of the mastics vertically upwards. This is due to the fact that the filler particles provide a filling effect with an increase in the filler concentration level. Since the general shape for the base bitumen and mastics is similar, the

viscoelastic trend of the mastics is inherited from the bitumen. However, a considerable increase in stiffening effect at high filler content indicates that the presence of a filler-filler interaction becomes increasingly more dominance on the rheological behavior for the 65% mastics. The 15% and 35% mastics are regarded as dilute suspension mastics, whereas the 65% mastics are regarded as concentrated suspension mastics.

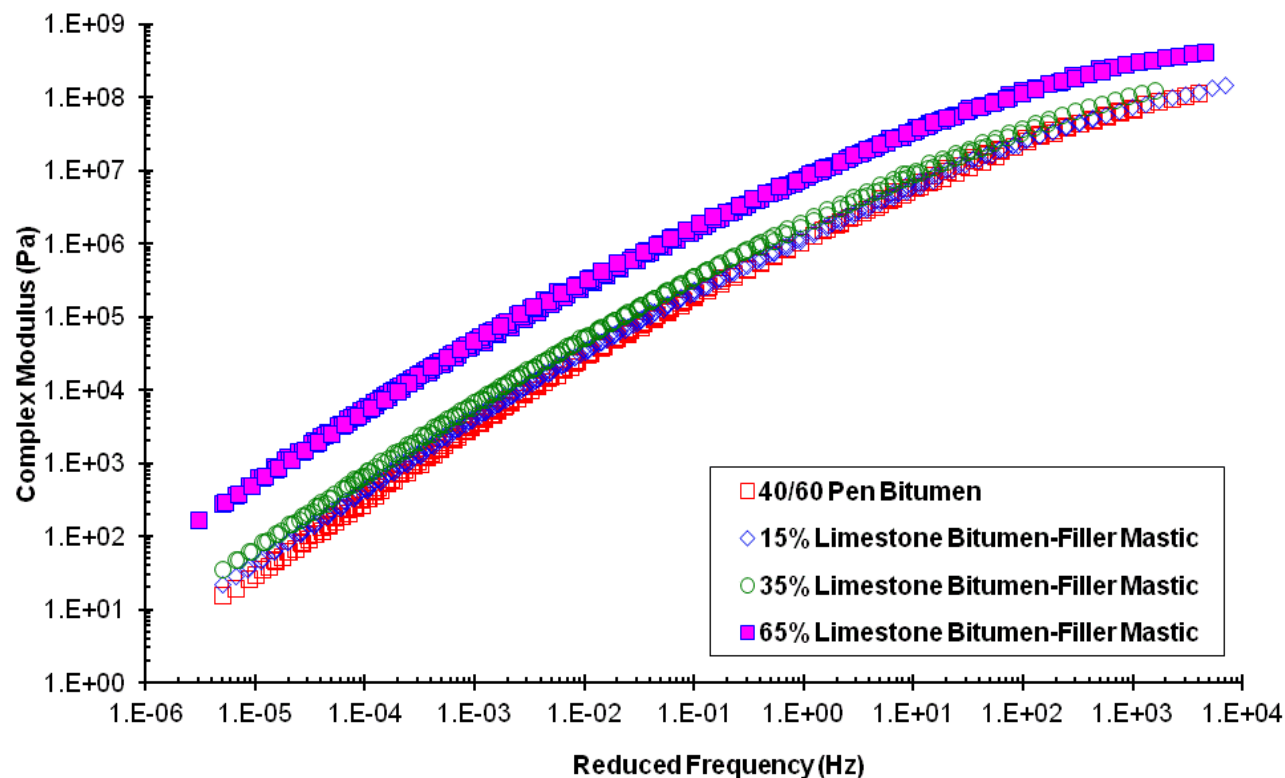


Figure 5.  $G^*$  master curves for base bitumen and limestone bitumen-filler mastics

Figure 5 also shows an increase in  $G^*$  as a function of frequency with an indication of a maximum  $G^*$  of approximately 109 Pa. Similar results are observed from the sets of master curves generated for other systems including cement and gritstone filler mastics. It is noted that the difference in stiffening effect is observed between low and high frequencies (high and low temperatures) for each mastic. According to the rheological data in this study, the range of stiffening ratio is between 1 and 18 at lower frequency (high temperature), while the stiffening ratios are between 1 and 3.6 at higher frequency (low frequency). The stiffening effect of filler on bitumen at higher temperatures is more pronounced than that at lower temperatures. This is due to the difference of the modulus between bitumens and fillers becoming larger at higher temperatures. Bitumen increasingly shows viscous flow while mineral filler still remains rigid.

Figure 6 shows the set of  $G^*$  master curves for the mastics including the base bitumen filled with limestone, cement and gritstone fillers. For the dilute suspension mastics, the difference in filler stiffening effect is minimal irrespective of filler type. This is probably due to the fact that there is an absence of any hydrodynamic interaction between filler particles. When the filler content is low, the filler inter-particle distance is larger compared to the mean particle size. The filler particles practically disperse in the bituminous matrix flow. The rheological behavior of the dilute suspension mastics is dominated by the base bitumen. However, there is a substantial effect of filler type on  $G^*$  for the concentrated suspension mastics. This is probably due to the difference in the filler effective volume as well as the

inter-particle interaction. The gritstone filler has the largest specific surface and Rigdon voids, leading to the largest effective volume in the bitumen-filler system. As the filler inter-particle distance gets closer, each particle is dependent on the presence of other neighboring particles. Therefore, the concentrated suspension mastic with gritstone filler has the highest  $G^*$  value, followed by the cement and limestone mastics.

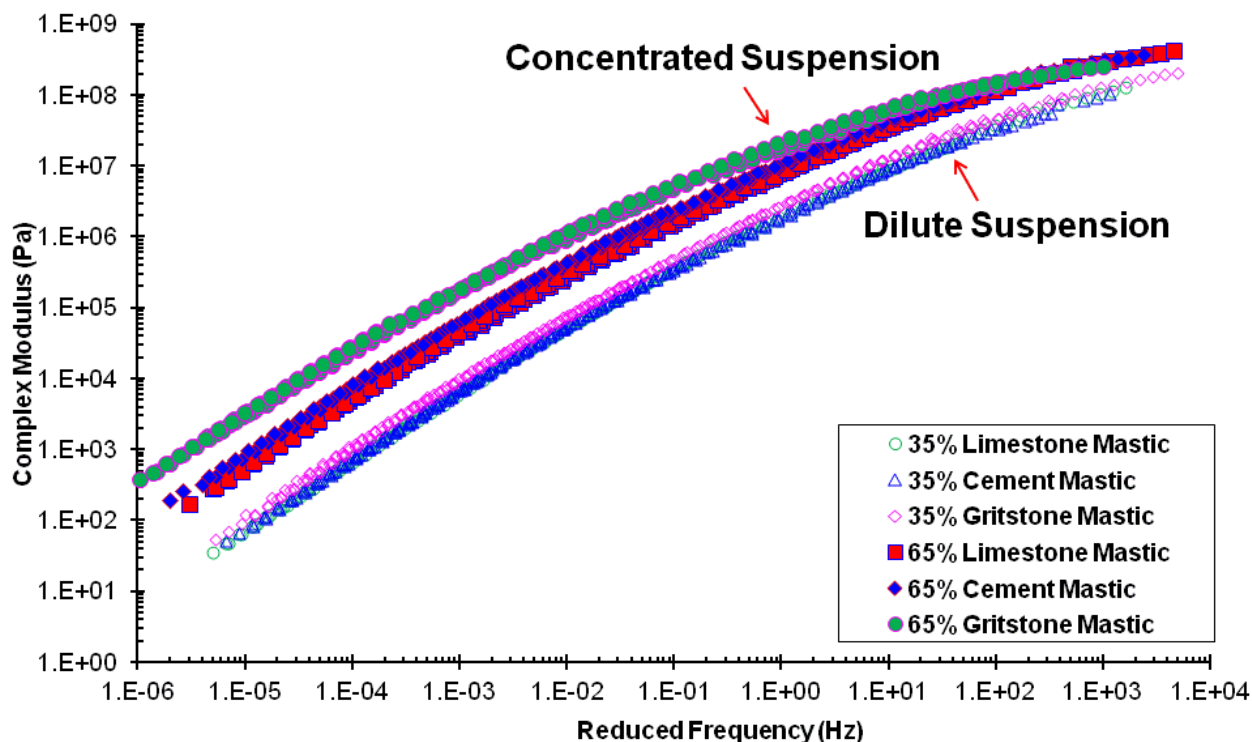


Figure 6.  $G^*$  master curves for the dilute and concentrated suspension mastics

As permanent deformation in an asphalt pavement is controlled by the binder's viscous flow and loading time, the complex modulus at low shear and high temperature is an important indicator. Figure 7 shows the effect of filler content and type on  $G^*$  stiffening ratio at a frequency of 0.1 Hz and a high temperature of 85°C for the base bitumen and mastics. There is an indication that an increase in the  $G^*$  stiffening ratio is linear until a certain filler effective volume concentration, where the rate of increase becomes nonlinear and reaches an asymptotic linear trend. The trend shows the effect of filler inter-particle interaction on the viscous properties of the bitumen-filler mastics. The  $G^*$  master curves also reflect the influence of the filler stiffening effect at low frequencies and high temperatures as shown in Figure 6.

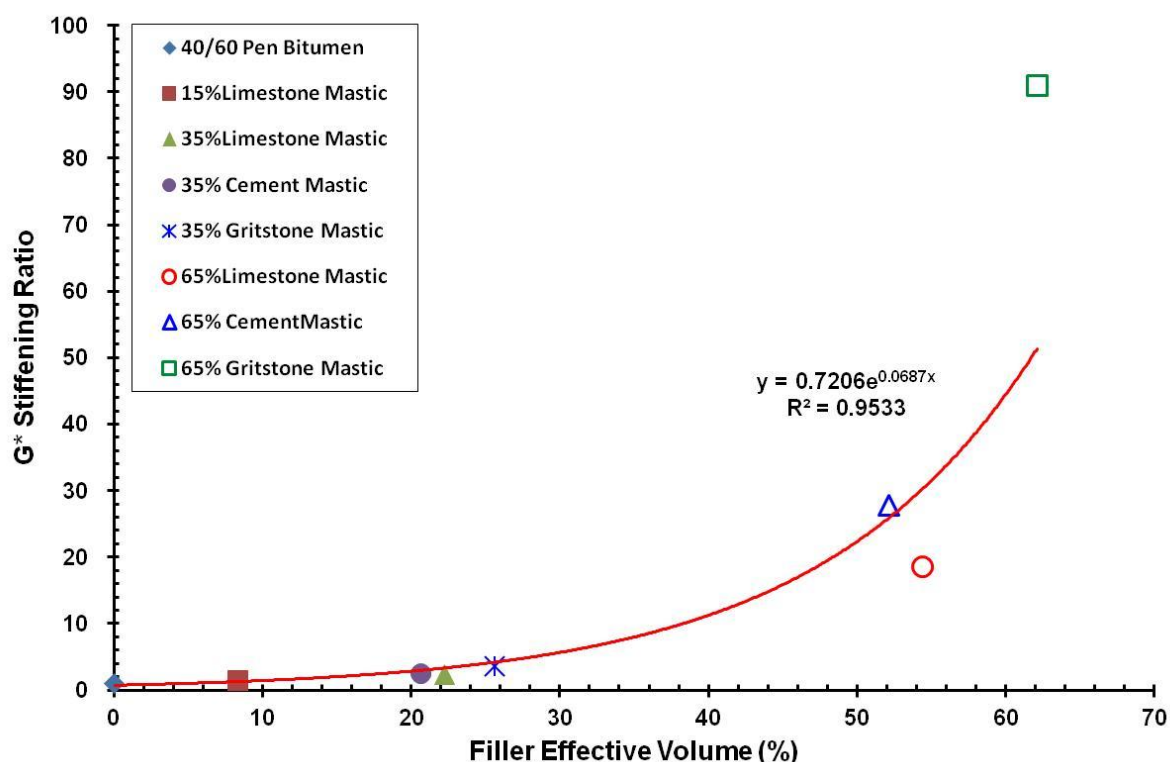


Figure 7. Stiffening ratios of filler contents and types at low shear condition for base bitumen and bitumen-filler mastics

Figure 8 shows the rheological properties of the base bitumen and limestone bitumen-filler mastics in terms of the phase angle,  $\delta$ , at different filler concentration levels. A reduction in  $\delta$  is observed with increasing filler content although little difference is observed between mastic and binder phase angle at low frequency (high temperature). This is due to the elastic component increasing with increasing mineral filler concentration in the bitumen-filler system. The base bitumen and dilute suspension mastics (15 and 35%) exhibit similar  $\delta$  as a function of frequency, while the limestone mineral filler significantly shifts the master curve of  $\delta$  for the concentrated suspension mastics (65%) downwards. The rheological behavior of the dilute suspension mastics is dominated by the base bitumen, while that of the concentrated suspension mastics is influenced by filler inter-particle interaction. Similar results are obtained from the set of phase angle master curves generated for other systems including cement and gritstone filler mastics.

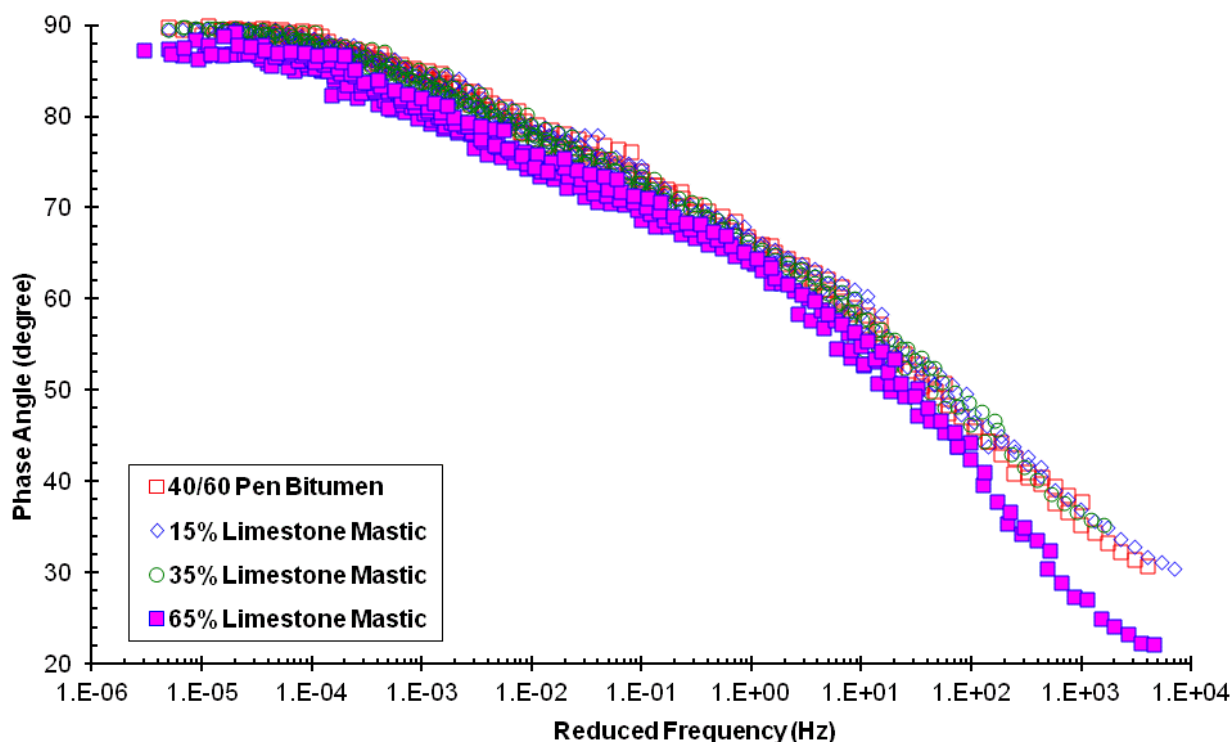


Figure 8. Phase angle master curves for limestone bitumen-filler mastics

Figure 9 shows the effect of filler type on phase angle at two filler concentration levels. For the dilute suspension mastics, the difference in stiffening effect is independent of filler type, whereas there is a considerable effect of filler type on  $\delta$  for the concentrated suspension mastics. It can be seen that the 65% gritstone mastic shows a partial breakdown of smoothness of the phase angle master curves. This represents a relatively complex rheological behavior for the 65% gritstone bitumen-filler mastic compared to the other mastics with a simple rheological behavior in terms of the phase angle. There might be due to the significant filler inter-particle interaction, leading to the relatively complex behavior.

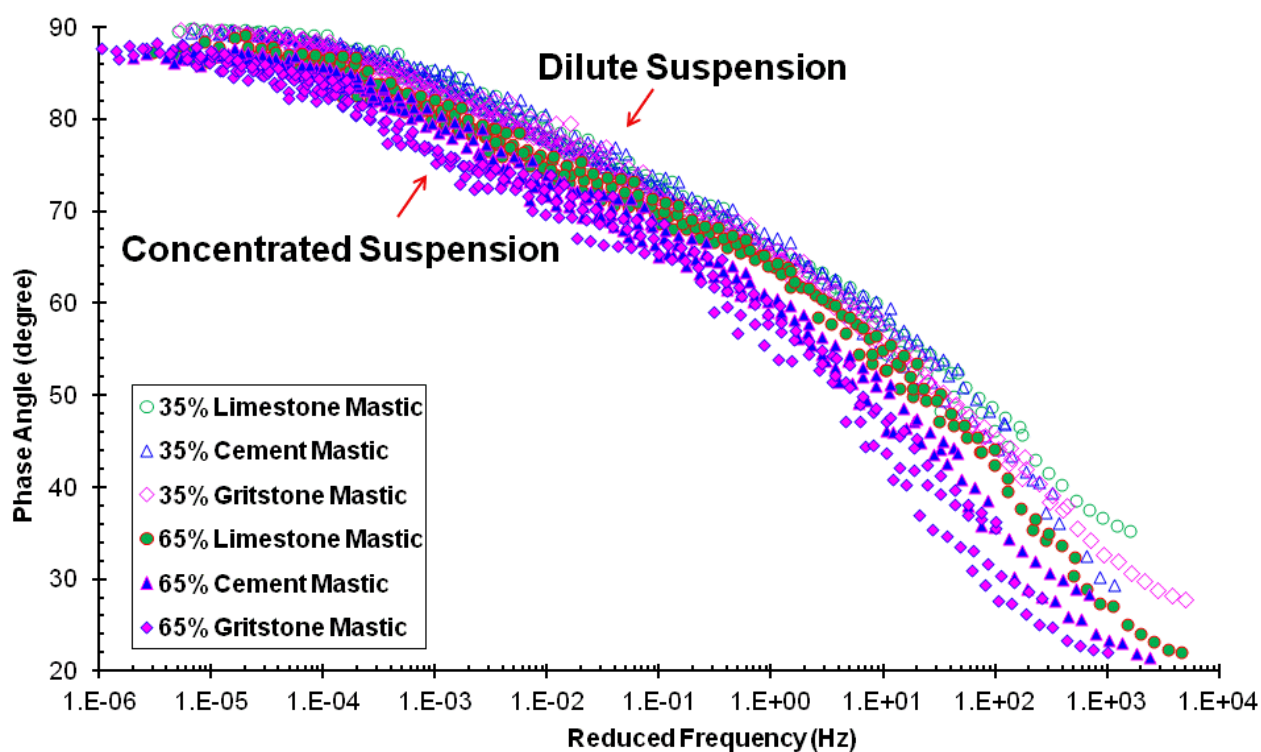


Figure 9. Phase angle master curves for dilute and concentrated suspension mastics

#### 4.3 Viscoelastic Properties by Christensen and Anderson model (CA model)

Figure 10 shows that the CA model was used to characterize the  $G^*$  master curves of the base bitumen and limestone bitumen-filler mastics. The CA model is reasonably accurate over a wide range of temperatures and frequencies for the base bitumen and 15% mastic. However, there is a slightly underestimated value for the 35% mastics at low frequencies. As the filler content increases up to 65%, the presence of filler inter-particle interaction leads to unsatisfactorily predicted  $G^*$  values in the viscous region. The model does not always predict results consistent with the experimental data as viscous flow is approached at low frequencies. Similar results and shapes of the master curves for cement and gritstone mastics are omitted in this section for brevity.



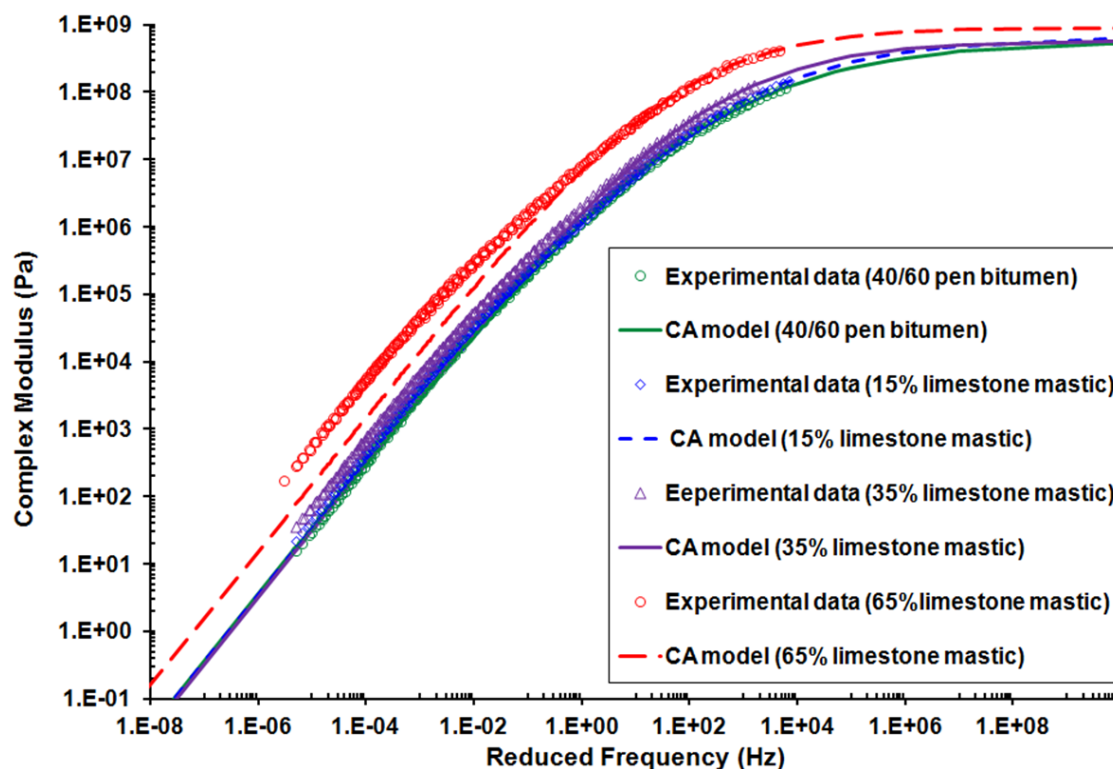


Figure 10. The CA model fit to  $G^*$  experimental data for limestone bitumen-filler mastics

Figure 11 shows a reduction in crossover frequency ( $\omega_0$ ) with increasing filler effective volume. There exists little difference between the base bitumen and dilute suspension mastics. The point where the viscous asymptote crosses the glassy modulus for the dilute suspension mastics is similar to that for the base bitumen. The crossover frequency indicates that the temperatures of viscoelastic solid to fluid transition are similar for the bitumen and dilute suspension mastics. However, the crossover frequencies for the concentrated suspension mastics are much lower because of the increase in elastic response. According to the results, the filler stiffening effect is significant for the concentrated suspension mastics compared to the dilute suspension mastics due to the crossover frequency being thought of as a hardness parameter. Therefore, the concentrated suspension mastic is much harder than the dilute suspension mastic. In addition, the experimental data at crossover ( $\tan \delta = 1$ ) are included in Figure 11 for comparison. There is only a minor difference in crossover frequency between the experimental and CA model data.

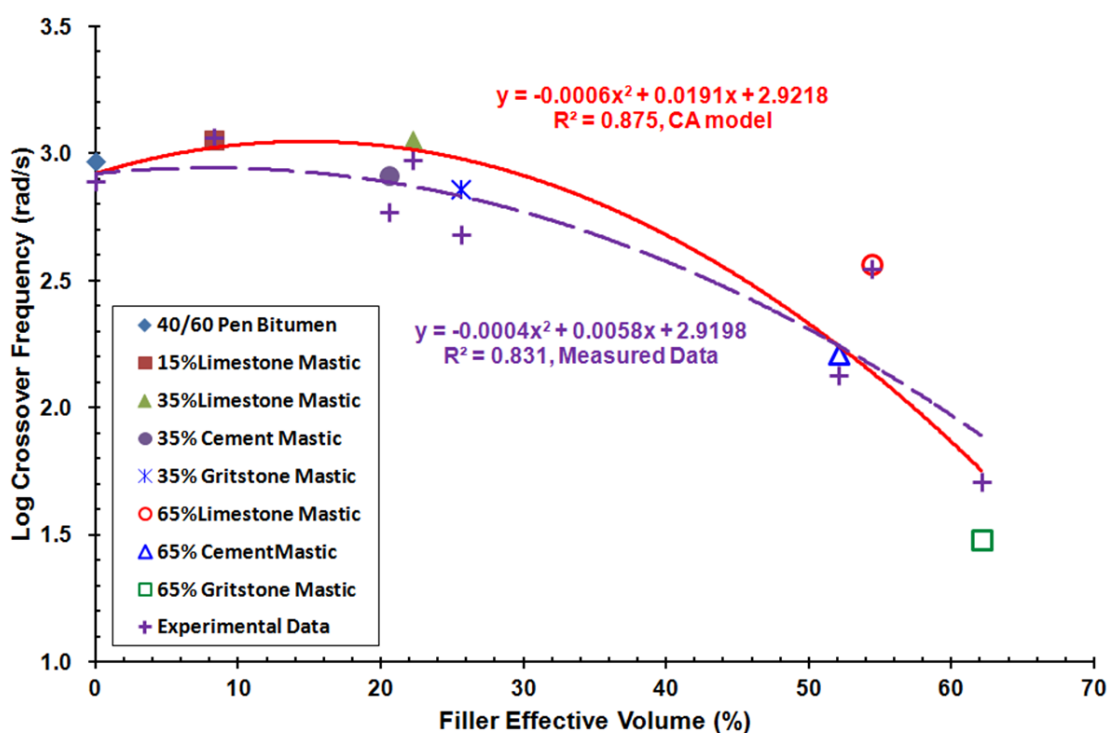


Figure 11. Effect of filler content and type on crossover frequency for base bitumen and bitumen-filler mastics

Figure 12 shows a decrease in rheological index,  $R$  with increasing filler effective volume. In general, the viscoelastic behavior of the concentrated suspension mastics change more slowly with time and temperature, followed by the dilute suspension mastics and base bitumen. According to the results in this study, the rheological indices for the base bitumen, dilute suspension mastics and concentrated suspension mastics are between 1.0 and 1.6. With higher values of rheological index, the concentrated suspension mastics show a slightly flatter master curve, and have an increased elastic behavior at intermediate loading times and temperatures compared to the dilute suspension mastics.



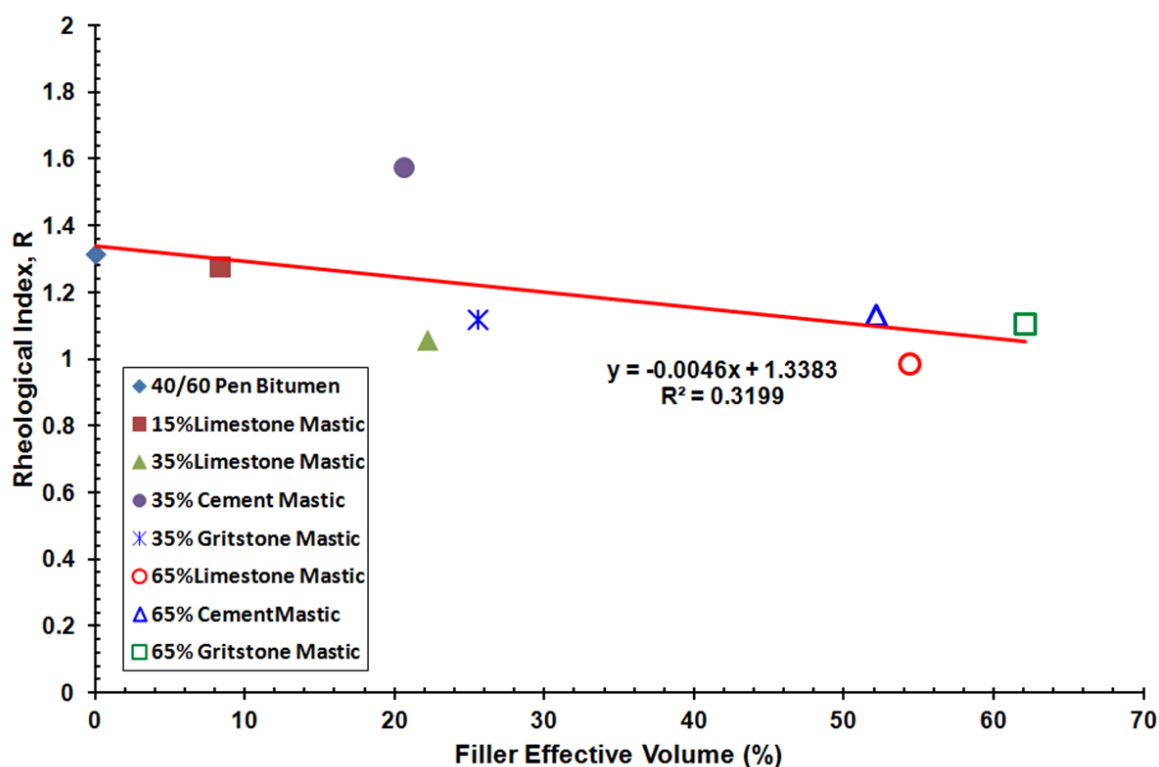


Figure 12. Effect of filler content and type on rheological index for base bitumen and bitumen-filler mastics

## 5. CONCLUSIONS

This paper describes the characterization of the linear viscoelastic behavior of bitumen-filler mastics by suspension rheology. Stress and frequency sweeps, using a dynamic shear rheometer, were performed on bitumen-filler mastics with dilute and concentrated filler suspension systems. The use of an empirical algebraic equation of the Christensen and Anderson model (CA model) was to characterize the LVE rheological behavior of the mastics. The following conclusions can be drawn from the test results and analyses:

- The LVE strain and stress limits as a function of  $G^*$  provide a clear indication that the LVE limits for the concentrated suspension mastics are more restricted than those for the dilute suspension mastics. The concentrated suspension mastics show a plateau and an upper strain limit of 1% at low frequency levels.
- In terms of the master curves of  $G^*$  and phase angle, the concentrated suspension mastics show the dominance of the filler inter-particle interaction. These filler particles cause a hydrodynamic interaction in this concentrated suspension system.
- The CA Model is reasonably accurate over a wide range of temperatures and frequencies for the base bitumen. However, the values are slightly underestimated values for the dilute suspension mastics at low frequencies. As the filler content increases up to 65%, the presence of filler inter-particle interaction leads to unsatisfactorily predicted  $G^*$  values in the viscous region.
- There is a significant stiffening effect on the viscoelastic behavior for the concentrated suspension mastics according to the crossover frequency and  $G^*$  stiffening ratio.
- Rheological results show that a significant difference in stiffening effect for the

concentrated suspension mastics exist due to the difference in the filler effective volume as well as the inter-particle interaction. For the dilute suspension mastics, the difference in filler stiffening effect is minimal irrespective of filler type.

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