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Rheological behaviors of waste polyethylene modified asphalt binder: statistical analysis of inter-laboratory testing results

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ABSTRACT

This paper investigated the effect of waste polyethylene (PE) on the modified asphalt binders' rheological behavior from a statistical point of view. The Interlaboratory testing results from the RILEM Technical Committee (TC) 279 WMR (Valorization of Waste and Secondary

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Materials for Roads) Task Group 1 (TG1) were used for this purpose. First, an unaged 70/100 penetration graded neat binder was selected as the reference material. Next, a single 5% content of waste PE additives (PE-pellets and PE-shreds) was mixed with a 95% neat binder to prepare two PE modified binders. Then, Dynamic Shear Rheometer (DSR) based temperature frequency sweep tests were performed over a wide range of temperatures and frequencies to evaluate the rheological properties of these three binders. Different rheological behaviors were observed in the isochronal plots at high temperatures. Based on a reproducibility precision requirement proposed for phase angle, 28 °C was set as the transition temperature across the rheological behaviors. Next, according to the three rheological behaviors defined in a previous study by the authors, statistical analysis was introduced to identify sensitive rheological parameters and determine the thresholds. Results indicate that the phase angle measured above 28 °C and 1.59Hz can be used as a sensitive parameter to discriminate the three rheological behaviors of PE modified binders. The thresholds among different behaviors were also calculated as an example for phase angle measured at the highest common testing temperature of 70 °C. Additional experimental evaluations on more types of PE modified binders, especially at intermediate and high temperatures, are recommended to better understand their influence on the rheological behavior of PE modified binders.

Keywords

Polyethylene (PE) Plastics, Modified binder, Dynamic Shear Rheometer (DSR), Rheological behavior, Statistical analysis, phase angle, *G-R* parameter

1. Introduction

Created about a century ago, polymeric, especially plastic, materials provided countless advantages to modern society. However, they became sources of several environmental issues due to rising production and consumption and inadequate disposal practices. As a result, pollution by plastic materials has become a serious environmental problem. It requires complementary approaches to mitigate this impact, such as consumption reduction, substituting new, easily degradable materials, and adequate solid waste disposal by sorting and recycling techniques. Although the volume of annually recycled plastics has increased regularly, the recycling rate is below the rate of virgin plastics being produced.[1] Since the 1950s, only approximately 9% of the cumulatively generated waste plastic has been recycled, while most were discarded in landfills or the natural environment.[2] The reuse and recycling of plastic waste materials are crucial for the transition to a circular economy. This good practice is essential given the peculiarity of plastic, its value chains, and accounting for its environmental and greenhouse gas footprint.[3]

Asphalt roads are one of the most relevant transportation infrastructures worldwide. Due to the increase in traffic volume and the resulting higher load caused by heavy vehicles, demand for better pavement performance and longer service life has made the asphalt industry adapt its materials during the past decades.[4] Asphalt binders require different types of polymer additives, fibers, or modifiers to improve the performance and durability of asphalt mixtures.[5] The additional cost of traditional synthetic or natural polymer is often compensated by the longer life of the materials and enables its use in asphalt pavement on a large scale. Thus, waste polymers have also been proven to improve asphalt properties compared to those attained with virgin polymers.[6] Using marginal and secondary materials in pavement construction could be viable with potential economic benefits. However, a complete evaluation can be achieved only through a life cycle cost assessment. Furthermore, such materials can be beneficial in increasing pavement performance and landfill reduction.[5] Different studies have been conducted on various waste polymers in road material pavement, evaluating the effects of polyethylene terephthalate (PET), polyethylene (PE), polypropylene (PP), polyurethane (PU), ethylene-vinyl acetate (EVA), acrylonitrile butadiene styrene (ABS), polyvinyl chloride (PVC), and different plastic fibers added into the asphalt.[7,8] Among these material sources, PE is one of the most commonly used.[9,10] Regarding the incorporation methods, dry and wet processes are widely used. In the wet process, waste plastic is incorporated directly into the binder by 0.5% to 10% weight of the binder at high temperatures.[11] Significant enhancement in the viscoelastic performance can be achieved at high temperatures, while comparable stiffness modulus was observed to the reference materials.[12] However, previous studies frequently observed scattered rheological responses at relatively high temperatures.[9,10,13] This inconsistency can be mainly attributed to the difference in density, viscosity, and incompatibility between recycled waste PE and binder.[12] Hence, the high temperature rheological behavior of PE modified binders needs to be carefully studied. In previous studies, almost all the results were measured by a single laboratory or limited laboratories. Therefore, specific testing conditions, including equipment and testing protocol, could significantly affect the experimental results questioning the validity and robustness of the research outcome.[9,10]

The Dynamic Shear Rheometer (DSR) based testing methods are commonly used to evaluate the rheological behavior of asphalt materials.[14,15] The temperature frequency sweep (T-f-sweep) test can effectively characterizes the asphalt binders' rheological response within the linear viscoelastic (LVE) range.[16,17,18] However, in previous studies, scattered rheological responses were frequently observed in PE modified binders at high temperatures.[19] Hence, rheological parameters and statistical analyses are necessary to be introduced to better understand the effect of PE modifiers. In the authors' previous works, it was found that the

Glover-Rowe parameter can be used to discriminate the materials' response at intermediate temperatures. In contrast, the crossover parameters (crossover temperature and crossover modulus) provide a sensitive tool over a wider range of temperatures.[17,20] In addition, the measured complex shear modulus and phase angle could also function as sensitive parameters.[16] The application of statistical analysis in the asphalt industry has become common practice for more than 4 decades. Different studies attempted to use it to evaluate and predict the performance properties of bituminous materials and the development of distresses.[21,22,23,24] Results indicate that statistical analysis is a useful and sensitive tool to discriminate different behaviors of bituminous materials.

Given such scientific background, RILEM established a Technical Committee entitled 279-WMR (Valorization of Waste and Secondary Materials for Roads) in 2017. Within the framework of this TC, Task Group 1 (TG 1) was generated to assess the possibility of using waste PE additives as modifiers of the asphalt binders and mixtures.[9,25] An interlaboratory testing protocol with eleven laboratories worldwide was conducted for this purpose. For the binder phase, conventional properties, including softening point temperatures and penetration values, and several DSR based rheological tests were conducted to evaluate the rheological properties of PE modified binders.[9] In this study, the results of temperature-frequency sweep (*T-f sweep*) tests were analyzed and discussed.

2. Objective and Research Approaches

This study evaluated the effect of PE additives on the rheological responses of modified binders. The transition temperature across rheological behaviors was firstly defined, and sensitive rheological parameters to discriminate the different rheological behaviors were analyzed via statistical analyses. The temperature-frequency sweep oscillatory tests were performed first over a wide range of temperatures and frequencies.[26,27,28] Two rheological parameters, complex shear modulus, $|G^*|$, and phase angle, δ, were recorded. Three parameters, $|G^*|/\sin\delta$, $|G^*|$, and δ measured at 1.59 Hz, were used to determine the rheological transition temperature. Next, based on previous inter-laboratory results, different rheological profiles (responses) were identified using the black diagram. In the present study, statistical analysis was applied to determine the potential sensitive rheological parameters for discriminating the rheological behavior. $|G^*|$ and δ results (at 1.59 Hz), which were recorded at temperatures higher than the transition temperature, together with crossover parameters (crossover temperature and crossover modulus)[20] and Glover-Rowe parameter,[29] were used for this purpose. Finally, the boundaries for different rheological profiles were calculated for the selected parameters.

3. Materials and Experimental Plan

In this research, a fresh 70/100 penetration graded[30] neat binder was selected as the reference material and designated as binder B. Two different PE additives (PE pellets and PE shreds) at 5% were blended with 95% neat binder to prepare the two PE modified binders, $B_{+pellets}$ and $B_{+shreds}$, respectively. PE pellets are produced by processing waste packaging materials primarily consisting of PE, while PE shreds are the by-product of the production process of the pellets.[12] Such PE content was decided in the authors' previous study;[9,12] specific details on the grinding and blending process can also be found in the same research. A remarkable increase in the softening point temperature (more than 15 ºC for $B_{+pellets}$ and more than 25 ºC for $B_{+shreds}$) and a decrease in the penetration values at 25 °C (more than 42 dmm for both $B_{+pellets}$ and $B_{+shreds}$) were observed in PE modified binders compared to the neat reference binder. Detailed information and analysis on the conventional properties can be accessed in the authors' previous works.[9,10]

In the present study, temperature-frequency sweep (*T-f sweep*) tests were performed with the DSR device. Complex shear modulus, $|G^*|$, and the phase angle, δ, were recorded. Two plate-plate geometries were selected for different temperature ranges over a wide range of frequencies (0.1 Hz to 20 Hz). 25 mm plate geometry with a 1 mm gap (PP25) was adopted for higher temperatures, between 34 °C and 82 °C, with a temperature interval of 6 °C. It should be noted that, in several laboratories, 70 °C is the highest measurement temperature. For the lower temperature range, 8 mm plate-plate geometry with a 2 mm gap (PP08) was selected (T=-6, 0, 4, 10, 16, 22, 28, 34, and 40 °C). All the *T-f sweep* measurements were performed within the linear viscoelastic (LVE) range with the suggested strain levels of 0.1% (PP25) and 0.05% (PP08), respectively. All eleven laboratories worked on $B_{+shreds}$, while a reduced number of participants performed binder B and $B_{+pellets}$ due to the limited amount of materials. More information about the testing protocols can be found in past research efforts.[9,10]

4. Results and Analysis

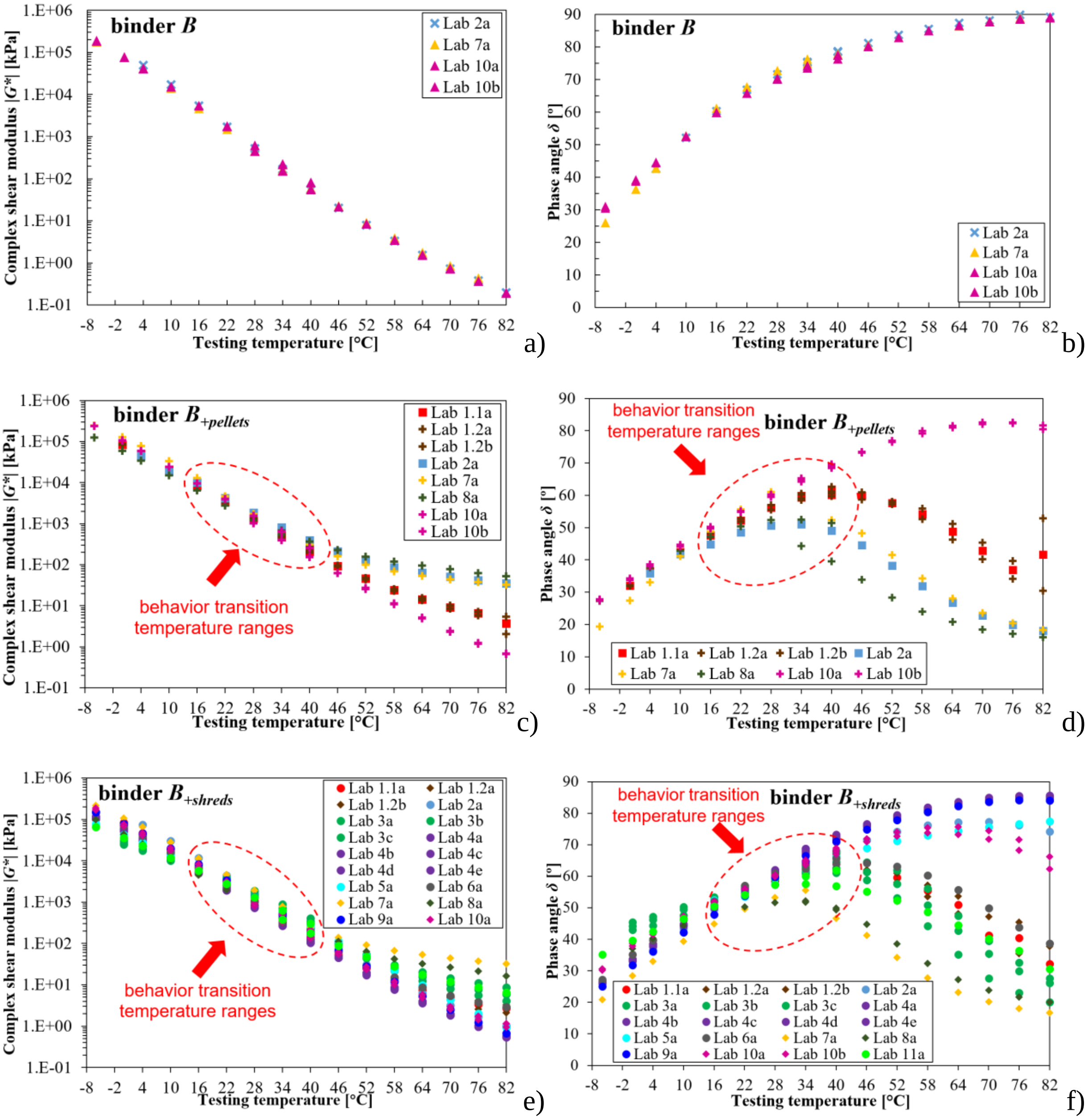
4.1 TRANSITION TEMPERATURE FOR THE RHEOLOGICAL BEHAVIOR

As a first step, the repeatability within laboratories and reproducibility among laboratories were conducted on the raw data. The precision of the data within a single laboratory was evaluated according to AASHTO T315-20.[14] The parameter $|G^*|/\sin\delta$ was used for this purpose; a maximum variation coefficient of 1s% (standard deviation) is fixed to 1.6% for unaged binders. Results indicate that only the neat binder fits the AASHTO repeatability criteria for single operator testing within a single laboratory; both PE modified binders' precisions fall beyond the limitations. This result is not surprising since such requirements were originally designed for neat binders. More specific analysis and discussion is reported in the authors' previous research.[10]

For the reproducibility among laboratories, visual comparisons (Figure 1) were conducted on all three binder types (B, $B_{+pellets}$, and $B_{+shreds}$), while quantitative comparison (Table 1) was performed for both PE modified binders. Figure 1 illustrates the isochronal curves of the complex shear modulus, $|G^*|$, and the phase angle, δ, for all three asphalt binders at 1.59 Hz. Not unexpectedly, the neat binders' results achieved very similar curves in $|G^*|$ and δ among all laboratories, indicating very similar rheological behaviors (Figures 1a and 1b). However, both PE modified binders exhibited different rheological behaviors, with testing temperature remarkably affecting their rheological response. Less variability was found at the relatively low testing temperatures (PP08), while remarkably different curves could be observed at high temperatures (PP25). In contrast, the transition in the data set occurred at the intermediate temperatures (range from 16 °C to 40 °C according to Figures 1c to Figure 1f). This variation may be attributed to the inhomogeneous distribution of plastic particles at high temperatures. Moreover, the greater plate-plate diameter and the lower measurement gap (1 mm) for PP25 may also lead to poor reproducibility among laboratories.

FIGURE 1

Isochronal plots at 1.59 Hz: a) $|G^*|$ of binder B; b) δ of binder B; c) $|G^*|$ of binder $B_{+pellets}$; d) δ of binder $B_{+pellets}$; e) $|G^*|$ of binder $B_{+shreds}$; f) δ of binder $B_{+shreds}$



The transition temperature between the data sets is critical in designing asphalt mixtures containing waste plastic materials. However, it is not easy to determine it through a simple visual comparison shown in Figure 1. The phase angle curves exhibited more scatter; however, the complex shear modulus results were plotted in a log scale; therefore, the actual differences (in percentage) may be even higher. Hence, a quantitative comparison was adopted for the three rheological parameters, $|G^*|/\sin\delta$, $|G^*|$, and δ. As previously mentioned at the beginning of this section, the |G*|/sinδ was developed and reported according to AASHTO T315-20; for evaluating multi-laboratory precision, a maximum variation coefficient of 1s% (standard deviation) is fixed to 3.6% for unaged unmodified binders among laboratories. However, such criteria were designed for unmodified binders, and they may not be necessarily suitable for this study. Hence, additional precision limitations developed by the RILEM TC-182 PEB (Performance testing and evaluation of bituminous materials) for both plain and modified binders were introduced in this study. The reproducibility precision requirements for |G*| and δ (coefficient of variation) were 10% and 5%, respectively.[31] Based on an active European standard,[15] the absolute precision of 2° for phase angle was also applied.

Table 1 lists the calculated reproducibility precisions for all three rheological parameters and both PE modified binders. It can be observed that the reproducibility standard deviation first decreased and then increased for PP08, while a monotonically increasing trend can be found in PP25. This tendency is true for all rheological parameters and both PE modified binders. This response may be attributed to the difference in stiffness between matrix (binder) and particles (plastic) experienced as the temperature increases when the binder starts to exhibit a more significant transition toward a viscous-like behavior.[32] Additionally, instrument compliance phenomena might appear at lower temperatures, making the measurements less consistent.[27,28] Hence, only results obtained at a temperature higher than 5 ºC were used for the analysis; overall increasing trends were observed in the reproducibility standard deviations. It is not surprising that parameters |G*|/sinδ in both PE modified binders were unable to meet the requirement for all temperatures because this parameter was developed for the neat binder. However, |G*| was also unable to meet the requirement for all temperatures; this may be attributed to the high modification of these two materials and the capability of available DSR devices. For δ, the reproducibility standard deviations (in both percentage and absolute value) meet the measurement requirements below 28 ºC; this is true for both modified binders. Hence, 28 ºC can be assumed as the transition temperature for rheological responses. According to the authors' previous study,[12] part of the PE particles did not melt, remaining in a micro-solid state in the binders. When the testing temperature increased to the transition temperature of the modified binders, the distribution of PE particles could not remain homogenous and start flowing. Hence, different behaviors were expected under different experimental configurations when the testing temperatures were higher than the transition temperature. This is especially true with the increase in temperatures. Such a transition temperature may differ from the experimental conditions and materials. Hence, it is not surprising that different transition temperatures were defined in the authors' previous studies. [9,10]

TABLE 1

Reproducibility analysis of $|G^*|/\sin\delta$, $|G^*|$ and δ at 1.59 Hz for $B_{+pellets}$ and $B_{+shreds}$

Material	**$\|G^*\|/\sin\delta$ [%]**		**$\|G^*\|$ [%]**		**δ [%]**		**δ [°]**	
	$B_{+pellets}$	**$B_{+shreds}$**	**$B_{+pellets}$**	**$B_{+shreds}$**	**$B_{+pellets}$**	**$B_{+shreds}$**	**$B_{+pellets}$**	**$B_{+shreds}$**
-6 (PP08)	*32.0*	*45.2*	23.7	31.1	13.8	12.5	3.5	3.4
0 (PP08)	*28.1*	*45.8*	22.6	36.5	6.7	12.3	2.2	4.5
4 (PP08)	*26.2*	*44.7*	22.9	37.9	4.4	8.6	1.6	3.4
10 (PP08)	***23.5****	***36.8****	**22.7***	**48.8***	2.7	4.5	1.1	**2.5***
16 (PP08)	***18.7****	***29.7****	**19.0***	**36.2***	3.3	3.8	1.6	1.9
22 (PP08)	***15.1****	***26.7****	**15.8***	**34.6***	4.4	3.5	2.0	1.9
28 (PP08)	***14.3****	***30.7****	**13.3***	**29.5***	**6.0***	**5.4***	**3.4***	**2.6***
34 (PP08)	***17.8****	***36.6****	**14.0***	**35.6***	**8.4***	**6.2***	**5.0***	**3.9***
40 (PP08)	***27.8****	***41.7****	**19.7***	**41.5***	**11.7***	**7.9***	**7.1***	**5.2***
28 (PP25)	0.4	**6.4***	0.5	**15.5***	0.2	0.4	0.1	0.2
34 (PP25)	**36.4***	**28.2***	**27.8***	**27.0***	**12.2***	**6.6***	**7.0***	**4.2***

40 (PP25)	**48.0***	**36.1***	**35.8***	**30.9***	**16.6***	**11.3***	**9.6***	**7.3***
46 (PP25)	**68.0***	**50.5***	**49.1***	**38.3***	**22.8***	**15.3***	**12.9***	**10.0***
52 (PP25)	**89.9***	**77.5***	**64.9***	**52.5***	**30.1***	**20.3***	**16.3***	**13.2***
58 (PP25)	**109.2***	**116.9***	**82.0***	**75.1***	**37.8***	**26.3***	**19.5***	**16.7***
64 (PP25)	**119.7***	**154.3***	**94.4***	**101.6***	**45.5***	**32.7***	**21.8***	**20.0***
70 (PP25)	**124.2***	**177.7***	**102.9***	**125.2***	**52.9***	**40.1***	**23.6***	**23.3***
76 (PP25)	**123.9***	**196.6***	**107.1***	**145.9***	**59.7***	**44.2***	**24.8***	**24.9***
82 (PP25)	**127.8***	**204.1***	**115.0***	**159.1***	**59.6***	**50.2***	**25.3***	**26.7***

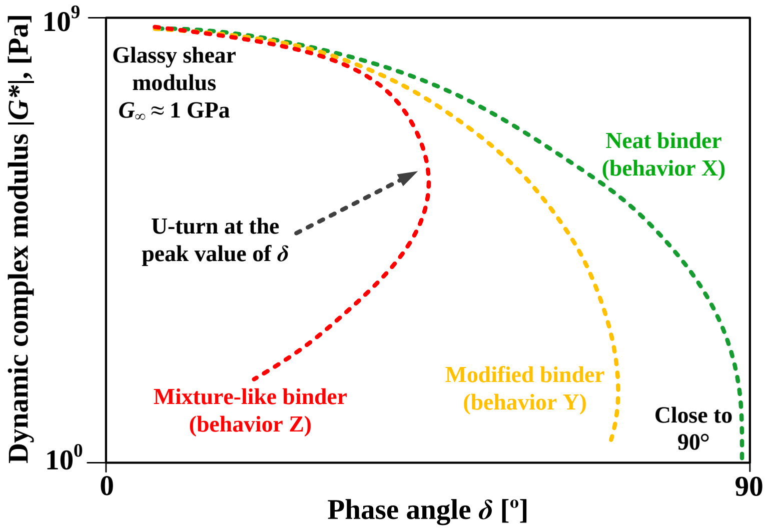
*: failed to pass the AASHTO T315-20[14] and EN 14770[15] reproducibility precision requirements.

4.2 SENSITIVE RHEOLOGICAL PARAMETERS TO DISCRIMINATE THE DIFFERENT RHEOLOGICAL BEHAVIORS

As shown in Figure 1, different rheological curves were visually detected in the isochronal curves at high temperatures. Based on the previous analysis, such differentiation starts from 28C. However, it is not easy to use isochronal profiles to classify different rheological behaviors since the complex shear modulus and phase angle data were plotted against temperature individually. In a previous study by Kim,[13] the black diagram showed the potential to discriminate different rheological profiles (responses) of bituminous materials. The range of δ and $|G^*|$ are from 0 to 90 degrees and 1kPa to 1GPa, respectively; such a range is independent of the binder types and aging conditions. Figure 2 presents an example of the black diagram incorporating the schematic of three major curve trends for binders depending on the degree of complexity and modification: neat binder (yellow), modified binder (orange), and complex modified binder (grey). The latter resembles the response commonly observed in asphalt composites such as asphalt mastic/mixture and is exemplified by the "U-turn" shape of the curve.[10,13]

FIGURE 2

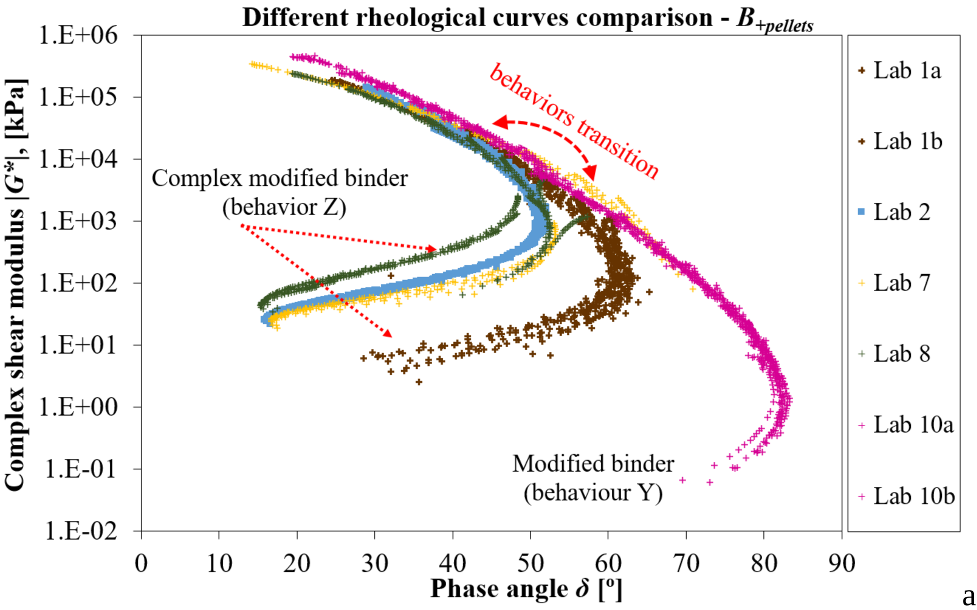
Illustration of different rheological curves in the black diagram



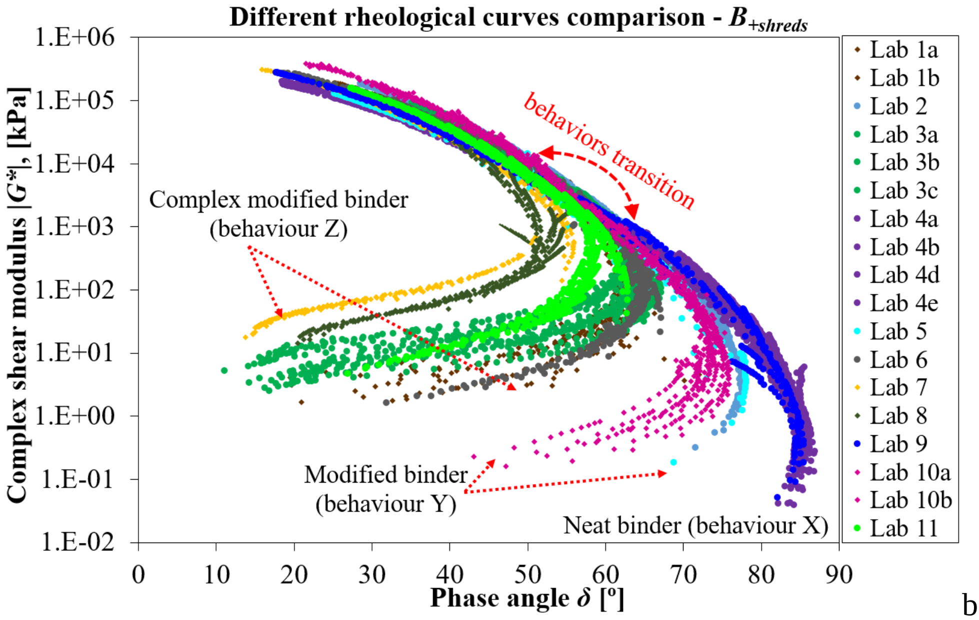
The raw data of two modified binders were plotted into the black diagram and shown in Figure 3. Due to the limited number of results, only two types of rheological behaviors were observed in $B_{+pellets}$ (Figure 3a), while three types of rheological behaviors were found in $B_{+shreds}$ (Figure 3b). Hence, only the results of $B_{+shreds}$ were used for further analysis. Three rheological behavior groups were defined for B+shreds based on the rheological behavior classification. Group X (behavior X: neat binder): laboratories 4a, 4b, 4d, 4e, and 9; Group Y (behavior Y: modified binder): laboratories 2, 5, 10a, and 10b, and Group Z (behavior Z: complex modified binder): laboratories 1a, 1b, 3a, 3b, 3c, 6, and 11.

FIGURE 3

Different rheological profiles observed in this study: a) $B_{+pellets}$ and b) $B_{+shreds}$



a)



b)

Statistical analysis was introduced to discriminate the different rheological profiles and responses observed in $B_{+shreds}$. Four rheological parameters were used for this purpose: crossover parameters (including crossover temperature and crossover modulus),[21] G-R parameters,[20] raw complex shear modulus, |G^*|, and phase angle, δ, recorded at 1.59 Hz. In the case of δ, all results measured higher than 28 °C were used. For |G^*|, because no transition temperatures were observed, only three temperatures (10 °C, 34 °C, and 70 °C) were selected based on the following criteria: 10 °C being the lowest testing temperature, i.e., higher than 5 °C; 34 °C being the transition temperature determined for phase angle, while 70 °C being the highest measurement temperature common to several laboratories. It should be noted that for |$G*$| and δ results measured under 34 °C and 40 °C, both PP08 and PP25 were used for analysis.

First, a Shapiro-Wilk Test was used to validate the normal distribution within groups for all the selected materials, with all the groups passing the validation. Then, analysis of variance (ANOVA) was applied to evaluate the statistically significant among three behaviors with a significance level α=0.05, outputs of p-value are listed in Table 2. Results indicate that most parameters (except crossover temperature, $T_{\delta=45^o}$, and |$G*$| measured by PP25 under 34 ºC) identify statistically different rheological behaviors. Finally, a multiple comparison statistical test based on the Tukey's HSD (honestly significant difference) method was conducted to evaluate each pair of rheological behaviors. The p-value of pairwise comparisons between each pair X vs. Y, X vs. Z, and Y vs. Z are shown in Table 2. Interestingly, only the phase angle data could sensitively discriminate the rheological behaviors from the statistical point of view; all the selected phase angle data measured above 28 ºC could function as such a tool.

TABLE 2

Analysis of the statistical significance of selected rheological parameters

	$G\text{-}R$	$T_{\delta=45^o}$	$\lvert G*\rvert_{\delta=45^o}$	$\lvert G*\rvert$ PP08		$\lvert G*\rvert$ PP25		δ PP08
				10 ºC	34 ºC	34 ºC	70 ºC	34 ºC
p-value	**0.00001**	0.27434	**0.02317**	**0.00335**	**0.01567**	0.34600	**0.00088**	**0.00020**
X vs. Y	0.26276	0.64516	**0.01835**	**0.00819**	0.06830	0.29077	0.67294	**0.01563**
X vs. Z	**0.00002**	0.26231	0.07461	0.96819	**0.01599**	0.59342	**0.00161**	**0.00018**
Y vs. Z	**0.00039**	0.74833	0.73931	**0.00512**	0.72605	0.83454	0.00841	**0.04739**

	δ PP08	δ PP25						
	40 ºC	34 ºC	40 ºC	46 ºC	52 ºC	58 ºC	64 ºC	70 ºC
p-value	**0.00005**	**0.00001**	**0.00001**	**0.00001**	**0.00001**	**0.00001**	**0.00001**	**0.00001**
X vs. Y	**0.00567**	**0.00072**	**0.00223**	**0.00477**	**0.01320**	**0.03009**	**0.03806**	**0.03253**
X vs. Z	**0.00004**	**0.00001**	**0.00001**	**0.00001**	**0.00001**	**0.00001**	**0.00001**	**0.00001**
Y vs. Z	**0.04115**	**0.00827**	**0.00024**	**0.00001**	**0.00001**	**0.00001**	**0.00001**	**0.00001**

*: parameters with statistical significance shown in bold

Based on the results shown in Table 2, the thresholds of three different rheological data sets were calculated using phase angle data; the values measured at 70 ºC and 1.59 Hz were selected as an example. The average value $\bar{x}$ and the mean value μ of the samples were calculated for different rheological behaviors. A 95% confidence interval was used for μ; the value can be calculated as:

$$\mu = \bar{x} \pm 2\times\sigma_n \qquad (1)$$

where, σ is the standard deviation, $\sigma_n = \sigma/\sqrt{n}$, n is the number of samples. Based on Equation 1, two μ values can be calculated, where μ_1 and μ_2 are the lower and upper thresholds, respectively. With these two μ values, the threshold of each rheological behavior with a 95% confidence interval can be calculated as: (μ_1-2×σ_n, μ_2+2×σ_n). The results are shown in Table 3. Considering the definition of behavior X (neat binder), the upper threshold corresponds to the limitation of phase angle 90º.

TABLE 3

Phase angle boundaries of three different rheological behaviors under 1.59 Hz and 70 ºC

	$\bar{x}$	σ	n	σ_n	μ_1	μ_2	μ_1-2×σ_n	μ_2+2×σ_n	**thresholds**
X	84.16	0.48	5.00	0.21	83.73	84.59	81.73	85.54 (90)	**[81.73, 90)**
Y	75.36	2.48	5.00	1.11	73.14	77.58	68.18	80.06	**[68.18, 80.06]**

Z	39.31	7.57	7.00	2.86	33.58	45.03	18.44	60.17	**[18.44, 60.17]**

5. Summary and Conclusions

As part of the RILEM technical committee TC-279 WMR Task Group (TG 1), a large interlaboratory activity was conducted based on the Dynamic Shear Rheometer (DSR) to characterize the rheological behavior of asphalt binders modified with PE. The tests were performed on a neat binder and two blended binders consisting of 95% neat binder blended with two types of 5% PE waste (pellets and shreds). The transition temperature of rheological behaviors was determined with the reproducibility precision criteria proposed by AASHTO and European standards. Statistical analysis was introduced to determine the sensitive rheological parameters to discriminate the three rheological behaviors observed. Phase angle data measured at high temperatures was used to calculate the thresholds of different rheological behaviors. The following conclusions can be drawn from the experimental results.

- The measured rheological properties of PE-modified binders at intermediate and high temperatures may differ by experimental conditions. This diversity can be attributed to the inhomogeneous distribution of particle PE caused by relatively high temperatures.
- A transition in the rheological data set was observed in the isochronal plots of $|G^*|$ and δ. Based on AASHTO and European standards, three different rheological parameters for evaluating the reproducibility precision were used to determine the transition temperature. The phase angle, δ, was selected as the optimal parameter, and 28 °C was determined as the transition temperature.
- Three main different rheological behaviors, named neat binder, modified binder, and complex modified binder, were defined based on the black diagram. The behavior of

complex modified binders exhibited a broader range, while the other two behaviors were relatively narrow.

- Sensitive rheological parameters, such as crossover temperature, crossover modulus, and *G-R* parameter, $|G^*|$ and δ measured under different temperatures at 1.59 Hz, were identified to discriminate the rheological behaviors of PE modified binder at intermediate and high temperatures. The phase angle measured above 28 °C showed to be sensitive in discriminating each pair of rheological profiles and could be used to determine the boundaries of these three behaviors.
- The statistical analysis was conducted based on the current interlaboratory results; the sensitive rheological parameters and boundaries may be updated and refined with additional tests.

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REFERENCE

1. A. Merrington. "Recycling of plastics," in *Applied Plastics Engineering Handbook*, ed. M. Kutz (William Andrew Publishing, 2017), 167-189. https://doi.org/10.1016/B978-0-323-39040-8.00009-2
2. R. Geyer, J. R. Jambeck, and K. L. Law, "Production, use, and fate of all plastics ever made," *Science advances*, 3(7), e1700782. (July 2017): 1-5. https://doi.org/10.1126/sciadv.1700782
3. K. R. Vanapalli, H. B. Sharma, V. P. Ranjan, B. Samal, J. Bhattacharya, B. K. Dubey, and S. Goel, "Challenges and strategies for effective plastic waste management during and post COVID-19 pandemic," *Science of The Total Environment*, 750, 141514. (January 2021):1-10 https://doi.org/10.1016/j.scitotenv.2020.141514
4. JTTE Editorial Officea, J. Q. Chen, H. C. Dan, Y. J. Ding, ... , and X. Y. Zhu, "New innovations in pavement materials and engineering: A review on pavement engineering research 2021," *Journal of Traffic and Transportation Engineering (English Edition)*, 8(6), (December 2021): 815-999 https://doi.org/10.1016/j.jtte.2021.10.001
5. Z. N. Kalantar, M. R. Karim, and A. Mahrez, "A review using waste and virgin polymer in pavement," *Construction and Building Materials*, 33, (August 2012): 55-62 https://doi.org/10.1016/j.conbuildmat.2012.01.009
6. S. Karmakar, and T. K. Roy, "Effect of waste plastic and waste tires ash on mechanical behavior of bitumen," *Journal of Materials in Civil Engineerin*g, 28(6), 04016006. (January 2016): 1-9 https://doi.org/10.1061/(ASCE)MT.1943-5533.0001484

7. Z. Zhao, F. P. Xiao, and S. Amirkhanian, "Recent applications of waste solid materials in pavement engineering". *Waste Management*, 108, (May 2020): 78-105 https://doi.org/10.1016/j.wasman.2020.04.024

8. L. D. Poulikakos, C. Papadaskalopoulou, B. Hofko, F. Gschösser, A. Cannone Falchetto, ... and M. N. Partl, "Harvesting the unexplored potential of European waste materials for road construction," *Resources, Conservation and Recycling*, 116, (January 2017): 32-44 https://doi.org/10.1016/j.resconrec.2016.09.008

9. M. Tušar, M. R. Kakar, L. D. Poulikakos, E. Pasquini, A. Baliello, M. Pasetto, L. Porot, D. Wang, A. Cannone Falchetto, D. Dalmazzo, D. Lo Presti, G. Giancontieri, A. Varveri, R. Veropalumbo, N. Viscione, K. Vasconcelos, and A. Carter, "RILEM TC 279 WMR round robin study on waste polyethylene modified bituminous binders: advantages and challenges," *Road Materials and Pavement Design*, (January 2022): 1-29. https://doi.org/10.1080/14680629.2021.2017330

10. D. Wang, A. Baliello, L. D. Poulikakos, K. Vasconcelos, M. R. Kakar, G. Giancontieri, E. Pasquini, L. Porot, M. Tušar, C. Riccardi, M. Pasetto, D. Lo Presti, and A. Cannone Falchetto, "Rheological properties of asphalt binder modified with waste polyethylene: an interlaboratory research," *Resources, Conservation and Recycling* (in review)

11. S. Wu, and L. Montalvo, "Repurposing waste plastics into cleaner asphalt pavement materials: A critical literature review," *Journal of Cleaner Production,* 280, 124355. (January 2021): 1-55 https://doi.org/10.1016/j.jclepro.2020.124355

12. M. R. Kakar, P. Mikhailenko, Z. Y. Piao, M. Bueno, and L. Poulikakos, "Analysis of waste polyethylene (PE) and its by-products in asphalt binder," *Construction and*

Building Materials, 280, 122492. (April 2021): 1-12 https://doi.org/10.1016/j.conbuildmat.2021.122492

13. Y. R. Kim, *Modeling of asphalt concrete*, 1st ed. (McGraw-Hill Education, ASCE, 2009).
14. American Association of State Highway and Transportation Officials - Standard Method of Test for Determining the Rheological Properties of Asphalt Binder Using a Dynamic Shear Rheometer (DSR), AASHTO M 315-20 (2021). (AASHTO, approved 2020).
15. Bitumen and bituminous binders - Determination of complex shear modulus and phase angle - Dynamic Shear Rheometer (DSR), EN 14770 (2012) (European Standard, approved May 16, 2022).
16. L. D. Poulikakos, A. Cannone Falchetto, D. Wang, L. Porot, and B. Hofko, "Impact of asphalt aging temperature on chemo-mechanics," *RSC advances*, 9(21), (April 2019):11602-11613 https://doi.org/10.1039/C9RA00645A
17. D. Wang, A. Cannone Falchetto, L. Poulikakos, B. Hofko, and L. Porot, "RILEM TC 252-CMB report: Rheological modeling of asphalt binder under different short and long-term aging temperatures," *Materials and Structure*, 52(4), (June 2019):52-73 https://doi.org/10.1617/s11527-019-1371-8
18. B. Hofko, A. Cannone Falchetto, J. Grenfell, L. Huber, X. H. Lu, L. Porot, L. D. Poulikakos, and Z. You, "Effect of short-term ageing temperature on bitumen properties," *Road Materials and Pavement Design*, 18(sup2), (March 2017): 108-117 https://doi.org/10.1080/14680629.2017.1304268
19. A. I. Al-Hadidy, Y. Q. Tan, "Effect of polyethylene on life of flexible pavements," *Construction and Building Materials*, 23(3), (August 2008): 1456-1464. https://doi.org/10.1016/j.conbuildmat.2008.07.004

20. L. Garcia Cucalon, F. Kaseer, E. Arámbula-Mercado, A. Epps Martin, N. Morian, S. Pournoman, and E. Hajj, "The crossover temperature: significance and application towards engineering balanced recycled binder blends," *Road Materials and Pavement Design*, 20(6), (August 2019): 1391-1412 https://doi.org/10.1080/14680629.2018.1447504

21. S. Mangiafico, H. Di Benedetto, C. Sauzéat, F. Olard, S. Pouget, S. Dupriet, L. Planque, and R. Van Rooijen, "Statistical analysis of the influence of RAP and mix composition on viscoelastic and fatigue properties of asphalt mixes," *Materials and Structures*, 48(4), (April 2015): 1187-1205 https://doi.org/10.1617/s11527-013-0225-z

22. K. Haslett, E. Dave, J. Sias, and E. Linder, "Statistical Analysis Framework to Evaluate Asphalt Concrete Overlay Reflective Cracking Performance," *Transportation Research Record*, (March 2022): 03611981221078570 https://doi.org/10.1177/03611981221078570

23. A. Amini, and R. Imaninasab, "Investigating the effectiveness of Vacuum Tower Bottoms for Asphalt Rubber Binder based on performance properties and statistical analysis," *Journal of Cleaner Production*, 171, (January 2018): 1101-1110 https://doi.org/10.1016/j.jclepro.2017.10.103

24. A. Kavussi, M. Qorbani, A. Khodaii, and H. F. Haghshenas, "Moisture susceptibility of warm mix asphalt: a statistical analysis of the laboratory testing results," *Construction and Building Materials*, 52, (February 2014): 511-517 https://doi.org/10.1016/j.conbuildmat.2013.10.073

25. L. D. Poulikakos, E. Pasquini, M. Tusar, D. Hernando, D. Wang, P. Mikhailenko, ... , and F. M. Navarro, "RILEM interlaboratory study on the mechanical properties of asphalt

mixtures modified with polyethylene waste," *Journal of Cleaner Production*, 134124, (November 2022): 1-10 https://doi.org/10.1016/j.jclepro.2022.134124

26. G. D. Airey, "Use of black diagrams to identify inconsistencies in rheological data," *Road Materials and Pavement Design*, 3(4), (September 2002): 403-424 https://doi.org/10.1080/14680629.2002.9689933

27. D. Wang, A. Cannone Falchetto, A. Alisov, J. Schrader, C. Riccardi, and M. P. Wistuba, "An alternative experimental method for measuring the low temperature rheological properties of asphalt binder by using 4mm parallel plates on dynamic shear rheometer," *Transportation Research Record*, 2673(3), (March 2019): 427-438 https://doi.org/10.1177/0361198119834912

28. D. Wang, A. Cannone Falchetto, C. Riccardi, and M. P. Wistuba, "Investigation on the low temperature properties of asphalt binder: Glass transition temperature and modulus shift factor," *Construction and Building Materials*, 245(118351), (June 2020): 1-12 https://doi.org/10.1016/j.conbuildmat.2020.118351

29. G. M. Rowe, "Interrelationships in rheology for asphalt binder specifications" (paper presentation, fifty-ninth annual conference of the canadian technical asphalt association (CTAA): Winnipeg, Manitoba. November, 2014)

30. European Standard for Bitumen and bituminous binders - Specification for paving grade bitumens, EN 12591 (2022) (European Standard, approved January 15, 2022).

31. D. Sybilski, A. Vanelstraete, and M. N. Partl, "Recommendation of RILEM TC 182-PEB on bending beam and rheometer measurements of bituminous binders," *Materials and structures*, 37(8), (October 2004): 539-546 https://doi.org/10.1007/BF02481578

32. R. A. Velasquez, *On the representative volume element of asphalt concrete with applications to low temperature*. Ph.D. thesis, University of Minnesota. Minnesota, USA. 2009.