

Viscosity Mixing Rules for Asphalt Recycling

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Forty-seven aged asphalt-softening agent pairs were blended at multiple levels of aged material content. The relationship between 60°C low-frequency limiting viscosity and aged material mass fraction for 45 of the asphalt-agent pairs can be described using the Grunberg model. The value of the viscous interaction parameter is a strong function of the viscosity difference between the aged asphalt and the softening agent. A normalized Grunberg model was developed to eliminate this dependency. An average normalized interaction parameter can be used to generate a "universal" mixing rule for commercial-type recycling agents. This new mixing rule was compared to the Epps mixing rule and the mixing rule specified in ASTM D4887. Comparison was based on the ability of each mixing rule to predict the quantity of softening agent required to produce blends with a specific target viscosity. It was concluded that for low-viscosity asphalt softening agents, the method specified in ASTM D4887 should be used. However, for supercritical fractions and commercial recycling agents, the universal normalized Grunberg mixing rule developed in this study is superior to the other two mixing rules.

Recycling of asphalt pavements is an environmentally and economically attractive proposition. To recycle an asphalt pavement efficiently, it is necessary to accurately predict the viscosity of the recycled binder or to perform time consuming trial and error blending. Asphalt is not a simple, pure liquid and it is nearly impossible, from a scientific standpoint, to predict the viscosity of a single asphalt, let alone a mixture of asphalts. Asphalts are mixtures of thousands of different chemical compounds, each having a separate and distinct viscosity. Furthermore, composition is not the same from asphalt to asphalt. It may be possible, from an engineering standpoint, to predict viscosity if these chemical compounds are grouped into only a few pseudocomponents. If this logic is followed to its natural conclusion, a mixture of two asphalts or a mixture of an asphalt and a recycling agent can be considered as a binary liquid mixture.

Irving conducted a survey of equations (1) proposed to describe effectively the viscosity of binary liquid mixtures. This survey identified more than 50 equations proposed to predict either the dynamic or kinematic viscosity of binary liquid mixtures. Irving also determined the effectiveness of the various mixture equations (2). Irving concluded that the following equation, proposed by Grunberg and Nissan (3), was the best overall mixing rule in terms of accuracy and simplicity for predicting the viscosity of nonaqueous binary systems.

$$\ln \eta_m = x_1 \ln \eta_1 + x_2 \ln \eta_2 + x_1 x_2 G_{12} \quad (1)$$

The interaction parameter G_{12} is usually considered to be a constant, however, G_{12} may be a function of x_i where x_i may be mole, mass,

or volume fraction. Irving determined that the viscosity of a mixture can be predicted to within 30 percent of the actual viscosity when an average, constant value of G_{12} is used for classes of mixtures (e.g., polar-polar). In addition, Irving's calculations (2) indicate that the choice of units for x_i (mole, mass, or volume fraction) make little difference in the accuracy of the model. Mehrotra has used the Grunberg equation to model bitumen-gas (4) and bitumen-solvent (5) systems. However, very little effort has been focused on using this equation to predict the viscosity of aged asphalt-softening agent mixtures. Instead, the majority of predictions are based on two other models.

The method proposed by Epps et al. (6) closely follows the Roelands mixing rule (7). The nomograph presented by Epps suggests that $\log \log \eta$ for the mixture is a linear combination of $\log \log \eta$ for the pure components in terms of mass fraction or volume fraction and the Roelands model uses $\log \log 10\eta$. Variations of the Roelands model have been proposed for recycled asphalts (8). Although Epps' rule has received much attention, the rule most commonly used to estimate a recycled asphalt binder's viscosity is the procedure specified in ASTM D4887. This procedure, also suggested by the Asphalt Institute (9), is the graphical representation of the Arrhenius equation (10). The Arrhenius equation is a special case of the Grunberg equation with G_{12} equal to zero. Irving (2) concluded that using the Grunberg model with G_{12} equal to zero resulted in errors larger than those obtained using an optimized or average value of G_{12} , if they are available. Large errors may require actual blending to determine a mixture's viscosity. Epps et al. (6) and ASTM indicate that some degree of trial and error blending may be necessary to achieve an accurate viscosity for a recycled binder.

Irving's results (2) indicate that it is possible to use an average interaction parameter for the Grunberg model to describe certain classes of mixtures. The present study was undertaken to determine whether the Grunberg equation can be used to describe aged asphalt-softening agent mixtures and whether an average interaction parameter can be used for aged asphalt-softening agent pairs.

EXPERIMENTAL METHODS

To produce viscosity mixing rules, tank asphalts were artificially aged and then blended with softening agents at multiple aged asphalt contents. Once the aged material had been produced, it was reheated in a laboratory oven and homogenized with a mixing paddle driven by a hand-held drill. Ideally, all of the aged material for a single asphalt was weighed at the same time so that all of the blends would have the same base material. For one asphalt, the sample was reheated, causing the viscosity to change. This viscosity change was taken into account and had no effect on the results of this study.

After the aged material was weighed into tins, the softening agent was added. Each blend contained at least 30 g of the softening agent. It was determined that 30 g would be sufficient for viscosity testing and also minimize problems with the homogeneity of blends. Each blend was mixed using a procedure similar to that specified in ASTM D4887.

The primary property of interest was the 60°C low-frequency limiting dynamic viscosity. All viscosity measurements were performed using a Carri-Med CSL-500 controlled stress rheometer with a 2.5-cm composite parallel plate and a 500 μm gap. The low-frequency limiting dynamic viscosity is obtained when the viscosity does not change with oscillation frequency in controlled stress measurements. To obtain the viscosity for some materials it was necessary to use the time temperature superposition principle (11). The average measured viscosities for the materials examined in this study are given in Table 1.

Compositional analyses of the softening agents were performed via high-performance liquid chromatography (HPLC) analysis using a Waters 712 sample processor and a 600E controller. Separation

was performed using a 125Å $\mu\text{Bondapak-NH}_2$ activated alumina column. The softening agent (asphaltene) content was determined by weighing the n-hexane precipitate, as described by Pearson et al. (12). The saturate content was determined from a calibration of the HPLC refractive index response, and the total aromatic content, the sum of naphthene and polar aromatic contents, was determined by difference (detailed composition data not included).

Infrared spectra were measured using a Mattson Galaxy Series 5000 FTIR with the attenuated total reflectance method as described by Jemison et al. (13). The carbonyl regions of the spectra were used to confirm the validity of the aging procedure for producing large quantities of hardened asphalt.

AGED ASPHALT PRODUCTION

Four asphalts were used in this study. Two of these asphalts were aged to multiple viscosities giving a total of seven aged materials. Three tank asphalts were obtained from the SHRP/LTPP MRL and one from the Coastal refinery in Corpus Christi, Texas. Two samples were aged in a pressure oxygen vessel (POV) at 82.2°C (180°F) and 20.7 bar (300 psia) pure oxygen (14). One sample was produced by aging in a laboratory oven. The majority of the aged material was produced in an air-bubbled (AB) reaction apparatus.

Small amounts of SHRP AAA-1 and SHRP ABM-1 were POV aged. The quantity of POV AAA-1 produced was sufficient to blend with only one softening agent, and the amount of POV ABM-1 was sufficient for blending with two softening agents. The Coastal asphalt was aged in 6-mm (1/4-in.) films on cookie sheets placed in a laboratory oven at approximately 110°C (230°F). The trays were rotated and the asphalt was stirred twice per day to encourage uniform aging. This oven-aged Coastal was blended with four softening agents. To produce the large amounts of material that were necessary for this study, a different aging procedure had to be developed.

An apparatus was built to age large quantities of asphalt in a uniform manner. The apparatus consists of a variable-speed 49.7-W (1/15-hp) motor that drives a mixing shaft 5.1 cm (2 in.) in diameter placed in a half-full gallon can of asphalt. The can is wrapped with a heating tape connected to a variable transformer and a thermocouple-actuated on-off controller. Building air passes through a surge tank, a filter, and a copper coil placed in a mineral oil temperature bath before being fed to the asphalt. The air is introduced to the asphalt through a sparging ring 12.7 cm (5 in.) in diameter made from 6-mm (1/4-in.) stainless steel tubing with 14 nearly uniformly spaced 1.6-mm (1/16-in.) holes. The inlet air temperature is controlled by adjusting both the temperature of the oil bath and the air flow rate. The operating temperature of the AB reaction vessel must be high enough for the oxidation to proceed at an appreciable rate, but not so high as to drastically alter the reaction mechanism or reaction products. Additionally, the temperature must be high enough to soften the asphalt so that the asphalt can be well mixed by the mixing paddle.

SHRP AAA-1 asphalt was aged at 148.9°C, 121.1°C, and 93.3°C (300°F, 250°F and 200°F) to study the effect of aging temperature on the reaction products. Samples were taken periodically to monitor the progress of oxidation. The viscosity and carbonyl areas (CAs) were measured and plotted in Figure 1. The hardening susceptibilities (HSs), defined as $\partial(\ln \eta)/\partial\text{CA}$, were determined and compared to the HS generated from samples aged in the POV. Lau et al. (14) showed that the POV HS is independent of aging tem-

TABLE 1 Representative Viscosities for Aged Asphalts and Softening Agents Studied

| Material | 60°C Viscosity (dPa·s) ^a |
|--------------|-------------------------------------|
| POV AAA-1 | 22,500 |
| AAA-AB7 | 22,900 ^b |
| AAA-AB8 | 36,600 |
| AAF-AB1 | 52,500 |
| AAF-AB2 | 20,900 |
| Oven Coastal | 100,000 |
| POV ABM-1 | 47,200 |
| NUSO 95 | 1.3 |
| Mobil 120 | 1.8 |
| Sun 125 | 3.0 |
| Cyclogen | 8.9 |
| AAF F2 | 12 |
| AAA F2 | 13 |
| YBF F2 | 38 |
| YBF F5 | 47 |
| AAF F3 | 70 |
| AAA F3 | 79 |
| ABM F2 | 98 |
| ABM F5 | 100 |
| YBF F3 | 138 |
| Shell F3 | 165 |
| ABM F3 | 650 |
| DS AC-3 | 310 |
| DS AC-5 | 500 |
| Shell AC-5 | 575 |
| SHRP AAV | 630 |
| SHRP ABH | 900 |

^a 1 dPa·s = 1 Poise

^b Initial value

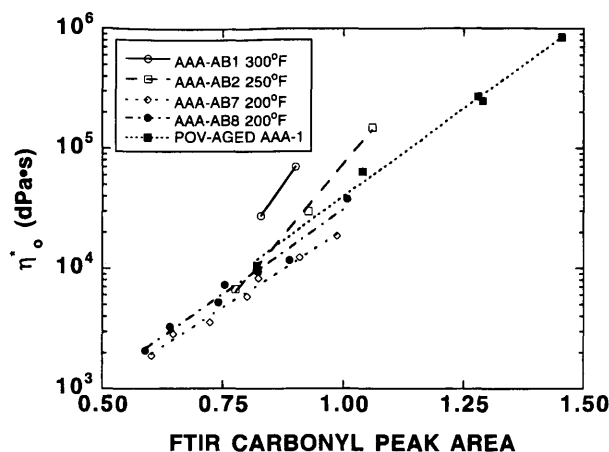


FIGURE 1 Effect of temperature on hardening susceptibility of AB asphalt.

perature for temperatures up to 93.3°C (200°F). In addition, the POV HS has been shown to be representative of the relationship between viscosity and CA in asphalt binder extracted from pavement samples (15,16).

Figure 1 shows clearly that the AB HS is a function of temperature, with greater deviation from the POV HS with increasing temperature. The 93.3°C (200°F) HS measured for two different samples was equal to the POV HS. As a result of these data, it was determined that the products of oxidation at 148.9°C (300°F) are not the same as those formed through oxidation at 93.3°C (200°F) with respect to the relationship between viscosity and CA.

Large quantities of SHRP AAA-1 and SHRP AAF-1 were produced in the AB apparatus. Both asphalts were aged to two different viscosity levels. The aged AAA-1 samples are designated as AAA-AB7 (SHRP AAA-1 air-bubbled Sample 7) and AAA-AB8, and the aged AAF-1 samples are designated as AAF-AB1 and AAF-AB2. To produce pavement-like materials, the reaction temperature was controlled at 93.3°C (200°F) initially. Extreme effort was not expended to maintain this temperature precisely; however, the temperature was never allowed to exceed 110°C (230°F).

SOFTENING AGENTS

The 21 different softening agents that were used in this study can be separated into two main classifications, low-viscosity asphalts and recycling agents. The recycling agents can be further separated into commercial agents and supercritical fractions. Additionally, the *n*-hexane maltene of one of the asphalts was used for one experiment.

Two asphalts, AAV and ABH, were obtained from the SHRP/LTPP MRL. An AC-3 and an AC-5 were obtained from the Diamond Shamrock (DS) refinery in Dumas, Texas, and an AC-5 was acquired from the Shell refinery in Deerpark, Texas. Four non-emulsified commercial agents were obtained: Sun Hydrolene 125, Witco Cyclogen, Exxon NUSO 95 and Mobil Mobilsol 120. The supercritical fractions were produced in the four stage asphalt supercritical extraction pilot plant at Texas A&M University.

The supercritical fractions were produced from five source asphalts using *n*-pentane as the supercritical solvent. The source asphalts for the supercritical fractionation were obtained from a

local pavement contractor, Shell, and the SHRP/LTPP MRL. The asphalt acquired from the local contractor is an AC-20 asphalt and is identified as YBF. The YBF, SHRP AAA-1, ABM-1, and AAF-1 asphalts were fractionated in two runs. The first run removed the asphaltenes and heavy polar aromatic materials and produced a large low-molecular-weight fraction rich in naphthene aromatics and saturates. The majority of this fraction was further fractionated into four additional fractions. The lightest of the fractions was designated Fraction 1 (F1) and the heaviest was designated Fraction 8 (F8). The majority of the supercritical fractions used in this study are either F2 or F3 from these two run fractionations; however, some of the lightest fraction from the primary fractionation (F5) was used as a recycling agent. The Shell asphalt, an AC-20, was fractionated in only one run. As a result, the fraction used in this study, F3, contained a small amount of asphaltenes.

EXPERIMENTAL DESIGN AND RESULTS

The first two experiments were performed to determine the validity of the approach and to test the AB aging technique. The first, preliminary experiment consisted of blending Sun Hydrolene 125 (Sun 125) with the POV AAA-1 in 10 percent increments of aged asphalt by mass. Figure 2 shows that 10 percent increments are not necessary to determine the relationship between viscosity and asphalt mass fraction. Furthermore, this experiment shows that the blends exhibit significant deviation from the viscosity predicted by the ASTM nomograph.

The second experiment was performed using Sun 125 as the recycling agent and AAA-AB7 as the aged asphalt. AAA-AB7 has approximately the same viscosity as the POV AAA-1 used in the first experiment. Aged material content varied from 0 percent to 100 percent in 20 percent increments. The values of the Grunberg interaction parameter for these two and all other experiments were determined by fitting the data in terms of $\ln \eta$. The values are tabulated in Table 2. Figure 2 shows that the Grunberg equation is capable of modeling the data for these first two experiments. The data for the AB-aged material show only minor differences from the data from the POV-aged material blends. The result of this experiment further supports the ability of the AB apparatus to produce quality aged material.

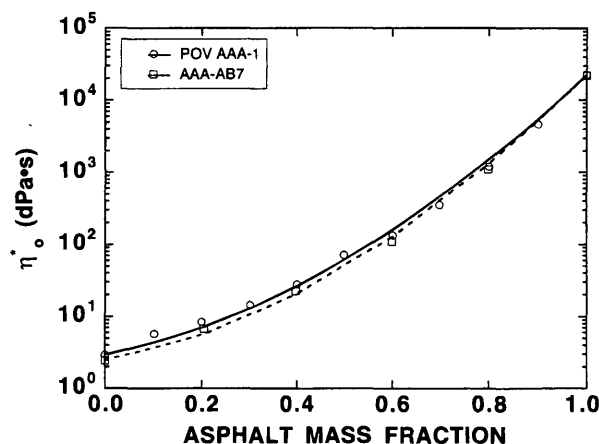


FIGURE 2 Viscosity as function of aged asphalt mass fraction for POV AAA-1 and AAA-AB7 blends with Sun 125.

TABLE 2 Aged Asphalt-Agent Grunberg Interaction Parameter G_{12}

| Asphalt | Agent | G_{12} | Asphalt | Agent | G_{12} |
|--------------|--------------------|----------|--------------|------------|----------|
| POV AAA-1 | Sun 125 | -5.80 | AAA-AB7 | Sun 125 | -6.31 |
| AAA-AB7 | Cyclogen | -6.28 | AAA-AB7 | YBF F2 | -5.42 |
| AAA-AB7 | YBF F5 | -4.28 | AAA-AB7 | ABM F2 | -4.63 |
| AAA-AB7 | YBF F3 | -3.45 | AAA-AB7 | ABM F3 | -4.10 |
| AAA-AB7 | SHRP ABH | 0.03 | | | |
| AAA-AB8 | Cyclogen | -6.33 | AAA-AB8 | AAA F2 | -5.47 |
| AAA-AB8 | YBF F5 | -4.03 | AAA-AB8 | AAA F3 | -4.77 |
| AAA-AB8 | AAF F3 | -4.52 | AAA-AB8 | DS AC-3 | --- |
| AAA-AB8 | DS AC-3 Maltene | --- | AAA-AB8 | Shell AC-5 | 1.14 |
| AAA-AB8 | SHRP AAV | -0.46 | AAA-AB8 | NUSO 95 | -6.23 |
| AAF-AB1 | NUSO 95 | -8.42 | AAF-AB1 | AAF F2 | -5.88 |
| AAF-AB1 | ABM F2 | -4.88 | AAF-AB1 | AAA F3 | -4.56 |
| AAF-AB1 | Shell F3 | -3.64 | AAF-AB1 | ABM F5 | -4.90 |
| AAF-AB1 | DS AC-5 | 2.57 | AAF-AB1 | SHRP ABH | 0.08 |
| AAF-AB1 | DS AC-3 | 2.18 | AAF-AB1 | ABM F3 | -3.99 |
| AAF-AB1 | Mobil 120 | -7.69 | | | |
| AAF-AB2 | Sun 125 | -6.24 | AAF-AB2 | Mobil 120 | -7.50 |
| AAF-AB2 | AAA F2 | -4.83 | AAF-AB2 | AAF F2 | -5.26 |
| AAF-AB2 | ABM F5 | -3.95 | AAF-AB2 | YBF F3 | -3.39 |
| AAF-AB2 | AAF F3 | -3.81 | AAF-AB2 | Shell F3 | -3.12 |
| AAF-AB2 | Shell AC-5 | -0.37 | AAF-AB2 | SHRP AAV | -0.88 |
| AAF-AB2 | DS AC-5 | 1.82 | | | |
| Oven Coastal | Sun 125 | -10.71 | Oven Coastal | Cyclogen | -9.28 |
| Oven Coastal | YBF F3 | -6.54 | Oven Coastal | YBF F5 | -6.90 |
| POV ABM-1 | ABM F3 | -1.48 | POV ABM-1 | ABM F2 | -3.39 |

* --- Data not applicable

The first aged asphalt to be studied systematically was AAF-AB2. This material was blended with three low-viscosity asphalts and eight recycling agents (six supercritical fractions and two commercial agents). Each AAF-AB2-softening agent pair was blended at levels from 0 to 100 percent in 20 percent increments. Figure 3 shows that the data for all AAF-AB2-softening agent pairs are adequately described by the Grunberg model. Although there is some deviation between the data and the fit through the data, a single parameter for each asphalt-softening agent pair is able to model the data. In addition, this parameter is a constant that is independent of the aged asphalt mass fraction. It is immediately obvious from these data that there is a negative deviation from the straight line that would connect the pure-component endpoints for the blends produced using recycling agents. Figure 4 shows that the data for the low-viscosity asphalt softening agents are near or above the straight

line representing the ASTM nomograph. This suggests that the recycling agent blends, both supercritical fraction and commercial agent, should be treated separately from the low-viscosity asphalt softening agent blends.

Table 2 shows the value of the interaction parameter for each asphalt-softening agent pair. The interaction parameter varies considerably depending on the softening agent, indicating that using an average value for the interaction parameter would result in substantial error. The only noticeable trend of these data is that the interaction parameter decreases (i.e., becomes more negative) as the agent viscosity decreases for the recycling agents. From this trend, it was hypothesized that some of the variation in this parameter is due solely to the viscosity difference between the softening agent and the aged asphalt. To eliminate this viscosity effect, it is necessary to normalize the data. The Grunberg equation may be

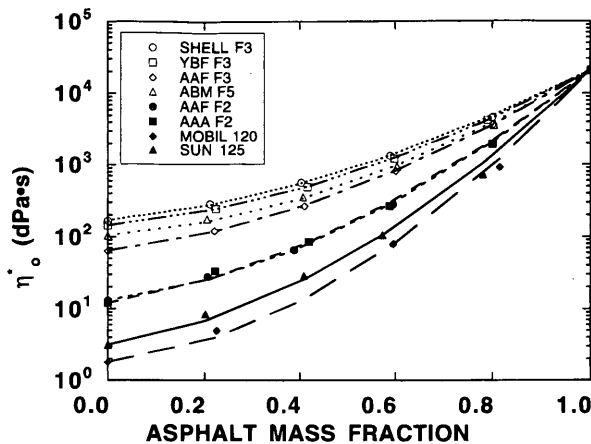


FIGURE 3 Viscosity versus mass fraction for blends of AAF-AB2 with eight recycling agents.

rearranged such that the pure component endpoints are zero for the pure softening agent and one for the pure aged asphalt. The normalized Grunberg model is given as Equation 2, with the aged asphalt as Component 2 and the recycling agent as Component 1.

$$DLV = \frac{\ln(\eta_m/\eta_1)}{\ln(\eta_2/\eta_1)} = \left(1 + \frac{G_{12}}{\ln(\eta_2/\eta_1)}\right)x_2 + \left(\frac{-G_{12}}{\ln(\eta_2/\eta_1)}\right)x_2^2 \quad (2)$$

The dimensionless log viscosity (DLV) can be fit as a second-order polynomial with respect to x_2 , aged-asphalt mass fraction. The coefficient on the second-order term can be viewed as the normalized Grunberg interaction parameter. Figure 5 shows the normalized viscosity plotted as a function of mass fraction for the AAF-AB2-softening agent pairs. The data for the aged asphalt-recycling agent pairs show remarkably little difference when analyzed in this manner. Again, the term "recycling agent" includes both supercritical fractions and commercial agents. Even though recycling agent saturate content varies from 8 to 23 percent and aromatic content varies from 77 to 92 percent, all of the recycling agents produce the same DLV for a given aged asphalt mass fraction. This result com-

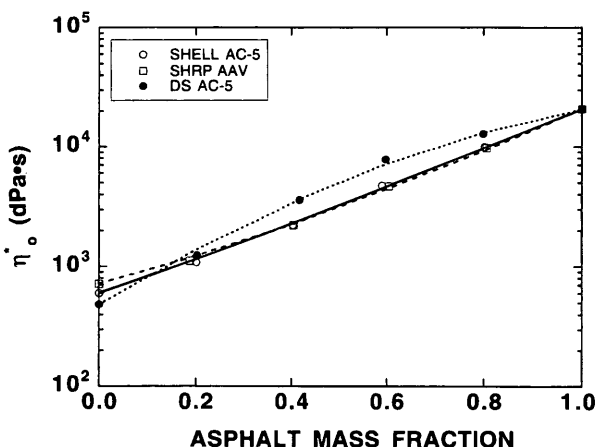


FIGURE 4 Viscosity versus mass fraction for blends of AAF-AB2 with three low-viscosity asphalts.

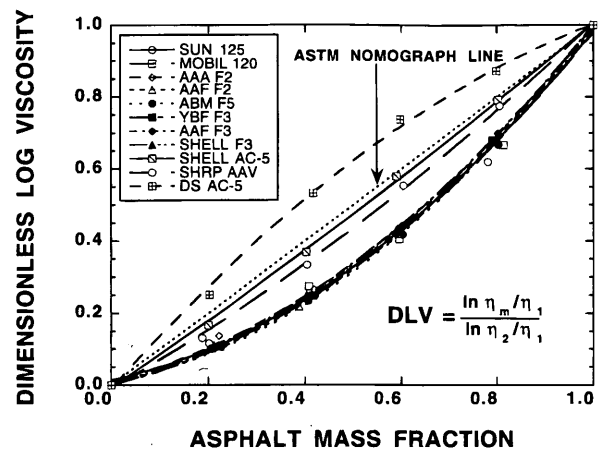


FIGURE 5 DLV as function of aged asphalt mass fraction for blends using AAF-AB2.

plicates the correlation of interaction parameter G_{12} with recycling agent chemical composition.

The low-viscosity asphalt softening agents do not collapse to a single grouping of data. SHRP AAV and the Shell AC-5 have similar interactions with AAF-AB2 and would be well predicted by the ASTM nomograph, but the DS AC-5 exhibits significant positive deviation. Even though these low-viscosity asphalt softening agents do not exhibit behavior similar to the supercritical fraction and commercial recycling agents, blends with all three low-viscosity asphalt softening agents can be modeled using the Grunberg equation in either the standard or normalized forms. In addition, the AAF-AB2 data show that it may be possible to use an average value for the normalized interaction parameter for aged asphalt-recycling agent systems.

The next aged asphalt studied was AAF-AB1. This material was blended with three low-viscosity asphalts and eight recycling agents (six supercritical fractions and two commercial agents). Each asphalt-softening agent pair was blended at levels from 0 to 100 percent in 20 percent aged asphalt increments. Of these 11 softening agents, 1 of the low-viscosity asphalts and 4 of the recycling agents were the same as those blended with AAF-AB2. One of the recycling agents, supercritical fraction ABM-1 F3, has a viscosity in the AC-5 range but with no asphaltene and a low saturate content.

The data for these aged asphalt-softening agent pairs are also well described by the Grunberg model. As Table 2 shows, the value of the interaction parameter varies considerably from softening agent to softening agent and is different for an agent blended with AAF-AB1 and that same agent blended with AAF-AB2. Without exception, the absolute value of the interaction parameter was larger for an AAF-AB1-agent pair than for an AAF-AB2-agent pair. Once again, this suggests that there is some effect due solely to the viscosity difference between the aged asphalt and the softening agent.

The normalized viscosity data for the AAF-AB1-softening agent blends are plotted in Figure 6. There is more variation in the data for the AAF-AB1/recycling agent blends than there is for the AAF-AB2-recycling agent blends, but there is still remarkably little difference. The AAF-AB1-recycling agent and AAF-AB2-recycling agent data are plotted together in Figure 7. It is clear that there is much similarity between the two sets of data. Blends of recycling agents (both supercritical fractions and commercial agents) with

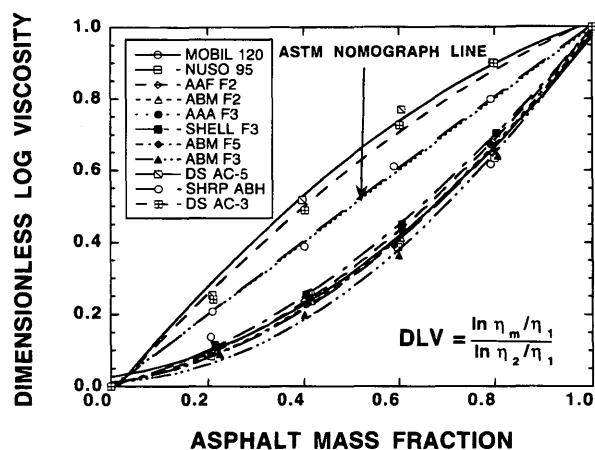


FIGURE 6 DLV versus mass fraction for blends using AAF-AB1.

20,900 dPa · sec (poise) AAF-1 and blends with 52,500 dPa · sec (poise) AAF-1 have essentially the same DLV for a given aged asphalt mass fraction, indicating that an average normalized interaction parameter can be used for AAF-1-recycling agent mixtures.

As was the case in the AAF-AB2 blends, the AAF-AB1-low-viscosity asphalt softening agent pairs do not collapse to a single grouping of data. Figure 6 shows that the DS AC-3 and DS AC-5 exhibit similar positive deviations but SHRP ABH shows no significant deviation from the behavior predicted by the ASTM nomograph. The behavior of the high-viscosity supercritical fraction ABM-1 F3 is similar to the behavior of the rest of the recycling agents, demonstrating that a high viscosity material can exhibit negative deviations. Once again, the Grunberg model seems adequate to describe all of the data.

Next, AAA-AB7, which was blended with Sun 125 in the second experiment, was blended with seven additional softening agents. The normalized Grunberg equation is able to model the AAA-AB7 data, as shown in Figure 8. Again, there is significant deviation between the recycling agents and the low-viscosity asphalt soften-

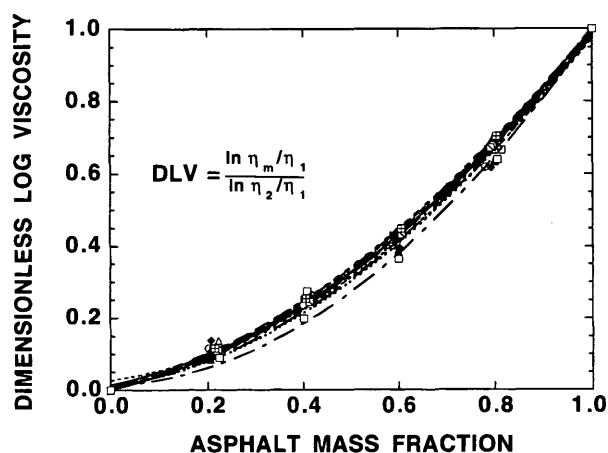


FIGURE 7 LV for blends of AAF-AB2 and AAF-AB1 with recycling agents.

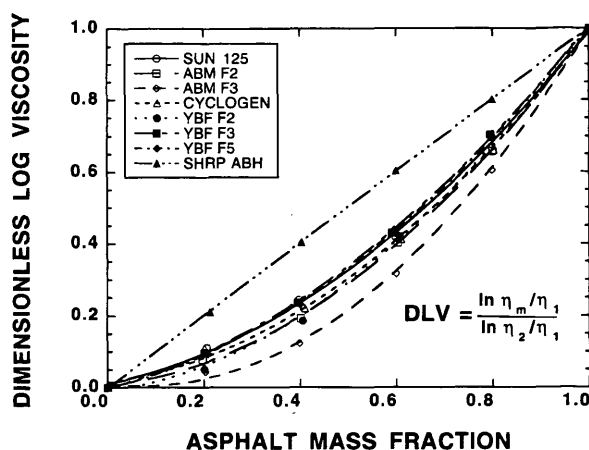


FIGURE 8 LV for AAA-AB7 blends.

ing agent. There are two important features of these experiments. The first is that the ABM-1 F3 agent, a high-viscosity supercritical fraction recycling agent, shows moderate deviation from the rest of the recycling agents. The second noticeable feature is that there is more scatter among the mixture data for AAA-AB7 blends than for AAF-AB2 blends, even though these aged materials have similar viscosities. This implies that AAF-AB2 blends will have similar DLVs independent of the recycling agent used, and that the mixture DLV behavior of AAA-AB7 can be slightly altered by the choice of recycling agent.

Aged material AAA-AB8 was blended with six recycling agents, three low-viscosity asphalts, and DS AC-3's maltene. Once again, the recycling agent blends form a narrow band with respect to DLV and the low-viscosity asphalt blends do not (Figure 9). Two significant results emerged from the AAA-AB8 data. First, the Shell AC-5 shows positive deviation from the ASTM nomograph with this aged asphalt that it did not with AAF-AB2, as is shown by the positive value of the interaction parameter in Table 2. Second, the Grunberg model fails miserably for the DS AC-3 and its maltene. In fact, the DS AC-3 and maltene data are highly sigmoidal, exhibit-

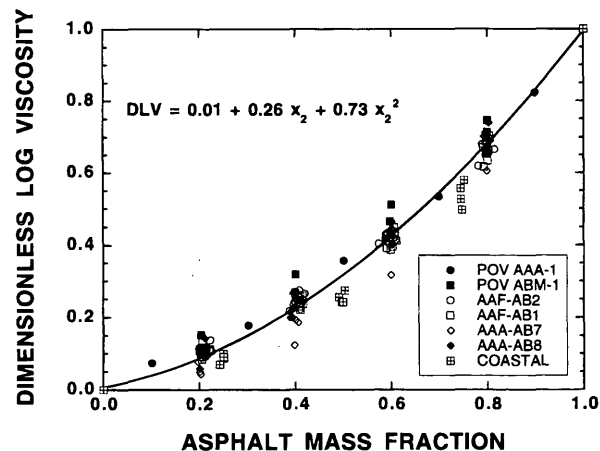


FIGURE 9 V as function of aged asphalt mass fraction for all recycling agent blends.

ing negative deviation at low AAA-AB8 levels and positive deviation at high AAA-AB8 mass fractions (data not shown). Additionally, the DS AC-3 blends had larger DLVs than the maltene blends. These results show that removing the asphaltenes from DS AC-3 has only a minor effect. This further complicates the correlation between viscous interaction and compositional parameters.

The oven-aged Coastal asphalt was blended with four different recycling agents (two supercritical fractions and two commercial agents). Aged asphalt content varied from 0 to 100 percent in 25 percent increments. The data from these blends also form a narrow band in terms of the DLV (Figure 9); however, this narrow band is significantly lower than the data for the other blends. Although these Coastal blends result in DLVs lower than the majority of the other asphalt blends, they are not as low as the data for the AAA-AB7-ABM-1 F3 blends. The data obtained for the POV ABM-1 blends are somewhat higher than the average for the rest of the data (Figure 9). In fact, the POV ABM-1-ABM-1 F3 blend data are closest to the diagonal line representing the ASTM-suggested mixing rule. Thus, blends made with ABM-1 F3 as the recycling agent form both the high and low boundaries of data collected in this study.

DISCUSSION OF RESULTS

All of the recycling agent (supercritical fraction and commercial agent) blend data collected in this study were placed on the same plot of DLV versus aged asphalt mass fraction. An overall mixing rule was determined by fitting the DLV data to a second order polynomial. The complete data and overall fit are shown in Figure 9.

This overall DLV mixing rule was used to predict the amount of softening agent necessary to obtain specification blends for all of the aged asphalt-softening agent pairs used in this study with the exception of the AAA-AB7-DS AC-3 material blends. The log log η mixing rules suggested by Epps and the ASTM nomograph were used for comparison. Two target viscosities were chosen for comparison. A target viscosity of 2000 dPa · sec (poise) was chosen because this is the specification for an AC-20 asphalt and the probable target viscosity for hot-mix recycling. A target viscosity of 5000 dPa · sec (poise) was also chosen. This is a reasonable value for an AC-20's viscosity after thin film oven treatment and a probable target viscosity for hot in-place recycling. The amount of softening agent required was calculated for each mixing rule and then the actual mixture viscosity was determined from the Grunberg interaction parameter for the individual aged asphalt-softening agent pair. If the predicted softening agent content was less than 10 percent, the data were considered unreliable and were not used for further analysis because unrealistically high actual viscosities resulted (mostly for the Epps rule). The resulting viscosities were calculated and an average value was obtained for the recycling agent blends as a group and for the low-viscosity asphalt softening agents as a group.

The average viscosities that would result from prediction using each model are given in Table 3. In addition to the average viscosity, the range of viscosities resulting from each model are listed. From these data, it is obvious that the DLV mixing rule using an average normalized interaction parameter is superior to the other two mixing rules at determining the proper amount of recycling agent (supercritical fraction or commercial agent) to use. This is to

TABLE 3 Comparison of Viscosities Resulting from Various Mixing Rules

| Model | Viscosity | | |
|---|------------|------|-------|
| | Average | Low | High |
| Commercial and Supercritical Recycling Agents; Target Viscosity 2000 dPa · s: | | | |
| DLV | 2040±390 | 1100 | 3000 |
| Epps | 1920±1200 | 780 | 6730 |
| ASTM | 700±370 | 160 | 2340 |
| Commercial and Supercritical Recycling Agents; Target Viscosity 5000 dPa · s: | | | |
| DLV | 5010±840 | 3120 | 7350 |
| Epps | 4380±1490 | 2140 | 9190 |
| ASTM | 1880±570 | 540 | 3460 |
| Low Viscosity Asphalt Softening Agents; Target Viscosity 2000 dPa · s: | | | |
| DLV | 5320±2200 | 2960 | 8800 |
| Epps | 3310±1190 | 1910 | 5090 |
| ASTM | 2430±680 | 1660 | 3410 |
| Low Viscosity Asphalt Softening Agents; Target Viscosity 5000 dPa · s: | | | |
| DLV | 11500±4300 | 6900 | 19200 |
| Epps | 8380±3000 | 4800 | 13000 |
| ASTM | 6180±2000 | 4030 | 9500 |

be expected, because the DLV mixing rule is based on the very data that it is predicting. However, the ability of the DLV mixing rule to produce AC-20 blends nearly 95 percent of the time in the aged asphalt-recycling agent blends is an extraordinary result given the extreme variation, both in terms of standard deviation and range, of the other two models. This shows that the current methods are inadequate at predicting proper recycling agent content. In fact, the ASTM nomograph results in completely unacceptable viscosities for better than 95 percent of the hypothetical mixtures. This substantiates the findings of Irving (2) as to the accuracy using G_{12} equal to zero. Use of the ASTM nomograph would certainly necessitate much trial-and-error testing to obtain the correct viscosity for these aged asphalt-recycling agent blends.

For prediction of the low-viscosity asphalt softening agent data, the DLV mixing rule does not perform very well. The average, deviation, and range are all larger than those obtained by the other mixing rules. Table 3 shows that the ASTM nomograph procedure is best at predicting the low-viscosity asphalt softening agent data. In fact, this method is remarkably good considering that these data include the blends formed by the DS asphalts and the Shell AC-5.

CONCLUSIONS

Forty-seven aged asphalt-softening agent pairs were blended at multiple levels of aged material content. For each asphalt-agent pair, 60°C low-frequency limiting viscosities were measured at each aged material content.

The relationship between mixture viscosity and aged material mass fraction for 45 of the asphalt-agent pairs can be described using the Grunberg model. Blends using low-viscosity asphalts as the softening agents exhibited significantly different behavior from blends using commercial recycling agents and supercritical fraction recycling agents. The low-viscosity asphalt softening agents had viscous interaction parameters close to or greater than zero. All of the blends using supercritical fraction and commercial recycling agents had interaction parameters less than zero.

The value of the interaction parameter G_{12} is a strong function of the viscosity difference between the aged asphalt and recycling agent. Normalizing viscosity in terms of the DLV reduces the difference between recycling agents. In fact, DLV data for all of the recycling agent blends show strikingly little variation between recycling agents regardless of chemical composition or aged asphalt used.

An average normalized interaction parameter was obtained by fitting all of the aged asphalt-recycling agent data. This overall fit was compared to the mixing rule of Epps (6) and the mixing rule specified by the ASTM (9). Comparison was based on the ability of each mixing rule to predict the quantity of softening agent required to produce blends with a specific target viscosity. If a low-viscosity asphalt is to be used as the softening agent to recycle an asphalt, the method specified in ASTM D4887 should be used. However, for prediction of the amount of recycling agent needed to produce the target viscosity, the DLV mixing rule developed in this study is superior to the other two mixing rules.

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