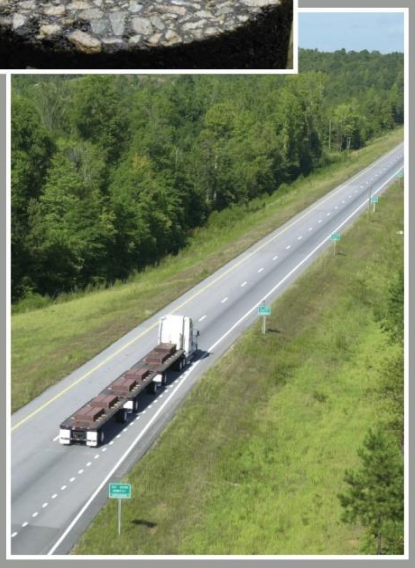

Storage Stability Testing of Asphalt Binders Containing Recycled Polyethylene Materials

A Draft Report *Submitted to*

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Introduction

This report summarizes the results and findings of the storage stability testing of asphalt binders containing recycled plastics [mainly recycled polyethylene (RPE)] that was conducted by the National Center for Asphalt Technology (NCAT) at Auburn University. Storage stability refers to the tendency of RPE to separate from asphalt binders and provides an indication of the degree of chemical compatibility between the two individual components. Storage stability is an important property to ensure the integrity of RPE-modified binders during storage and handling in the field.

The Phase I study by the Asphalt Technologies, LLC found promising results regarding the use of RPE for asphalt modification (*1*). RPE-modified binders showed improved elasticity, rutting resistance, and fatigue resistance as compared to unmodified asphalt binders. However, the study also identified several limitations of using RPE for asphalt modification. For example, the inclusion of RPE reduced the low-temperature cracking resistance of asphalt binders due to increased embrittlement and reduced relaxation properties. Furthermore, it was observed that RPE-modified binders had phase separation issue; the polymers tended to separate from the asphalt binders, due to the difference in density and viscosity as well as incompatibility between the two components, and floated and agglomerated at the top of the modified binders. Therefore, this study was undertaken to evaluate the storage stability of RPE-modified binders and explore the use of compatibilizers in potentially mitigating the phase separation issue.

Experimental Plan

Figure 1 presents the experimental design of the study. Two unmodified asphalt binders with the same performance grade (PG) but from different crude sources were included. Asphalt binders from different sources typically have different chemical composition and may react differently with RPE. Both asphalt binders had a PG of 58-28, which was softer than the two control binders used in the Phase I study (i.e., PG 64-22) and were selected deliberately to counteract the stiffening effect from adding RPE. Binder 1 was from Strathcona Refinery in Alberta, Canada, and Binder 2 was from Ergon Refinery in North Carolina. As per the request of the sponsor, four RPE samples were tested and provided to NCAT in pellet form (as shown in Table 1). Each RPE sample was added at a dosage of 5.0% by weight of the asphalt binder, which is similar to the typical dosage of polymer modifiers used for asphalt modification. Finally, two compatibilizers, which were also provided by the sponsor, were evaluated in terms of their ability to improve the storage stability of RPE-modified binders. One compatibilizer (i.e., CA1) was provided in pellet form while the other (i.e., CA2) was in powder form. Both compatibilizers were included at the recommended dosage provided by the material suppliers; CA1 and CA2 were added at 0.075% and 0.5% by weight of the asphalt binder, respectively. A total of 12 RPE-modified binders were prepared and tested for stability storage in the study.

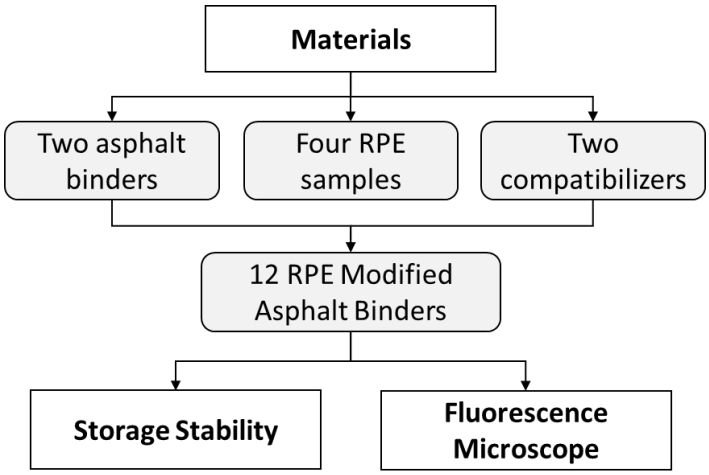
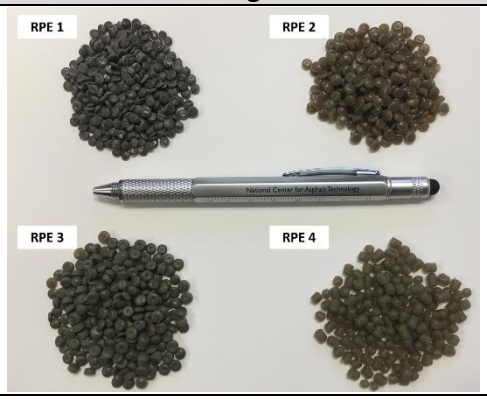


Figure 1. Experimental Design

Table 1. Description of RPE Samples Tested in the Study

Sample ID	Resin Make-up	Form	Images
1	LLDPE+LDPE	Pellet	
2	LLDPE+LDPE	Pellet	
3	LLDPE+LDPE, HDPE	Pellet	
4	LDPE	Pellet	

To prepare an RPE-modified binder, the unmodified asphalt binder was preheated in an oven for two hours at 180°C. In cases where a compatibilizer was used, it was first added to the binder and blended for 10 minutes at 180°C using a high shear mixer (3,000 rpm). Then, the RPE sample was added to the binder and blended for another hour at 180°C. Because the temperature used to prepare RPE-modified binders was higher than the melting point of the RPE samples tested in the study (i.e., 120 to 130°C), all RPE samples were well dispersed in the binders after blending with no coalescence of undissolved RPE observed.

The American Society for Testing and Materials (ASTM) D7131, *Standard Practice for Determining the Separation Tendency of Polymer from Polymer Modified Asphalt*, was used to determine the storage stability of RPE-modified binders. The test procedure is briefly described as follows. A measured quantity of RPE-modified binder in a sealed aluminum tube is conditioned in a vertical position for 48 hours at a temperature of 163°C. After the static heat conditioning, the sample is submitted to a freezing cycle at -10°C for four hours. The top and bottom portions of the sample are then separated and subjected to further testing to determine the degree of separation. The tests chosen for this purpose typically depend on the polymer modification system being evaluated and the type of information desired by the user.

In this study, the softening point test per ASTM D36, *Test Method for Softening Point of Bitumen*, was selected to quantify the difference between the top and bottom portions of RPE-modified binders. Asphalt binder is a viscoelastic material without a sharply defined melting point. It gradually becomes softer, less viscous, and more liquid as the temperature increases. The softening point indicates the tendency of an asphalt binder to flow at elevated temperatures encountered in service and is often used to specify the classification and uniformity of the material. The softening point test uses the ring-and-ball apparatus as shown in Figure 3. Two horizontal disks of asphalt binders are casted in shouldered brass rings and then heated at a controlled rate in a liquid bath while each supports a 3.5 g steel ball. As the temperature increases, the asphalt binders become softer and allow the steel balls to fall because of self-gravity. The temperature at which the steel balls touch the bottom plate of the apparatus is recorded as the softening point. According to Georgia Department of Transportation (GODT) specifications, a difference of 10°C or less in the softening point between the top and bottom samples indicates that the modified binder is storage stable.

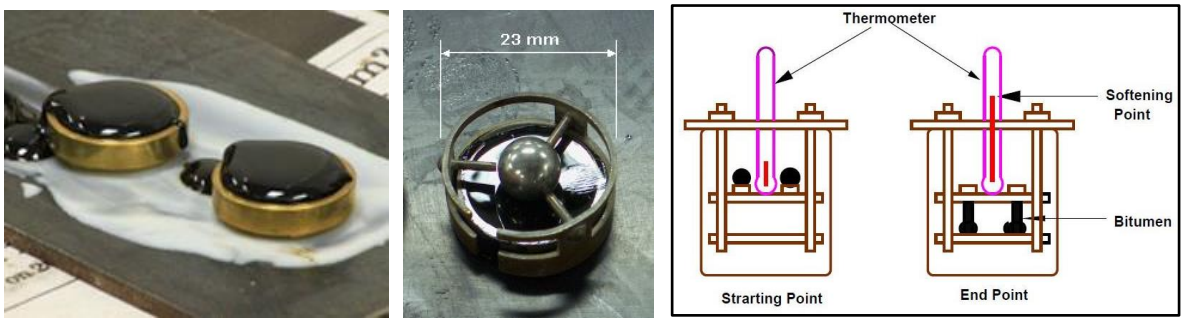


Figure 3. Softening Point Test Apparatus and Illustration

Test Results

Table 2 summarizes the storage stability test results of all RPE-modified binders examined in the study. Blends 1 to 8 refer to RPE-modified binders without compatibilizers, and Blends 9 to 10 and 11 to 12 are modified binders with CA1 and CA2, respectively. As shown in the table, the top sample of all RPE-modified binders had a softening point above 80°C. The specific softening point was higher than the upper temperature range of the ASTM low softening point thermometer (i.e., -2 to 80°C), and thus, could not be determined. On the other hand, the softening point of all bottom samples was in the range of 43 to 50°C, which was significantly lower than that of the corresponding top sample. Considering that the two PG 58-28 asphalt binders had a softening point of 38 and 39°C, all the bottom samples seemed to be slightly modified and contained a small amount of RPE samples. Based on the difference in softening point between the top and bottom samples, none of the RPE-modified binders tested in the study satisfied the GDOT requirement of 10°C or less. Although the four modified binders with compatibilizers (i.e., Blends 9 to 12) showed slightly better storage stability than the corresponding control binders (i.e., Blends 2 and 3), they still had a difference beyond GDOT's maximum acceptable limit.

Table 2. Summary of Storage Stability Test Results

Blend ID	Blend Description	Softening Point, °C			Pass/Fail (Max. 10°C)
		Top Sample	Bottom Sample	Difference	
-	Binder 1	38		-	-
-	Binder 2	39		-	-
1	Binder 1 + 5% RPE1	80+	43	37+	Fail
2	Binder 1 + 5% RPE2	80+	47	33+	Fail
3	Binder 1 + 5% RPE3	80+	48	32+	Fail
4	Binder 1 + 5% RPE4	80+	44	36+	Fail
5	Binder 2 + 5% RPE1	80+	49	31+	Fail
6	Binder 2 + 5% RPE2	80+	44	36+	Fail
7	Binder 2 + 5% RPE3	80+	45	35+	Fail
8	Binder 2 + 5% RPE4	80+	43	37+	Fail
9	Blend 2 + 0.075% CA1	80+	49	31+	Fail
10	Blend 3 + 0.075% CA1	80+	49	31+	Fail
11	Blend 2 + 0.5% CA2	80+	50	30+	Fail
12	Blend 3 + 0.5% CA2	80+	50	30+	Fail

In addition to the storage stability test, fluorescence microscopy was also employed to evaluate the chemical compatibility of two selected RPE-modified binders (i.e., Blend 2 and 3) and characterize the distribution and network formation of RPE particles. Fluorescence microscopy is capable of investigating heterogeneous surfaces where components have different ultraviolet (UV) light excitation responses and has been widely used to analyze polymer distribution in asphalt binders. Figure 4 presents the fluorescent micrograph of the two RPE-modified binders under a Zeiss Axiovert 200 Inverted Microscope. The lighter color represents the polymer (i.e., RPE)-rich phase and the darker color represents the asphalt-rich phase. Both modified binders did not show a uniform distribution of RPE particles. There were several isolated polymer coalescences caused by the physical separation and agglomeration of RPE particles from the asphalt binders. These results further indicated the poor compatibility and storage stability of RPE-modified binders, which agrees with the observations of the Phase I study and existing literature (*1-3*).

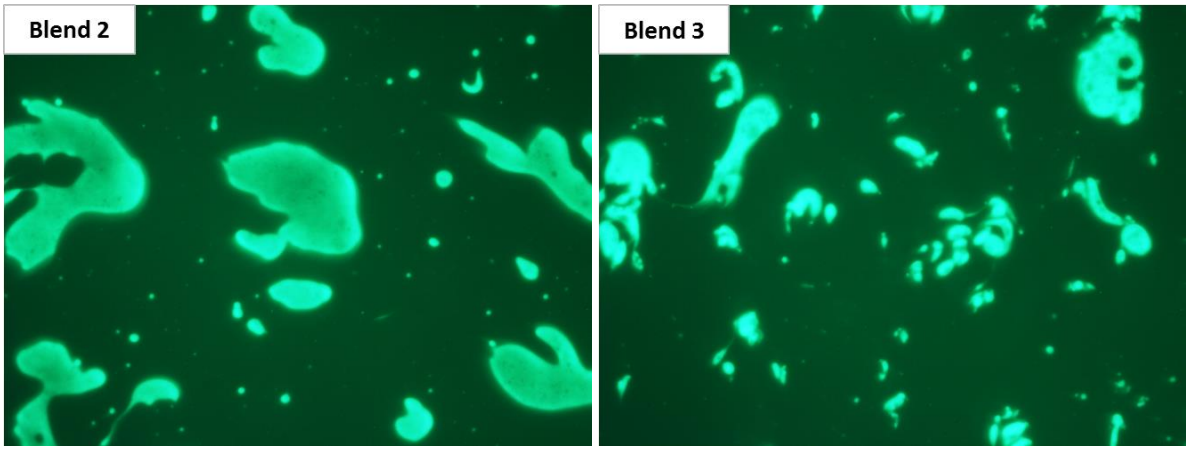


Figure 4. Fluorescent Micrograph of Two RPE-Modified Blends

Conclusions and Recommendations

Based on the results of the study, the following conclusions were made:

- None of the RPE-modified binders tested in the study satisfied the storage stability requirement per GDOT specifications. Phase separation was observed on all tested samples.
- Coalescence of RPE particles was observed in the fluorescent micrograph of two selected RPE-modified binders, which confirmed the poor compatibility between the RPE samples and asphalt binders tested in the study.
- The two compatibilizers evaluated in the study did not significantly improve the storage stability of RPE-modified binders.

It is recommended for future work to evaluate additional compatibilizers to mitigate the phase separation and chemical incompatibility of RPE-modified binders. The research team identified several potential compatibilizers with promising results reported in literature (*4-8*), and recommended the following two for further testing:

- The stabilization of RPE into asphalt binders could potentially be achieved by the addition of a copolymer that would act as a steric stabilizer, hindering the coalescence of RPE particles. Ethylene Acrylate Copolymer is an example of a reactive polymer that could increase the storage-stable behavior of RPE-modified binders (*9*).
- Another potential solution to improve the storage stability of RPE-modified binders is the addition of trans-polyoctenamer rubber (TOR). TOR is a semi-crystalline polyolefin additive known as a facilitator for the dispersion of crumb rubber in asphalt by increasing the surface wetting of rubber particles. Furthermore, TOR enhances the interaction between crumb rubber and asphalt binder via crosslinking reactions (*10*).

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