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The Use of Inter-conversion Equations on Bituminous Binder Data

(Penggunaan Persamaan Antara-Penukaran ke Atas Data Pengikat Berbitumen)

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ABSTRACT

According to the classical theory of viscoelasticity, a linear viscoelastic (LVE) function can be converted into another viscoelastic function even though they emphasize different information. In this study, dynamic tests were conducted on different conventional penetration grade bitumens using a dynamic shear rheometer (DSR) in the LVE region. The results showed that the dynamic data in the frequency domain can be converted into the time domain functions using a numerical technique. This was done with the aid of the non-linear regularization (NLREG) computer program. The NLREG software is a computer program for solving nonlinear ill-posed problem and is based on non-linear Tikhonov regularization method. The use of data interconversion equation is found suitable for converting from the frequency domain into the time domain of conventional penetration grade bitumens.

Keywords: Complex modulus; creep compliance; master curve; relaxation modulus

ABSTRAK

Menurut teori klasik likat kenyal, satu fungsi linear likat kenyal (LVE) boleh ditukar kepada fungsi likat kenyal yang lain walaupun ia menekankan maklumat yang berbeza. Dalam kajian ini, ujian dinamik dijalankan ke atas bitumen biasa yang berlainan gred penembusan dengan menggunakan reometer ricih dinamik (DSR) di dalam sempadan LVE. Keputusan menunjukkan bahawa data dinamik dalam domain frekuensi boleh ditukar kepada fungsi-fungsi dalam domain masa dengan menggunakan teknik berangka. Ini dilakukan dengan menggunakan bantuan perisian komputer pengaturan tidak linear (NLREG). Perisian NLREG adalah satu program komputer yang digunakan untuk menyelesaikan masalah tidak linear yang buruk yang timbul dan ia berdasarkan kaedah pengaturan Tikhonov tidak linear. Penggunaan persamaan antara-penukaran data adalah didapati sesuai untuk menukarkan domain frekuensi kepada domain masa terhadap bitumen bergred penembusan biasa.

Kata kunci: Lengkung utama; modulus kelonggaran; modulus kompleks; pematuhan rayap

INTRODUCTION

It is widely known in the classical theory of viscoelasticity that by using specific mathematical inter-relationship equations, one linear viscoelastic (LVE) function can be converted into another viscoelastic function even though they emphasize different information (Plazek & Echeverria 2000). For example, the dynamic data in the frequency domain (storage modulus, G' and loss modulus, G'') can be converted into other functions in the time domain (creep compliance, $J(t)$ and relaxation modulus, $G(t)$). Goodwin and Hughes (2000) discussed several factors that attracted rheologists to transform the material responses. Interested readers can consult these works for a detail discussion and are not shown here for brevity. The mathematical inter-relationship equations among the LVE functions have been established and normally presented either in a numerical or approximation method. Ample studies have been conducted in establishing the numerical methods including the works of Baumgaertel and Winter (1989), Park and Schapery (1999), Park and Kim (2001) and Mun et al. (2007). However, this method involves complex mathematical calculations.

On the other hand, classical approximation methods such as the Kopelman, Christensen, Ninomiya and Ferry and Schwarzl and Struik are still in use, mostly in the industrial laboratories (Emri et al. 2005). In rheological studies, the data interconversion method suffers from a drawback called the ill-posed problem (not well posed, fuzzy problem). In this problem, small changes in the data can cause arbitrarily large changes in the result. The ill-posed problem often occurs when the values of some model parameters must be obtained from the observed data. The ill-posed problem, however, can be avoided by keeping the number of relaxation modes small (Baumgaertal & Winter 1989). There are several methods found in literature for transforming from the frequency domain to the time domain and vice versa. The first method uses a Fourier transformation technique. However the extrapolation process is highly arbitrary particularly at higher and lower frequencies due to the limited experimental data. The second method utilises empirical correlations which are successfully used for wide classes of materials. For complex materials, one would like to have more general conversion method (Baumgaertal & Winter 1989). The third method is based on minimising the sum square of error between the measured and predicted (model) data. Over the years, a reasonable number of papers have been published, dealing with the ill-posedness problem in rheology (Baumgaertal & Winter 1999; Honerkamp & Weese 1990, 1993; Mun et al. 2007; Park & Kim 2001; Park & Schapery 1999). In addition, some computer programs such as the Non-Linear REGularization (NLREG), Fast TIKhonov REGularization (FTIKREG), Interactive Rheological Software (IRIS), CONTIN and Rheology Analysis (RHEA) are commercially available (Honerkamp & Weese 1990, 1993; Provencher 1982; Rowe & Sharrock 2000). The RHEA software for instance, is available to simplify the process involved in the construction of master curves (Anderson 2010).

Among the computer programs available, the nonlinear regression with regularization is a reliable and practical method to surmount the ill-posed problem. The NLREG program, which was developed by Honerkamp and Weese (1993), is a computer program for solving nonlinear ill-posed problem and is based on nonlinear Tikhonov regularization method. Honerkamp and Weese reported that the NLREG software takes the noise into account and yields a smooth relaxation and retardation spectra curves (Dealy & Larson 2006). In addition, to conduct creep, stress-relaxation and oscillatory tests simultaneously is time consuming, expensive and requires skill of personnel if complete LVE material profile is to be obtained. The use of data interconversion equations is found to be a valuable alternative procedure particularly in polymer studies. However, not many studies have been conducted using data inter-conversion equations on binders. The aim of this study was to express the dynamic mechanical data of penetration grade bitumens (frequency domain) with continuous spectra to allow the LVE behaviour in the time domain to be described. The reliability of the chosen technique is discussed.

Experimental Design

Four binders: 10/20 (hardest), 35/50, 40/60 and 160/220 (softest) penetration grade bitumens were used in this study. All samples underwent the dynamic tests using a dynamic shear rheometer (DSR) conducted in the LVE region (Figure 1). The test procedure and sample preparation method that were used with the DSR have been described in the previous publication and did not shown here for brevity (Airey 1997).

First, amplitude sweep tests were conducted to determine the LVE region of the binders based on the point where $|G^*|$ had decreased to 95% of its initial value (Airey 1997). After obtaining the limiting strain, the frequency sweep tests were carried out under the following test conditions on each sample:

Mode of loading	: controlled-strain
Temperature	: 10 to 80°C (with the interval of 5 °C)
Frequencies	: 0.01 Hz to 10 Hz
Spindle geometries	: 8 mm (diameter) and 2 mm gap (10 to 35°C) and 25 mm (diameter) and 1 mm gap (25 to 80°C)
Strain amplitude	: within the LVE response, dependent on $|G^*|$ of each material

The dynamic mechanical data obtained was presented in terms of master curves. For the construction of master curves, a reference temperature, T_{ref} was arbitrarily taken at 10°C. The curve was shifted randomly, without assuming any shift factor function for the construction of master curves.

Results and Discussion

Amplitude Sweep Test

The temperatures of the amplitude sweep tests are taken close to the lowest temperature of each testing geometry because the linear limit of the material decrease with the temperature. Table 1 shows the LVE strain limit obtained from each samples using the 8 mm spindle at 10°C and 25 mm spindle at 40°C, respectively. To ensure that the test is conducted in the corrected region, a target strain from 8 mm spindle at 10°C is selected for all the experiments.

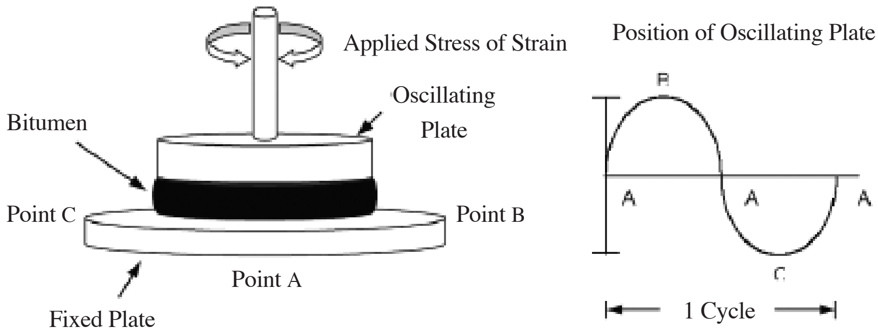

FIGURE 1. Dynamic shear rheometer set-up

TABLE 1. Linear viscoelastic (LVE) strain limits

Pen. Grade	10/20	35/50	40/60	160/220
8 mm at 10ºC	0.6%	0.8%	1.3%	1.5%
25 mm at 40ºC	1.0%	1.0%	2.0%	4.0%

DYNAMIC TEST

Figure 2 shows the complex modulus, $|G^*|$ master curves for all samples. The conventional bitumens generally can be classified as thermo-rheologically simple materials with their rheological properties being temperature and frequency (or time) equivalent, provided that they are determined within the LVE region (Airey 2002). The four $|G^*|$ master curves of conventional bitumens reach a limiting value at high frequencies, called the glassy modulus, G_g between 0.5 and 1 GPa. However, these values depend on the hardness of a specimen. The 10/20 penetration grade bitumen show the lowest value of G_g. This phenomenon could be attributed to the compliance (testing) errors associated with the rheometer. On the other side, all bitumens tend to show the Newtonian (viscous) flow at high temperatures and/or low frequencies. Meanwhile, a Black diagram, a graph of the magnitude (or norm) of $|G^*|$ versus the phase angle, δ obtained from the dynamic tests is unique. It is observed that all samples merge into a single curve. Therefore, the Black diagram can be thought as the 'fingerprint' to check inconsistencies in the experimental data (Airey 2002; Figure 3).

According to the theory of elasticity, the $|G^*|$, G', G'' and δ are related to each other and shown as the following:

$$\left|G^*\right| = \sqrt{G'^2 + G''^2}, \tag{1}$$

or in complex notation as:

$$G^* = G' + iG'', \tag{2}$$

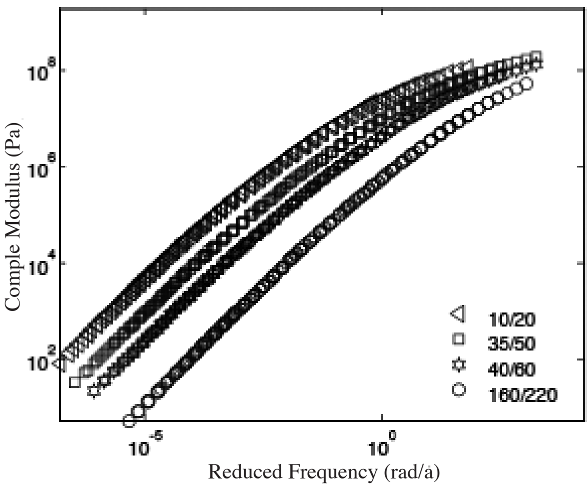

FIGURE 2. Complex modulus master curves

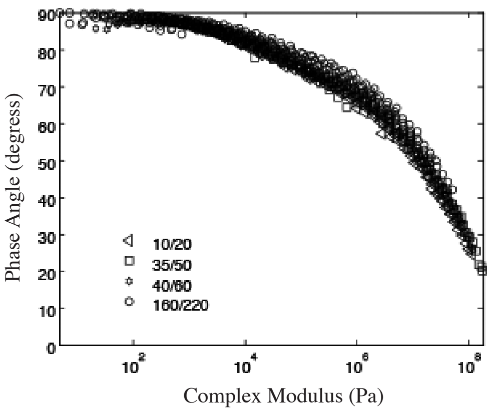

FIGURE 3. The Black diagrams

with

$$\delta = \tan^{-1}(G''/G'), \tag{3}$$

where i is the complex number ($i^2 = -1$), $G' = |G^*| \cos\delta$ and $G' = |G^*| \sin\delta$. The other symbols are as previously defined. G' and G'' in the NLREG software can be shown as the following equations:

$$G'(\omega) = G_e + \int_{-\infty}^{\infty} H(\tau)\left((\omega\tau)^2 / 1 + (\omega\tau)^2\right) d\ln\tau, \tag{4}$$

and

$$G''(\omega) = \int_{-\infty}^{\infty} H(\tau)\left((\omega\tau)^2 / 1 + (\omega\tau)^2\right) d\ln\tau, \tag{5}$$

where $H(\tau)$ is a relaxation spectrum, G_e is the equilibrium modulus, ω is frequency and τ is relaxation time. The pre-smooth technique is necessary to eliminate the noise and waviness in the experimental data. No critical difference could be observed on $|G^*|$ data before and after pre-smoothing technique. However, the curves are not shown here for brevity. Therefore it is inferred that (2) and (3) can be used for the following calculations. The $|G^*|$ master curves can also be shown in the form of the dynamic compliance, J^* master curves. In the NLREG software, J^* is defined as follows:

$$J^* = \sqrt{J'^2 + J''^2}, \tag{6}$$

with:

$$J'(\omega) = J_g + \int_{-\infty}^{\infty} L(\tau) \times \left(1/1 + \omega^2\tau^2\right) d\ln\tau, \tag{7}$$

and

$$J''(\omega) = \int_{-\infty}^{\infty} L(\tau) \times \left(\omega\tau/1 + \omega^2\tau^2\right) d\ln\tau, \tag{8}$$

where J_g is the instantaneous compliance and $L(\tau)$ is retardation spectrum. The other parameters are as previously defined. J^* master curves are depicted in Figure 4. These curves indicate deformation susceptibility of materials at high frequencies and/or low temperatures. It is worth mentioned that the J^* data can be converted into the creep compliance, $J(t)$ using the conversion of ω=1/T and therefore, they are expected to carry out similar information of each other. The discussion will only be presented for the $J(t)$ master curves with the understanding that the commentary applies to the J^* master curves as well.

CREEP COMPLIANCE

The creep compliance, $J(t)$ is calculated using the following expression:

$$J(t) = J_g + \int_{-\infty}^{\infty} L(\tau)\left(1 - e^{-t/\tau}\right) d\ln\tau + (t/\eta_0), \tag{9}$$

where the parameters are as previously defined. Figure 5 shows the $J(t)$ master curves for all the samples used in this study. In general, the $J(t)$ master curve consists of three regions namely the glassy, transition and terminal. At short loading times, $J(t)$ tends to converge approximately between 10^{-8} and 10^{-9} Pa^{-1}. This value is called the glassy compliance, J_g. Therefore, $J(t)$ values are relatively independent on the characteristics of the binders at short loading times. In the transition region, a dramatic change of several decades may take place in $J(t)$ with the rises by several powers of 10. As time and temperature increase, the terminal (or viscous region) is dominant. At this region, the slope of most curves of conventional bitumens approach infinity, showing that the viscous flow is achieved in the observed range of time scale (Einaga et al. 1971). It is expected because at the long time of loading, $J(t)$ includes a contribution from viscous flow and therefore it increases without limit (Ferry 1980).

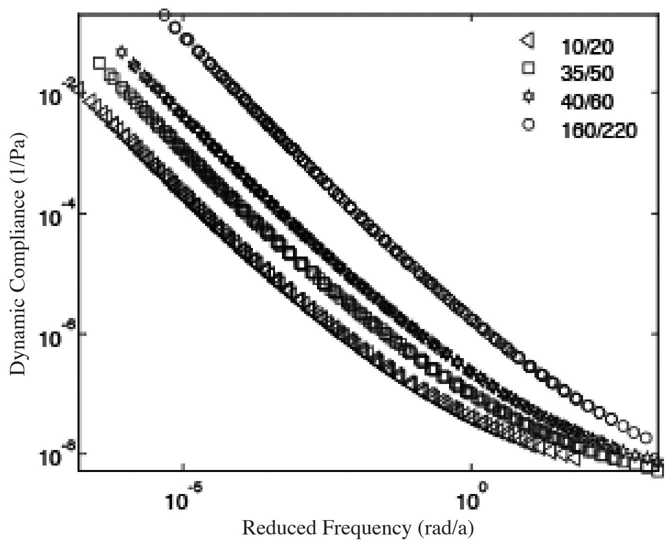

FIGURE 4. Dynamic compliance master curves

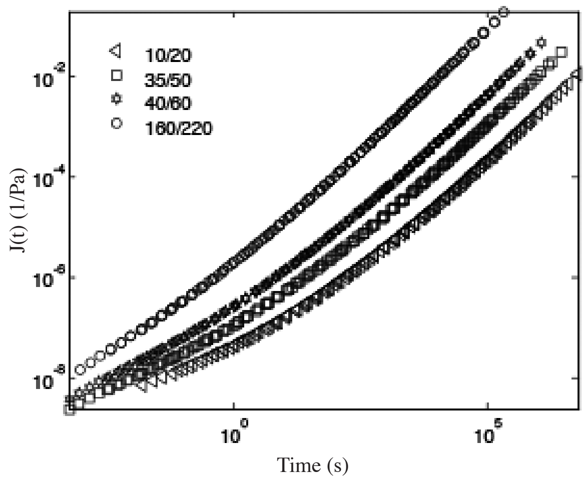

FIGURE 5. Creep compliance master curves

As discussed by Mayama (1997), the higher value of compliance indicates the easier it is for the material to deform. By considering $J(t)$ as a measure of deformation susceptibility, it is clear that the 10/20 penetration grade bitumen has a very high deformation resistance, followed by the 35/50, 40/60 and 160/220 penetration grade bitumens. The presence of higher asphaltenes content in harder bitumen provides higher resistance on deformation. This result in an increase in polar, high molecular weight fraction at the expense of the lower molecular weight resins (Mayama 1997). An anomaly, however, still seen particularly for the 10/20 penetration grade bitumen at the low temperatures. This phenomenon could be attributed to the reason where $J(t)$ data are exposed to the compliance errors from a rheometer at the glassy region, as similarly observed for the $|G^*|$ master curve.

Table 2 shows the η_0, J_g and J_e^o values obtained from the NLREG software. The higher value of η_0 indicates the higher resistance to rutting (permanent deformation). From the study, it is found that the η_0 values decrease from hard to soft penetration grade bitumens. Therefore, it can be inferred that harder bitumen is more suitable to be used for high in-service temperature regions. It is also observed that J_g is similar for all tested bitumens. At long times, $J(t)$ increases without limit since it includes a contribution from a viscous flow. If the viscous flow is subtracted, the remainder $J(t)–t/\eta_0$ approaches a limiting value, J_e^o. When examining the J_e^o data obtained form the NLREG software, there is not such an obvious trend could be observed as all tested binders show almost similar values.

Meanwhile, the $L(\tau)$ curves are shown in Figure 6. The $L(\tau)$ equation used in the NLREG software is shown as:

$$L(\tau) = H(\tau) \Big/ \left(\left(G_e - \int_{-\infty}^{\infty} \left(H(u) / (\tau / u) - 1 \right) d \ln u \right)^2 + \pi^2 H(\tau)^2 \right), \tag{10}$$

where the parameters are as previously defined. The $L(\tau)$ is represented as a continuous form in the $J(\tau)$ equation. It is recommended not to use the discrete element as it involves representing the spectrum by an empirical equation involving a set of coefficients that have no basic physical meaning. Moreover, the information present in the original data is always lost in the conversion to a discrete spectrum (Dealy & Larson 2006).

At long times, $L(\tau)$ should vanish when the viscoelastic material reaches the Newtonian flow (Ferry 1980). It is observed that the continuous $L(\tau)$ spectrum for the 160/220 penetration grade bitumen reaches the pinnacle point approximately at $\tau = 3 \times 10^3$ and after that, $L(\tau)$ dropped to the lower values, indicating the changes from the transition region to the terminal region. The peak point remarks the start of the terminal zone of that bitumen. It is expected since the 160/220 penetration grade bitumen is the softest sample and easily reached the viscosity flow at high temperature. Other bitumens, however, need longer time before reaching the pinnacle point.

RELAXATION MODULUS

The relaxation modulus, $G(t)$ is defined as:

$$G(t) = G_e + \int_{-\infty}^{\infty} H(\tau) e^{-t/\tau} d \ln \tau, \tag{11}$$

where the parameters are as previously defined. The $G(t)$ master curves are shown in Figure 7. In general, bitumen relaxes stresses a bit slower at low temperatures and/or at short loading times; however, it becomes quicker as the temperature increases and/or at long loading times. Like $|G^*|$, $G(t)$ is also the time and temperature dependent.

It is observed that the lower the viscosity and penetration index, the faster the stress relaxation (Mayama 1997). In general, the 160/220 penetration grade bitumen shows the fastest stress relaxation, followed by the 40/60, 35/50 and 10/20 penetration grades. This finding is in a good agreement with the work of Mayama (1997) even though he used asphalt mixture samples in the study. The rapid stress of 160/220 penetration grade bitumen is recommended from the point view of thermal stress relaxation; however this bitumen is at a disadvantage under

TABLE 2. Parameters obtained from the NLREG software

Pen. Grade	η_0 (Pa.s^{-1})	J_g (Pa)	J_e^o (Pa)
10/20	5.52×10^8	0.00	7.95×10^{-4}
35/50	9.72×10^7	0.00	1.95×10^{-3}
40/60	2.48×10^7	0.00	1.98×10^{-3}
160/220	1.12×10^6	0.00	1.41×10^{-4}

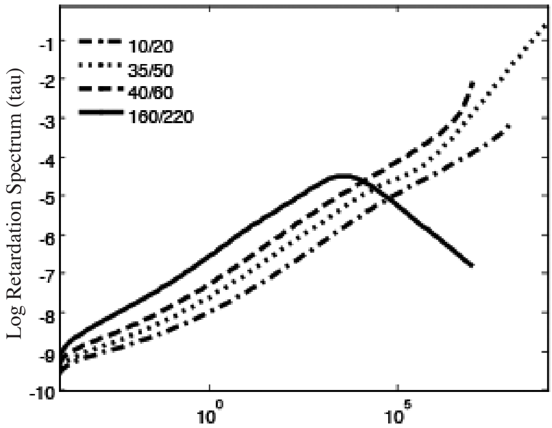

FIGURE 6. The retardation spectrum curves

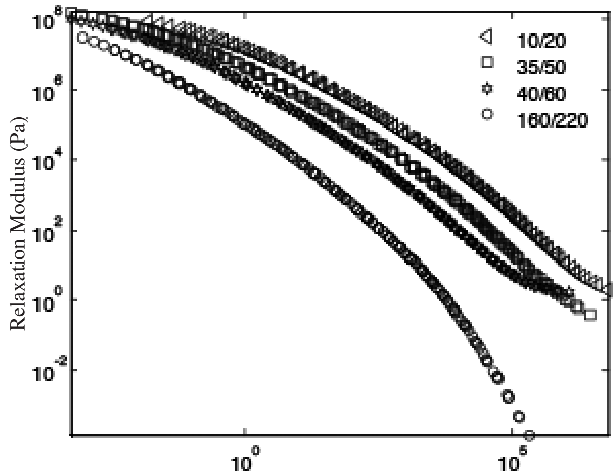

FIGURE 7. Relaxation modulus master curves

heavy standing or slow moving traffic, for example at bus stops and traffic lights (Mayama 1997).

Meanwhile, the relaxation spectrum, $H(\tau)$ is shown as the following:

$$H(\tau) = L(\tau) \Big/ \left(\left(J_g + \int_{-\infty}^{\infty} \left(L(u) / 1 - (\tau / u) \right) d \ln u - (\tau / \eta_0) \right)^2 + \pi^2 L(\tau)^2 \right), \quad (12)$$

where the parameters are as previously defined. The $H(\tau)$ curves for all bitumens are shown in Figure 8.

The $H(\tau)$ curves are almost similar with the $G(t)$ in shapes as they are related to each other (11). According to Ferry (1980), $H(\tau)$ weights contributes to modulus and $L(\tau)$ contributes to the compliances. In general, short time processes are revealed in more detail in $H(\tau)$ and long time processes in $L(\tau)$. However, this condition could possibly be changed with the presence of other substances such as polymer, higher asphaltenes and waxy element contents. Further studies will be directed in this way.

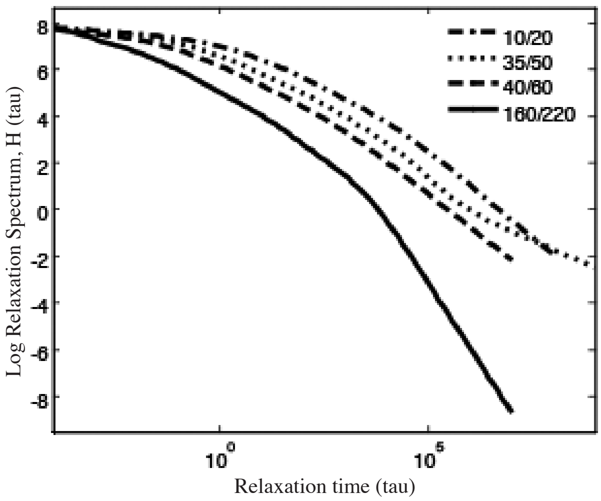

FIGURE 8. Relaxation spectrum curves

CONCLUSION

Penetration grade bitumens can be classified as thermorheologically simple materials with their rheological properties being temperature and frequency equivalent. Meanwhile, the Black diagrams are useful to check the inconsistencies of the measured dynamic mechanical data. A smooth curve indicates that penetration grade bitumen obeys the time-temperature superposition principle (TTSP). It is found that the creep compliance values increase as the temperature and time of loading are increased. The creep compliance master curve can be divided into three main regions namely the glassy, transition and terminal. The 10/20 penetration grade bitumen has the highest resistance to deformation, followed by 35/50 and 40/60 penetration grade bitumens. As expected, the 160/220 penetration grade bitumen is severely susceptible to deformation. It is observed that the lower the viscosity and penetration index, the fastest the stress relaxation with the 160/220 penetration grade bitumen shows the fastest stress relaxation, followed by the 40/60, 35/50 and 10/20 penetration grade bitumens. When the functions of relaxation and retardation spectra available, it is easy to obtain the relaxation modulus and creep compliance data. The use of data inter-conversion equations from the frequency domain into the time domain is found adequate for conventional penetration grade bitumens.

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