ALTERNATE SHIFT FACTOR RELATIONSHIP FOR DESCRIBING THE TEMPERATURE DEPENDENCY OF THE VISCO-ELASTIC BEHAVIOUR OF ASPHALT MATERIALS

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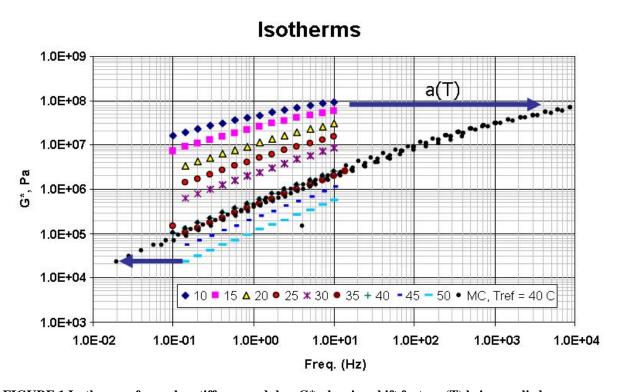
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ABSTRACT

Traditionally, various forms of shift factors such as Arrhenius, Williams-Landel-Ferry (WLF) and polynomials have been used with asphaltic materials. In addition, shift factors have also been estimated using binder viscosity parameters. Successful extrapolation of viscoelastic functions requires a robust form of shift factor-temperature relationship. This is important if trying to perform calculations at extremes of temperature found in practice. Rowe and Sharrock (1) presented data showing a preliminary analysis of complex modulus E* data of mixtures obtained from the MEPDG database which demonstrated that the Kaelble form of shift factor could more accurately describe the functional form of the shift factor than either the Arrhenius, WLF or polynomial fitting functions. However, the Kaelble shift function as originally described uses the same temperature as a reference temperature and as an inflection temperature. This creates a problem when trying to implement the function in a design method or when comparing materials at a given temperature. Since 2008 additional work has been conducted to investigate the use of this shift function to describe the properties of asphalt materials, particularly mixes and those that require a very wide range of property description (both above and below the glass transition or some other defining point). In particular, a modified form of the Kaelble function has been implemented in analysis software (2) thus making multiple calculations more rapid. We have performed additional analysis working with materials from MEPDG E* database and have observed that shifting works best with the Kaelble modification of the WLF equation. In addition, the same method has been applied to a wide variety of other asphalt materials.

INTRODUCTION

Master curves that describe the dependence of viscoelastic behavior on temperature provide a useful tool for engineers and scientists. If the curves can be represented in functional form they can more readily be applied to the analysis and design of asphalt pavements. For asphalt materials this has gained significantly more importance in the past 15 years since the adoption of the Strategic Highway Research Project (SHRP) through the Superpave program and the development of Mechanistic Empirical Pavement Design Guide (MEPDG). However, it may be noted that historically the application of master curves to asphalt materials originated as early as the 1950's (3). In the construction of master curves, isotherms of modulus tested at multiple temperatures are shifted by applying a multiplier (shift factor) to the frequency (or time) at which the measurement is taken so that the individual isotherms of stiffness combine to form a single smooth curve of frequency or time versus stiffness (the master curve). The amount of shift used at each particular temperature is analyzed to determine a shift factor - temperature relationship. This process is illustrated in FIGURE 1 and FIGURE 2. Through the use of the master curve and shift factor relationships it is possible to interpolate stiffness at an expanded range of frequencies and temperatures compared with those at which the data was collected. If functional forms are fitted to the shape of the master curve plot and to the shift factor relationship this interpolation becomes rapid and easy to apply in computer software. In addition, if a functional form with some thermodynamic basis is used then the resulting equations can be employed to extrapolate the data beyond the observed range of temperatures and frequencies.



 $FIGURE\ 1\ Is otherms\ of\ complex\ stiffness\ modulus,\ G^*,\ showing\ shift\ factor\ a(T)\ being\ applied$

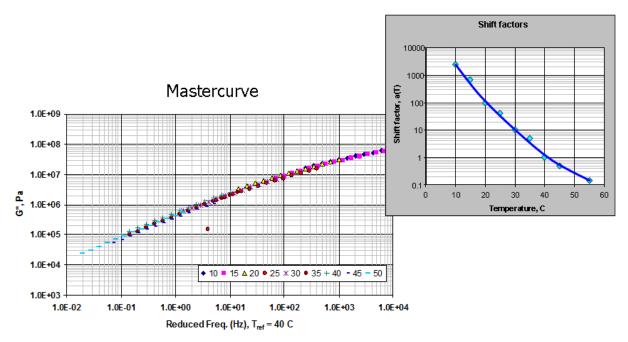


FIGURE 2 Master curve and shift factor plot

BACKGROUND

In the development of master curves two basic approaches can be considered, which we describe as "free shifting" and "constrained shifting." In the "free shifting" approach the shifts are determined from successive pairs of isotherms to form a smooth curve with the resulting shape of the master curve and shift factor functions being independently derived each time a master curve is constructed. In the constrained shifting approach an underlying model is used to force the master curve and/or shift factors to fit a predefined functional form. Particularly when the nature of the temperature / frequency susceptibility of a material is unknown, the "free shifting" type of analysis is much preferred because it enables the shape of the individual isotherms to strongly influence the shape of the master curve. The constrained analysis on the other hand can be very useful if the data is relatively noisy with correspondingly ill-defined isotherm shapes (provided that a particular functional form of master curve is known beforehand to be appropriate).

With asphalt materials historically both types of shifting have been employed but more recently in the USA constrained shifting has become the norm with data fitted typically to variations of the Christensen-Anderson (CA) (4) model for binder work whereas mixture data has been fitted to a symmetric sigmoid as originally developed by Witczak and reported by the Asphalt Institute (5).

For asphalt materials different shift factor forms have been used in the analysis methods. In the AASHTO PP42 (6) method an Arrhenius form of shift factors is adopted. This is generally consistent with much of the work conducted during SHRP from which it was concluded that the Arrhenius shift factor worked well at the colder temperatures. Anderson et al (7) discuss the definition of a "defining temperature" above and below which the WLF and Arrhenius functions are considered to apply. For the SHRP core asphalt the defining temperature ranges from -20.4 to 6.0° C (average values) depending upon the source, grade and aging condition of the asphalts. The conclusion made in this document is that the Arrhenius equation describes the behavior below the glass transition whilst the WLF equation provides an adequate representation above this temperature. This statement implies that the defining temperature thus obtained approximates a parameter similar in nature to a glass transition temperature but which could better be described as a transition to where Arrhenius or WLF better describes the shift factors generated. Consequently, from analysis of the methods considered for binder we note that two different shift factor temperature relations are used with a defining temperature, denoted as T_d , separating them.

Asphalt mixes have been the subject of master curve analysis for over forty years with much of the pioneering work being conducted by Professor Matt Witczak to develop the Witczak Modified E* equation (5). This method originally developed in the late 1970s/early 1980s has been updated for use with the MEPDG by analysis of additional mixtures to produce a more robust method (8). The shift factors employed with this method are considered in two manners. First a second order polynomial expression for the log of the shift factor is used in

the determination of the coefficients of the symmetric sigmoid used. This when combined with the coefficients for the master curve produces seven coefficients to describe the variation of E* with temperature and frequency (expressed as time within the MEPDG method). The second method adopted within the MEPDG framework for describing the shift factor is to make use of the log-log viscosity-temperature relationship (A+VTS relationship) and to replace the shift factor with a function which is dependent upon this equation.

The data collected in this manner allows for reasonable interpolation within the data set but concerns always exist with the use of extrapolation when polynomial fits are made to the data. However, it would appear that the extrapolation within the MEPDG software relies on the log-log viscosity versus temperature relationship. It is interesting to note that analysis of master curves generated using a A+VTS relationship produces a shift functional form close to a WLF type as shown in FIGURE 3 due to the fact that a log-log relationship versus temperature is used (9). While this method may provide a reasonable analysis route which enables dynamic data to be converted to viscosity information that was first used with the development of the MEPDG data it should be noted that the shape of the two shift factor formats used is different in the methods used.

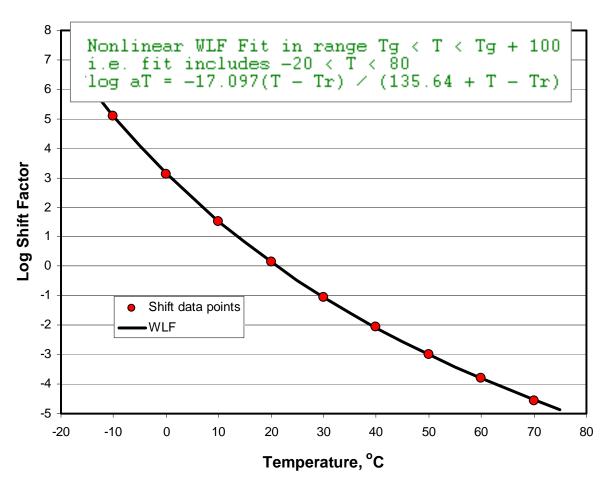


FIGURE 3 Shift factor developed from isotherms produced by assuming a log-log A+VTS relationship with fitted WLF relationship

The discussion given above notes that in both binder and mixture data the modeling of a shift factor -temperature relationship is complicated. In the binder the structural changes that occur around or close to the glass transition temperature result in recommendation by the SHRP researchers to use two types of shift factors around a defining temperature. In mixture work a second order polynomial was adopted as being descriptive of the shift factors since it can better describe the shape of the shift factor curve but then this has been dropped in the further analysis of this method with shift factors based upon a log-log type relationship which represents a different shape format for the shift factor curve.

DEVELOPMENT OF RHEOLOGICAL DATA

In the development of the data contained within this paper we have used the software developed by Rowe and Sharrock (1), to perform the shifting and develop the shift factors reported. The shift procedures for producing a master curve have been developed by various researchers. This software contains an implementation of Gordon and Shaw's (10) various computerized methods that can be applied for the characterization of visco-elastic materials and have formed the basis of the shifting techniques employed in the software. For the production of master curves these involve steps as follows:

- Determine an initial estimate of the shift for each pair of isotherms using a linear fit and checking against a modified WLF calculation using Ferry's original "universal" constants
- Iteratively refine the initial pairwise shifts using weighted least squares polynomial fits
- The order of the polynomial is an empirical function of the number of data points and the decades of time/frequency covered by the isotherm pair
- This gives optimized shift factors for each successive pair, which are summed from zero at the lowest temperature to obtain a distribution of shifts with temperature above the lowest
- The shift at T_{ref} is interpolated and subtracted from every temperature's shift factor, causing T_{ref} to become the origin of the shift factors

In the software implementation of Gordon and Shaw's methods a cubic spline fit has been used for the final shift to the reference temperature. The time-temperature shifting routine as described in Gordon and Shaw's book (10) and implemented in this work (1) can shift data sets which contain gaps (no overlap). The default gap discussed within Gordon and Shaw's software code is controlled by a "Wild" shift parameter with default gap of up to 4-decades. Our experience with this code shows that this method works very well with gaps in data. Vertical shifting is not considered with the exception that a density correction is applied as follows. The modulus data is normalized to account for the different density at the reference temperature compared to the temperature of measurement (Rouse (11)), thus:

$$G(T_R, t) = \frac{T_R}{T} \frac{\rho(T_R)}{\rho(T)} G\left(T, \frac{t}{a_T}\right)$$

SHIFT FACTOR EQUATIONS

Conventional forms for shift factors used for asphalt material analysis can be considered as Arrhenius, WLF or polynomial, as discussed earlier. The Arrhenius shift factor as applied to asphalt materials typically takes the form:

$$\log \mathbf{a}_T = a \left(\frac{1}{T} - \frac{1}{T_{ref}} \right)$$

We typically add a constant to this equation thus making a linearized relation between the shift factors and the reciprocal of temperature, as follows:

$$\log \mathbf{a}_T = a + b \left(\frac{1}{T} - \frac{1}{T_{ref}} \right)$$

The WLF equation and second order polynomial (as used in the Witczak approach) are as follows:

$$\log a_T = -\frac{C_1(T - T_g)}{C_2 + (T - T_g)}$$
 (WLF equation)

$$\log a_T = aT^2 + bT + c$$
 (second order polynomial)

ANALYSIS

The use of these shift factors for a heavily SBS polymer modified binder (12) referenced as 125-35-0 is shown in FIGURE 3. This data analysis combines bending beam rheometer (BBR) data with that obtained from the dynamic shear rheometer (DSR).

The data is combined by converting the BBR stiffness to storage and loss moduli which involves several analysis steps. First the BBR apparent stiffness master curve is fitted with the CA, CAS and CAM functional models (Rowe et al. (13)) within the software and a determination is made as to which model produces the lowest error. Christensen-Anderson (CA) model (3) is defined as follows:

$$S(\xi) = S_{glassy} [1 + (\xi / \lambda)^{\beta}]^{-1/\beta}$$

In this model S_{glassy} is a constant $(3*10^3 \text{ MPa})$ and λ and β are fitted. This type of model enables the low temperature properties of asphalt binders to be modeled with reasonable accuracy. However, it should be noted that models of this form should not be applied to the total binder master curve since no change in the slope of [d log $S(\xi)$ /d log (ξ)] is possible at higher temperatures (14). This is because at long times $S(\xi) \approx S_{glassy} (\xi/\lambda)^{-1}$ and at short loading times $S(\xi) \approx S_{glassy}$. The CA model can be considered to be a restricted form of a more general equation:

$$S(\xi) = S_{glassy} [1 + (\xi/\lambda)^{\beta}]^{-\kappa/\beta}$$
 where, S_{glassy} , λ , β and κ are fitted.

With this equation four parameters are fitted. However, while this method works well with data collected over a wide time range the limited time range data from the BBR test is best fitted using only a three-parameter model. Typically BBR data covers only two decades of stiffness and loading time. Consequently, two three parameter models can be defined which are referenced as CAM or CAS. In the CAM model (15) S_{glassy} is taken as a constant (3*10³ MPa) and λ , β and κ are fitted whereas in the CAS model (Christensen-Anderson-Sharrock) S_{glassy} , λ and β are fitted and κ is assumed as -1 (13).

This allows the retardation master curve to be interpolated smoothly so that the Hopkins and Hamming (16) algorithm can be used to convert the retardation master curve to the relaxation modulus E(t). The resulting E(t) data is then refitted to a CAM model using the Glassy modulus determined from the previous fitting. This gives a functional form which describes the E(t) fit. A discrete spectrum is calculated for the E(t) fitted function using the method described by Baumgaertel and Winter (17) and the reciprocal of the observed times are then substituted into the function to estimate the E', E" data points. The data points are shifted using the original shift values obtained along with a reverse density correction to obtain dynamic isotherms corresponding to the original data. Finally, extensional data is then output to a data file converting to G in the process with an optional Poisson's ratio (we have used the default value of 0.5 in this work). The combination of both BBR and DSR data enables a much greater range of temperatures to be studied than could be easily obtainable from DSR testing alone. In this analysis we have included isotherms that cover a temperature range from -24 to 135°C.

In this example the polynomial form of the shift factor work well, but it can be clearly seen that at the colder temperatures the WLF starts to depart from the determined shift factors whereas the Arrhenius does not fit the data at the extremes on mid sections. It should be noted that the rules governing how the WLF is fitted to a data set effect this aspect. The WLF is fitted to data that lies within the temperature range of the glass transition temperature (Tg) to $Tg+100^{\circ}C$. In addition, a minimum of five shifted isotherms are required to perform the fit (10). For this data set a Tg of -20 was assumed (12).

This data set clearly shows differences that are obtained with the different methods. The use of an appropriate Tg is important if trying to make use of the WLF method. Typically, the departure in the WLF from the measured data occurs at or close to the defining temperature as defined by Anderson et al. (7) or close to the glass transition temperature.

While in this example the WLF or Arrhenius gives a reasonable solution this tends not to be the case when mixtures and or filled systems are analyzed. For example a typical mixture result is given in FIGURE 5 which shows that with five isotherms of stiffness a poor fit to the Arrhenius function is obtained. It should be observed that although in this case the reported r-squared regression coefficient is still quite high (r^2 =0.9202), the fit is visibly very poor and this points to the dangers in using this particular error parameter alone. Inspecting the data it is clearly seen that a curved line is required to fit the data rather that the straight line represented by the Arrhenius type equation.

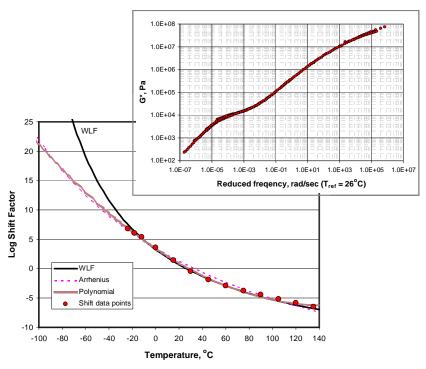


FIGURE 4 Three types of shift factor as applied to SBS modified binder 125-30-0 (Tg -20 $^{\circ}$ C), master curve shown in upper right corner at reference temperature of 26 $^{\circ}$ C

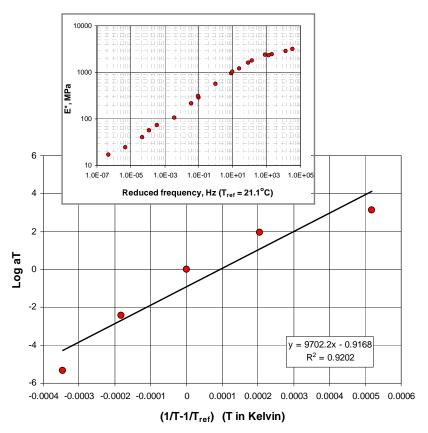


FIGURE 5 Arrhenius shift factor applied to Maryland Port Authority hot-mix asphalt with AC20 binder taken from MEPDG database, master curve shown at reference temperature of $21.1^{\circ}\mathrm{C}$

When encountering data of this type we sought to investigate different shift factor relationships to determine if a more appropriate method existed which maintained a good physical meaning and interpretation of the data. At this time we were conducting significant work with various asphalt materials such as roofing mastics, asphaltic concretes, and heavily polymer modified materials to name but a few. The data analysis showed that a symmetric sigmoid model could not be adopted for any of the materials with accuracy and that the shift factor relationships in normal use were not appropriate. After some research work of available methods we noted that a generalized logistic function better described the shape of the master curve of the filled systems, including asphalt concrete mixtures, as represented by the following equation:

$$\log E^* = \delta + \frac{\alpha}{\left[1 + \lambda e^{(\beta + \gamma(\log \omega))}\right]^{1/\lambda}}$$

This information has been published in RILEM in 2009 (2) and also in that paper we explored a preliminary use of a description of the shift factors using a modification to the WLF equation as proposed by Kaelble (18). The modification to the WLF relationship is the inclusion of a magnitude term in the denominator of the right hand side fraction:

$$\log a_T = -\frac{C_1(T - T_d)}{C_2 + |T - T_d|}$$

The use of the magnitude term in the relationship introduces an inflection point in the curve at the temperature T_d , and changes the overall shape from hyperbolic to sigmoidal. It will be observed though, that above the inflection point temperature the curve is identical to a WLF curve. Below the inflection point temperature, the WLF log a_T increases rapidly, approaching a vertical asymptote at $T = T_d - C_2$. The Kaelble log a_T instead slowly approaches a horizontal asymptote. However, the equation in this form is difficult to apply to data since the form of the equation implies that the defining temperature and the reference temperature are the same. This difficulty can be overcome by the addition of a constant term separating T_d from T_D as follows:

$$\log a_T = -C_1 \left(\frac{T - T_d}{C_2 + |T - T_d|} - \frac{T_r - T_d}{C_2 + |T_r - T_d|} \right)$$

The software was then subsequently extended with this modification, enabling rapid processing of data. In the example discussed above the root mean square error (rms) (%) reduces from 0.8872 (Arrhenius) to 0.2367% (Kaelble) with a significantly better fit as shown in FIGURE 6 when this method is applied. It should be noted that the software implementation did not permit the computation of the WLF curve since the number of data points requirement was not met. However, we obtained a fit using the SOLVER function in EXCEL (ignoring the normal limiting rules) which is difficult and poor compared to the data points due to the shape function of the WLF with the log a_T increasing very rapidly at low temperatures. It can clearly be seen by inspection that WLF does not fit this data set. Also shown in the plot is the polynomial fit, which works well in this instance.

In FIGURE 7 we have compared the modified shift factor to the three standard shift factor types discussed earlier. On close examination it can be observed that this form of shifting better describes the data. The rms error calculated for WLF, Arrhenius and modified Kaelble methods are 0.3329, 0.4669 and 0.0854 respectively. It should be noted that the error term is a root mean square in decades of a log shift. Consequently, a reduction in error by a factor of 4 in this parameter can be regarded a significant improvement. It should be further noted that the use of r^2 parameters, which is often employed with master curves, has very limited meaning due to the large range in the magnitude of the numbers.

In FIGURE 8 we have shown the analysis applied to binders contained within the ALF experiments conducted by the FHWA (19). In this case little difference can be observed between the WLF and Kaelble fits when a glass transition temperature of -20 has been adopted for the analysis of the AC5 binder. The root mean square errors computed are 0.3352, 0.3918 and 0.3339 for the WLF, Arrhenius and Kaelble methods respectively. The inflection point produced in the analysis is -24.0 $^{\circ}$ C which is very close to the Tg estimated in the analysis. This analysis demonstrates how the T_d produced by the Kaelble analysis produces a numerical value very close to that of the defining temperature as prescribed in the SHRP work conducted by Christensen and Anderson (4).

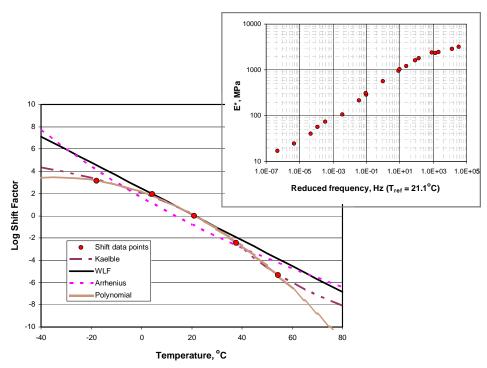


FIGURE 6 Modified shift factor applied to Maryland Port Authority hot-mix asphalt with AC20 binder - taken from MEPDG database, master curve shown in upper right corner at reference temperature of 21.1°C

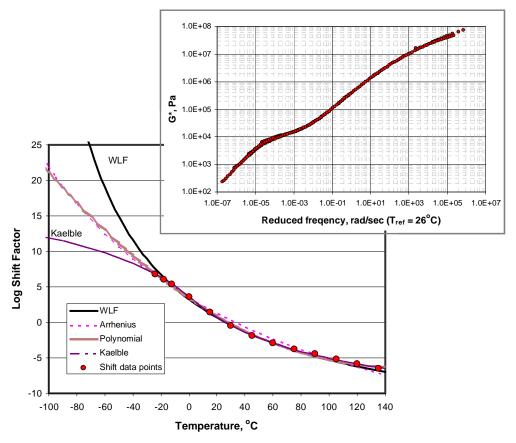


FIGURE 7 Modified Kaelble compared to Arrhenius, WLF and Polynomial shift factors as applied to SBS modified binder 125-30-0, master curve shown in upper right corner at reference temperature of 26° C

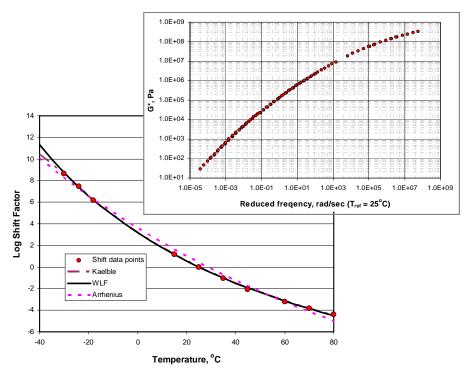


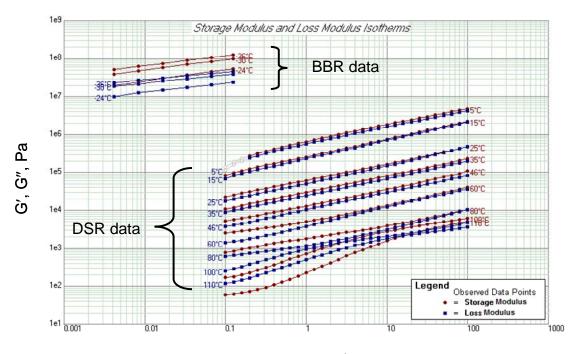
FIGURE 8 Modified Kaelble compared to Arrhenius and WLF and shift factors as applied to AC-5 binder from ALF study, master curve shown in upper right corner at reference temperature of 25°C

DISCUSION

The analysis described above has been applied to a large number of modified materials using the software implementation. A complete example of an analysis is shown below. In this example a concentrated thermoplastic polymer additive Rosphalt (20) has been added to a sample of the binder to produce a series of stiffness isotherms as shown in FIGURE 9. This data includes converted BBR data using the method described earlier in this paper. The G* data has been shifted as shown in FIGURE 10 and the shift factor determinations made. The gap in the data does not present any problems with the shifting techniques described earlier in this paper and given in Gordon and Shaw's (10) method which can typically consider a gap as large as 4-decades. Again by inspection we can observe that the Arrhenius representation of the data does not describe the functional form well. The WLF in this case does a better job of producing a functional fit but clearly the best fit is obtained from the Kaelble modified format. The relative error of the Kaelble fit compared to the WLF is less than one third. This type of result is typical of data sets involving a wide range of temperatures and frequencies.

The robustness of this type of analysis is that it can be applied to many materials and situations. For example, while not an asphalt material, we have applied this analysis to polystyrene which forms part of the basic data set used in evaluation of software changes, as shown in FIGURE 11. The difference in this analysis is produced by the limitation of the shifting function when WLF shifts are applied in accordance with Gordon and Shaw's method. If this convention is ignored and the $Tg+100^{\circ}C$ limited removed, the WLF shift will be identical to that produced by the Kaelble analysis. It should be noted that Kaelble (18) produced a similar approach and in his book commented that the modification to the WLF is based upon the extension of the free volume concept to temperatures well below the glass transition temperature.

As discussed earlier, we have noted that in the analysis of asphalt binders the value obtained for the defining temperature in the modified Kaelble equation can be close to that represented by T_d in the work of Anderson et al. (7). For asphalt mixtures this temperature tends to be higher (in the general range 10 to 35°C) but is dependent upon the data set being analyzed. In some cases, when low temperature data is of poor quality, an inflection point cannot be readily obtained and the resulting best fit defaults to the WLF form (which is a special case of the more generalized modified Kaelble fit) but with the inflection temperature assigned to the lowest temperature isotherm. The use of a single functional form to describe the full range of time-temperature supposition would appear to offer advantages to engineers attempting to model pavement structures making use of extrapolation functions.



Frequency, rad/sec

FIGURE 9 PG64-22 modified with 20% Polymer – isotherms including converted BBR data

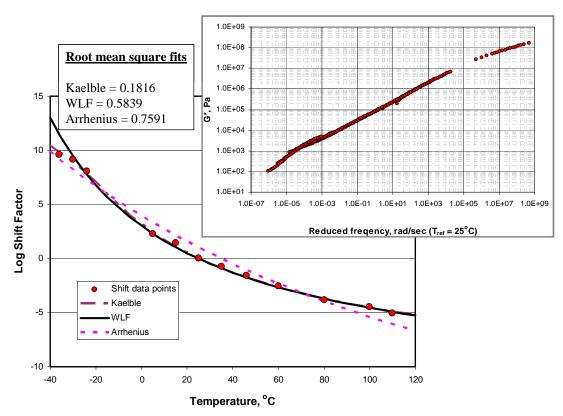


FIGURE 10 PG64-22 modified with 20% Polymer – Shift factors with (Kaelble, WLF and Arrhenius formats with relative errors) and G^* master curve including converted BBR data at a reference temperature of $25^{\circ}C$

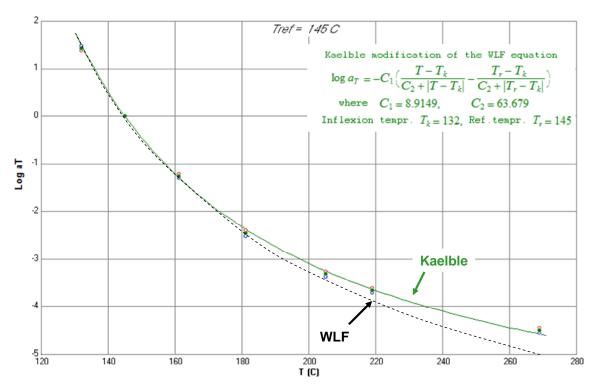


FIGURE 11 Modified Kaelble applied to polystyrene material

SUMMARY

An alternative form for describing the shift factor: temperature relations that exist when performing time-temperature superposition has been evaluated and implemented in computer software. The initial stage of work involved a minor modification to the Kaelble equation which in turn is a modification of the WLF equation. The equation offers a single functional form to describe the shift factors of asphalt materials from a high to low pavement temperature that does not rely upon polynomial fitting and provides a stable relationship. The relationship has been applied with success to a wide range of materials in the evaluation including those used for polymer modified binders for roofing and bridge deck applications, conventional asphalt and hot mix asphalt (with modified or conventional asphalt binder).

While this method has been in existence for approximately 25-years it has not been previously applied to asphalt materials. The method offers significant advantageous but some additional study should be performed to check the robustness of the procedure with mix and binders samples. With regard to application of this method we would note the following:

- ullet Asphalt binders the T_d parameter as defined by Kaelble has a very similar meaning to the defining temperature parameter as prescribed by the Christensen-Anderson model but avoids the need for two different functional forms to describe the shift factor relations above and below the glass transition temperature.
- Filled binders/modified binders the Kaelble provides a better functional form than either the WLF or Arrhenius formats.
- Asphalt mixtures the Kaelble provides a better fit and functional form than the polynomial or the WLF
 equation. In the case of the application of the A+VTS method a WLF fit is effectively applied to the data
 which can often be considered questionable.

The Kaelble formulation of the shift factor provides an elegant method for extrapolating and interpolating data when used with models such as the Christensen-Anderson (or CAM and CAS formats) for the of binder data, and with the sigmoid models (Witczak or Richards) for mix data. We recommend that the analysis techniques be studied further to demonstrate the effectiveness of the error reduction in building master curves.

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