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Review Article

State of the Art Study on Aging of Asphalt Mixtures and Use of Antioxidant Additives

Okan Sirin,[1] **Dalim K. Paul**,[2] **and Emad Kassem**[3]

[1]*Department of Civil and Architectural Engineering, Qatar University, P.O. Box 2713, Doha, Qatar*
[2]*Ashghal Research and Development Center, Public Works Authority, Doha, Qatar*
[3]*Department of Civil and Environmental Engineering, University of Idaho, 875 Perimeter Drive MS 1022, Moscow, ID 83844, USA*

Correspondence should be addressed to Okan Sirin; okansirin@qu.edu.qa

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The detrimental effects of hardening in asphalt pavements were first recognized by pioneering pavement engineers in the 1900s and have been studied extensively during the last 70 years. This hardening process, referred to as asphalt aging, is generally defined as change in the rheological properties of asphalt binders/mixtures due to changes in chemical composition during construction and its service life period. Aging causes the asphalt material to stiffen and embrittle, which affects the durability and leads to a high potential for cracking. This paper presents the state of the art on asphalt and asphalt mixture aging and use of antioxidant additives to retard the aging. A picture of complex molecular structure of asphalt and its changes due to atmospheric condition and various protocols used to simulate aging in laboratory environment are also discussed. Emphasis is given on recent studies on simulation of aging of asphalt mixtures as there has been limited research on mixtures compared to the asphalt binder. Finally, this paper presents the application of antiaging techniques and its mechanism, use of various types of antioxidant additives to retard aging of asphalt and, hence, improve the performance of asphalt pavements.

1. Introduction

Asphalt is the most widely used binding material in road pavements all over the world. Approximately 95% of asphalt that are produced worldwide each year is applied in the paving industry [1]. Asphalt essentially acts as a binder for mineral aggregates to form asphalt mixes, also called asphalt concrete or bituminous mixes. The first use of asphalt in road construction during the era of Nabopolassar, King of Babylon (625–604 BC), was mentioned by Abraham [2]. However, bitumen essentially disappeared from the pavements until the discovery of European sources of natural bitumen which led to the development of the modern applications for this material [1]. Asphalt paved roads have been in operation in Europe since the 1850s [3] and in the United States for about 125 years [4]. Pioneering pavement engineers [2, 5] observed a strong effect of temperature on its consistency and soon realized that hardening or aging of asphalt occurs during mixing, construction, and operation affecting the performance of asphalt pavement [6].

The term aging may be applied to describe multiple mechanisms in asphalt binder/mixture. Hence, it seems necessary to clarify the terminology used by pavement engineers. In pavement engineering, change in the rheological properties of asphalt binders/mixtures is due to changes in chemical composition during construction and its service life period.

Aging of asphalt binders occurs during the production of asphalt mixtures and while in service when exposed to the surrounding environment. The first stage of aging occurs at a very fast rate when asphalt mixture is produced at a very high temperature. This stage is often referred to as short-term aging. During this stage, a very thin film of asphalt is exposed to air at elevated temperatures, leading to a significant change in the rheological properties of the asphalt binders. Such changes are presented in high viscosity and

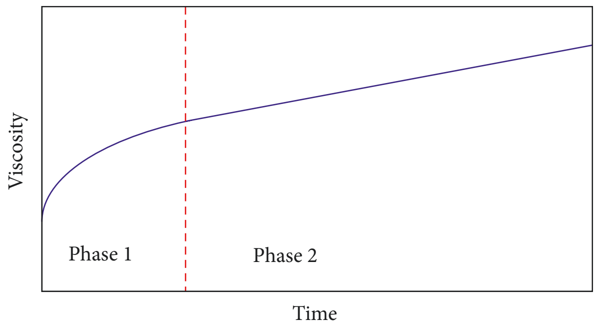


Figure 1: Typical hardening response for asphalt binder (after Glover et al. [8]).

increased stiffness [7]. The second stage of aging occurs when the asphalt is exposed to the environment as in-service pavement at a relatively lower temperature for a long duration. The rate of hardening depends on the in-place air void content and surrounding environment.

Figure 1 shows a typical hardening response for unmodified asphalt binder. There is a jump in the viscosity of the asphalt binder due to short-term aging (Phase 1), while there is a linear increase at a lower constant rate with time (Phase 2). Phase 2 represents hardening due to long-term aging.

There are several factors that influences asphalt aging. External factors include plant type, mixing temperature and silo storage time during short-term aging, and in-field conditions (i.e., temperature, ultraviolet (UV) ray, and rainfall) and time during long-term aging. The rate and extent of aging also depends on mixture properties such as source and type of asphalt, aggregate gradation and absorption, void content/permeability, and the thickness of asphalt binder film over the aggregate. Table 1 summarizes various factors and their effects on the short- and long-term aging of asphalt. A recent study by Morian et al. [9] reported that mixtures' effective binder content has provided the strongest indicator on the aging characteristics of asphalt mixture, irrespective of the type of granular aggregate.

Aging causes several changes in asphalt mix properties which are reflected in the performance of asphalt pavement. Results from experimental studies [33] demonstrated that ductility and penetration of asphalt binder are reduced while softening point and ignition temperature are increased as a result of aging. Ultimately, viscosity of the asphalt is increased and becomes a stiffer asphalt mixture. Increase in viscosity up to 10 times due to 5-year field aging in the Middle East conditions was observed as shown in Figure 2 [34].

Regarding mechanical properties, the stiffness modulus also increases due to aging (Figure 3), and this increase can be up to 4 times depending on the type of asphalt [35]. This may cause the mixture to become excessively hard and brittle and susceptible to disintegration and fatigue cracking at low temperatures [36–38]. Aging may also render the mixture less durable than the original mixture, in terms of wear resistance and moisture susceptibility [39]. As a result, damage tolerance of asphalt layer reduces, and many highway and airfield pavements in service fail prematurely. However, aging is not necessarily a negative phenomenon since the resistance of the asphalt mixture to permanent deformation and the load-bearing capacity is improved due to increased stiffness and cohesion. In some cases, aging may also help a mixture to achieve optimum properties [6].

This paper presents a comprehensive review of aging of asphalt paving materials with the following key focus areas:

(1) Comprehensive definition of asphalt aging and an overview of asphalt chemistry

(2) Critical discussion on aging mechanisms, corresponding changes in molecular structure, and its effect on the properties of asphalt materials

(3) Existing test methods, protocols, and techniques for assessing the aging of asphalt paving materials with an emphasis on recent studies on aging of asphalt mixtures

(4) Antiaging techniques and different types of additives and their mechanism to retard aging of asphalt to improve performance of asphalt pavement.

2. Asphalt Chemistry and Aging Mechanisms

Asphalt is either derived from natural deposits or obtained as a residue of crude petroleum or a product of solvent extraction of petroleum. It has a variable and complex elemental composition which depends primarily on its crude source. Asphalt is primarily composed of carbon (typically 80–88%) and hydrogen atoms (10–12%) which gives a hydrocarbon content of around 90% [41, 42]. The remaining portion consists of two types of atoms: heteroatoms and metals. Heteroatoms include nitrogen (0–2%), oxygen (0–2%), and sulphur (0–9%). Metal atoms are vanadium, nickel, and iron, and these atoms are present in trace quantities, typically far less than 1% [43, 44]. Table 2 shows elemental analysis of 8 different core asphalts of various crude origins.

The hydrocarbons constitute the basic structure of asphalt whereas the metal atoms provide indication or characteristic of asphalt crude source. Heteroatoms contribute to many of asphalt's unique chemical and physical properties by interacting with molecules. For example, sulphur reacts more easily than carbon and hydrogen to incorporate oxygen into the asphalt structure which leads to oxidative aging of asphalt [45].

According to Corbetts's method [46], these chemical elements combine to form the four main components or fractions of asphalt cement: asphaltenes, saturates, naphthalene aromatics, and polar aromatics (or resins), each of which provides different characteristics to asphalt. Asphaltenes and saturates are normally incompatible compounds and are brought together by aromatics. Asphaltenes are mainly responsible for viscosity (i.e., hardening effects) whereas abundance of aromatics and saturates decreases the ductility (i.e., elastic effects). Some researchers divide the asphalt into two broad chemical groups according to Rostler's methods of precipitation [47], namely asphaltenes and low molecular weight maltenes.

Table 1: Factors affecting aging of asphalt mixture.

	Factors	Findings	References
Short-term aging	Binder chemistry	Major effect	Traxler [10]
	Binder type and source	Significant effect on field aging	Lund and Wilson [11, 12]
	Binder type and source	Significant effect on lab aging; reduced aging with polymer	Topal and Sengoz [13], Zhao et al. [14], Morian et al. [15]
	Asphalt binder film thickness	Significant effect	Kandhal and Chakraborty [16]
	Aggregate gradation	No effect	Chipperfield and Welch [17]
		Important effect	Morian et al. [15]
	Aggregate absorption	Major effect	Traxler [10]
		Important effect	Aschenbrener and Far [18], Morian et al. [15]
	Inclusion of recycled materials and reheating	Significant effect	Mogawer et al. [19]
	Plant type	Significant effect	Terrel and Holen [20], Chollar et al. [21]
	Production temperature and silo storage	Significant effect	Mogawer et al. [19], Daniel et al. [22]
Long-term aging	Aggregate source	No effect on lab aging	Morian et al. [15]
	Aggregate porosity	Significant effect	Kemp and Predoehl [23]
	Binder source	Significant effect	Morian et al. [15]
	Asphalt content	Significant effect	Kari [24]
		No effect	Rolt [25]
	Air voids	Significant effect	Kemp and Predoehl [23] Harrigan [26], Houston et al. [27]
		No effect	Rolt [25]
	Pavement permeability	Significant effect	Kari [24]
	Pavement depth	Field aging not limited to the top 25 mm of the pavement; field aging gradient observed	Farrar et al. [28]
		Aging decreases over depth	Sirin et al. [29]
		10-year field aging can result in deterioration to the 2nd layer	Wu et al. [30]
	In-service temperature	Significant effect	Kemp and Predoehl [23], Rolt [25], Epps Martin et al. [31], Sirin et al. [28]
	Exposure time	Significant effect	Rolt [25]
	Ultraviolet ray	Significant effect	Lee [32]

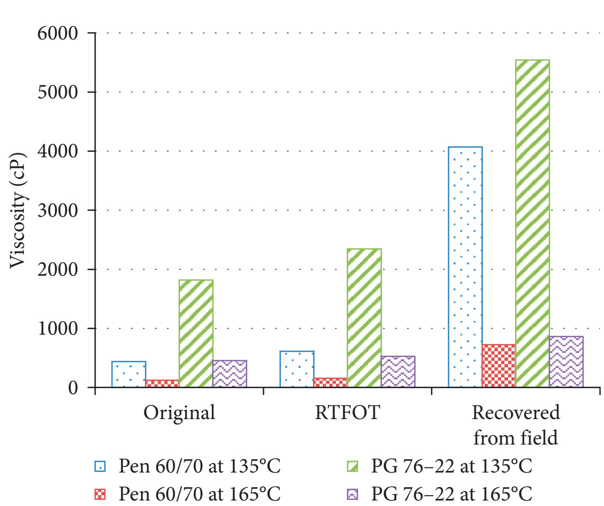


Figure 2: Effect of 5-year aging on the viscosity of asphalt binders.

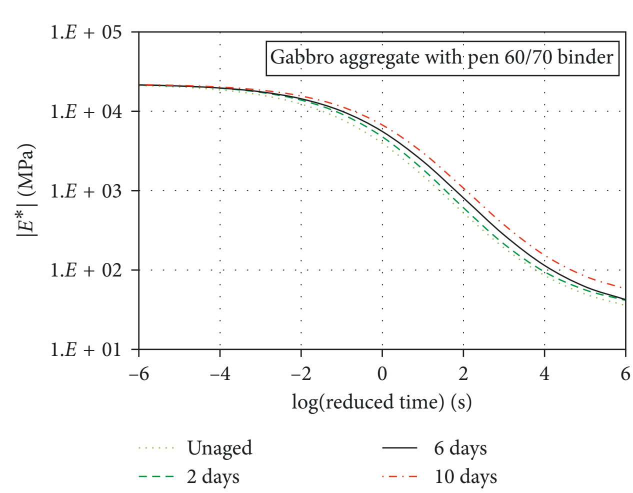


Figure 3: Effect of long-term aging duration on dynamic modulus [40].

Table 2: Elemental and component analysis of SHRP core asphalts by Asphalt Institute (from Mortazavi and Moulthrop [42]).

Asphalt code and crude source	AAA-1 Canada	AAB-1 USA	AAC-1 Canada	AAD-1 USA	AAF-1 USA	AAG-1 USA	AAK-1 Venezuela	AAM-1 USA
Elemental analysis								
C (%)	83.9	82.3	86.5	81.6	84.5	85.6	83.7	86.8
H (%)	10.0	10.6	11.3	10.8	10.4	10.5	10.2	11.2
H + C (%)	93.9	92.9	97.8	92.4	94.9	96.1	93.9	98.0
O (%)	0.6	0.8	0.9	0.9	1.1	1.1	1.0	0.5
N (%)	0.5	0.5	0.7	0.8	0.6	1.1	0.7	0.6
S (%)	5.5	4.7	1.9	6.9	3.4	1.3	6.4	1.2
V (ppm)	174	220	146	310	87	37	1480	58
Ni (ppm)	86	56	63	145	35	95	142	36
Fe (ppm)	<1	16	—	13	100	48	24	255
Component analysis								
Asphaltene (%)	18.3	18.2	11.0	23.0	14.1	5.8	21.1	3.9
Saturates (%)	10.6	8.6	12.9	8.6	9.6	8.5	5.1	1.9
Polar aromatics (%)	37.3	38.3	37.4	41.3	38.3	51.2	41.8	50.3
Nanthelene aromatics (%)	31.8	33.4	37.1	25.1	37.7	32.5	30.0	41.9

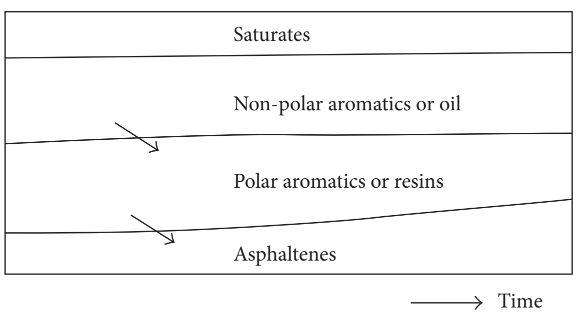


Figure 4: Effect of aging on the chemical composition of the asphalt (from Heneash [55]).

Maltenes are viscous liquids composing of resins and oils [48]. A complex mixture system of asphalt is formed upon chemical and physical interaction between these fractions [49–52]. A component analysis of different types of asphalt is presented in Table 1.

Researchers [53, 54] used high pressure-gel permeation chromatographic (HP-GPC) to separate asphalt into different fractions and studied independently the effect of aging process on asphalt components. Studies regarding chemical composition of asphalt through aging indicate that the asphaltenes content is increased, while the resins and aromatics contents decrease. As a result of increase in asphaltenes content, the asphalt become harder (i.e., stiffer), and that could be easily manifested as decreasing of penetration and increasing softening point and viscosity [55]. Figure 4 shows the effect of aging on the chemical composition of a typical asphalt binder. Researchers also indicated that due to aging, the asphaltenes/maltenes ratio changes causing an increase in bitumen viscosity, becoming more hard and brittle [1].

Physical and chemical properties of asphalts change over time due to the exposure to various environmental conditions in the field during their service life. Since the 1930s, research has continued to develop an understanding of the mechanisms contributing to short- and long-term aging [56]. Mechanisms causing binder aging include oxidation, volatilization, thixotropy (or steric hardening), polymerization due to actinic light, and condensation polymerization due to heat [6, 10, 51, 57]. Among them, oxidation, volatilization, and steric hardening are considered the main mechanisms associated with the aging process of asphalt mixtures [51, 57–59]. During production, laying, and compaction, asphalt mixture is subjected to higher temperature which causes aging due to oxidation and loss of volatile compounds. On the contrary, long-term aging during service periods takes place at lower temperature primarily due to oxidation mechanism [60].

2.1. Oxidation. Many researchers have addressed binder oxidation chemistry [8, 52, 61–63]. Oxidation is the irreversible chemical reaction between oxygen molecules and the component species of bulk asphalt resulting in significant alterations to the desired physical and/or mechanical properties of asphalt. Oxidative aging of asphalt is believed to be caused by the generation of oxygen-containing polar chemical functionalities on asphalt molecules, which in turn can cause agglomeration among molecules due to increased chemophysical associations such as hydrogen bonding, van der Waals force, and Coulomb force [41, 64, 65].

The effect of binder oxidation in pavement on its performance is rather contradictory. Asphalt's complex organic components react with atmospheric oxygen and ultraviolet (UV) radiation, and as a consequence, the pavement surface is hardened which leads to cracks. Coons and Wright [66] reported that binder oxidation occurs only in the top inch of the pavement and that below the top inch; the binder is left virtually unaffected by years of use and years of environmental exposure. Recently developed Mechanistic Empirical Pavement Design Guide [67] also assumes in its calculation that binders oxidize only in the top inch. As a consequence, binder oxidation and the resulting increase in pavement stiffness actually can have a positive, beneficial impact on pavement fatigue life [8].

However, Walubita et al. [68] and Walubita [69] indicated that binder oxidation in pavements can have a very significant negative impact on pavement fatigue life. More profound evidence of pavement hardening well below the surface has been reported based on extensive data by Glover et al. [70] and Al-Azri et al. [71] where a large number of Texas pavements were cored, the binder extracted and recovered, and then tested to determine binder stiffness as a function of age of the pavement. Increase in stiffness and a decrease in the ductility of the asphalt mixtures is reported due to oxidation, which could reduce its resistance to fatigue cracking [72].

Asphalt oxidation produces changes in the chemical composition of asphalt. Saturates remain essentially unchanged due to their low chemical reactivity whereas other three fractions exhibit significant variations [73, 74]. As a result, functional groups (i.e., carbonyl and sulfoxide groups) are formed in asphalt molecules leading to decreased aromatic fractions and increased asphaltenes fractions [1]. Many attempts have been taken to quantify oxidation for better understanding of asphalt aging. Liu et al. [75] indicated that as the area of carbonyl region (CA) in FT-IR spectra is a direct measure of binder oxidation and the percentage of carbonyl compounds can be used to assess the changes due to oxidative aging [8]. The content of carbonyl depends on the temperature and oxygen partial pressure.

The carbonyl reaction rate is described by [8]

$$\frac{d\mathrm{CA}}{dt} = r_{\mathrm{CA}} = AP^{\propto} e^{-E/\mathrm{RT}}, \tag{1}$$

where $d\mathrm{CA}/dt$ = carbonyl reaction rate, A = frequency factor, P = pressure, $\propto$ = reaction order, E = activation energy, R = gas constant, and T = absolute temperature. Studies show that the values of A, E, and $\propto$ differ for different asphalt types.

2.2. Volatilization. Volatilization is another important mechanism that occurs during hot mixing and construction of asphalt cement. At high temperatures, lighter molecular weight can vaporize and escape into the atmosphere [1, 10]. This may be more significant in the preparation of modified asphalt binders where oil-like compounds are evaporated from the asphalt. When thin asphalt film comes into contact with aggregates at temperatures of 150°C or higher, aromatic fractions rapidly evaporate, and asphaltene fractions generally increase between 1 and 4% [76]. Fumes and steams are generated as a result of this reaction depending on the contact surface area between the asphalt film and the aggregates [77]. As a result of weight loss, asphalt flow properties are reduced, that is, viscosity is affected by volatilization, especially given the speed with which volatilization takes place [78, 79]. Researchers [6, 80] have found that viscosity increases from 150 to 400%. Significant increase of modulus and decrease of phase angle were observed because of volatilization [81]. Anderson and Bonaquist [60] suggested that quantifying the amount of volatile compound loss is essential for better understanding of asphalt hardening during short-term aging.

2.3. Steric Hardening. Steric hardening, also known as physical hardening, occurs over time when asphalt cements are exposed to low temperature. In this process, molecular structure of asphalt is reorganized, affecting its asphaltene fractions [82]. Consequences of steric hardening are increased viscosity, slight volume contraction, and ultimately the hardening of asphalt [10, 83]. Steric hardening is more pronounced at temperatures close to 0°C and must be considered while testing asphalt at very low temperature. As this hardening is a result of structural reorganization of the molecule at low temperatures [51], it can be reversed through heat or mechanical work [84].

3. Laboratory Accelerated Aging and Evaluation Methods

Asphalt is aged in laboratory environment at a faster rate through heat and air application to simulate field aging and hence to predict performance of asphalt pavement. Earliest work to simulate aging in laboratory was by Dow [5] who used extended heating test. Since then, many research efforts [6, 10, 27, 50, 57, 85–99] have been devoted to assess effect of aging on the performance of asphalt materials. After aging acceleration treatment, samples are usually studied to quantify changes in the asphalt binder/mixture properties before and after the aging treatment (commonly known as aging index). Properties examined during aging studies are generally weight loss, viscosity, penetration, ductility, tensile strength, and stiffness modulus.

Treatment of asphalt or tests related to aging of asphalt materials can be broadly divided into two categories, namely, tests performed on asphalt binder and tests performed on asphalt mixtures. Therefore, discussion of the work is represented in the following two sections: binder studies and mixture studies.

3.1. Binder Studies. Researchers have devised several testing methods to characterize properties of asphalt binders by simulating aging of asphalt in a hot mix plant and during service period life of pavement. Most of these researches have used thin film ovens to age asphalt by applying extended heating and air blowing (or oxidation) procedure. The most commonly used and standard tests for simulation of asphalt hot-mix aging are the Rolling Thin-Film Oven Test ((RTFOT) ASTM D2872 [100], AASHTO T240 [101]) and Thin-Film Oven Test ((TFOT) ASTM D1754 [102], AASHTO T179 [103]). Pressure aging vessel (PAV) is used to simulate long-term aging of asphalt binder that is experienced in the field [104]. In the current Superpave binder specifications, an asphalt binder to be evaluated is to be subjected RTFOT for short-term aging at 163°C for 85 minutes followed by a PAV process in order to simulate several years of field aging.

The TFOT was first introduced by Lewis and Welborn [105] to simulate short-term aging by applying a temperature of 163°C on asphalt of film thickness of 3.2 mm for 5 hours. However, researchers criticized TFOT due to a film thickness much thicker than that is commonly observed in

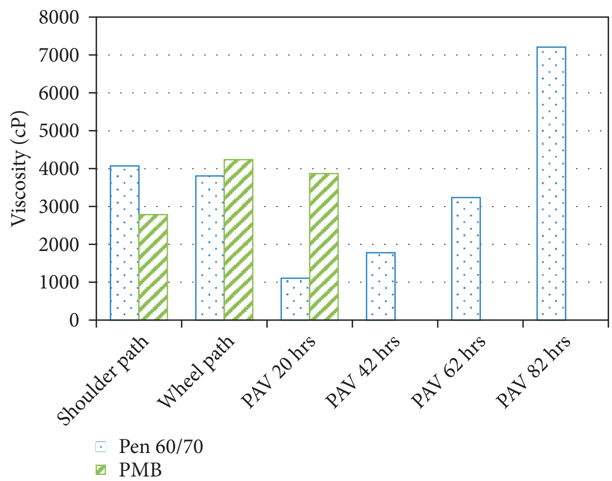


Figure 5: Comparison of PAV-aged binders with binders recovered from 5-year old pavement (after Sirin et al. [34]).

the field and nonuniform aging throughout the depth of asphalt [58]. Many researchers attempted to develop or improve testing methods to age asphalt with more representative film thickness. One such effort was the Modified thin film oven test by Edler et al. [106] who utilized a film thickness of 100 μm with an additional increased exposure time of 24 hours. Researchers also proposed some other testing methods such as Shell microfilm test [107], Rolling microfilm oven test [108], Tilt-oven durability test [23], and Thin film accelerated aging test [92] for better simulation of asphalt binder aging.

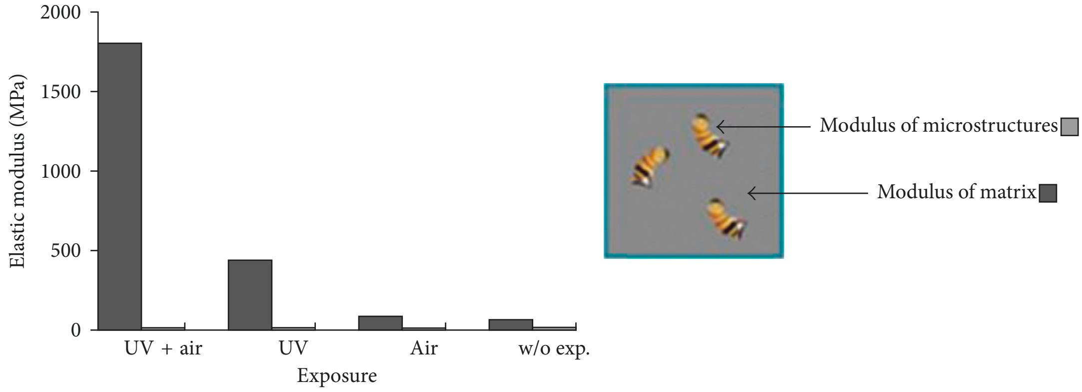
The most significant modification of the TFOT was the RTFOT, developed by the California Division of Highway [109], where eight glass bottles each containing 35 gm of asphalt are aged by applying heat and oxidation on thin films of 1.25 mm. This method ensures uniform aging of asphalt with no skin formation and correlates asphalt hardening reasonably well with that observed in hot-mixing process [110]. However, several researchers [111–113] identified a number of deficiencies (e.g., spilling out from RTFOT bottles) in RTFOT especially while testing modified asphalt binders. To overcome these limitations, researchers developed improved testing methods such as Modified Rolling Thin Film Oven Test ((RTFOTM) Bahia et al. [96]), Modified German Rotating Flask [111, 114] (MGRF), and Stirred Air Flow Test [115] (SAFT) to evaluate short-term aging of neat and modified asphalt binder.

Although thin film oven tests can adequately simulate short-term aging of asphalt binder, they lack of accuracy in predicting long-term aging during service period life of pavement. To predict long-term aging, a number of attempts have been made by combining thin film oven tests with oxidative aging such as Iowa durability test [32], Pressure oxidation bomb [106], Accelerated aging test device [116], PAV [80, 117], and High pressure aging test [118]. Among them, PAV treatment is believed to be the most reliable method to simulate long-term aging. In this process, RTFOT aged asphalt is subjected to 100°C for 20 hours at 2.07 MPa pressure to reproduce field aging effects. It generally simulates aging of 8–10 years of pavement service life according to USA standards [79]. However, 20 hours conditioning in PAV may not be sufficient for severe weather conditions like in the Middle East where up to 70 hours of conditioning may be needed to simulate the field aging of a 5-year old asphalt pavement (Figure 5).

In a recent NCHRP study (Project number 9-36), Anderson and Bonaquist [60] attempted to develop an improved procedure to replace both RTFOT and PAV by a single apparatus for simulating short- and long-term aging. They examined both MGRF and SAFT, but with different operating conditions. Attempts with MGRF were not successful however SAFT with a modified impeller was proved to be successful to some extent to simulate both the short-term and long-term aging of asphalt binder.

Atomic force microscopy (AFM) is often used to study the aging of asphalt binder in microscopic level and evaluate the change in micromechanical and microrheological properties. AFM is a nondestructive imaging tool that can deliver the surface topography, stiffness, tackiness information, and molecular interaction at microlevel of materials [119, 120]. In AFM images, a bee-shaped (black and yellow stripes) structure is noticed, which indicates the asphaltene phase in bitumen [121, 122]. The presence of such microstructures somewhat dictates the macroscale properties of bitumen, such as stiffness, viscoelasticity, plasticity, adhesion, fracture, and healing characteristics. Evolution of these microstructures with ageing and related to the resulting mechanical response is in focus to better understand the long-term properties of asphalts.

In recent days, AFM has become a popular technique and utilized by many researchers [123–126] to characterize the effect of short-term, long-term ageing, and ultraviolet (UV) radiation on the morphology of asphalt binders. An increase of bee-shaped microstructure with PAV aging was reported by Huang and Pauli [127], Wu et al. [128], and Zhang et al. [123]. Zhang et al. [123] showed that lab-simulated aging affects the bitumen morphology significantly, and these changes in morphology are strongly correlated with the physical properties as well as chemical compositions of the binders before and after aging. The overall surface stiffness increased, and the bitumen surface became more solid-like [123]. Both asphaltenes content and the size of microstructures play a role in determining asphalt micromechanical properties [129]. Important relationships between microstructural changes depicted in AFM images and changes in composite viscoelastic properties obtained from the measurements were reported by Allen et al. [124]. Das et al. [126] found reduction in binder tackiness with aging, and as a result, adhesion of the asphalt binder specimens was negatively impacted causing an adhesive bond failure between binder and aggregates. The researchers reported the modulus of microstructure always being higher than the modulus of the matrix at the measured temperature, as shown in Figure 6. It was also noted that increase in modulus due to UV radiation exposure is higher than oxidation, and the highest value was always obtained after the combined exposure of UV and oxidation. Similar observation was found for 3 different binders of different sources.



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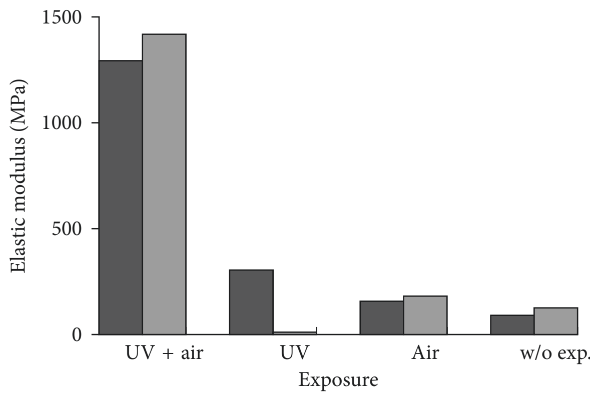

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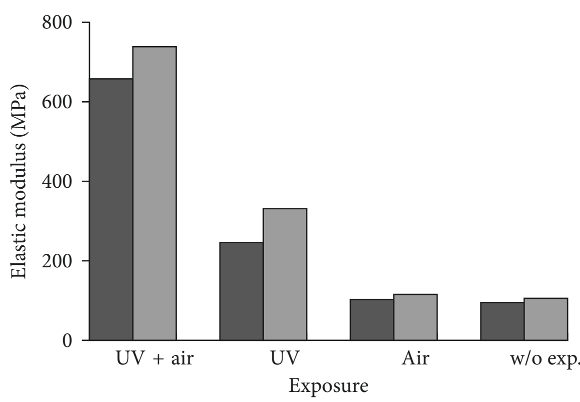

(c)

Figure 6: Variation in modulus measured using AFM at different levels of exposure for 3 different binders of different sources: (a) Bit-A, (b) Bit-B, and (c) Bit-C (from Das et al. [126]).

AFM had also been used to investigate the effect of aging on modified asphalt binders compared with the control binders [127, 128, 130].

3.2. Mixture Studies. Compared to research on asphalt binder, there has been relatively little research on the aging of asphalt mixtures. Much of the early work on asphalt aging was conducted solely on the binders, without involving mixtures [6, 131]. Eventually, efforts have been seen to analyze aging of the asphalt-aggregate mixture system by measuring changes in penetration and viscosity of the extracted and recovered binders [91, 132–134]. Studies reported in NCHRP Project 9-6 [134] involved measurements and comparison of viscosity and penetration of the binders extracted and recovered from mixtures aged in the laboratory under various conditions with those from field-produced mixtures. Limited resilient modulus testing was also conducted on the laboratory-compacted samples. However, long-term aging properties, simulating 5 to 10 years in service, had to be extrapolated from the 2-year data available. More realistic approach to simulate asphalt mixture aging is to subject the asphalt mixtures to various

Table 3: Temperature and time duration for different conditioning types.

Conditioning type	Temperature	Time
Conditioning for mixture design	Varies*	2 hr
Short-term aging	135°C	4 hr
Long-term aging	85°C	5 days

*Mixture's specified compaction temperature and type of mixture (plant-produced, reheated, etc.).

aging conditions, measure the physical properties of the aged mixtures, and then compare with field aged specimens [57, 135, 136].

In recent studies on asphalt mixtures, the researchers indicated that nonuniform field aging of asphalt mixtures over depth and the surface of the asphalt pavement is found to be aged faster than the bottom [34, 137]. Embrittlement of asphalt mixture due to aging was reported by Rahmani et al. [38] and Elwardany et al. [138]. Brittleness increases with conditioning period under all aging modes, and over time in in-field service conditions [139]. As a result, fatigue cracking resistance and durability of asphalt mixtures is affected, which would be more notable as the temperature increases [140]. Gao et al. [141] showed that degradation in asphalt mixture elastic modulus increases with the increase in aging period. Azri and Mohseni [142] showed that different asphalt mixtures age in very different ways, and this will significantly affect their short-term and long-term rutting performances. Aging increased the permanent deformation resistance in terms of flow number, as reported by Islam et al. [139] and Babadopulos et al. [143].

3.2.1. Asphalt Mixture Aging Simulation Protocol. The current practice recommended by the American Association of State Highway and Transportation Officials (AASHTO) is to cure asphalt mixtures for a few hours and days for short-term and long-term aging, respectively. The test procedure, based on the work done by Von Quintus et al. [90], covers three types of conditioning and presented in the AASHTO R30 standard procedure [144]:

(i) Mixture conditioning for volumetric mixture design
(ii) Short-term conditioning to simulate the aging that occurs during mixture mixing and placement
(iii) Long-term conditioning to simulate aging that occurs after the construction process and over the life of pavement

In this standard practice, the mixture is conditioned in a forced-draft oven for different periods of time and at different temperatures, as shown in Table 3.

3.2.2. Short-Term Aging Protocol. Research works have been conducted to evaluate the short-term aging protocol to simulate the aging of asphalt mixture during the production, laying, and construction of asphalt pavement layer. Although the performance test results of lab and plant-produced mixture were not an exact match, there was a general acceptance in the past that the laboratory aging was representative of field aging [145]. However, due to recent development in asphalt technology and changes in mixture components, mixture processing, and plant design, validity of current mix design methods in order to meet performance expectations is questioned.

In a comprehensive study, Bell et al. [57] evaluated the aging of asphalt mixture and found that the AASHTO short-term aging protocol simulates asphalt mixture aging adequately well except few conservative predictions. Research conducted by the University of California at Berkeley in conjunction with Oregon State University and Austin Research Engineers, Inc. [57] also found the protocol to be adequate based on resilient modulus and indirect tensile test results. Aschenbrener and Far [18] conducted extensive investigation throughout Colorado, conditioned the mixtures at field compaction temperature for different durations (0–8 hours), and found the short-term aging protocol equivalent to 2–4 hours based on theoretical maximum density and asphalt absorption and 1–3 hours based on Hamburg wheel tracking test results. The researchers recommended conditioning the laboratory-produced mixtures for 2 hours at the field compaction temperature in order to simulate asphalt aging and absorption during plant production. Epps Martin et al. [31] also evaluated different short-term aging protocols, and the final recommendation was to condition lab mixture at 135°C for 2 hours before compaction.

3.2.3. Long-Term Aging Protocol. Researchers have used different conditioning procedures (extended heating, oxidation, and UV/Infrared treatment) to investigate the long-term aging of asphalt pavement. Conditioning may also be done either on loose mixture or on the compacted specimen. Oven aging on compacted specimen is commonly used to simulate the long-term aging of asphalt mixtures. However, in the compacted specimen, existence of oxidation gradient in radial direction and along the height of specimen was reported [27]. Hence, researchers [90, 138, 146–149] sometimes preferred conditioning loose mixture at elevated temperature due to homogeneity and efficiency of aging. However, compacting loose mix conditioned sample often was found to be problematic as mixture become too stiff due to loss of volatility of binder [148]. A significantly high number of gyration thus higher shear stress was required to compact loose mixtures specimen resulting in degradation in the aggregate structures hence changes the mixture properties [148, 150]. The temperature at which aging is executed is also important. Higher temperature (>95°C) might cause slump/distortion and affects air distribution in the compacted specimen [148]. Loose mixture aging temperature more than 135°C results in a significant change in the relationship between asphalt binder rheology and chemistry and affects the mixture performance [151]. The optimal loose mixture aging temperature of 95°C is suggested by the researchers [138].

A number of studies [90, 138, 146, 147, 149, 152] showed that the long-term aging protocol can vary depending

Table 4: Studies on the simulation of long-term aging of asphalt mixture.

References	Aging	Findings
Bell et al. [57]	0, 2, 4, and 8 days at 85°C 1, 2, and 4 days at 100°C	2 days at 85°C or 1 day at 100°C = 1–3 years field aging 8 days at 85°C or 4 days at 100°C = 9 years of field aging
Brown and Scholz [154]	4 and 5 days at 85°C	5 days at 85°C simulates long-term aging of UK pavements; 4 days at 85°C simulates 15 years old pavement in the US
Harrigan [26] and Houston et al. [27]	5 days at 80, 85, and 90°C	5 days at 85°C = 7–10 years of field aging
Epps Martin et al. [31]	1 to 16 weeks at 60°C	4–8 weeks at 60°C = first summer of field aging
Islam et al. [139]	1, 5, 10, 15, 20, and 25 days of oven aging at 85°C	1-day laboratory aging is close to 1-year of field aging
Yin et al. [155]	2 weeks at 60°C, 3 days at 85°C and 5 days at 85°C	2 weeks at 60°C = 7–12 months field aging 5 days at 60°C = 12–23 months field aging
Sirin et al. [29]	0, 3, 7, 15, 30, 45, 60, 90, and 120 days at 85°C on compacted specimen	45 and 75 days at 85°C = 5 years field aging in Middle East condition for wearing and base course, respectively
	0, 1, 2, and 3 days at 135°C on loose mixtures	2-3 and 1-2 days at 135°C = 5 years field aging in Middle East condition for wearing and base course, respectively

on the climatic conditions, laboratory aging method, laboratory aging temperature, or asphalt type. Furthermore, most of these studies only estimate the long-term aging of asphalt mixture without proper validation with field results, especially in the component level. The standard protocol to simulate the field aging is conditioning the compacted specimen at 85°C for 5 days in accordance with AASHTO R30.

The protocol uses a single temperature and does not account for different environmental conditions or mix properties. Thus, the applicability of the protocol to different climatic conditions (e.g., like in the Middle East) is questionable without field validation. Asphalt pavement experiences severe weather conditions at high temperatures (often exceeds 40°C during the summer months) in the Arabian Gulf region. Additionally, there is no precipitation during the summer and very little during the remainder of the year. These elevated temperatures increase binder oxidation significantly, which could lead to fatigue cracking and eventually pavement failure with heavy and repeated traffic loading. Previous studies also illustrate a need to develop an aging protocol that considers climate conditions, traffic volume, and mix properties [27, 57, 136, 153]. These studies recommended considering these changes while in the design stage to better improve performance analysis of asphalt pavements.

Table 4 presents the key researches focused on the long-term aging simulation protocol. Bell et al. [57] included different climate zones to evaluate the long-term aging protocol of asphalt mixtures. Experimental results suggested conditioning the compacted specimen for 2 days at 85°C or 1 day at 100°C to simulate long-term aging of new pavements (1 to 3 years old). Mixture needed to condition for longer time (4 to 8 days for 85°C or 2 to 4 days for 100°C) to predict aging of 9-10 years of field aging. However, the authors suggested avoiding the higher temperature of 100°C since conditioning the mixtures at this temperature could cause damage to the specimens. More importantly, the researchers recommended further research to achieve better validation and simulation for a wider range of climate zones. The researchers also recommended developing a model to simulate field aging using inputs that describe climate zones and traffic. Possible inputs could include traffic volume, maximum and minimum air temperature, average rainfall, age of pavements, and age of laboratory mixtures.

Romero and Roque [156] indicated that the use of the long-term aging procedures involving compacted mixes may not be better than the currently used short-term oven aging procedures and therefore long-term oven aging using the compacted asphalt samples should be discontinued. Houston et al. [27] performed a long-term aging study for different sites across the United States and for different aggregates and binders. The researchers considered conditioning the specimen at multiple temperatures (80°C, 85°C, and 90°C) for 5 days. High variability in the data from the selected sites were reported, and due to this variability and inability to account for various variables such as environmental conditions and mix properties, the researchers were not able to develop a new procedure or revise the current one for long-term conditioning of asphalt mixtures. It was concluded that the current standard procedure is not sufficient to truly simulate and predict the long-term aging of asphalt mixtures in the field. Developing a new procedure that accounts for different environmental conditions and mix properties such as air void content is highly desirable. In addition, they recommended including different types of materials: unmodified binders, modified binders, rubber binders, and reclaimed asphalt pavement. In a recent study, Yin et al. [156] proposed long-term aging protocols of 2 weeks at 60°C and 5 days at 85°C produced mixtures with equivalent in-service field ageing of 7–12 months and 12–23 months, respectively, considering WMA technology, recycled materials, aggregate absorption, polymer modified binder, and production temperature. Sirin et al. [29] indicated a severe aging of asphalt pavements in Middle East region due to harsh environmental condition. For such condition, it would require 45 and 75 days at 85°C on the compacted specimen to simulate 5 years field aging for wearing and base course, respectively. To avoid such a long conditioning period, the researchers suggested conditioning of the loose mixture as an alternative and found that it would take 2-3 and 1-2 days at 135°C to simulate the same level of aging for wearing and base course, respectively.

4. Antioxidant Additives

The control of asphalt aging is important because aging causes stiffening and brittleness that can lead to cracking and premature failure of asphalt pavement. As discussed in previous sections, there are several asphalt hardening mechanisms. Oxidation during asphalt mixture production, compaction, and in service is a major one and believed to be the most understood and the easiest to simulate in the laboratory [157, 158]. Therefore, researchers attempted to reduce/minimize oxidative hardening using chemical additives in order to obtain a longer lasting pavement and substantial savings in life cycle cost.

Additives that are used to modify asphalt and retard age hardening are called antioxidants. When antioxidants are added to asphalt as modifiers, they control oxidation by trapping or scavenging free radicals which are responsible for initiating and/or propagating oxidation. These antioxidants (e.g., lead diamyldithiocarbamate (LDADC)) act as sacrificial species that get oxidized instead of the asphalt binders [158, 159]. Some other antioxidants function by reacting with polar compounds and/or oxidation catalysts such as metals present in asphalts.

There are many antioxidants available for asphalt bitumen in the market. Based on the mode of controlling oxidation, antioxidants may be classified into four main groups: primary antioxidants, secondary antioxidants, metal chelators, and light stabilizers [158, 160]. The primary antioxidants have reactive OH or NH groups and function as free radical scavengers by either donating or accepting electrons from free radicals and thereby breaking the oxidation chain reactions. The secondary antioxidants include sulfur and phosphorous compounds such as sulphides, thioesters, disulphides, and phosphates. They function as peroxide or hydroperoxide decomposers by reducing them to stable compounds. Metal chelators operate by trapping trace metals such as vanadium, nickel, and iron which are believed to accelerate the formation of free radicals by acting as catalysts to the propagation step [160]. Finally, light stabilizers are used to prevent degradation by absorbing harmful radiant energy.

4.1. Studies on Antioxidant Additives to Retard Aging of Asphalt Mixture. Several studies reported the benefits of using antioxidant-modified binders. Although most of these studies are fairly old, some antioxidants (i.e., hydrated lime, lead antioxidants, and carbon black) hold promising results [50, 88, 160–164]. In these studies, the researchers used different additives to retard the oxidative hardening of asphalt binders and evaluated antioxidant systems by determining the degradation of asphalt physical properties, mainly the viscosity and ductility. However, most of these systems for retarding oxidative hardening have not performed satisfactorily in the field due to problems such as degradation, volatility, and loss of the antioxidant from the asphalt system.

A few recent studies were conducted to examine the effect of using antioxidant additives on binder performance (Table 5). Mohamed [165] evaluated the potential of CRABit (CR30 and CR50) as antioxidant modifier for use in dense asphalt mixtures (ACW14). The researcher conducted the study in two phases; first phase was to test the rheological characteristics of the new product by using wet mix through dynamic shear Rheometer (DSR) and the second phase included preparation of ACW14 mixture containing base and modified bitumen by dry mix and testing them to determine fundamental properties (i.e., resilient modulus, indirect tensile, creep, and fatigue resistance) before and after aging the specimen. The researcher found an improvement in the engineering properties and the performance with the modification, particularly with CR30.

Apeagyei et al. [166] evaluated the cracking potential of asphalt mixtures containing various levels of antioxidants. The researchers considered two levels of aging in a forced-draft oven to simulate short-term and long-term oven aging (STOA and SLOA, resp.) conditions. In addition, two levels of antioxidant modification were used: furfural (an aromatic aldehyde) and Dilauryl thiodipropionate (DLTDP—an antioxidant and thermal stabilizer) with the asphalt binders. Mixing percentages varied from 0.2% to 10% w/w of the base asphalt were incorporated into the base asphalt using a Barnant blender with a 2-inch blade operating at a speed of 750 rpm. The results showed that antioxidant-modified asphalt mixtures performed better than those with unmodified asphalt mixtures. The antioxidant-modified binders showed at least about 50% lower flexural stiffness relative to an unmodified asphalt binder at a low temperature (about −4°C to about −58°C), which indicated improved fatigue resistance. The modification also found to yield at least about 18% higher stiffness relative to an unmodified asphalt binder at a high temperature (from about 46°C to about 82°C) indicating better rut resistance. In a separate study, Apeagyei [167] evaluated AOXADOUR as an antioxidative additive with PG 64-22 base binder and found higher dynamic modulus, improved rut resistance in terms of creep compliance, higher tensile strength at low temperature (−10°C), and less effect of aging on fracture behavior for both STOA and LTOA conditioned specimen. AOXADUR-modified asphalt mixture showed less reduction in predicted life with aging and took longer time to critical cracking compared to control (Figure 7).

Table 5: Studies on antioxidant additives.

References	Antioxidant	Properties evaluated	Findings
Mohammed [165]	CRABit (CR30 and CR50)	Resilient modulus, indirect tensile strength, creep, and fatigue resistance	Improvement in the engineering properties and the performance with the modification, particularly with CR30
Apeagyei et al. [166, 167]	Furfural and DLTDP; AOXADUR	Cracking potential, dynamic modulus, creep compliance, and tensile strength	Antioxidant-modified asphalt mixtures performed better than those with unmodified asphalt mixtures
Apeagyei [59]	DLTDP/furfural, hydrated lime, vitamin E, carbon black, Irgafos P-EPQ, and Irganox 1010	Stiffness modulus, cracking potential	Furfural and DLTDP additives provided a 40 percent reduction in aging compared to unmodified binders. Antioxidant-modified binders had a lower stiffness modulus and flexural stiffness compared to untreated binders
Reyes [168]	Vitamin E as an antioxidant modifier; hydrated lime and fly-ash as stabilization agents	Viscosity, stiffness, fatigue resistance, rutting potential	Vitamin E–modified binder showed better resistance against fatigue cracking but there is concern on rutting resistance
Pan et al. [52]	Coniferyl-alcohol lignin	Viscosity and ductility	Coniferyl-alcohol lignin can slower oxidation and hardening
Williams [169]	Agriculturally derived lignin-containing ethanol coproducts 3–12%	Rheological properties from DSR and BBR	Lignin-containing coproducts showed beneficial antioxidant activity and stiffened the binder at all stages of aging
University of Illinois at Urbana-Champaign [158]	AOXADUR which consists of three additives: aldehyde, thioester, and a caltalyst.	Stiffness, thermal stress and cracking potential	Dramatic increase in high-temperature stiffness and a substantial decrease in low-temperature stiffness
Dessouky and Diaz [170]	Copolymers solution ethylene-butylene/styrene (SEBS) and solution styrene-butadiene rubber (SSBR) with enhanced antioxidant agents	Brittleness	Copolymers improved the rutting and moisture resistance of the modified asphalt mixtures but they decreased the fatigue life compared to the control mixture

Combinations of various antioxidants were also evaluated by Apeagyei [59] to determine whether synergistic behavior existed between any of the antioxidants. These additives included DLTDP/furfural, hydrated lime, vitamin E, carbon black, Irgafos P-EPQ, and Irganox 1010. The DSR was used to examine the rheological properties of untreated and antioxidant-modified binders. The findings of this study illustrate that the combination of furfural and DLTDP additives had the lowest aging index compared to the other modifiers. This specific combination provided a 40 percent reduction in aging/hardening compared to unmodified binders. In general, the antioxidant-modified binders had a lower stiffness modulus and flexural stiffness compared to untreated binders, which is expected to have better cracking resistance. The author recommended further research to validate the results using additional binders and to evaluate the properties of both asphalt mixtures and binders.

Reyes [168] assessed the potential of using vitamin E as an antioxidant modifier with two types of binder: unmodified (PG 64-22) and modified (PG 70-22). As vitamin E has low viscosity, two calcium-based additives (Hydrated Lime and Fly-Ash) were used as stabilization agents to increase the stiffness of the binder. The researcher used high shear rate blender at 2100 rpm for 1 hour for mixing each samples. The results of this study show that the use of vitamin E reduced the viscosity of the binders. Also, the use of stabilization agents such as fly ash and hydrated lime improved the stiffness of the antioxidant vitamin E–modified binders. The modified binders with vitamin E had desirable characteristics that would resist fatigue cracking; however, there was a concern about rutting resistance. The antioxidant vitamin E–modified binder had reduced stiffness modulus and increased phase angle. The author suggested that experiments be conducted to determine optimum antioxidant and stabilization agent percentages to achieve better performance with aging.

Pan et al. [52] performed an atomistic-based chemo-physical analysis to facilitate the fundamental understanding of the aging and antioxidation mechanisms and thereby to develop antiaging strategies. In this study, the chemical and physical bases of asphalt oxidation as well as the anti-oxidation mechanism of coniferyl-alcohol lignin was investigated. The researchers developed a quantum chemistry-based chemophysical environment and studied various chemical reactions between asphalt components and oxygen and the resulting physical changes on the contrary to the

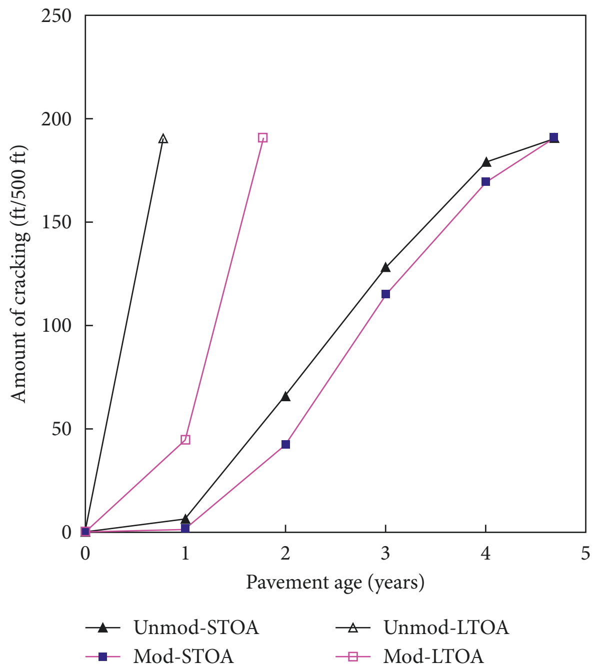

Figure 7: Effect of aging on thermal cracking of AOX-modified and unmodified asphalt mixtures (from Apeagyei [167]).

traditional evaluation method of the degradation of asphalt physical properties (i.e., viscosity and ductility). Two distinct stages of asphalt aging were identified; asphalts initially exhibit a high chain-breaking trend and a high reactivity with oxygen, causing a rapid spurt in the formation of light-molecular-weight alkanes, ketones, and sulfoxides, and the spurt is followed by a slower rate of oxidation and hardening. The authors suggested that coniferyl-alcohol lignin can be used as antioxidant for petroleum asphalt, with the maximum radical-scavenging effectiveness achieved in a nonoxidative condition of the lignin (e.g., <130°C under 1 ATM oxygen partial pressure).

Williams [169] evaluated the potential of agriculturally derived lignin-containing ethanol coproducts for use as antioxidant in asphalt binder. The researcher used four coproducts mixed with four different types of asphalt binders in the range of 3–12% resulting in 52 treatment combinations. Three coproducts contained lignin processed from corn where the fourth one had its lignin removed and acted as a control to measure the antioxidant activity of the three other lignin coproducts. The performance testing of each combination consisted of DSR and bending beam Rheometer (BBR) testing coinciding with field simulative aging using a RTFOT and PAV. The results showed that the lignin-containing coproducts had beneficial antioxidant activity and stiffened the binder at all stages of aging. The researcher suggested that more separation testing should be conducted to evaluate the effect of variables such as physical size and chemical composition of the coproducts.

Depending on climatic condition, two distinct types of phenomena due to aging is observed in asphalt pavement. At low temperature, stiffness of asphalt is increased, and as a result, flexibility of the asphaltic concrete reduces, causing cracking in pavement due to fatigue or thermal stresses. Higher temperature, on the other hand, softens the asphalt and consequently reduces the stiffness of asphaltic concrete making the mix more susceptible to rutting. Some antioxidant treatments are promising in reducing binder stiffness but still prone to softening at higher temperature, stiffening at lower temperatures or leaching out over time.

In 2006, a team of researchers at the University of Illinois at Urbana-Champaign produced an antioxidant treatment using AOXADUR which consists of three additives: aldehyde, thioester, and a caltalyst. A condensation reaction of aldehyde with asphalt to form novolacs, which can act as antioxidants, results in a reduction of age-susceptible polar aromatics in the binder. The thioester serves as a secondary antioxidant, which is highly effective against oxidative degradation of hydrocarbons. Laboratory testing of over 40 binders at the University of Illinois showed that the AOXADUR-modified binder produces the lowest aging index and a dramatic increase in high-temperature stiffness and a substantial decrease in low-temperature stiffness. The researchers reported improvement in binder properties at both high and low temperatures results in less thermal stress and reduced cracking potential.

5. Conclusions

The following are the main points discussed in this paper.

(i) Asphalt aging is a complex phenomenon and affects performance of asphalt pavement by causing functional damage to asphalt. It is generally defined as change in the rheological properties of asphalt binders/mixtures due to changes in chemical composition during the construction and its service life period. Aging is influenced by intrinsic and extrinsic variables: intrinsic variables include asphalt binder mixture types, aggregate, void content, and film thickness and extrinsic variables are mixing temperature and environmental conditions. Aging affects asphalt pavement in many ways and makes it embrittle, of reduced damage tolerance and less durable. As a result, pavement becomes susceptible to disintegration and cracking failures at low temperatures.

(ii) Complex molecular structure of asphalt and its chemical components are changed as a result of exposure to temperature variation and atmospheric conditions resulting in alteration in asphalt properties. The main mechanisms for asphalt aging are identified as oxidation, volatilization, and steric hardening. During construction, asphalt binder is subjected to higher temperature which causes aging due to oxidation and loss of volatile compounds. On the contrary, long-term aging during service periods takes place at lower temperature primarily due to oxidation mechanism. Steric hardening occurs during long-term aging at relatively lower temperature.

(iii) Extended heating by thin film oven and oxidation by air blowing are main methods to simulate aging of asphalt binder in laboratory environment. The most commonly used tests for simulating asphalt binder aging are the RTFOT and PAV tests. In this process, an asphalt binder to be evaluated is to be subjected RTFOT for short-term aging at 163°C for 85 minutes followed by a PAV process at 85°C for 5 days in order to simulate several years of field aging.

(iv) The standard protocol to simulate aging of asphalt mixture is to cure asphalt mixtures for 4 hours at 135°C for short-term aging and 5 days at 85°C for long-term aging. However, these standard aging protocols have limitations and cannot be applied for different environmental conditions. Therefore, development and validation of a new aging simulation procedure that accounts for different environmental conditions and mix properties such as air void content is highly desirable.

(v) Various antioxidant additives have been used to retard aging of asphalt pavement and thereby improving performance of flexible pavement and substantial savings in life cycle cost. One of the most desirable properties of asphalt mixture is to perform well at higher temperature against rutting as well as at lower temperature against cracking due to fatigue. Results from experimental research indicated that several additives performed well at higher temperature while showing poor performance at lower temperature or vice versa. Further research on a variety of antioxidant additives is warranted in order to obtain a more effective and sustainable asphalt mixture that can perform equally well both at high and low temperatures.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

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