



Further exploration of the pavement oxidation model – Diffusion-reaction balance in asphalt

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HIGHLIGHTS

- Pavement oxidation model simulate asphalt oxidative aging including both diffusion and reaction.
- Asphalt oxidative reaction results asphalt hardening.
- Diffusion transports oxygen to reaction, but suffers from asphalt hardening.
- Asphalt oxidative aging could be controlled by either diffusion or reaction or both.

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ABSTRACT

Oxygen diffusion and oxidative reaction are two main factors in asphalt oxidative aging discovered by experiment, but the dynamic balance between them could not be easily studied by experimental methods due to its comprehensiveness. A pavement oxidation model was utilized to simulate this process in asphalt: oxygen molecules penetrate into the asphalt film and then react with the asphalt molecules.

In some special cases, diffusion or reaction can become the determination step of asphalt oxidative aging. Another factor, asphalt hardening, which is a result of oxidative reaction, slows the oxygen diffusion seriously and consequently. With the help of the model simulation, the efforts of binder reaction kinetics, hardening properties and temperature on this balance have been studied and discussed in detail. A new standard for asphalt binder, solely from oxidation aspect, includes high activation energy, low hardening susceptibility and high hardening intercept. A new idea of anti-oxidant development was proposed. By decreasing oxygen diffusion, asphalt oxidation can be slowed down, but only works for oxygen diffusion determined cases.

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1. Introduction

Over the years, there have been efforts to predict the pavement long term performance in need of the design and maintenance purposes and a series of models have been developed. The driving force, which encourages the scientists to continue their research on those models, is the increasing number of findings on pavement aging.

First, it is important to accurately predicting the change of the viscoelastic properties of an asphalt binder on the road from oxidative aging for predicting short-term and long-term performance

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and improving the design of new pavements [2]. The asphalt pavement collapses due to the long-term oxidation aging of bituminous binder. It can be restored using regeneration principles and appropriate technologies [22]. The required amount of asphalt mix components and the lowest price can be calculated using optimization methods [23]. A binder deterioration model (BDM) was developed by establishing a relationship between the apparent viscosities of the binder with pavement age [3]. The apparent viscosity data generated using the BDM were compared with that computed using the bitumen hardening model developed by Oliver [18]. The two models show a consistent trend in the binder deterioration.

Except binder deterioration, a numerical model to predict the diffusion and reaction of oxygen in petroleum bitumen films was developed by Herrington [8]. Model parameters for the diffusion-oxidation process was obtained by correlating oxygen uptake of

bitumen solutions to changes in carbonyl infrared spectral absorption and viscosity.

Farrar evaluated the potential of using the evolution of the binder crossover modulus with time and depth in the pavement to determine the change in linear viscoelastic properties of the binder during the life of the pavement. The crossover modulus is one of three parameters in the Christensen-Anderson complex shear modulus model. The crossover modulus, when described by a kinetic model that expresses the evolution with time in an Arrhenius reaction rate form, allows the determination of the change in linear viscoelastic properties of the binder during the life of the pavement. The Christensen-Anderson model was used by Farrar et al. [5] to develop complex modulus and phase angle master curves of the binder as a function of field time, depth, and temperature.

Alavi et al. [1] demonstrated and evaluated a new approach for quantifying the effect of asphalt binder oxidative aging on the viscoelastic properties of the asphalt mixture. The oxidative aging was measured for the carbonyl area (CA) in the recovered asphalt binder.

Lopes et al. [15] proposed a new protocol to characterize aging of binders located at the surface of the mixture. This protocol is based on sampling two particles at the top of the layer for studying and characterizing aging by means of an FTIR analysis on the solubilized binder. Han et al. [7] developed a one-dimensional model to predict pavement temperature nationwide based on heat-transfer fundamentals. The model employs recorded hourly solar radiation, daily average wind speed, and interpolated hourly air temperature.

In order to predict pavement aging, Lunsford proposed a pavement aging model in 1994 [16], which includes many important concepts, such as asphalt oxidative reaction kinetics, asphalt binder content and air voids structure. Prapaitrakul et al. [20] refined this model by introducing a new concept, average shell thickness, and using hourly pavement temperature input. Han [6] replaced the average shell thickness with five pore radii from X-ray CT measurements of pore size distribution and also quantified the effort of asphalt viscosity on oxygen diffusion. The latest and most advanced model is developed by Jin et al. [9] with two significant improvements: 1) introducing the concept of diffusion depth to better define the oxygen diffusion region in the asphalt-aggregate mastic; 2) incorporating both fast-rate and constant-rate asphalt oxidation kinetics to better describe the aging process in the pavement.

A common in previous studies is the only use of the model was to predict the pavement aging status and the results described particularly in the literatures already show its capability and accuracy. Of course, the role of the model should not be limited just to this. Of even greater importance to the scientists is to understand the factors in asphalt aging.

Seven fundamental components in the model are believed to play most important roles in pavement aging:

1. Pavement design information, including basic information such as designed air void and asphalt binder content;
2. Accessible air voids, derived from X-ray CT analysis on field cores, providing the exposed surface area of asphalt to air;
3. Hourly full depth pavement temperatures, calculated by pavement temperature model [7];
4. Asphalt oxidation kinetics, obtained from laboratory aging tests on original binder. With the kinetics parameters, the formation rate of carbonyl could be described as a function of temperature and oxygen partial pressure;
5. Asphalt hardening properties, obtained from laboratory aging tests on original binder, reflect the physicochemical relationships between carbonyl components and binder's stiffness. The hardening process can be tracked with measurement of the binder viscosity or DSR F_n ;

6. Oxygen diffusivity in asphalt, determined by both temperature and binder viscosity;
7. Oxygen concentration, determined by oxygen transport-reaction process. The concentration changes as a function of position and time in the asphalt thin film, influenced by oxygen diffusion in asphalt and oxidative reaction with asphalt.

The understanding on those aging components are still limited; accelerated aging conditions are widely used in laboratory but it has been proved not be able to reproduce the same relative rankings of a group of asphalts as aging under milder pavement conditions. This is because of the complex dependency of oxidation kinetics and the consequent changes in physical properties on aging temperature and pressure as well as oxidation time and pavement structure. However, there can be a fortuitous cancellation of errors that produces better results than might be expected [11,4]. Thus, the effect of each component on asphalt aging has been studied in depth while the effects of other factors have been eliminated. But, in deeper levels of thinking, it is necessary to consider those factors simultaneously and to further discover the relative importance of those factors. With current methods, this issue could not be easily addressed without designing a series of extremely complicated experiments due to the comprehensiveness in this problem. At this time, the pavement oxidation model could be utilized as a good tool to solve this problem, because of those two reasons: First, the model already combines all the individual factors; Second, much less time and workload is required by model simulation than actually doing the experiments.

Among all the topics of interest on asphalt aging, one of them is of extreme importance. Between oxidative reaction and oxygen diffusion, the question of which factor has more determination in the whole aging process, has been discussed all the time but no real experimental provident has been post out yet.

In this study, an exploration of the model application is focused on the interactions between oxygen diffusion and oxidative reaction. There are four concerns is this topic.

First, oxygen diffusion and oxidative reaction occur simultaneously. Oxygen molecules need to penetrate through the asphalt film to a certain depth and then react with the asphalt at that position. If the oxygen diffusion runs much faster than the reaction, there should be plenty of oxygen molecules in the asphalt film that makes the reaction runs at its maximum level at all depth and all time. On the other hand, if the reaction runs much faster than the diffusion, the oxygen concentration keeps at a very low level or even almost no oxygen molecule in the asphalt film. It will seriously limit the reaction as feedback. Moreover, in the latter case, the oxygen "black hole" (low oxygen concentration in asphalt) would create a delta of oxygen concentration between the atmosphere and the asphalt film. This delta gives a high diffusion driving force, which enforces the diffusion as feedback. Of course, there is still a big chance that the rates of diffusion and reaction are at a similar level; therefore, neither of them fully controls the aging process.

Second, this study only focuses on the aging process in a long time range, although the balance between oxygen diffusion and oxidative reaction changes every second. The reaction hardens the asphalt and increased asphalt stiffness results the diffusivity decreasing over time. In turn, decreased diffusion rate slows the reaction (if the aging is not in the reaction domination stage). This negative feedback makes the research interest not focus on an instantaneous correlation at a given moment, but an accumulated effort in a period of 10 years.

Third, another issue needs to be considered is the effect of temperature. Since temperature plays extremely important roles in both diffusion and reaction, the sensitivity of the diffusion-reaction balance to temperature needs to be studied. In most of

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