Comparison between Viscoelastic Mechanical and Chemical Kinetic Aging Model of Asphalt Concrete

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Abstract. The objective of this study was to fully characterize and validate the mechanical-based aging model parameters and compared with the chemical kinetic model. This study found that level of aging applied to both asphalt binder and asphalt concrete showed differences, as verified by all of the p-values from regression analysis test, were greater than 0.05. The aging variable value decreased with the aggregate involved. The aging history dependent (k2) could be simulated the actual aging behavior as a polynomial curve. The Pearson correlation test showed that there is a strong relationship between the carbonyl content and the aging model variable.

Keywords: Aging, Asphalt concrete, Asphalt binder, Carbonyl content, Mechanistic model, Chemical kinetic model

1. Introduction

Asphalt aging is one of the significant factors influencing the performance of asphalt pavements [1]. Aging could result in two major effects: first, load-bearing capacity and permanent deformation resistance may increase due to the pavement becoming stiffer; and second, it can lead to the formation of cracks in the pavement due to reduced pavement flexibility [2].

The most fundamental issue affecting binder aging in pavements is the binder oxidation chemistry. The cause of the oxidative aging was due to the formation of carbonyl compound that transform the light molecular weight compound into large molecular weight within the asphalt binder [3]. Asphalt’s carbonyl compound and formation rate depend on the temperature and oxygen pressure [4]. The chemical reactions have resulted in enhanced stiffness, viscosity, and brittleness of asphalt mixture. It will cause more vulnerability to fatigue cracking.

Another way to characterize binder oxidative aging is through mechanical based modeling of asphalt material behavior [5, 6]. Michalica et al. (2008) modeled the impact of aging using loading time and creep compliance as a function of time; Masad et al. (2008) modeled the impact of aging by introducing aging time shift factor in the nonlinear viscoelastic model. On the other hand, a mechanical based oxidative aging modelling by introducing oxidative aging variable which is conceptually model the carbonyl formation rate of asphalt materials during the long-term aging process [7].
Numerous studies conducted to understand constitutive modelling of oxidative aging and its effect on the mechanical response of asphalt concrete. There are three methods can use to determine relationship between oxidative aging and mechanical response, specifically: aging-time shift factor, creep compliance/relaxation moduli, and continuum-based modeling of asphalt concrete [8]. Among the three methods that are very widely used now is continuum-based modeling of asphalt concrete. Because this method can show physically-based oxidative aging constitutive relationship coupled with the viscoelastic response of asphalt concrete due to the process of aging.

Although the constitutive model proposed by Al-Rub et al. (2013) incorporated an aging variable to capture the effect of long-term aging on asphalt concrete’s viscoelastic response, the fully characterization and validation of such aging model is not available yet. Several studies have attempted to characterize the mechanical-based aging model parameters, but their work has only limited on characterizing the internal aging variable and temperature-dependent parameter [8, 9]. Therefore, it is necessary to fully characterize and validate the mechanical-based aging model parameters and compared with the chemical kinetic model.

The main objective of this study was to characterize and validate the mechanical-based aging model parameters and compared with the chemical kinetic model. The asphalt concrete and the asphalt binder aged to five different level of aging. Dynamic modulus test, linear frequency sweep test was used to characterize the mechanical response of asphalt concrete and binder respectively. Fourier transform infrared spectroscopy (FTIR) test was employed to investigate the change of chemical compound in the asphalt binder [10]. The result then used to characterize and validate the linear viscoelastic mechanical-based aging model parameters.

2. Material and Methods

Plant mixed polymer modified asphalt mixes were obtained from asphalt plant and materials were aged to various level in the laboratory. Mechanical test of asphalt concrete, as well as rheological and chemical tests of asphalt binder, have been conducted in the study to characterize the evolution of viscoelastic and chemical kinetic properties of the material. The aging variable models derived from the viscoelastic test result were compared to the carbonyl formation rate model from the chemical kinetic tests.

2.1 Material

A plant mix 19 mm polymer modified coarse well grade mix was used in this study. The asphalt binder used is a Polymer Modified Asphalt Type III (PMAIII) binder. Table 1 shows the viscosity of asphalt binder in various temperature. Also from the test results obtained that SG = 1.033 and \( \rho_{\text{binder}} = 1033 \text{ kg/cm}^3 \).

| Temp (°C) | Rotor Model | Rotation Speed (rpm) | Torque Value (%) | Viscosity CP Poise |
|-----------|-------------|----------------------|------------------|-------------------|
| 160       | SC4-18      | 75                   | 20.2             | 689 6.89          |
| 180       |             | 9.3                  | 317              | 3.17              |
| 200       |             | 4.9                  | 167              | 1.67              |

Aggregate used in the study was crushed river gravel with 19 mm maximum aggregate size. The physical properties tests of aggregate have been conducted as seen in Table 2. The designs result were found to be suitable for Taiwan specifications as illustrated in Table 3.
### Table 2. Physical Properties of the Aggregate

| Test Item                                               | Test Value | Specification Value | Test Standard                  |
|---------------------------------------------------------|------------|---------------------|--------------------------------|
| Aggregate Abrasion Value at 500 revolutions (%)         | 21         | ≤ 40                | CNS 490 (2009) ASTMC 131       |
| Fractured Particle in Course Aggregate at one face (%)  | 92         | ≥ 90                | CNS 15312 (2010)               |
| Fractured Particle in Course Aggregate at one face (%)  | 90         | ≥ 75                | ASTM D5821-13                  |
| Sand Equivalent Value                                   | 69         | ≥ 45                | CNS 15346 (2010) ASTM D2419-14 |
| Ratio of Width to Thickness of Aggregate Retained In    | 4.7        | ≤ 15                | CNS 15171 (2008)               |
| #4 Larger Than 3 Times of The Ratio (%)                 |            |                     | ASTM D4791-10                  |
| Soundness of Aggregates (%)                            | 1.2        | -                   | CNS 1167 (1995)                |

### Table 3. Asphalt Mixture Properties Used in the Mixture Design

| Term                          | Experimental Value | Standard Value |
|-------------------------------|--------------------|----------------|
| Binder Content (%)            | 4.7                | -              |
| Maximum Specific Gravity (Gmm)| 2.467              | -              |
| Bulk Specific Gravity (Gmb)   | 2.379              | -              |
| Density (kg/m³)               | 2379               | -              |
| Stability (lbf)               | 5120               | ≥2700 lbf      |
| Air Void (%)                  | 4                  | 3–5            |
| Voids in Mineral Aggregate (VMA, %) | 13.1               | ≥12.7          |
| Voids Filled with Asphalt (VFA, %) |                |                |

#### 2.2 Methods

The BRRC aging method was followed for both binder and asphalt mixes to simulate the long-term aging of materials [11]. Both binder and loose mixes were conditioned in temperature (60 and 95 °C) and time (5 and 10 days) to simulate different levels of aging.

The prony series model was used to fit the dynamic modulus test results to obtain the viscoelastic parameter of asphalt concrete. The SGC specimen was cored and cut into a cylindrical specimen with 101.6 mm in diameter and 152.4 mm in height. The test was conducted at four temperatures (15°C, 25°C, 40°C, 55°C) and six loading frequencies (25, 10, 5, 1, 0.5, 0.1 Hz) at each temperature.

The dynamic shear rheometer (DSR) was employed for frequency sweep test of asphalt binder to characterize the rheological behavior of asphalt binder at intermediate and high service temperature. The frequency sweep tests were performed over a range of temperature 22°C to 34°C representing the intermediate service temperatures and 40°C to 76°C to represent high service temperature with 6°C intervals.

In FTIR test, a sample subjected to infrared light of varying frequencies to determine how well it absorbs the light at each wavelength. A Thermo-Nicolet 6700 FTIR spectrometer was employed to produce the spectra. The background spectra and sample spectra are scanned using wavelength range of 400 to 4000 cm⁻¹ and a scan resolution of 4 cm⁻¹. Data analysis then produced using the Omnic 8 Software.

### 3. Characterization of Asphalt Aging Material

#### 3.1 Viscoelastic Model Parameters for Unaged Materials

This section describes an organized procedure to obtain time-temperature shift factor and the linear viscoelastic model parameters and aging variables. Results obtained from the dynamic modulus test
were dynamic modulus \((E^*)\) and phase angle \((\theta)\) at different frequencies and temperatures. Then, complex compliance \((D^*)\) calculated as Eq. (1).

\[
|D^*| = \frac{1}{|E^*|} \tag{1}
\]

The sigmoidal function regarding complex compliance then utilized to fit experimental data and obtain the time-temperature shift factor as shown in Eq.(2)- (3) respectively. Horizontal shifting then applied to create a dynamic modulus master curve.

\[
|D^*| = \delta + \frac{\alpha}{1+\exp[\beta + \gamma \log(\omega_r)]} \tag{2}
\]

Where, \(D^*\) is complex creep compliance, \(\delta\) is maximum value of the dynamic compliance, \(\delta + \alpha\) = minimum value of the dynamic compliance, \(\beta + \gamma\) = shape parameters, and \(\omega_r\) = reduced frequency.

\[
a_T = \frac{\omega_r}{\omega} \tag{3}
\]

Where \(\omega\) is the frequency and \(a_T\) is the time-temperature shift factor.

This study employed Schapery’s integral form to calculate the strain response due to an applied stress \[12\]. In general, the total strain of an asphalt mixture decoupled into storage compliance \((D'_\text{exp})\) and loss compliance \((D''_\text{exp})\). Both values computed by utilizing the creep compliance and phase angle. The storage and loss compliance \((D'_\text{model} \text{ and } D''_\text{model})\) that related to Prony series then calculated by minimizing the error between the experimental and model data in Eq. (4)- (5). The error function used is shown in Eq. (6). The Prony series then formulated to represent the creep compliance \(\Delta D\) as a function of time. It should be mentioned that the result of the Linear Frequency Sweep (LFS) test of binder (complex modulus \(G^*\) and phase angle \(\delta\)) also followed the same procedures to get the time-temperature shift factors and linear viscoelastic parameters.

\[
D'_\text{exp} = |D^*| \cos \theta; \quad D''_\text{exp} = |D^*| \sin \theta, \tag{4}
\]

\[
D'_\text{model} = D_0 + \sum_{i=1}^{N} \frac{D_i}{1+\omega^2\tau_i^2}; \quad D''_\text{model} = \sum_{i=1}^{N} \frac{D_i\omega\tau_i}{1+\omega^2\tau_i^2} \tag{5}
\]

\[
\text{error} = \frac{1}{M} \sum_{\tau=1}^{N} \left[\left(\frac{D'_{\text{fit}}}{D'_{\text{exp}}} - 1\right)\right]^2 + \left[\left(\frac{D''_{\text{fit}}}{D''_{\text{exp}}} - 1\right)\right]^2 \tag{6}
\]

Where \(N\) is the number of prony series coefficient that could be assumed between 5 to 9, \(M\) is the total data set, \(\tau\) is 1/\(\lambda\), \(D_n\) and \(\lambda_n\) are the nth coefficients of Prony series and retardation time respectively.

3.2 Viscoelastic Model Parameters for Aged Materials

The term of the aging variable \((A)\) was referred as the oxidative aging variable that increases as the level of aging increases. To identify the aging variable \((A)\), the aged asphalt mixture followed the same procedure to get time-temperature shift factors \((a_T)\), storage compliance \((D'_\text{exp})\) and loss compliance \((D''_\text{exp})\). To relate the aged and unaged viscoelastic properties, the loss and storage compliance model \((D'_\text{model} \text{ and } D''_\text{model})\) of the unaged Eq. (5) was recalled and incorporated with Eq.(7), so that a new relationship \[7\] was created as shown in Eq.(8).

\[
D_n^A = (1-A)^k_2 D_n; \quad \lambda_n^A = (1-A)^k_2 \lambda_n, \tag{7}
\]

\[
D'_\text{model}^A = D_0 + \sum_{i=1}^{N} \frac{(1-A)^k_2 D_{i0}(1-A)^k_2 \tau_i}{1+\omega^2(1-A)^k_2 \tau_i^2}; \quad D''_\text{model}^A = \sum_{i=1}^{N} \frac{(1-A)^k_2 D_{i0}(1-A)^k_2 \omega \tau_i}{1+\omega^2(1-A)^k_2 \tau_i^2} \tag{8}
\]

The new model certainly indicated a relationship between both compliances and retardation times of aged material altered by the aging level. Since the test covered all of the variable needed except \(A\), the use of Microsoft Excel solver was required to minimize the error Eq. (6) to obtain the aging variable \(A\).
3.3 Chemical Properties of Asphalt Binder

In this study, the test conducted to evaluate the evolution of carbonyl formation groups’ chemical composition in a different level of aging. To calculate infrared absorption ratio of the carbonyl content and corresponding functional groups in the asphalt binders following to Yao’s method [13]. The denominator in this equation is between wavenumber 600 and 2000 cm\(^{-1}\) where the aging components in the asphalt binder formerly concentrate.

3.4 Statistical Analysis

To verify the effect of different aging levels on both materials, mixture, and binder, the statistical analysis was conducted for the dynamic and complex modulus test result. The regression analysis was then performed to test the data with 95% confidence level. To evaluate significance, p value lower than 0.05 determined as significant which means between two sets of data measured, it has significantly different. The same observation procedure also conducted to complex modulus data from binder test.

4. Results and Discussion

4.1 Mechanical Properties of Asphalt Mixture

4.1.1 Viscoelastic Properties of Asphalt Concrete

Using time-temperature superposition principle, Figure 1 shows the dynamic modulus master curve of asphalt concrete in a different level of aging. The results demonstrated that the dynamic modulus interval between different aging level was greater in the low frequency rather than in high frequency. In addition, to verify the difference between each dynamic modulus test result at different aging level (AL), regression test was conducted at confidence level 95% and p-value set to evaluate the significance. The results showed that all of the test data had significantly different since their p-value were all lower than 0.05. Next, the dynamic modulus master curve was further fitted with prony series to determine viscoelastic model parameters. Using iteration method to minimizing the error between the experimental data and the prony series model, Prony series parameters can be obtained. The fact that using more prony series coefficients made the error between the experimental data and the prony series model will come to smaller number.

![Figure 1. Dynamic modulus master curves at 25 °c for asphalt concrete at various aging level](image-url)

4.1.2 Rheological Properties of Asphalt Concrete

The dynamic shear modulus master curve of asphalt binder also showed a similar trend to the dynamic modulus master curve of asphalt concrete where the interval between different aging level was greater in the low frequency rather than in high frequency as shown in Figure 2. The result also showed that when the oxygen pressure reduced to 0.8 atm, the dynamic shear modus master curves at all aging level showed lower values than that in the 1 atm oxygen condition.
To verify the difference between each dynamic shear modulus test result at the different aging level and oxygen pressure, regression tests were conducted at confidence level 95% and p-value set to evaluate the significance. The analysis showed that both within same atmospheric pressure and between different atmospheres, there are significant differences between various dynamic modulus master curves. The result of all the tests showed p-value smaller than 0.05. It is noteworthy that the procedure to get time-temperature shift factors, master curves and Prony series also followed the same procedure as derived for asphalt concrete.

4.1.3 Calculation of Aging (A)
Once the dynamic modulus master curves and viscoelastic model parameters of asphalt concrete/binder were obtained, the internal state aging variable (A) can be determined using Eq. (4)-(8). Figure 3 showed the result of aging variable A from asphalt concrete and Figure 4 showed the results from asphalt binder. For instance, in Figure 3, a minimal change of aging variable when asphalt concrete was aged under 60°C. The aging variable changed from 0 (aging level I) to 0.005 (aging level II). In the other hand, when the materials were aged at 95°C, a much higher rate of aging was observed. The aging variable was 0.101 at aging level III and 0.257 at aging level IV. Figure 4 showed that asphalt binder aging variable changed from 0 (aging level I) to 0.214 (aging level I) to 0.271 (aging level II). Otherwise, when the materials were aged at 95°C, a much higher rate of aging was observed. The aging variable (A) has a limitation range value between 0 and 1 where A=0 stands for unaged material and A=1 means materials have been fully aged so that the carbonyl formation was completely
stopped [7]. Practically, the aging variable will never reach 1.0 due to the long-term aging is very slow through the service life of the asphalt pavement [6].

From the results of the aging variable of asphalt concrete and binder, different trends were observed. The aging variable from the asphalt concrete has lower values than that of the asphalt binder. Also, the increasing rate of an aging variable was higher in the asphalt binder than that in the asphalt concrete.

4.2 Chemical Properties of Asphalt Mixture

The impact of oxidative aging could be determined by evaluating the changes regarding carbonyl and two functional carbonyl groups those are carboxylic acids and ketones that have peak integral value of IR absorbance at 1670-1820 cm\(^{-1}\), 1700-1725 cm\(^{-1}\), 1665-1685 and 1680-1700 cm\(^{-1}\) respectively.

Figure 5 shows the FTIR test result at all aging levels. Figure 6 to Figure 8 shows the ratio calculations.
for different bonds with additional 1 atm oxygen pressure. For instance, in Figure 6, a minimal change of aging variable when asphalt binder was aged at 60°C. The aging variable changed from 0.0386 (aging level 0) to 0.044 (aging level I) to 0.048 (aging level II). In the other hand, when the materials were aged at 95°C, a much higher rate of aging was observed. The figures indicate that the carbonyl increased due to the oxidative aging. Both Carboxylic Acids and Ketones as the main product of oxidation. Yao et al. (2015) indicated that the Carboxylic Acids and Ketones are the main components that correlate well with pavement rutting and fatigue resistance. The chemical reactions between the oxygen and the resins as well as the asphaltenes during the aging of asphalt that would produce the aromatic hydrocarbons and the polar functional group that leads to increasing of carboxylic acid and ketones bonds within the asphalt binder.

**Figure 5.** IR spectra of asphalt binder at different aging level

**Figure 6.** Ratio of carbonyl within the asphalt binder

**Figure 7.** Ratio of carboxylic acid within the asphalt binder
4.3 Comparison of Mechanical and Chemical Aging Variable
In this study, to characterize those parameters, two temperatures (60°C and 95°C), and two different days (5 and 10 days) for asphalt mixture and additional two different oxygen content for asphalt binder were applied to be the aging condition in different aging levels. From the aged asphalt binder, with the obtained aging variable values as seen in Figure 4, the \( k_1, k_2, k_3 \) and \( \Gamma_0 \) parameters achieved by minimizing the error between aging variable from experiment test result and model prediction. The summary of the model parameters for asphalt binder was presented in Table 4. To capture the evolution of time rate of aging parameter \( A \) at different relevancies such as temperature, oxygen content and aging variable, the rate of aging and fitted model parameters were used. The model prediction of Aging variable then calculated and was shown in Figure 9. The aging variable had two rates, which was fast rate at the beginning of the aging period and slower rate by the end of the aging period.

Table 4. Aging Model Parameters for the Asphalt Binder

| \( \Gamma_0 \) | \( A_0 \) | \( k_1 \) | \( k_2 \) | \( k_3 \) | \( T_0 \) |
|---|---|---|---|---|---|
| 0.0269 | 0 | 8.57 | 8.64 | 1.60 | 25 |

Figure 9. Aging variable model

Besides, using the same procedures of asphalt binder to determine the model parameters and using the \( k_1 \) value from asphalt binder as the \( k_1 \) value for the asphalt concrete aging model, the rest of the aging model parameters were calculated and it was shown in Table 5. Figure 10 then showed the evolution of time rate of aging parameter (A) at different relevancies and Figure 11 described the prediction model of aging variable A for asphalt concrete.
### Table 5. Aging Model Parameters for the Asphalt Concrete

| \( \Gamma^o \) | \( A_0 \) | \( k_1 \) | \( k_2 \) | \( k_3 \) | \( T_0 \) |
|--------------|--------|--------|--------|--------|--------|
| 0.0013       | 0      | 8.57   | 5.98   | 0.68   | 25     |

**Figure 10.** Rate of aging parameters of asphalt concrete, (a) at different temperature, (b) at different oxygen content, (c) at different aging variable

Aging Variable: \( k_2 = 8.570 \)
Figure 10 (a) showed that temperature change has a contribution to the rate of aging variable, especially in higher temperature. It has the same objection with the previous study that said asphalt as a temperature-dependent material and by this study, it was validated that the aging model could predict the aging rate in term of temperature change very well. Previous studies showed the fact that the rate of aging variable was also related to the oxygen content available within the asphalt concrete since the amount of it could lead to higher rate of chemical reaction during the oxidative aging process. That being said, the prediction model of the aging variable also captured the phenomenon was shown in Figure 10 (b) and was validated by the decreasing rate of aging variable during the service life Figure 10 (c). It leads to an interesting finding that by using the assumption of aging history variable \( k_2 = 1 \), the rate of the aging variable was decreased with the linear curve which is not realistic to the real aging phenomenon since the rate of aging tends to decrease with increasing level of aging and gradually reach a plateau. In this study, the \( k_2 \) parameter was obtained amount 8.570. Employed the obtained \( k_2 \) into the rate of aging resulted in a decreasing rate of aging variable by polynomial curve which was a closer relation of the oxidative aging process in reality. Figure 12 showed the comparison between \( k_2=1 \) and \( k_2=8.570 \) for the aging variable vs. aging rate.

Figure 11. Aging variable model

Figure 12. Comparison of aging variable rate with assumption and evaluated aging history parameter \( k_2 \) value

It was noteworthy about the aging model that it was more related to simulation of the aging of asphalt concrete rather than asphalt binder. Abu al-Rub et. al. (2013) mentioned that the aging variable value was formulated as a function of the diffused oxygen content and temperature evolution that linked to the mechanical response of aged material during the oxidative aging of asphalt concrete. Babadopulus et. Al. (2014) and Rahmani et. Al. (2017) studies that related to the same aging variable also showed
that there is a closer rate of an aging variable due to aged asphalt concrete rather than aged asphalt binder found from this study. Lastly, FTIR test result was taken to verify the aging model. The Pearson Correlation Analysis applied to obtain the relationship between the aging variable rate of asphalt concrete and the carbonyl rate of aged asphalt binder. Based on the results of this study, aging variable of asphalt concrete was strongly related to the rate of carbonyl content with the result of $r = 0.976$ and p-value = 0.0046. Figure 13 showed the result of the correlation test.

**Figure 13.** Correlation test result between carbonyl and aging variable

5. **Conclusion**

The findings of this study summarized as follow:

- The level of aging applied to both asphalt binder and asphalt concrete showed differences, as verified by all of the p-values from the regression analysis test, were greater than 0.05.
- The aging variable values were decreased with the aggregate involved
- The aging model parameters value of asphalt binder were 0.0269, 8.570, 8.640, 1.598 for $\Gamma^a$, $k_1$, $k_2$, $k_3$ respectively, where asphalt concrete were 0.0013, 8.570, 5.978, 0.780.
- Aging history dependent ($k_2$) found in this study allow the model to simulate the actual aging behavior as a polynomial curve.
- Carbonyl content was used to validate the aging model variable feasibility using the Pearson correlation test resulting $r = 0.976$ and p-value = 0.0046 which means there was a strong correlation between both of them.

6. **References**

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