MODELLING THE LINEAR VISCOELASTIC RHEOLOGICAL PROPERTIES OF BASE BITUMENS

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Abstract: A laboratory work has been carried out on four different unaged pure bitumens to determine the properties of the 2S2P1D (combination of two springs, two parabolic elements and one dashpot) model. Two types of tests namely the oscillatory dynamic and creep tests, were conducted in order to obtain the rheological data by means of the dynamic shear rheometer (DSR). Then, a method based on inter-conversions function was proposed to model the creep behaviour of base bitumen. It was found that the model which consists of seven parameters simulates in an excellent way the linear visco-elastic properties of the bitumens over a wide range of temperature and frequencies. Furthermore, a simple approximation applied to the 2S2P1D model allows to model the creep data satisfactorily. The goodness of fit statistical analysis showed that the model had a good correlation compare to the measured dynamic modulus data.

Keywords: modelling; complex modulus; phase angle; and creep

1.0 Introduction

The behaviour of bitumen has traditionally been evaluated using empirically based tests as part of penetration or viscosity graded specifications. The novel purpose of these specifications is to grade the bitumen according to its consistency and, therefore they do not address specific distress modes or ensure long term field performance. However, with the increase use of bitumen modifications such as polymer-modified bitumens, there is a need for fundamental testing as the conventional tests have proved inadequate for describing the linear viscoelastic (LVE) properties needed for a complete rheological evaluation of bitumens (Van der Poel 1954). Therefore, the dynamic shear rheometer (DSR)

was introduced during the Strategic Highway Research Programs (SHRP) campaign. The DSR is a very powerful tool to characterise the LVE rheological properties of bitumens (Airey 1997, 2002).

However, the DSR machine also has its limitation where it can only be conducted at a limited range of temperatures and frequencies in accordance with several problems encountered during experimental work and also from the machine itself (Airey 2002). Thus the introduction of modelling work is seen to be very useful for paving technologists in order to predict the behaviour of bitumen that cannot be reached by the experimental campaign. In early years, the invention of non-linear multivariable method was used to represent the linear viscoelastic rheological properties of bitumens. However, this method becomes obsolete these days due to the invention of computational techniques and was then replaced by the empirical algebraic equation and mechanical element approach.

In the empirical algebraic (also known as mathematical or constitutive or phenomenological) approach, one is simply adjusts any mathematical formulation whatsoever to the experimental main curve, with the quality adjustment being the sole criterion of choice of formulation. Meanwhile, in the mechanical element approach (or analogical model), use is made of the fact that the behaviour of linear viscoelastic material can be represented by a combination of simple spring and dashpot mechanical models, resulting in a particular mathematical formulation (Eurobitume 1995). The predictive models have been developed circa 1950s with the invention of Van Der Poel's non-linear multivariable model (Van Der Poel 1954). Most of the models rely on the construction of stiffness/complex modulus (S_b or $|G^*|$) and phase angle (δ) master curves and they do imply that the time temperature superposition principle (TTSP) for bitumens (Christensen and Anderson 1992; Lesueur *et al.* 1996).

Among the models observed form the literature, the 2S2P1D model was found to be a unique model where this model is capable of predicting the LVE behaviour for both bitumens and asphalt mixture. Moreover, each of the model's parameter brings a significant physical meaning. Therefore, this study was conducted to envisage the capabilities of the 2S2P1D model to predict the LVE behaviour of base bitumens. An extension to the modelling of creep tests, involving the combination of the 2S2P1D model with inter-conversions functions, is also presented. Finally the correlation between measured and predicted values was assessed using the goodness of fit statistical analysis.

2.0 The 2S2PID Model

The 2S2P1D, an abbreviation of the combinations of two springs, two parabolic creep elements and one dashpot, is a unique rheological model to predict the LVE properties for bitumens and asphalt mixtures (Olard et al. 2003; Olard and Di Benedetto 2003; Delaporte et al. 2007; Pellinen et al. 2007). This model is based on the generalization of the Huet-Sayegh model and is shown in Figure 1.

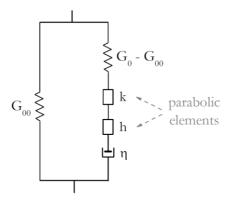


Figure 1: Repesentation of the 2S2P1D model (Delaporte et al. 2007)

The introduced model consists of seven parameters and the complex modulus, $|G^*|$ can be shown in the following mathematical expression:

$$|G^*| = G_o + \frac{G_{oo} - G_o}{1 + \delta (\omega \tau)^k + (\omega \tau)^h + (\omega \beta \tau)}$$
 ... (1)

where i is complex number defined by $i^2 = -1$, ω is frequency (Hz), k and h are exponents with 0 < k < h < 1, δ is constants, G_{oo} is the static modulus when $\omega \to 0$, G_o is the glassy modulus when $\omega \to 0$, β is constant and defined by $\eta = (G_{oo} - G_o) \beta \tau$, η is the Newtonian viscosity and τ is characteristic time, function of temperature.

The τ evolution maybe approximated by a shift factor law such as the William, Landel and Ferry (WLF) and Arrhenius equations in the range of temperature observed in the laboratory. It accounts for the TTSP (Delaporte et al. 2007):

$$\tau = \mathbf{a}_{\mathrm{T}} \bullet \mathbf{b}_{\mathrm{o}} \qquad \dots (2)$$

where a_T is the shift factor at temperature, T_i and τ_o is $\tau(T_{ref})$ determined at the reference temperature (T_{ref}) . The a_T can be determined using the WLF equation:

$$\log a_{T} = \frac{-C_{1} \P - T_{ref}}{C_{2} + \P - T_{ref}} \qquad ... (3)$$

where C_1 and C_2 are dimensionless parameters and the other parameters are as previously defined.

3.0 Experimental Design

3.1 Materials

Four different base bitumens have been tested, namely as bitumens 10/20, 35/50, 40/60 and 160/220 penetration grades in order to characterise the linear viscoelastic behaviour of bitumen and its modelling. The choice of these bitumens was dictated with the following targets: determines the properties of the 2S2P1D model and verify its validity for different types of base bitumens.

3.2 Dynamic test

The dynamic tests have been conducted using the Bohlin Gemini controlled strain Dynamic Shear Rheometer (DSR). The DSR is set by two plates sandwiching the bitumen. Both plates have the same dimensions, but the bottom one is fixed whereas the upper one is mounted on an axis allowing it to rotating, as shown in Figure 2.

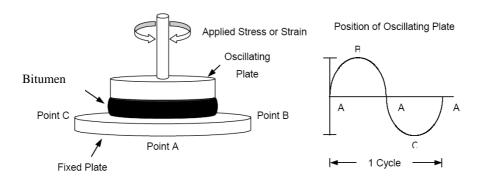


Figure 2: Principles of the dynamic shear rheometer (Airey, 1997)

The test is conducted by applying a torque on the top spindle, which creates oscillating shear stress (σ) and strain (ϵ), measured in the disc of bitumen. That provides for a given frequency, ω (Hz) and temperature, T (degrees Celsius), the value of the complex modulus $|G^*|$ (Pa) and the time lag between the applied load and the response, also known as phase angle, Φ (degrees) and defined by:

$$\left|G^*\right| = \frac{\sigma^*}{\varepsilon^*} = \frac{\tau_{\text{max}}}{\gamma_{\text{max}}} e^{i\varphi} \tag{4}$$

Two plates geometries used depend on the testing temperature: the 8mm and 25mm plates are used for temperatures from 0°C to 50°C and 35°C to 80°C, respectively (Airey, 1997). Between 35 °C and 50°C, the most accurate results were considered, depending on the tested specimens. The limit of the linear domain is determined for each binder at 10°C for the 8 mm plate and at 40°C for the 25 mm plates and then used to perform the test: it will be imposed a strain such that the behaviour is actually in LVE region.

3.3 Creep test

The creep test was performed by applying a constant loading on a specimen. The testing temperatures were selected between 10° C and 80° C. Like the dynamic test, two plate geometries were used: 8 mm from 0° C to 50° C and the 25 mm plates from 35° C to 80° C. The constant loading is applied during a given time, and the sample deformation is measured. This allows to get the stiffness G(t) (Pa) and the creep compliance J(t), which is the inverse of the G(t) in Pa⁻¹, of

the specimen. For every test, a constant loading $\sigma_o = 2kPa$ was applied. This value chose to obtain linear creep behaviour; generally observed for stress lower than 50kPa. The loading was applied during 100s to make sure sample reaches the steady state creep and consequently avoiding from the break.

3.4 Goodness of fit

Two statistical methods were used to indicate the goodness of fit between the measured and predicted results and can be shown as the following (Tran and Hall 2005; Garcia and Thompson 2007):

3.4.1 Standard error ratio (S_e/S_v)

The standard error of estimation, S_e and standard error of deviation, S_y can be defined as the following:

$$S_{e} = \sqrt{\frac{\sum \left(\mathbf{\hat{Y}} - \hat{\mathbf{\hat{Y}}} \right)^{2}}{\left(\mathbf{\hat{I}} - \mathbf{\hat{K}} \right)^{2}}} \tag{5}$$

and

$$S_{y} = \sqrt{\frac{\sum \left(\mathbf{V} - \overline{\mathbf{Y}} \right)^{2}}{\left(\mathbf{h} - 1 \right)^{2}}} \tag{6}$$

where n is sample size, k is number of independent variables in the model, Y is tested dynamic modulus, \hat{Y} is predicted modulus and \hat{Y} is mean value of tested dynamic modulus. The smaller the value, the better the prediction (Tran and Hall 2005).

3.4.2 Coefficient of correlation (R^2)

$$R^{2} = 1 - \frac{\left(\mathbf{A} - \mathbf{k} \right)}{\left(\mathbf{A} - 1 \right)} \times \left(\frac{\mathbf{S}_{e}}{\mathbf{S}_{y}} \right)^{2} \tag{7}$$

where the parameters are as previously defined and for the perfect fit, $R^2=1$. The criteria for the goodness of fit statistic parameters are shown in Table 1 (Tran and Hall 2005; Garcia and Thompson 2007):

Criteria	R^2	S_{e}/S_{y}
Excellent Good Fair Poor Very Poor	≥ 0.90 $0.70 - 0.89$ $0.40 - 0.69$ $0.20 - 0.39$ ≤ 0.19	≤ 0.35 $0.36 - 0.55$ $0.56 - 0.75$ $0.76 - 0.89$ ≥ 0.90

Table 1: Criteria for goodness of fit statistics

4.0 Results and Discussion

4.1 Complex modulus test

From the experimental campaign, it was found that all the tested samples could be categorized as *thermo-rheological simple* material due to several reasons:

- A shifting procedure done with shift factors a_T on the isothermal plots of $|G^*|$ allows to get a single, continuous curve at the T_{ref} =10°C. For brevity, only an example for the bitumen 35/50 penetration grade is shown in Figure 3. This property is called time temperature superposition principle (TTSP) and is typical from a *thermo-rheologically simple* material. Meanwhile, Figure 4 illustrates the shift a_T is used and the WLF equation has been employed in order to construct $|G^*|$ master curve. The $|G^*|$ master curves for all tested samples can represented in Figure 5.
- The Black diagrams, which is a graph of the magnitude (norm) of $|G^*|$ versus the Φ obtained from a dynamic test, are unique (Figure 6).

These plots exhibit the classical rheological properties of pure bitumen. Increase of temperature and decrease of frequency causes bitumens more viscous, and bitumen becomes more elastic when the temperature is decreases and frequency is increases. The four $|G^*|$ master curves reach a limiting glassy modulus (G_o) between 0.5 GPa to 1GPa depending on the specimen. In addition, it also can be seen that the $|G^*|$ values are higher as sample becomes harder (Figure 5).

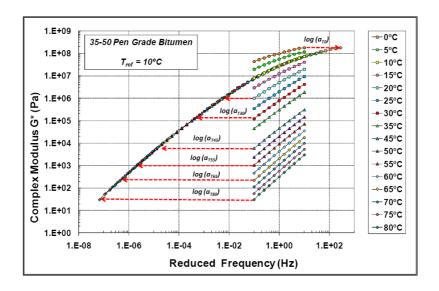


Figure 3: Shifting procedure for the $|G^*|$ master curve

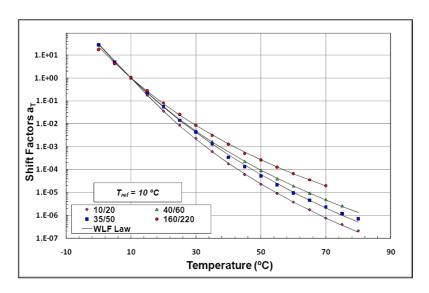


Figure 4: Shift factors curves using the WLF equation

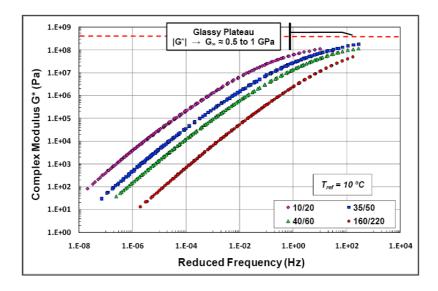


Figure 5: The |G*| master curves

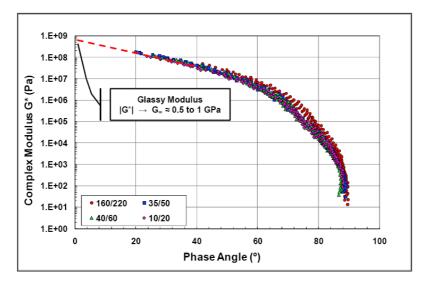


Figure 6: The Black diagrams

This phenomenon could attribute to the presence of more asphaltene (polar molecules) content increases the hardness of bitumen. All these observations are classical properties of base bitumen and conform to the expectations and can therefore be used to study the 2S2P1D model.

4.2 Creep test

The isothermal plots were shifted by an amount b_T close to the inverse of the shift factors a_T obtained from the complex modulus testing results (Figure 7). This is another characteristic property of the *thermo-rheologically simple* behaviour of pure bitumen. Then, as expected, as the hardest binder (10/20) has the lowest creep compliance and the softest bitumen (160/220) has the highest creep compliance (Figure 8).

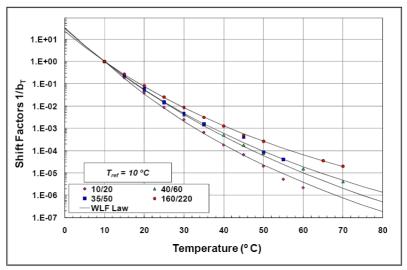


Figure 7: Shift factors for creep compliance data

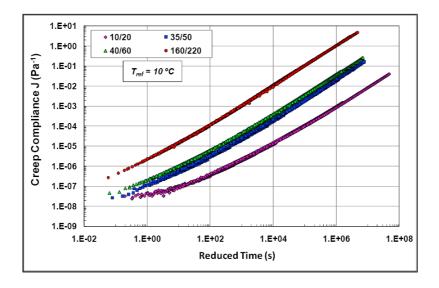


Figure 8: Creep compliance master curves

4.3 Model calibration

4.3.1 Modelling the complex modulus data

The 2S2P1D model was calibrated for every sample using the DSR complex modulus data at 10°C. The calibration was done manually using the MS Excel spreadsheet. Table 2 shows the 2S2P1D model parameters for the base bitumens.

Table 2: The 2S2P1D model's parameters

Pen Grade	G _{oo} (Pa)	G _o (GPa)	k	h	δ	τ	β
10/20	0	0.45	0.21	0.61	2.3	2.30 E-02	75
35/50	0	0.55	0.21	0.61	2.3	2.00 E-03	75
40/60	0	0.55	0.21	0.61	2.3	5.50 E-04	75
160/220	0	1.00	0.21	0.61	2.3	1.50 E-05	75

Interestingly, it was found that the G_{oo} , k, h, δ and β values can be taken as similar for all samples regardless the penetration grade of bitumens. The 2S2P1D curve can be shown in Figure 9. The k, h and δ values were found to be in good agreement with the prevoius researcherses (Olard et al. 2003; Olard and Di Benedetto 2004; Delaporte et al. 2007). The β values are similar due to the fact that they have similar slope at low frequencies or high temperature. As expected, the τ values different for each sample since it has a direct relationship with the T_{ref} . Moreover, the model's parameters can be reduced to six as the G_{oo} values can be taken as nought. The measured Φ also showed good agreement with the 2S2P1D model.

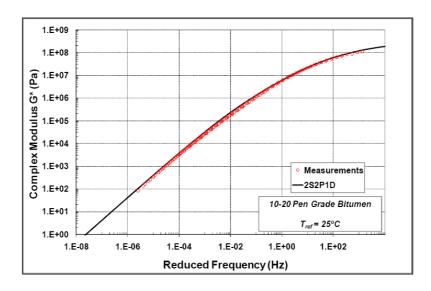


Figure 9: Comparison between measured and predicted $|G^*|$ master curve

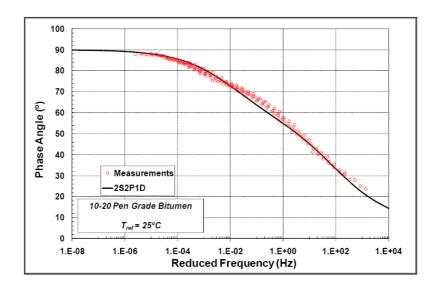


Figure 10: Comparison between measured and predicted Φ master curve

4.3.2 Modelling the creep compliance data

The previously modelled complex modulus values have been converted into creep testing results by means of the following inter-conversions functions (Marasteanu, 1999):

Kopelman equation;

$$J \bigcirc |J^* \bigcirc J^{-\frac{1}{\alpha}}|$$
 (7)

Christensen equation;

$$J \bigcirc |J' \bigcirc |_{\underline{\underline{a}}}$$
 (8)

Schwarlz and Struik equation;

$$J \blacktriangleleft .25t \geqslant \begin{bmatrix} J' + 0.5303J'' + 0.5282\omega \\ +0.021J'' + 0.0849\omega - 0.0418J'' + 0.37\omega \end{bmatrix}_{t=\frac{1}{\omega}}$$
(9)

where t is time (sec), J is the creep compliance (Pa⁻¹), J^* is the complex creep compliance (Pa⁻¹), J' is the real part of J^* , J'' the absolute value of the imaginary part of J^* and ω the frequency (Hz).

As shown in Figure 11, the results obtained show a good correlation between measured and predicted data particularly with the Kopelman and the Schwarlz and Struik equations. However, the predicted seems deviate from measured data when Christensen equation, therefore this equation did not take into account for the statistical analysis. The statistical analysis undertaken is summarized in Table 3.

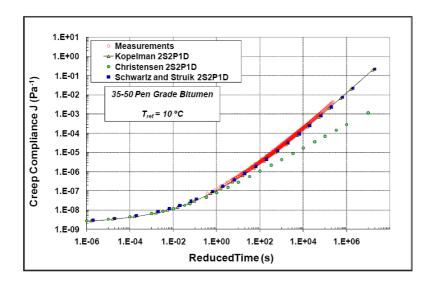


Figure 11: Comparison between measured and predicted creep compliance

Table 2: Goodness of fit statistical analysis

Test		Complex modulus			Creep		- Evaluation
	2S2P1D		Kopelman		Schwarzl & Struik		
Pen Grade	\mathbb{R}^2	S_e/S_y	\mathbb{R}^2	S_e/S_y	R^2	S_e/S_y	
10/20	0.994	0.075	0.988	0.110	0.972	0.169	excellent
35/50	0.999	0.028	0.991	0.097	0.998	0.049	excellent
40/60 160/220	0.990 0.997	0.010 0.055	0.995 0.998	0.069 0.042	0.985 0.999	0.123 0.034	excellent excellent

From Table 2 above, it was observed that the 2S2P1D model produced the promising results as it can predict the complex modulus excellently. Meanwhile, the Kopelman and Schwarlz and Struik equations show an excellent correlation between measured and predicted data. However, it is profoundly recommended to use the Kopelman equation compare to the others due to this equation is less complex in nature.

4.0 Conclusion

Several conclusions can be drawn from this study:

- 1) The 2S2P1D model is quite simple in its formulation, as a combination of springs, parabolic elements and dashpot. The calibration of the model is not a very difficult tasks and the model can provide very good modelling of the bitumens within a large range of temperatures and frequencies.
- 2) The experimental campaign, including dynamic shear rheometer complex modulus and creep tests on four different unaged pure bitumens allowed getting suitable results for modelling and studying the properties of the 2S2P1D model.
- 3) The k, h, δ and β parameters can be taken similar for all tested sample, regardless the penetration grade of bitumen.
- 4) It can be said that the conversion of the 2S2P1D complex modulus values allow to get a correct modelling of dynamic moduli tests. Meanwhile the Kopelman and Schwarlz and Struik equations show an excellent correlation between measured and predicted creep compliance data. However, it is suggested to employ the Kopelman equation since it is easier to be used.

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