MODELING THE PHASE ANGLE MASTER CURVES OF MODIFIED BITUMINOUS BINDERS: ESTIMATING THE PLATEAU ZONE USING HEAVISIDE STEP FUNCTION

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ABSTRACT

Phase angle master curves of modified binders in the linear viscoelastic range show more variations compared to the |G| master curves. They were not however subjected to modeling purposes that much. In this research the Heaviside step function is combined with a Sine function to introduce a new model, capable of modeling the irregularities in the phase angle master curves of modified binders. Three neat and seven modified binders are used for validating the model. It is shown that the model is capable of predicting the plateau zone and fitting the master curves of modified binders in general. A comparison between the new model and another model recently generated based on the same approach is performed. It is shown that the new model is able to fit master curve irregularities at high temperature range while the old model is capable of modeling asymptotic behavior, especially at infinitely high frequencies.*

Keywords: Master curve; Modified binder; Phase angle model; Plateau

INTRODUCTION

Modeling the mechanical behavior of asphalt binders and mixtures has been subject of research since 1950s. Improvements in the dynamic testing techniques led to the construction of master curves of dynamic mechanical parameters like the norm of complex modulus (e.g. |G*|), or the loss and storage moduli [1-3]. Generation and use of phase angle δ master curves however have received less attention, since their construction, interpretation and application are not as straight forward and common as that of other parameters. Unlike |G*| master curves, the phase angle master curves of polymer modified binders (PMBs) may substantially differ from those of neat binders. A common characteristic of many of the PMBs master curves is occurrence of a plateau region which can be caused by formation of entanglements in the bitumen-polymer network in elastomer modified binders, or the presence of a rigid network in plastomer modified binders [4, 5]. Modeling of the plateau zone has not been sufficiently addressed in asphalt literature. However it is recently tried to capture this feature by introducing a Double-Logistic (DL) mathematical model [6]. This paper presents a new mathematical model for |G*| and phase angle master curves by applying the Heaviside step function into a sinusoidal function. The introduced models are aimed to be capable of predicting the plateaus and other irregularities that usually occur for PMB phase angle master curves. A comparison with the DL model has also been performed in order to clarify the weaknesses and strengths for each model.

THEORY

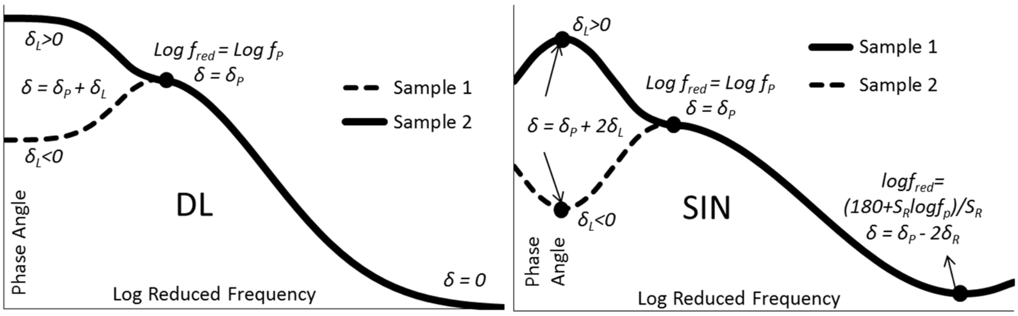
Fig. 1 shows the general form of the DL model. The model introduces a parameter δ_P which represents the phase angle at the plateau and f_P represents the frequency in which the plateau occurs. Phase angle falls down at the right side of the f_P so that it reaches the asymptotic value of zero at very high frequencies. At the left side of f_P the phase angle can rise or fall by an amount of δ_L. The rates of the changes at the left and right side of the plateau region are determined by two parameters S_L and S_R. The general equation of the DL model is:

$$\delta = \delta_p - \delta_p H(f_{red} - f_p)\left[1 - e^{-\left(S_R \cdot log\left(\frac{f_{red}}{f_p}\right)\right)^2}\right] + \delta_L H(f_p - f_{red})\left[1 - e^{-\left(S_L \cdot log\left(\frac{f_p}{f_{red}}\right)\right)^2}\right] \qquad (1a)$$

$$H(u) = \begin{Bmatrix} 0 & if\ u < 0 \\ 1 & if\ u \geq 0 \end{Bmatrix} \qquad 0 < \delta_p \leq 90 \qquad 0 < \delta_p + \delta_L \leq 90 \qquad 0 < S_L\ ,\ S_R \qquad (1b)$$

where the phase angle δ (in degrees) is modeled as a function of reduced frequency f_{red} using five parameters δ_P, δ_L, f_P, S_L and S_R, and the Heaviside step function as defined in equation (1b). Each of the Heaviside step functions in equation (1a) controls the master curve at one side of the plateau region. At the right side of the plateau ($f_{red}>f_P$), the first Heaviside step function is activated (becomes 1) and reduces the phase angle from δ_P down to zero. The reduction takes place by the mathematical term in the bracket which represents a simplified bell-shape Gaussian function. The general shape of the master curve in this region coincides with this function. At the left side of the plateau ($f_{red}<f_P$), the second Heaviside step function comes into play. The bell-shaped curve can rotate upward and the rise (or fall) value by a variable amount of δ_L occurs. A condition in equation (1b) keeps the value of δ_P between 0° and 90°. The horizontal asymptote of the master curve at low frequencies can be determined by $\delta=\delta_P+\delta_L$. If δ_L is positive, the expression yields the maximum value of the phase angle that should be less than 90° to produce the correct phase angle values. Negative values of δ_L also make the maximum value of the model equal to δ_P at the peak, which should be less than 90°.

Figure 1. General form of Phase angle master curve of DL and SIN models.



The Heaviside step function can be similarly applied to any other bell-shape mathematical function to get a specific PMB master curve. The capability of using the Sinusoidal function for this purpose is examined. The phase angle sinusoidal (SIN) model for master curves of modified binders could be written as:

$$\delta = \delta_P - \delta_R H(f_{red} - f_p)\left[1 - cos\left(S_R log\left(\frac{f_{red}}{f_p}\right)\right)\right] + \delta_L H(f_p - f_{red})\left[1 - cos\left(S_L log\left(\frac{f_{red}}{f_p}\right)\right)\right] \quad (2a)$$

$$0 \leq \delta_P \leq 90 \qquad 0 \leq (\delta_P - 2\delta_R) \qquad 0 \leq (\delta_P + 2\delta_L) \leq 90 \qquad 0 \leq \delta_R \quad (2b)$$

$$\max(\log f_{red}) \leq (180 + S_R \log f_P)/S_R \quad (2c)$$

where δ (in degrees) is modeled as a function of reduced frequency f_{red} using six parameters δ_P, δ_R, δ_L, f_P, S_L, S_R, and the Heaviside step function. All parameters are defined as the parameters for the DL model except that the phase angle drop at the right side of the plateau (δ_R) is not fixed on δ_P in this model. Fig. 1 also exhibits a good description of SIN model and its parameters. The right side of the plateau (low temperature or high frequency range) is modeled using δ_P, δ_R, f_P and S_R. δ_P represents the phase angle value at the plateau and controls the vertical displacement of the master curve. It is thus limited to between 0° and 90° as shown in equation (2b). Lower δ_P value can be expected for highly modified binders with a lower plateau phase angle value. The parameters δ_R and δ_L control the domain of the rise and fall on the right and left sides of the plateau. The parameters S_R and S_L control the period of the sine function at two sides of the plateau and then represent the slope of the curve after the plateau; the parameter f_P controls the horizontal displacement of the model over frequency axis.

The lowest value of the phase angle occurs at the right side of the SIN model, where the value of the sine operator equals -1. This occurs at $\log f_{red} = (180+S_R\log f_P)/S_R$, where the phase angle value is (δ_P -2δ_R) (Fig. 1). The maximum value of the phase angle is (δ_P +2δ_L). This occurs when δ_L is positive. To prevent the model from yielding inappropriate values for the phase angle or the corresponding parameters, it is recommended to restrict the maximum and minimum values of the phase angle to 90°and 0°, respectively. Also, the right side limit of the reduced frequency should be set at the point where the phase angle SIN model begins to produce an increase. This will prevent the phase angle model value from rising after reaching its minimum value. These conditions are expressed in equations (2b) and (2c). A positive δ_R value is also required.

The same parameters in phase angle equation can be used to generate a model for the |G*| master curve. Based on Kramers-Kronig equation modified by Booij and Thoone [7] shown in equation (3), the |G*| formula can be calculated by integrating the phase angle equation through the parameter of angular frequency ω.

$$\delta(\omega) \cong 90 \times \frac{dlog(G^*(\omega))}{dlog\omega} \rightarrow log(G^*(\omega)) \cong \frac{1}{90}\int \delta(\omega) dlog\omega + C \qquad (3)$$

Equation (3) has been previously validated and used by others [1, 8, 9]. Equation (3) has been also applied for DL model to get the |G*| formula. Integrating equation (3) yields to the following direct function for the |G*| master curve:

$$log|G^*| = log|G_0^*| + \frac{\delta_P}{90} log f_{red} - H(f_{red} - f_p)\frac{\delta_R}{90}\left[log f_{red} - \frac{sin\left(S_R log\left(\frac{f_{red}}{fp}\right)\right)}{S_R} - \left(\frac{\delta_R+\delta_L}{\delta_R}\right) log f_P\right] + H(f_p - f_{red})\frac{\delta_L}{90}\left[log f_{red} - \frac{sin\left(S_L log\left(\frac{f_{red}}{fp}\right)\right)}{S_L}\right] \qquad (4)$$

The |G*| master curve of binders can be modeled using equation (4). A parameter of $|G_0^*|$ was added to the parameters of the phase angle master curve and controls the vertical displacement of the |G*| master curve.

MATERIALS AND METHODS

Three neat and seven modified binders were used to generate master curves. The neat binders were a Pen.85/100 (Neat-1), an aged Pen.40/50 (Neat-2) and another Pen.85/100 binder from a different source (Neat-3). The modified binders comprise of one commercial (Styrelf 13-80), and six laboratory-made PMBs. These PMBs were produced using the binder Neat-1 and different types and amounts of modifiers. The modifiers were 0.5% and 2.5% Polyphosphoric Acid (PPA), 9% and 18% crumb rubber (CR), 7% Styrene-Butadiene-Styrene (SBS), and 6% Ethylene Vinyl Acetate (EVA). The required data for generating master curves were all gathered using a Dynamic Shear Rheometer (DSR) machine MCR301, in a wide range of temperatures from -30°C up to 88°C. The frequency sweep data in the frequency range of 0.1Hz to 10Hz were obtained in a strain-controlled mode and within the linear viscoelastic range. The master curves were generated using US200/32 V2.3 commercial software of Anton-Paar Company. Model fitting was performed by simultaneous fitting of the models over G' and G" and minimization of the summation of square of relative errors (SSRE).

RESULTS

The results of applying the SIN model to all binders are shown in Tab. 1. The DL model results are also presented for comparison. The table presents the SSRE, the parameter values and the number of data points used for master curve generation for all binders.

Table 1. Fitting results of SIN and DL model for 10 binders

Para-meter	Unit	Mod-el	Neat-1	Neat-2	Neat-3	PPA 0.5	PPA 2.5	Styrelf 13-80	EVA6	SBS7	CR9	CR18
δ_P	Degrees	SIN	90	90	90	90	49.8	69.0	56.8	63.6	52.8	51.4
		DL	90	90	90	90	47.7	68.4	56.5	64.0	59.1	53.1
f_P	Hz	SIN	3E-04	7E-06	9E-04	3E-05	1E-02	5E-02	3E-02	9E-03	3E+01	2E+00
		DL	5E-04	1E-05	1E-03	4E-05	3E-01	1E-01	1E-01	9E-03	1E+00	4E-01
δ_R	Degrees	SIN	40.4	39.9	43.6	39.6	24.4	31.0	26.4	27.2	21.8	20.1
		DL	--	--	--	--	--	--	--	--	--	--
δ_L	Degrees	SIN	0	0	0	0	7.6	10.1	-28.4	-10.6	16.1	12.4
		DL	0	0	0	0	24.3	37.8	-56.5	-22.2	24.7	19.4
S_R	--	SIN	0.221	0.207	0.220	0.210	0.181	0.269	0.214	0.246	0.349	0.307
		DL	0.114	0.105	0.122	0.105	0.111	0.143	0.120	0.118	0.142	0.126
S_L	--	SIN	0.221	0.207	0.220	0.210	1.230	0.868	0.812	1.750	0.776	1.127
		DL	0.114	0.105	0.122	0.105	0.273	0.257	0.237	0.985	0.898	1.553
$\|G^*_0\|$	GPa	SIN	3E+04	2E+05	3E+04	8E+04	2E+05	1E+05	3E+04	4E+04	2E+04	5E+04
		DL	3E-05	2E-04	4E-05	8E-05	2E-04	2E-04	2E-05	2E-05	3E-05	7E-05
SSRE	--	SIN	4.9	12.8	3.7	2.2	0.9	1.2	0.9	0.4	3.9	1.5
		DL	5.4	8.8	10.2	1.3	1.0	1.0	1.1	0.6	4.0	3.4
n	--	--	121	143	127	131	158	143	123	143	143	142

Figure 2. Apparent viscosity as a function of shear rate.

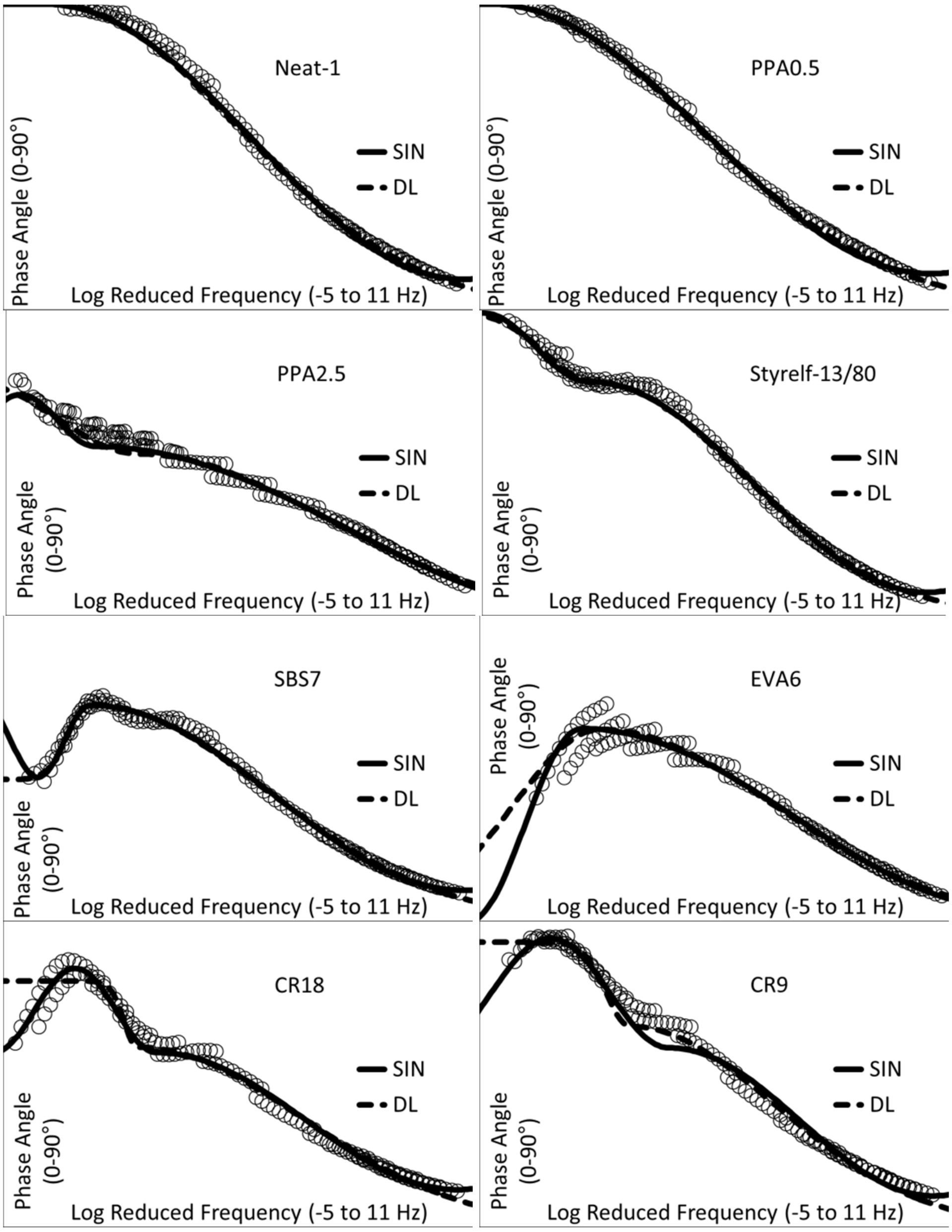


Fig. 2 presents the fitting results for 8 binders. Two neat binders are not shown for the sake of brevity. Neat binders usually show a general form of a simple logistic function. The phase angle plateau values for the neat binders δ_P approach 90° for both models in Tab. 1. The entire phase angle master curve range is modeled using the parameters in the right side of the plateau. This is also the case for PPA0.5 which is a slightly modified binder and does not show any irregularities in the master curves. It can be seen that SSRE values of both models for neat binders are not as good as for PMBs because, in fact, a smaller number of parameters has been used for them.

PPA2.5 depicts a phase angle master curve with low frequency dependency. It should be noted that it is difficult to visually determine an exact region in which the plateau occurs. Both models show δ_P values of about 50°. The visual inspection also confirms this selection. The phase angle master curve of Styrelf 13-80 consists of an intermediate plateau region at about 70°, a rise toward 90° at low frequencies, and a decline toward zero at high frequencies. Both DL and SIN models can precisely fit the master curves as shown in Fig. 2. The values of δ_P in Tab. 1 also confirms the plateau values in the figure.

Two PMBs, SBS7 and EVA6, follow similar patterns of phase angle master curves as shown in Fig. 2. Both models were able to properly fit the master curve for the two binders. They both show a peak at intermediate frequencies followed by a decline toward lower frequencies. Both models resulted in negative values for parameter δ_P which means that these two binders do not follow more viscous behavior at high testing temperatures.

CR18 and CR9 also follow similar patterns of phase angle master curves. On the left side of the plateau which is about 50° for CR18 (60° for CR9), a rise toward 80° (85° for CR9) happens and then a decline occurs. This is the situation which cannot be observed in other binders of this research. For CR18, the SIN model fits this master curve with the exact pattern, as described, while the DL model is not capable of modeling the last decline at low frequencies, as shown in Fig. 2. The main advantage of the SIN model over the DL model could be exactly in the case of binders which show such a behavior, like, e.g. CR18. In this case, the SSRE value of the SIN model is also less than that of the DL model. The low frequency return of the curve of CR9 is much lower, and the SSRE values for both models do not differ a lot. The Parameter δ_L is positive in both models, in spite of SBS7 and EVA6, because the phase angle does not immediately decline at the left side of the plateau.

There is another difference between the two models in the high frequency part of the phase angle master curves for most of the binders shown in Fig. 2. In this region, the DL phase angle model continuously declines as it approaches zero in the infinity, but the SIN model almost reaches its minimum value. This is an advantage of the DL model over the SIN model, in which the pure elastic behavior at infinitely high frequencies for bituminous materials prevails.

CONCLUSIONS

It is concluded that both SIN and DL models, introduced for estimating the plateau zone using Heaviside step function, are able to properly predict master curves of neat and modified binders in the widespread temperature and frequency ranges. The SIN model is able to predict more variations that usually occur at high temperatures in the phase angle master curves. On the other hand, the DL model is a simpler model which shows better high and low frequency asymptotic values and is more appropriate for extrapolation purposes, especially at low temperatures. The phase angle master curves of modified binders have been better estimated than those of neat binders for both models.

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