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Rheological, microscopic, and chemical characterization of the rejuvenating effect on asphalt binders

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HIGHLIGHTS

- The rejuvenating effect for aged binders was thoroughly evaluated at both macro- and micro-scales.
- Results suggest that changes in chemical compositions contributed to mechanical properties variations.
- Characterization of the rejuvenating effect benefits the effort for better recycling bituminous materials.

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ABSTRACT

With the increasing environmental awareness and rising costs of virgin binders, reclaimed asphalt pavement (RAP) has been used as an alternative for energy and cost saving in asphalt pavements. However, RAP binders have been aged to different extents during pavements' service life and adding rejuvenating agents provides a practical means for restoring the mechanical properties of the aged binders reducing the needed additional virgin binder. In many studies, the rejuvenating effect has been evaluated in terms of the improvement of rejuvenated binders' rheological properties whereas the fundamental rejuvenation mechanism remains unclear. In this research, two different asphalt binders from the Materials Reference Library of the Strategic Highway Research Program (SHRP) were aged, and rejuvenated by complete blending with two commonly used rejuvenators. The rheological properties of the virgin, aged, and rejuvenated binders were tested using the dynamic shear rheometer and the bending beam rheometer. Furthermore, in order to better understand the rejuvenating effect, surface microscopic properties and chemical composition of the binders were measured using atomic force microscopy (AFM) and SARA (Saturates, Aromatics, Resins, Asphaltenes) fractionation, respectively. Results indicated that the bulk mechanical properties (complex modulus and viscosity) of the rejuvenated binders were in between those of the virgin and aged binders. Aging and rejuvenation led to morphological changes as compared to their virgin binders; however, the rejuvenated binders did not always reproduce the microstructures of the virgin binders. Microscopic measurements on adhesion and dissipation of virgin, aged, and rejuvenated samples were qualitatively consistent with the bulk rheological results. SARA separation results suggested that changes in chemical fractions were responsible for the stiffening effect of aging and the improvement of mechanical properties with the addition of the rejuvenators. Such a systematic approach of characterizing the rejuvenating mechanism will benefit the effort of producing more sustainable RAP-containing asphalt pavements.

1. Introduction

As of 2011, more than 40 state agencies in the US allow to use above 30% reclaimed asphalt pavement (RAP) in mix design of Hot Mix Asphalt (HMA) [1], however, currently the average RAP use is still between 10% and 20% [2]. According to the Superpave mix design specification (AASHTO M 323), when more than 15% RAP is used in the mix design, it is required to reduce the binder performance grade (PG) by one to compensate for the aged RAP binder. Furthermore, when adding more than 25% RAP, virgin binder PG has to be determined based on the properties of extracted RAP binder. Such requirements are set to ensure that the virgin binder compensates for the deteriorated mechanical properties of the

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RAP binder in order to avoid cracking failures in the asphalt pavement. However, this also leads to additional expenses for contractors in terms of purchasing unconventional binder grade, installing additional hot storage tanks, maintaining a laboratory with extraction and testing equipments or outsourcing of the required additional testing. In many cases, this discourages the contractors to produce high RAP content mixtures because the savings from a relatively small increase in RAP content are outweighed by the increased expenses.

The use of rejuvenators (i.e., products that are used for restoring the mechanical properties of aged binders) is a relatively poorly understood alternative for mix design of RAP-containing HMA. This is primarily due to concerns associated with their ability to diffuse in the RAP binder and to provide the required long term performance for another service period of the pavements [3]. However, introducing rejuvenators provides major benefits as compared to merely bumping the virgin binder grade as follows:

- Unrestricted RAP content using a single rejuvenator;
- Cheap storage, since in most cases rejuvenators do not require heating;
- Simple addition to the mixture using volumetric pump or existing liquid additive dosage system;
- Ability to add the precise required dose based on the RAP binder properties.

These advantages would help increase the average amount of RAP used in HMA asphalt pavements if the rejuvenating mechanism is better understood. Previous studies suggest that rejuvenators should replenish the volatiles and light chemical fractions that have been lost during aging of pavements [4] by providing a homogeneous system where asphaltenes are well dissolved and prevented from precipitation or flocculation [5]. Other research has shown that a better rejuvenating effect can be attained with high amounts of resin or aromatic fractions [6]. However, a systematic approach of exploring how the rejuvenator modifies the chemical, microscopic and mechanical properties of the aged asphalt binders is still absent.

In this study, two virgin asphalt binders from different crude sources were aged using both Rolling Thin Film Oven (RTFOT) and Pressurized Aging Vessel (PAV) methods. In order to verify the capability of rejuvenators to restore the mechanical properties of the aged binders, two different rejuvenators were added into the aged binders. For better understanding of the rejuvenating mechanism at both macro and micro scales, rheology, surface microscopic properties, as well as chemical fractions of the virgin, aged and rejuvenated asphalt binders were characterized using dynamic shear rheometer (DSR), bending beam rheometer (BBR), atomic force microscopy (AFM), and SARA (Saturates, Aromatics, Resins, Asphaltenes) fractionation.

2. Materials and methods

2.1. Materials

2.1.1. Virgin asphalt binders

Two types of asphalt binders, AAD (PG 58–28) and ABD (PG 58–10), were obtained from the Materials Reference Library of the Strategic Highway Research Program (SHRP). They were chosen because of variations in their crude source, SARA fractions, chemical elemental analysis, physical and mechanical properties, as indicated in Tables 1 and 2. Between the two virgin binders, AAD is a "softer" binder with a higher wax content and higher asphalthene content in comparison to ABD.

Table 1
Crude source, SARA fractions, and elemental analysis of the asphalt binders according to reference [7].

Binder Crude source		AAD (PG 58–28) Califonia Coastal	ABD (PG 58–10) California Valle
SARA fractions (%)	Saturates	8.6	10.4
	Aromatics	25.1	28.4
	Resins	41.3	52.7
	Asphaltenes	23.9	10.2
Elemental analysis	Carbon	81.6%	86.8%
	Hydrogen	10.8%	10.7%
	Oxygen	0.9%	1.2%
	Nitrogen	0.8%	1.2%
	Sulfur	6.9%	1.6%
	Vanadium	310 ppm	62 ppm
	Nickel	145 ppm	123 ppm
	Iron	13 ppm	54 ppm
Wax content (%)		1.94	0.81

Table 2
Physical and viscoelastic properties of the asphalt binders [7].

Aging state	Property	AAD binder	ABD binder
Unaged	Viscosity at 135 °C	3.09 poise	2.41 poise
	Viscosity at 60 °C	1055 poise	2112 poise
	Penetration at 25 °C	135 × 0.1 mm	47 × 0.1 mm
	Softening point	47.8 °C	48.9 °C
	$G^*/\sin\delta$ at 58 °C	1.47e−3 MPa	2.68e−3 MPa
RTFO residue	Mass change	−0.81%	−0.12%
	$G^*/\sin\delta$ at 58 °C	4.29e−3 MPa	4.74e−3 MPa
PAV residue	$G^*\cdot\sin\delta$ at 20 °C	2.8 MPa	n/a*
	$G^*\cdot\sin\delta$ at 30 °C	n/a*	4.1 MPa
	Stiffness at −10 °C	83 MPa	279 MPa
	m-value at −10 °C	0.38	0.28

* Data are not available.

2.1.2. Rejuvenators

Two generic rejuvenators, aromatic extract and waste vegetable oil (WV oil), were used in the study. Table 3 shows their viscosity and specific gravity measured by the authors and other relevant properties obtained from the manufacturers. A brief description of the two rejuvenators is also provided below. Based on previous studies [8,9], 12 wt.% of each rejuvenator was added to the binders.

Aromatic extract is a traditional rejuvenator with dominant polar aromatic molecules. According to the manufacturer, aromatic extract contains approximately 75% of aromatic oil and resin compounds with small amount of saturate oil. Chemical compositions of the aromatic extract were determined in this study using SARA fractionation and discussed later (Section 3.3).

WV oil is increasingly used for bio-diesel production. It is derived from fast and convenience food frying oil and also referred to as "yellow grease". This product usually has low free fatty acid content (<15%), and MIU (Moisture, Impurities, Unsaponifiables) (<2%) [11]. According to the manufacturer, WV oil used in this study consists predominately of peanut, sunflower, and canola oils, with large concentrations of oleic and linolic acids.

2.2. Methods

2.2.1. Binder aging

The two virgin asphalt binders were aged using both Rolling Thin Film Oven (ASTM D2872-2004) and Pressurized Aging Vessel (ASTM D652-2008) methods. The combined aging procedures are expected to simulate the field aging of asphalt binders at later stages of pavement life when asphalt has to be removed and needs