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Effect of recycling agents in recycled asphalt binders observed with microstructural and rheological tests

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HIGHLIGHTS

- Performance of rejuvenated binders was investigated by AFM and DSR.
- AFM provided indications about molecular mobility and degree of association.
- Surface roughness, phase contrast and dispersed domain appearance were considered.
- Improved flow properties due to recycling agents were observed with DSR and AFM.
- Aging of rejuvenated blends caused lower molecular mobility and higher brittleness.

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ABSTRACT

In this study, the effect of recycling agents on recycled blends obtained from base and recycled asphalt binders and the performance of the rejuvenated binders with aging was investigated by means of Atomic Force Microscopy (AFM) and Dynamic Shear Rheometer (DSR). This study arises as the amount of recycled materials in asphalt pavements continues to increase. Recycled binders are significantly stiffer than base binders; however, the performance grade can be restored by inclusion of recycling agents. Nevertheless, intermediate temperature cracking performance evaluated by the Glover-Rowe (G-R) parameter indicated that the effect of the recycling agents is not sustained with subsequent aging, which could jeopardize its effectiveness in service. The AFM images provided microstructural information, which offer indications about changes in molecular mobility and degree of association with aging, presence of agglomerations, and the dispersive effect of recycling agents.

1. Introduction

Using reclaimed asphalt pavements (RAP) and recycled asphalt shingles (RAS) in asphalt pavements is a popular practice in the asphalt industry given their environmental and economic benefits. The recycled binder from the RAP is highly aged, and its stiffness depends on the source and grade of the base binder, the climate where the pavement was located, and the period between construction and the time the pavement was reclaimed. Typical sources of RAS include tear-off asphalt shingles (TOAS) and manufacturer waste asphalt shingles (MWAS). TOAS are obtained during re-roofing or roof removal projects and thus represent the largest portion of shingle waste. MWAS consist of the excess material obtained during production, and therefore are less prevalent. The binder in shingles becomes highly oxidized during production (MWAS), before exposure on the roof for 20–30 years under the sun, which in turn causes the binder to be even stiffer and more oxidized (TOAS) [1]. Private and public sectors have invested in the development and implementation of technologies to enable the use of increased amounts of recycled materials in asphalt pavements, implying higher recycled binder ratios (RBR) or proportion

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of recycled to total binder. The RBR is the percentage of recycled binder from RAP and/or RAS by weight with respect to the total binder in the asphalt mixture. Recycled binders are significantly stiffer than base binders; however, their performance grade (PG) can be restored by inclusion of recycling agents, also called rejuvenators [2].

Asphalt binders can be represented by a colloidal model consisting of a highly polar asphaltene phase dispersed in a maltene phase. The concept of binder compatibility refers to the balance between soluble and insoluble fractions in the colloid, which controls its flow properties. A more compatible binder has increased mobility and improved flow properties at a given load and temperature condition. From a rheological perspective, a compatible binder is more ductile, and binder phase angles are higher at any given modulus. The inclusion of recycling agents in recycled binders improve molecular mobility by partially restoring proportions of the lost lighter oils and separating agglomerations caused by aging [3]. Depending on the chemical composition of the recycling agent, the mechanisms by which the recycling agent restores rheological properties may differ. Yet, rejuvenation mechanisms are not fully understood and long-term aging presents challenges to rejuvenation effectiveness [4].

This Atomic Force Microscopy (AFM) study was motivated by the need to further investigate the mechanisms of rejuvenation, reflected as improved flow properties (compatibility) observed in rheology experiments, and the loss of rejuvenation effectiveness with aging. Prior to this AFM study, a separate study by authors of this paper compared SAR-AD™ (Saturates, Aromatics, Resins, Asphaltene Determinator) and Differential Scanning Calorimetry (DSC) experiments to rheology measurements (PG and Glover-Rowe) [5]. While DSC and rheology measurements showed a softening effect upon rejuvenation, the SAR-AD™ compatibility indices did not show clear benefit from incorporation of recycling agents. Findings suggested that the mechanisms of rejuvenation by inclusion of recycling agent T1 may involve a strong polar interaction between the asphaltenes and the fatty acids in the tall oil. SARA, SAR-AD™ and other chemical tools commonly evaluate asphaltenes as molecular agglomerations held together by polar bonds rather that individual molecules [6]. Understanding rejuvenation and aging requires experimental and modeling tools that go beyond quantifying chemical composition. Techniques such as AFM and others, which are capable of providing mapping of the associations/domains within the binder or verifying molecular distribution within the sample, could offer valuable information to further understanding of rejuvenation mechanisms and compatibility.

AFM has been previously applied to investigate the microstructure of asphalt binders, both original and after aging using standard methods, and with novel aging protocols [7–12]. Microstructural investigations with AFM have also been performed on recycled binders and blends of base and recycled binders [13–15]. The surface microstructure of asphalt binders provides information about molecular mobility, as it depends on the molecular interactions among different chemical species, and could offer evidence about the ability of the recycling agents to diffuse into the recycled binders. Rejuvenated binder blends using different recycling agents have been analyzed with AFM in previous studies. Bio-oils were observed to have different rejuvenation effects in terms of microstructure in different aged binders [16]. Nahar, Qiu, Schmets, Schlangen, Shirazi, Van de Ven, Schitter and Scarpas [3] observed that different recycling agents, an emulsion type and a liquid type rejuvenator, caused different microstructures in the same laboratory aged binder, hinting that the mechanism leading to the softening and rejuvenation of the aged binder varies according to the recycling agent type. Yu, Zaumanis, Dos Santos and Poulikakos [17] investigated the microstructure of two laboratory aged binders before and after addition of two recycling agents, an aromatic extract and a waste vegetable oil, and observed that the rejuvenated blends did not necessarily reproduce the microstructures of the virgin binders. The addition of the rejuvenators resulted in more significant morphological changes than the aging effect.

2. Objectives

The objectives of this study were to (1) investigate the effect of blending of base binders with recycled binders extracted from different sources (RAP, MWAS, and TOAS) on rheology and microstructure and (2) explore the effect of different types of recycling agents on these recycled blends before and after aging. The AFM analysis was performed in order to observe and analyze changes in microstructure and possibly detect the presence of agglomerations in the recycled blends with and without recycling agents. The microstructural analysis could clarify the role of different types of recycling agents in breaking and/or dispersing the agglomerations in the recycled blend. In addition, microscopic investigations may help understand the rejuvenation mechanisms observed macroscopically by decreased viscosity and improved ductility of the rejuvenated blends. In addition, this study provides results regarding the properties of the rejuvenated blends after laboratory long-term aging. This is a necessary step in the prediction of the performance of rejuvenated blends in service. Rheological properties were also characterized and compared with the microstructural analysis.

3. Experimental plan and materials selection

Two base binders with different chemical, rheological, and aging characteristics were characterized in this study: a PG 64-22 from Texas (TX) and a PG 64-28 from New Hampshire (NH). The NH PG 64-28 binder was more compatible than the TX PG 64-22 binder based on its wide PG range and good relaxation properties at low temperatures. The NH PG 64-28 binder had a ΔT_c of +1.2 whereas the TX PG 64-22 had a ΔT_c of −4.6. The ΔT_c is the difference between continuous grade temperature for stiffness and relaxation properties (i.e., the critical temperature when S = 300 MPa minus the critical temperature when m-value = 0.30), per AASHTO M 320 specification (reported in Table 1). A high ΔT_c (i.e., positive or less negative) has demonstrated correlation to binder durability [18] and is therefore desirable. Three types of recycled materials were considered (RAP, MWAS, and TOAS) from Texas with a high-temperature PG (PGH) of 107, 133, and 178, respectively. Three types of recycling agents at different dosages by weight of total binder were also considered: a tall oil (T1), which is a residue from the paper industry, a vegetable oil (V1) obtained from corn following ethanol production, and an aromatic extract (A1), which is a petroleum derivative with a composition similar to the polar aromatics of the maltene phase in asphalt binders. Recycled and rejuvenated blends were prepared using a variety of materials at different proportions. Table 1 summarizes the components and characteristics of the binder blends evaluated in this study.

The control recycled blend with up to 30% recycled binders (0.3 RBR) did not require inclusion of recycling agents as indicated in current Texas Department of Transportation (TxDOT) specifications [19]. The use of recycling agents enabled the inclusion of increased amounts of recycled materials (higher RBR). The 0.3 control recycled blend was used as reference to compare against other blends with higher RBR, different RAP/MWAS/TOAS proportion, and various recycling agent dosages. The two distinct TX PG 64-22 and NH PG 64-28 binders were used to produce the 0.3 RBR and 0.5 RBR binder blends.