



# Evaluation of the sensitivity of asphalt concrete modulus to binder oxidation with a multiple length scale study



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## HIGHLIGHTS

- This paper presents the sensitivity assessment of asphalt concrete modulus to oxidation.
- Three different binders and two different aggregates are evaluated.
- The study is multiscale study with binder, mastic and Fine Aggregate Matrix (FAM).
- The binders were aged prior to mixing with the aggregates to avoid the blunting effects of aggregate interaction with binder.

## ARTICLE INFO

### Article history:

Received 30 January 2017

Received in revised form 20 June 2017

Accepted 4 July 2017

### Keywords:

Asphalt binder

Mastics

FAM

Physico-chemical

Multiscale

Oxidation

Sensitivity

## ABSTRACT

It is well known that the properties of asphalt concrete mixture are affected by the oxidation of asphalt cement. However, the precise relationship between these two length scales remains largely uncharacterized. In the present study, a multiple length scale evaluation approach is applied to study and quantify the sensitivity of the mechanical properties of asphalt concrete mixture to asphalt cement oxidation. The study involves temperature and frequency sweep experiments on unaged and aged asphalt cement (to establish baseline properties), asphalt mastic (to consider physico-chemical aspects), and Fine Aggregate Matrix (FAM – to consider air voids and aggregate interaction effects). The multiscale approach separates effects of aggregate-binder physico-chemical interactions from those caused by air voids and physical aggregate interactions. Also, all asphalt cements were pre-aged to specific aging levels before preparing respective aged mastics or FAM samples. The methodology adopted for assessment of sensitivity was based on the theory of crossover modulus and second order rate kinetics of asphalt binder oxidation. The results from the analysis indicate that the mechanical properties of mastics are more sensitive to binder oxidation than FAM. Also, mechanical properties of mastics and FAM materials prepared with softer binders are more sensitive to oxidation than those with higher modulus asphalt cements. Finally, it is found that if a laboratory aging procedure is found to match the rheological properties of in-service level of asphalt oxidation at a given level of accuracy, then the expected accuracy in matching the resulting modulus of an asphalt mixture tested after being subjected to that laboratory process will be 1.5–3.6 times higher.

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## 1. Introduction

Oxidation of asphalt cements has long been recognized to affect performance of asphalt concrete pavements by causing the material to stiffen and embrittle, which leads to a high potential for cracking. Laboratory investigations of oxidation have been traditionally performed using two approaches. In the first, asphalt binders are subjected to various durations of extended heating with or

without pressure and then subsequently tested to identify their mechanical properties as a function of exposure condition [1–3]. In the other approach, asphalt mixtures are subjected to extended heating and then their corresponding mechanical properties are measured [4–6]. In both approaches, some parameter/metric based on mechanical properties is defined to quantify and evaluate the effect of oxidation.

Many have used the ratio of aged modulus to unaged modulus as this indicator [1,7,8]. Huang et al. [3] likened the effect of aging time on modulus of binder to the reverse effect of temperature and instead used similarities or differences in variation of slope of modulus as evaluated by shifting isothermal modulus curves at

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different aging times using the Williams-Landel-Ferry (WLF) equation. Aspects of binder oxidation chemistry as reported by Petersen et al. [9] and Glover et al. [10] were also explored to quantify aging based on carbonyl area (CA) growth [11,12]. The increase of low shear rate limiting viscosity in direct proportion to carbonyl growth is well documented [13–15], with the proportionality factor termed as hardening susceptibility. Aging has also been quantified based on changes in crossover modulus,  $G_c^*$  [16], which is simply the modulus at the point where storage modulus  $G'$  and loss modulus  $G''$  are equal.

Studies have been performed relating the changes in binder oxidation metrics, like CA, to the significance of aggregate type, age of the mixture etc. and its subsequent effect on resultant mixture viscoelastic responses [12] and also thermal cracking [15]. Daniel et al. [5] investigated the effect of aging on dynamic modulus, phase angle, and also viscoelastic properties such as creep compliance, relaxation modulus. Baek et al. [6] used a damage model to evaluate fatigue performance of laboratory prepared and oxidized asphalt concrete mixtures. More recent studies, have developed micromechanical models that link binder oxidation magnitude and rate with the resulting change in mechanical properties of asphalt concrete materials with different volumetric properties. Caro et al. [17] developed a two dimensional micromechanical model to study the influence of air void structure on oxidative hardening mechanism in asphalt concrete.

Owing to the importance of asphalt oxidation and aging with respect to in-service performance, there has also been considerable interest in developing laboratory based simulation procedures for long-term oxidation process in asphalt mixtures [18–22] and for understanding how asphalt binder and asphalt mixture properties are related. The current standard oxidation method (given in AASHTO R30) is based largely on experiments that compared modulus values from in-service pavements with laboratory samples that had been exposed to varying conditioning time [18]. A potential shortcoming with this approach is that changes in asphalt concrete properties over time can occur because of other factors besides oxidation, particularly moisture and load induced damage. A more direct evaluation of oxidative impacts can be made through study of asphalt binder alone, specifically by comparing properties of field aged binder with those from binder extracted from mixtures after applying different simulative aging methods. However, because there does not currently exist a means to accurately map changes in asphalt binder to resultant changes in mechanical properties of asphalt mixtures, it cannot be determined how closely the laboratory procedure needs to replicate the field aged binder properties.

## 2. Objective

The main objective of this paper is to present results and discussion on sensitivity of modulus of asphalt concrete to asphalt binder oxidation. Sensitivity refers to the impact of oxidation induced changes in the asphalt binder on modulus of an asphalt concrete. The study was motivated by the need to balance the accuracy that laboratory aging processes should replicate in-service aging and still yield acceptable simulation of field aged mechanical properties. The approach taken to meet this objective involves a multiple length scale testing program involving different material sources, in order to isolate independent contributions of physico-chemical influences and aggregate contacts.

## 3. Materials and methods

The experimental plan to investigate this sensitivity involved testing asphalt binders, asphalt mastics, and Fine Aggregate Matrix (FAM). Asphalt mastic consists of asphalt binder and aggregate particles finer than 0.075 mm while FAM consists of

asphalt binder, filler, and fine aggregates smaller than approximately 1.18–0.6 mm depending on the mixture type. Conclusions inferred upon asphalt concrete, are based on results obtained from FAM tests, which has been used as a surrogate for asphalt concrete in light of findings from relevant studies [23,24]. It has been shown that a substantial amount of total internal structure that exists within asphalt concrete mixture resulted from structural configuration of FAM, and thus experiments with this FAM could provide useful insight into behaviors of asphalt concrete [24]. It has also been shown that modulus and fatigue properties of these two materials can be linked through analytical and computational models [23,25,26]. In this study both asphalt mastic and FAM have been fabricated with pre-aged asphalt binder. By pre-aging asphalt prior to mixing, the degree of oxidation in the binding medium was known and thus any changes in modulus of the composites could be quantified with respect to changes in asphalt binder aging parameter (AP). It is worth noting that one of the possible implications of using a pre-aged asphalt is the slight variation in the adhesive properties of virgin binder vs aged binder. However, literature, SHRP-A-341 [27], has shown that binder chemistry is of little importance in asphalt – aggregate interactions and instead that these interactions are largely driven by the aggregate mineralogy and aggregate chemistry. In the present study, all FAM samples have been fabricated with only one aggregate source. The primary reason for using pre-aged binder for the FAM samples is simulate the similar aged binder for both mastic and FAM materials and also to avoid the damage of the specimen that could result from aging of compacted FAM samples.

### 3.1. Study materials

The study material matrix included three binders, two aggregates and two gradations as summarized in Table 1. Mastics were prepared based on the volume content of filler ( $V_c$ ), as calculated from volumetric characteristics of each aggregate type and gradation. FAM tests were conducted at different air void contents to represent 4.5% and 7.5% air voids in the asphalt concrete mixture. It has been estimated that between 40 and 70% of air voids that are present in asphalt concrete are part of the FAM phase [28], and here it is assumed that 55% of total voids exist in the FAM material and remaining air voids are considered to exist in formation of AC material with FAM and coarse aggregates. It should be noted that the estimated air void content is based on the authors definition for FAM and based on previously published literature [23,28] as there doesn't exist to date any acceptable standardized definition and design for FAM materials. Table 1 summarizes the study materials and the adopted naming convention.

### 3.2. Aging methodology

The study first involved creating mastics and FAM using asphalt cements that were pre-aged to levels that represent short (STA), medium (MTA), and long-term (LTA) oxidation. These pre-aged asphalts were first oxidized in Rolling Thin Film Oven (RTFO) at 163 °C for 85 min. Then, depending on the binder and the desired oxidation level, they were subjected to 20 h of conditioning in a pressure aging vessel (PAV). The methodology employed in the study involves creating asphalts that approximate 7, 15, and 22 years of pavement service (the exact numbers are arbitrary but generally the goal is to cover STA, MTA, and LTA conditions).

The aging temperatures were estimated from the regression model developed in the NCHRP 09–23 study, Eq. (1). This equation predicts the required PAV temperature,  $T_{PAV}$ , in order to simulate a specified aging time,  $t_{aging}$  (in years), for a given location. Additional terms needed to predict this temperature include, viscosity of RTFO residue at 60 °C,  $\eta_{RTFO,60^\circ C}$ , as constructed air void content of the mixture ( $VA_{orig}$ , assumed equal to 7%), and mean annual air temperature (MAAT), which is expressed in degrees Fahrenheit.

$$T_{PAV} = \left[ \frac{\left( 2.132432 + 0.193560 \left( \log \log \eta_{RTFO,60^\circ C} \right)^2 MAAT \right) \times}{\ln(t_{aging} \times 12) + 109.9632 - 78.2945 \left( \log \log \eta_{RTFO,60^\circ C} \right)^2} \right] \times \left( 0.445445 \times VA_{orig}^{0.378370} \right) \quad (1)$$

To use this function, it is assumed that 7, 15, and 22 years represent asphalt cement at STA, MTA, and LTA. It is also recognized that viscosity and MAAT values are correlated, specifically that asphalt binder used in regions with high MAAT is more likely to have a high viscosity and those used where MAAT is low are more likely to have low viscosity. For this assessment it is important to consider the correlation between climate (specifically the MAAT) and PG grade selection. This correlation can be seen by subjectively comparing a map of the high temperature PG grade distribution to a map of MAAT values across the country, Fig. 1.

However, to the author's knowledge the exact correlation has not been expressly evaluated in the published literature, and thus an analysis was performed to identify and quantify the correlation. The precise relationship is established by identifying correlation between the climate based PG grade and MAAT since viscosity can be predicted from PG grade [29]. Two sets of data are obtained for this purpose; 1) the 98% reliability based PG grades and latitude/longitude of all available weather stations (7358 stations) in LTPPBINDV3.1 and 2) the Global Historical

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