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Rheological properties of asphalt binder modified with waste polyethylene: An interlaboratory research from the RILEM TC WMR

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ABSTRACT

Significant volumes of waste materials such as plastics is generated every year worldwide with a potentially harmful impact on the environment. At the same time, the demand for asphalt modifiers has seen an increase over the year with consequent higher costs for these types of additives. Therefore, combining large amounts of available waste plastics with asphalt binder as an extender or modifier would potentially improve the paving material properties while limiting the disposed waste. While several rheological studies have been performed in the past, they were restricted to single research efforts hindering a consistent comparison among the valuable results of these investigations. For this reason, the Task Group 1 of the RILEM Technical Committee 279-WMR established a research activity with 11 international institutions to conduct interlaboratory research to evaluate the possibility of using waste polyethylene (PE) as an additive in asphalt binder. The study addressed the combined impact of PE materials and experimental conditions on the rheological properties of asphalt binder. For this purpose, conventional tests (penetration value, softening point temperature, and Fraass breaking point temperature) and the linear viscoelastic characterization using the Dynamic Shear Rheometer (DSR) were adopted. An unaged pen grade 70/100 neat binder was selected as the reference binder; PE-pellets and PE-shreds, produced from the recycled waste polyethylene materials, were used as the polymer additives. A single content (5%) of PE-pellets and PE-shreds was used to blend the PE material with the reference binder (95%) and prepare the two PE modified binders. Results indicate that the use of plastic modifiers leads to an overall higher complex shear modulus and softening point temperature while decreasing the penetration value. Higher dispersion in the results, especially in phase angles, was observed for blended binders at high temperatures. The PE modified binders exhibited poor reproducibility among laboratories and a low level of repeatability. Such a scatter in the data could result from an uneven dispersion of plastic material at high temperatures. In contrast, plastic shapes and batches appeared to have a limited impact. Three different rheological behaviors, neat binder, modified binder, and complex modified binder, were visually identified among the interlaboratory results and based on a simple statistical analysis of variance. Further analysis of the data suggested that the Glover Rowe (*G-R)* parameter can be used as a sensitive tool to classify the rheological behaviors of PE modified binders. Further experimental evaluation on specific testing conditions, such as measurement gaps of DSR at high temperatures, is

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recommended to advance the understanding of their influence on the rheological behavior of PE modified binders.

1. Introduction and background

Currently, approximately 9 billion tonnes of waste are generated annually worldwide. The list includes municipal, agricultural and animal, medical, hazardous, industrial non-hazardous, and construction and demolition debris (Wilson and Velis, 2015). However, due to the mainly linear material processes used in economic activities, most of the wastes end up in landfills, dumps, or the natural environment (UNEP, 2021), which ultimately lead to serious environmental issues and consumption of nonrenewable resources. This situation is especially true in developed industrial countries, producing more waste than the worldwide average. Hence, there is an urgent need to promote waste reuse and recycling and phasing out disposal or incineration of waste, for example, in Europe (Nugent, 2017). An ambitious European strategy was proposed in 2020 to promote smart, sustainable, and inclusive growth by adopting drastic actions (Europe 2020 Strategy, 2020). The same document states: "*strong dependence on fossil fuels such as oil and inefficient use of raw materials expose European consumers and businesses to harmful and costly price shocks, threatening economic security and contributing to climate change*". Hence, the reuse or recycling of fossil-based products is a top priority. Among these products, plastic occupies 8% – 10% of the total consumption (1 bag at a time - Plastic Bags and Petroleum 2021), while more than 99% of plastic products are made from nonrenewable resources (CIEL, 2021). Moreover, considering the specificity of plastic, its value-chains, and accounting for its environmental footprint, the reuse or recycling of waste plastic appears to be essential for the transition toward a circular economy (Closing the loop-An EU action plan for the Circular Economy, 2015).

Several ambitious attempts have been made in Europe to recover and recycle plastics in the past decades, but this situation varies significantly among countries. Even though some countries recover almost 100% of the collected post-consumer waste plastics, around 32% of collected waste plastic are recycled, and about 25% (more than 14 million tons) goes to landfills (Preventing plastic waste in Europe, 2019; Plastics-the Facts 2020, 2020; Larrain *et al.*, 2021). Therefore, finding new ways to turn waste plastic into a valuable resource or secondary raw materials is essential within the circular economy.

Roads, especially asphalt pavements, are one of the most relevant components of the European infrastructure. From 2009 to 2019, the total annual production of asphalt mixture was about 270 million tonnes. The corresponding consumption of fossil-based asphalt binder and additives was around 14 million tonnes (Asphalt in Figures 2019, 2020). Despite this massive material production, different polymer additives, fibers, and modifiers are needed to improve the performance and durability of asphalt materials. However, the high cost of the conventional synthesis or natural polymer restricts its large-scale application in asphalt pavements. Hence, using inexpensive polymers from waste plastics in road construction is a promising option for the sustainable development of plastic materials and asphalt pavements (Haider *et al.*, 2020). In asphalt pavement, the reuse of waste plastic, polyethylene terephthalate (PET), dates back to the early 1990s (Ledesma and Isaacs, 1991). Since then, different shapes of waste plastics (pellets, shreds, or granulates) from multiple sources, such as PET, polyethylene (PE), polypropylene (PP), polyurethane (PU), ethylene-vinyl acetate (EVA), polyvinyl chloride (PVC) and different types of plastic fibers have been developed to be incorporated into asphalt pavement (Poulikakos et al., 2017; Office *et al.*, 2021; Abdy *et al.*, 2022). Among them, PE is one of the most available waste sources (Tušar *et al.*, 2021).

Two approaches exist to incorporate waste plastics in asphalt pavement: dry and wet processes. Plastics are added directly to the mixture as an aggregate/filler replacement or mixture modifier in the dry process (Radeef *et al.*, 2021). It is worth noting that part of the melted plastic works as binder polymer additives. Hence, the main obstacle in implementing the dry process concerns the lack of consistency of the final produced mix. In the case of the wet process, plastics are added directly to the asphalt binder and then incorporated into the mixture (Ma *et al.*, 2021). Scattered results were observed at high temperatures. However, such a process also presents limitations due to the poor storage stability of the plastic modified binders, where the recycled polymers tend to separate from the asphalt binder due to the difference in density and viscosity and the incompatibility between the two components (Kakar et al., 2021). Hence, the effect of experimental conditions should be carefully studied, especially on the high temperature properties.

Research has made significant progress in incorporating waste plastics into building and construction materials. However, the studies focused mainly on cement and concrete materials (Shoubi *et al.* 2013; Ganesh Prabhu *et al.* 2014; Saikia and de Brito 2014; Sharma 2017; White, Reid 2018), while little work can be found on the application to bituminous materials (Gürü *et al.* 2014; Dalhat and Al-Adbul Wahhab 2017; Leng *et al.* 2018; White and Reid 2019).

The possibility of using waste plastic from different sources (industrial plastics, plastic packages) as a binder extender/modifier in both mixture and binder scales has been studied for asphalt materials. Most of the studies on the mixture phase showed very encouraging results (Al-Hadidy *et al.*, 2009a, 2009b; Attaelmanan *et al.*, 2011; Gawande *et al.*, 2012; Kalantar *et al.*, 2012; Rokade, 2012; Gawande, 2013; Zhang and Hu, 2016; Nouali *et al.*, 2020). The addition of recycled waste plastics enhances thermal stability (Fuentes-Audén *et al.*, 2008; Naskar *et al.*, 2010), Marshall stability (Hassani *et al.*, 2005; Abdullah *et al.*, 2017), resistance to permanent deformation (Sabina *et al.*, 2009; Köfteci *et al.*, 2014), fatigue performance (Gawande *et al.*, 2012; Chee *et al.*, 2014), and moisture resistance (Ahmad 2014; Singhal *et al.*, 2016). In the binder phase, the waste plastic modified binder indicates better rheological properties than the original neat binder (Nasr and Pakshir, 2019). A higher softening point temperature and viscosity, lower penetration value, and improved viscoelastic properties such as higher complex shear modulus and lower phase angle are observed (Al-Hadidy *et al.*, 2009a; Abdelaziz and Mohamed Rehan, 2010; Habib *et al.*, 2011; Kumar and Garg, 2011; Sadeque and Patil, 2013; Singhal *et al.*, 2016; Hu *et al.*, 2018). Therefore, depending on the waste plastic source and approaches, dry or wet processes used to incorporate the plastic in the asphalt material, the modified binders and mixtures exhibit different overall performances. Consequently, more research is needed to understand better the impact of recycled plastics on materials' performance.

According to previous studies (Kalantar *et al.*, 2012; Poulikakos *et al.*, 2017), understanding the rheological properties of the modified binder is a critical task in selecting the appropriate materials for asphalt pavement construction. More profound knowledge in this area is also instrumental for improving the prediction of the mechanical properties and performance of asphalt mixture and asphalt pavement. The Dynamic Shear Rheometer (DSR) is a well-established testing tool to evaluate the viscoelastic properties of plain and modified binders over a wide range of frequencies and temperatures. The device was initially developed to determine the viscoelastic properties at intermediate and high temperatures (AASHTO T315, 2020; AASHTO M320, 2021). More recently, several research efforts have demonstrated that DSR could also be used for low temperature characterization (Farrar *et al.*, 2015; Wang *et al.*, 2019a). With such a device, different tests, including temperature-frequency sweep (T-f sweep), multiple stress creep recovery (MSCR), and Linear Amplitude Sweep (LAS) tests, can be performed. This range of experimental methods allows one to evaluate the binders' performance inside and outside the linear viscoelastic range to predict the pavement's performance. In particular, different studies (Hofko *et al.*, 2017; Poulikakos *et al.*, 2019; Wang *et al.*, 2019b) showed that T-f sweep tests are a powerful test method to characterize the asphalt binders' rheological behavior within the linear viscoelastic (LVE) range. In addition, other rheological parameters, such as crossover temperature (Garcia Cucalon *et al.*, 2019) and Glover-Rowe parameter (Glover *et al.*, 2005; Rowe, 2014), can be further used to evaluate the rutting and cracking resistance of asphalt binders.

Given this background, the large availability of waste plastic such as PE and its potential for being incorporated into paving materials present the double benefit of reducing waste disposal while possibly improving the properties of asphalt binders and mixtures. Despite several efforts devoted to investigating the use of PE in asphalt material, the results of these studies remain challenging to compare across the different research institutions, hindering the full exploitation at a broader scale. To overcome this limitation, the RILEM Technical Committee TC-279 WMR (Valorisation of Waste and Secondary Materials for Roads) Task Group TG 1, in its ongoing activities, addressed how waste plastic could be incorporated as a polymer modifier in asphalt binder (Tušar *et al.*, 2021). For this purpose, interlaboratory activity with 11 participating international institutions was established to provide a solid research outcome supported by a robust consistency of the experimental approach and scientific practices, eventually leading to reliable results and a widely accepted research outcome. These could further encourage the paving industry to adopt a technology that can increase the sustainability of the construction process toward a circular economy where the transformation process of the waste PE into asphalt paving materials promotes a wiser use of natural resources and encourage material innovation of asphalt additives through the application of inexpensive PE modifiers. At the same time, reduced waste disposal, less expensive road construction, and a more resource-efficient process can ultimately benefit the entire society in the awareness that a higher level of sustainability can ultimately improve individuals' well-being.

2. Objective and research approach

As mentioned in the introduction, the application of waste plastic materials in asphalt pavement construction is a timely technology with several potential economic and environmental benefits (Ahmadinia *et al.*, 2011; Poulikakos *et al.*, 2017). However, it was shown that the current technology readiness level (TRL) is between 5 and 7 (European Commission, 2014), indicating that pilot projects have been implemented in the field and the widespread industrial implementation is still facing barriers (Piao *et al.*, 2021). Hence, the use of waste plastics needs more reliable and scrutinized investigation. PE is one of the most promising and commonly used materials among different waste plastic sources. Therefore, RILEM TC-279 WMR established a TG to evaluate the possibility of utilizing waste PE as an additive to enhance the asphalt binders. The following research objectives were identified to address this goal:

- Characterize the rheological properties of PE modified asphalt binders over a wide range of temperatures and frequencies by using DSR.
- Identify parameters to discriminate the rheological behaviors between unmodified and PE modified binders and possibly within plastic-modified binders.

An interlaboratory testing protocol with eleven participating laboratories was designed for this purpose. In this study, plastic pellets (PE-pellets), produced for recycling from waste packaging materials, and their by-products, plastic shreds (PE-shreds), were selected as the binder additives. The effect of waste plastic shape, blended batches, sample preparation, and experimental conditions was evaluated. Fig. 1 summarizes the research approach adopted in the present study. First, an unaged 70/100 penetration grade (EN 12591, 2015) neat binder, as reference material, and two PE waste sources (PE-pellets and PE-shreds), as polymer additives, were chosen respectively. Two laboratories prepared the testing materials by blending PE-pellets or PE-shreds with the reference neat binder at a single content of 5%. B, $B_{+pellets}$, $B_{+shreds}$ are used to represent the neat binder, neat binder blended with PE-pellets,

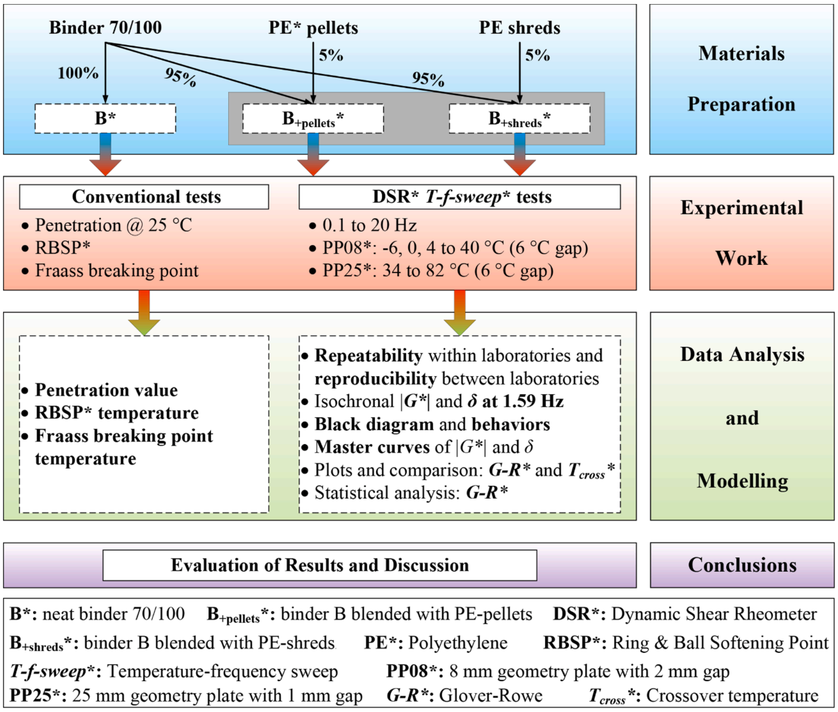

Fig. 1. Research approach.

and neat binder combined with PE-shreds, respectively. Next, conventional empirical tests, including needle penetration (EN 1426, 2015), softening point (EN 1427, 2015), and Frass breaking point (EN 12593, 2015), were conducted on these three binders. Moreover, temperature-frequency sweep (T-f sweep) tests over a wide range of temperatures, −6 °C – +82 °C, and frequencies, 0.1 – 20 Hz, were carried out to measure the complex shear modulus, $|G^*|$, and phase angle, δ. Then, isochronal curves of $|G^*|$ and δ at 1.59 Hz were generated to evaluate repeatability (within laboratory) and reproducibility (among laboratories). Following the visual analysis of the black diagram and master curve, numerical and statistical analysis was also conducted on the calculated rheological parameters and master curves' fitted parameters. The effect of plastic shape, sample preparation, and experimental conditions was also discussed. Finally, recommendations on the testing protocols were also proposed in the perspective of follow-up research.

3. Materials and testing

3.1. Materials preparation

In this project, an unaged penetration graded 70/100 neat binder (EN 12591, 2015) (Venezuelan origin), provided by a European binder supplier, was selected as reference material. The recycled waste PE materials were collected from a Swiss plastic recycling company. PE pellets are produced by processing waste packaging materials primarily consisting of PE; PE shreds are the secondary waste and a by-product from the production process of the pellets. The pellets are further used to produce other items such as pipes, while the shreds are burned as fuel by the cement plants. An important aspect is that the cement industry benefits from financial support to burn shreds instead of fossil-based energy, as they are accepting a waste product (Imbabi *et al.*, 2012). On the other hand, there is a potential to use these materials for other sustainable value chain purposes, such as producing asphalt materials. It should be noted that the main difference between these two PE additives is the shape while they came from a single source.

A single 5% content of PE additive was used to prepare the blended binder with the neat 70/100 binder (95%). To avoid any uncertainty during material preparation, two laboratories produced these blended binders (Table 1) and distributed them to the eleven laboratories involved in the interlaboratory test. The following protocol was followed to prepare the blended binders. For the shred plastic, it was first grounded to facilitate the blending process with the neat binder. Next, the neat 70/100 binder was heated at 170 °C for one hour in the oven, and then the PE additives were directly introduced for blending. The blending process was performed using a high-shear mixer for one hour at 3500 rpm. More specific details on the grinding and blending process and chemical and morphological effects of these additives can be found in the pre-study of this current work (Kakar *et al.*, 2021; Tušar *et al.*, 2021). According to this earlier study, the inclusion of PE waste does not change the composition of the bitumen; the incorporation occurs physically. They also addressed the storage stability of the PE blended binders. The material designation used is as follows: the number is the lab number, subscript a, b indicates repeated tests by the same laboratory followed by the binder type: B, $B_{+pellets}$, $B_{+shreds}$. The two batches are identified using a dot followed by the batch number; for example, 1.1 B indicates neat binder results by laboratory 1, first batch, while 1.2 B identifies neat binder results by laboratory 1, second batch. Table 1 summarizes the binders tested from the different laboratories. In an authors' previous study (Tušar *et al.*, 2021), it was found that different batches only lead to limited influence on the rheological properties of PE modified binders. Hence, only batch 1 blending at laboratory 1 was used for analysis in this study.

3.2. Conventional empirical tests

Conventional tests, including needle penetration (EN 1426, 2015; ASTM D5/D5M, 2020), and softening point (EN 1427, 2015), were conducted in this study. Due to the limited amount of materials, only laboratories 7 and 10 performed both tests on all three binders (Table 2). All penetration values were consistent between these two laboratories, while both PE modified binders decreased the penetration values by about 50%. However, remarkably different softening point temperatures were observed between these two laboratories. The difference in softening point temperature between $B_{+pellets}$ is only 3 °C, while $B_{+shreds}$ reached 30°C. As previously mentioned, PE-shreds are the secondary waste from the production of PE-pellets; hence, it is expected that the components of PE-shreds and their distribution in the blended binder $B_{+shreds}$ are more diverse in comparison to the one in $B_{+pellets}$ (Kakar *et al.*, 2021). Therefore, the inhomogeneous distribution may lead to a much larger difference in the results; this phenomenon is especially true for low viscosity binders at relatively high temperatures. Hence, it is not surprising that more scattered results were observed in the $B_{+shreds}$ compared to $B_{+pellets}$. Consequently, poor repeatability results were observed within a single laboratory; for example, the case of Batch 1-$B_{+shreds}$ by laboratory 1. Fig. 2 visually demonstrates this diverse response where the left ball of the ring and ball device had already reached the bottom plane while the right one remained on the ring. Moreover, poor reproducibility trends among laboratories were also found for the measurements obtained from the Ring and Ball test on $B_{+shreds}$ ranging from 45 °C to 110 °C (Tušar *et al.*, 2021).

Besides the penetration value at 25°C and softening point temperature tests, Fraass breaking point temperature (EN 12593, 2015) and the storage stability (EN 13399 2017) were determined by one laboratory (laboratory 7). Fraass breaking point temperature increased in comparison to the neat binder when adding plastic waste (+7 °C, and +5 °C in the case of B+pellets and B+shreds). Further explanation is provided in the detailed analyses reported in a different work (Kakar *et al.*, 2021; Tušar *et al.*, 2021). To expand the information obtained on PE modified binder from the initial set of conventional tests, temperature-frequency sweep (T-f sweep) tests were conducted.

3.3. DSR testing protocol

In this study, temperature-frequency sweep tests were conducted by eleven laboratories on the neat binder and the different batches of blended binders by using the DSR devices. Two plate-plate geometries were selected for the different temperature ranges. The 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. While the 8 mm plate-plate geometry with 2 mm gap (PP08) was selected for the lower temperature range ($T = -6$, 0, 4, 10, 16, 22, 28, 34, and 40 °C). Fig. 3 presents samples from the three binders at the conclusions of the

Table 1
Overview of the laboratories, binder batches and materials used.

Laboratories		1	2	3	4	5	6	7	8	9	10	11
B			√					√			√	√
Batch 1 (2018)	$B_{+pellets}$		√									
	$B_{+shreds}$	√	√	√	√	√	√			√		√
Batch 2 (2019)	$B_{+pellets}$	√						√	√		√	
	$B_{+shreds}$	√						√	√		√	

Table 2
Example of penetration and ring and ball temperature for the bitumen blends from Batch 2.

Laboratories	B Penetration value [0.1 mm]	RBSP [°C]	Batch 2 $B_{+pellets}$ Penetration value [0.1 mm]	RBSP [°C]	Batch 2 $B_{+shreds}$ Penetration value [0.1 mm]	RBSP [°C]
7	88	45.0	36	63.4	40	109.0
10	81	45.8	38	60.8	39	79.0

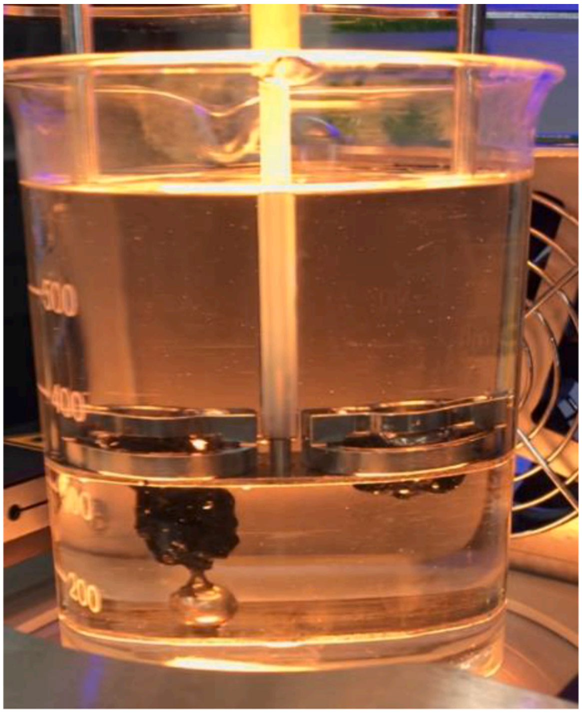

Fig. 2. Example of the softening point experiments of $B_{+shreds}$ (Laboratory 1 batch 1) at end of the test showing that one ball descended the ring while the second one was still on the ring.

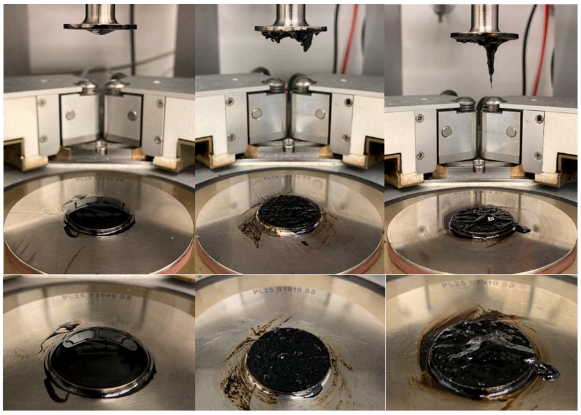

Fig. 3. Laboratory 10 Bitumen samples at the conclusion of the DSR measurements with the PP25 geometry at 100°C front view and top view: left: B; middle: $B_{+pellets}$; right: $B_{+shreds}$.

DSR measurements when the temperature was still 100 °C (laboratory 10). The snapshots distinctly visualize a rough and heterogenous composition in the waste PE modified binder, and a different appearance is visible between $B_{+pellets}$ and $B_{+shreds}$. As discussed previously, an inhomogeneous distribution of PE particles may occur, especially at relatively high temperatures. Hence, the inhomogeneous distribution and binder morphology may significantly differ for PP25 results within and among laboratories. This trend may not be the case for PP08 results, where the structure is more driven by the binder matrix's viscosity, which leads to more stable distribution and similar results within and among laboratories at relatively low temperatures. The specific influence of PE shape and batch, and testing temperatures will be discussed based on the DSR results.

All the DSR T-f sweep measurements were performed within the linear viscoelastic (LVE) range. The suggested strain levels of 0.1% and 0.05% were used when adopting the corresponding testing geometry PP25 and PP08, respectively. Alternative strain/stress levels within the LVE range were also determined through the amplitude sweep test by laboratory 10 to obtain reliable results; such a mixed stress/strain control mode was validated in several previous studies (Wang *et al.*, 2019a, 2021). During the testing, the gap/normal force control was applied to adjust the measurements under different testing temperatures for each laboratory. Although all laboratories followed the general guidelines of a common testing protocol, differences in the experimentation were observed. More information about the testing protocols, such as the conditioning time, testing duration, the order of temperature and frequency application, and replicates for blended binders, were collected from each laboratory and summarized in Table 3. Besides the experimental conditions, the differences between the materials themselves, PE

Table 3
DSR experimental conditions applied in each laboratory.

Laboratories	Low temperatures (PP08) t_c*	 T*	 f *	High temperatures (PP25) t_c*	 T*	 f *	Replicates ($B_{+pellets}$ and $B_{+shreds}$)
1	20	High to low	Low to high	15	High to low	Low to high	2
2	30	Low to high	High to low	30	Low to high	High to low	1
3	10	Low to high	Low to high	10	Low to high	Low to high	3
4	20	Low to high	High to low	20	Low to high	High to low	5
5	10	Low to high	Low to high	10	Low to high	Low to high	1
6	10	High to low	High to low	10	Low to high	High to low	1
7	10	High to low	Low to high	10	Low to high	Low to high	1
8	5	Low to high	Low to high	5	Low to high	Low to high	1
9	10	Low to high	Low to high	15	Low to high	Low to high	1
10	40	Low to high	Low to high	20	Low to high	Low to high	2
11	30	High to low	Low to high	30	High to low	Low to high	1

t_c*: conditioning time at each temperature, unit: minutes; T* and f*: order of temperature and frequency sweep.

shapes (PE-pellets and PE-shreds), and materials sources (Batches 1 and 2) may influence the final results. The combined effect of materials and experimental conditions are analyzed in the next section, followed by discussions and recommendations.

4. Results and analysis

4.1. Preliminary inspection of the data

As a first step, data analysis was performed by constructing the isochronal curves of the complex shear modulus, $|G^*|$, and the phase angle, δ, at the reference frequency $f = 1.59$ Hz (corresponding to 10 rad/s). In particular, the interlaboratory tests' reliability was evaluated by assessing the repeatability among the involved participants. The precision of the data for a single-operator testing (within the same laboratory) for neat binder and PE blends was checked according to AASHTO T315-20 (2020). The standard prescribes the acceptance limits for the standard deviation of $|G^*|/\sin\delta$ parameter (AASHTO T315, 2020) for unaged materials, calculated according to ASTM C670-15 (2015). The maximum variation coefficient *1s%* (standard deviation) is fixed to 1.6%.

The results of data replicates are presented in Fig. 4. Results indicate that AASHTO repeatability criteria for a single-operator testing (repeatability within a single laboratory) were met only when testing unmodified neat binder B. Moreover, for the blended PE modified binders, the scattering of the data became significant, leading to a much greater *1s%*. $B_{+pellets}$ analyzed by laboratory 10 tended to be still acceptable in the majority of the tested temperatures. However, $B_{+pellets}$ tested by laboratory 1.2 (Laboratory 1 Batch 2) mainly exhibited *1s%* meeting the acceptance criteria given by the reference standard. $B_{+shreds}$ blends exceeded the allowable errors regardless of the testing laboratory. This deviation may be attributed to the more diverse distribution of the shred particles at high temperatures (Kakar *et al.*, 2021). It should be noted that the precision limitation based on the AASHTO standard is designed for the neat binder tested above 6 °C (AASHTO T315, 2020). Both blended binders used in this paper are highly polymer modified binders. Moreover, the lowest testing temperature reached −6 °C. Hence, such precision limitations and the applicability for these conditions should be treated with caution. *1s%* here is only used to evaluate the effect of plastic shapes, and experimental conditions instead of fulfilling the required bias. Despite this consideration, the results reported in Table 4 cover the entire spectrum of temperatures. This decision was made to evaluate the experimental data based on an accepted standard parameter that includes information on the consistency of measurements by incorporating both |G*| and δ when estimating the repeatability. The authors are aware that, at lower temperatures, a small variation in phase angle may significantly affect the value of $|G^*|/\sin\delta$, and negatively influence repeatability. Nevertheless, this parameter provides a broader view of the measured data concerning repeatability than the single |G*| and δ. In Table 4 and the following tables, the measurements with the PP08 geometry are reported in italic.

In general, it was noticed that testing temperature had a direct and significant effect on the results; less variability was found for all materials at low testing temperatures (PP08), especially when considering $B_{+pellets}$ and $B_{+shreds}$. The modified binders showed a similar complex shear modulus to the neat binder in the cold temperature regime. In contrast, all the laboratories could see an increase in the complex shear modulus in comparison to the neat binder in the high temperature regime. The phase angle showed a more elastic behavior in the cold temperature domain. On the other hand, in the high temperature regime, the effect of the plastics was seen in a reduction of the phase angle compared to the neat binder indicating a more elastic response. In this temperature range, a greater difference was also observed in the results of the participating laboratories. These findings could be an

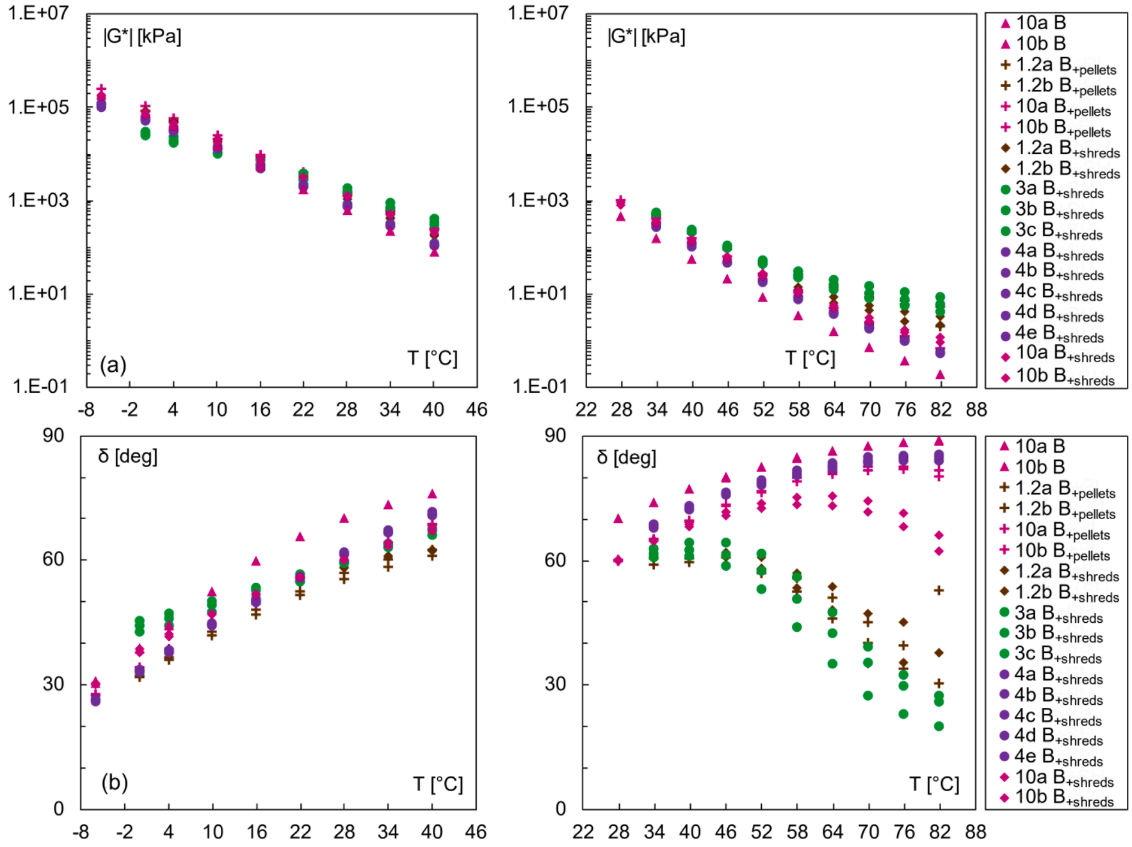

Fig. 4. Isochronal plots of Binder B, $B_{+pellets}$, and $B_{+shreds}$ blends at the reference frequency of 1.59 Hz (10 rad/s). a) $|G^*|$ at low (left) and high (right) testing temperature; b) δ at low (left) and high (right) testing temperature.

Table 4

Repeatability statistics of *1s%* for $|G^*|/\sin\delta$ precision [kPa] at the reference frequency 1.59 Hz (10 rad/s): single-operator precision: B, $B_{+pellets}$, and $B_{+shreds}$.

Material	B	$B_{+pellets}$		$B_{+shreds}$			
laboratories	10	1.2	10	1.2	3	4	10
-6 (PP08)	1.7*	-	0.3	-	-	5.4*	5.4*
0 (PP08)	2.4*	0.8	0.7	0.6	10.0*	6.3*	6.4*
4 (PP08)	1.3	0.6	0.8	1.0	13.2*	6.5*	7.1*
10 (PP08)	0.7	0.3	0.9	1.1	13.4*	5.7*	7.6*
16 (PP08)	0.0	1.5	1.1	2.1*	15.5*	6.9*	8.0*
22 (PP08)	0.8	2.1*	1.2	1.9*	15.2*	5.6*	0.2
28 (PP08)	0.6	3.4*	1.0	1.9*	11.8*	5.7*	3.9*
34 (PP08)	0.7	4.5*	1.2	2.2*	10.6*	5.6*	4.0*
40 (PP08)	0.5	3.8*	1.2	3.2*	12.2*	5.3*	4.5*
28 (PP25)	0.3	-	0.4	-	-	5.9*	6.4*
34 (PP25)	0.4	3.0*	0.6	2.8*	3.4*	5.3*	5.0*
40 (PP25)	1.1	3.8*	0.6	3.5*	4.5*	1.3	4.7*
46 (PP25)	0.7	4.1*	0.3	3.9*	7.7*	4.3*	6.4*
52 (PP25)	0.8	5.2*	3.3*	6.9*	12.4*	3.6*	6.5*
58 (PP25)	0.8	7.2*	3.0*	9.9*	20.4*	3.3*	8.8*
64 (PP25)	0.7	9.3*	2.6*	17.3*	30.3*	2.5*	8.5*
70 (PP25)	0.6	13.8*	2.1*	25.4*	37.9*	2.4*	8.6*
76 (PP25)	1.0	16.7*	1.4	32.3*	42.0*	2.2*	9.8*
82 (PP25)	0.9	61.4*	2.2*	35.5*	44.1*	-	11.2*

- : non-available data; *1s%*: standard deviation.

*: data not meeting AASHTO T315-20 criteria – max(1s%): 1.6.

indication that the plastic particles are, in fact, being measured instead of the whole blend due to the lower viscosity at the higher testing temperatures (softer binder), revealing the sample's inhomogeneity and subsequent fluctuations of results (depending on the volume of plastic particles in the specimen). The greater diameter and lower gap of the samples used for high temperature analysis (PP25) could be an additional factor increasing such effect. Reasonable sample stability was observed during tests of all three bituminous blends, regardless of the testing temperature.

Based on the data reported in Fig. 4 and Table 4, no correlations between the scattering of the data and curves' shapes were identified. Interestingly, different forms of the phase angle curves at high testing temperatures (Fig. 4b right) were found. These diverse trends may indicate a differentiation in rheological behavior that will be discussed later.

4.2. Black diagram and classification of binders' behavior

As mentioned in the previous section, different rheological behaviors were visually detected in the phase angle vs. temperature plots. However, the varied polymer additive types and contents may lead to diverse curve shapes/trends. Several previous studies (Bahia *et al.*, 2001; Kim, 2009; del Barco Carrion *et al.*, 2020; Wang *et al.*, 2020) found that δ and $|G^*|$ should vary from 0 to 90 degrees, and 0 to 1GPa, respectively. In this sense, the black diagram is a convenient representation: the x and y axes report δ and $|G^*|$, respectively. Therefore, this plot provides a simple instrument to visually discriminate the rheological behavior of bituminous materials. Fig. 5 presents an example of the black diagram incorporating the schematic of three major curve-trends commonly observed in literature 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 mixture and is exemplified by the "U-turn" shape of the curve (Brinson *et al.*, 1991; Kim, 2009; Yusoff *et al.*, 2013).

Asphalt binder is a temperature-susceptible material. It behaves like an elastic material at very low temperature, where the phase angle tends to 0, and the complex shear modulus reaches a glassy state (around 1 GPa). This behavior is valid for all asphalt binders; hence, in Fig. 5, the curve shapes of the three different materials are very similar in the left top corner. Neat binders without modification exhibit a smooth curve (yellow curve on the right) with the phase angle monotonically increasing toward 90 as the temperature increases. This phenomenon was reported in many studies (Di Benedetto *et al.*, 2004; Kim, 2009; del Barco Carrion *et al.*, 2020; Wang *et al.*, 2020). For the modified binder case (orange curve in the middle), an overall slightly lower phase angle can be observed compared to the neat binder, associated with a slight reduction in the phase angle at higher temperatures (low complex shear modulus) (Di Benedetto *et al.*, 2004; Yusoff *et al.*, 2013; del Barco Carrion *et al.*, 2020;). For the complex modified binder curve (grey line on the left) (Bahia *et al.*, 2001; Di Benedetto *et al.*, 2004; Kim, 2009), the behavior is closer to a solid viscoelastic material (asphalt mixture). The phase angle increases from 0 to a peak significantly lower than 90 degrees to bend back (U-turn) toward lower phase angle values commonly experienced at very high temperature and/or low frequency.

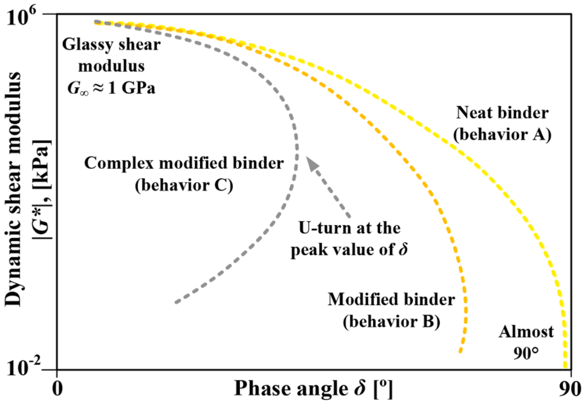

Fig. 5. Illustration of neat binder, modified binder, and complex modified binder's behaviors trend in black diagram.

Given the above considerations, the black diagram of three different binders, B, $B_{+pellets}$, and $B_{+shreds}$, is compared to evaluate the variability over a wide range of testing temperatures and frequencies after removing any outliers. Fig. 6 shows the black diagram of the three different binders in which the defined rheological behaviors are exhibited. In a previous study conducted during SHRP (Anderson *et al.*, 1994), to assure good performance, a minimum acceptable and meaningful complex shear modulus (1 kPa) for bituminous binders was highlighted with a horizontal line in the plot (Fig. 6). Nevertheless, the results lower than 1 kPa were still included in the present analysis. After an initial visual evaluation, some of the data was identified as outliers and therefore excluded for further analysis (marked in red dotted ellipses or rectangles in Fig. 6a, b, and c). The following criteria were used for these outliers: points have no physical meaning; outliers beyond the overall shape trend; and poorly matched curve measured by the PP08 and PP25 at the same temperatures. In Fig. 6a, binder B curves (behavior X) show very similar rheological behavior over the entire range of temperatures. However, in the upper left corner (high temperatures and low frequencies), laboratory 2 and 11′s phase angle results reach 90° indicating a purely Newtonian fluid behavior (Kim, 2009). Moreover, several small wings (data beyond the LVE range) were found at intermediate temperatures under high frequencies, reflecting a relatively poor time temperature superposition. These data were defined as outliers and removed. In Fig. 6b and c ($B_{+pellets}$ and $B_{+shreds}$, respectively), the curves present similar rheological behavior at low temperatures (until 28 °C) among the different laboratories, while diverging tendencies appeared at higher temperatures. Several outliers were found in laboratory 1′s results (marked in red dotted ellipses) and then removed. A closer observation could detect poor overlapping curves between PP08 and PP25 results for laboratory 7 (orange curve highlighted by the black ellipses) in both blended binders. Hence, laboratory 7′s results were also excluded from the analysis. In the case of laboratory 8, relatively poor overlapping curves were observed in $B_{+pellets}$ (green curve

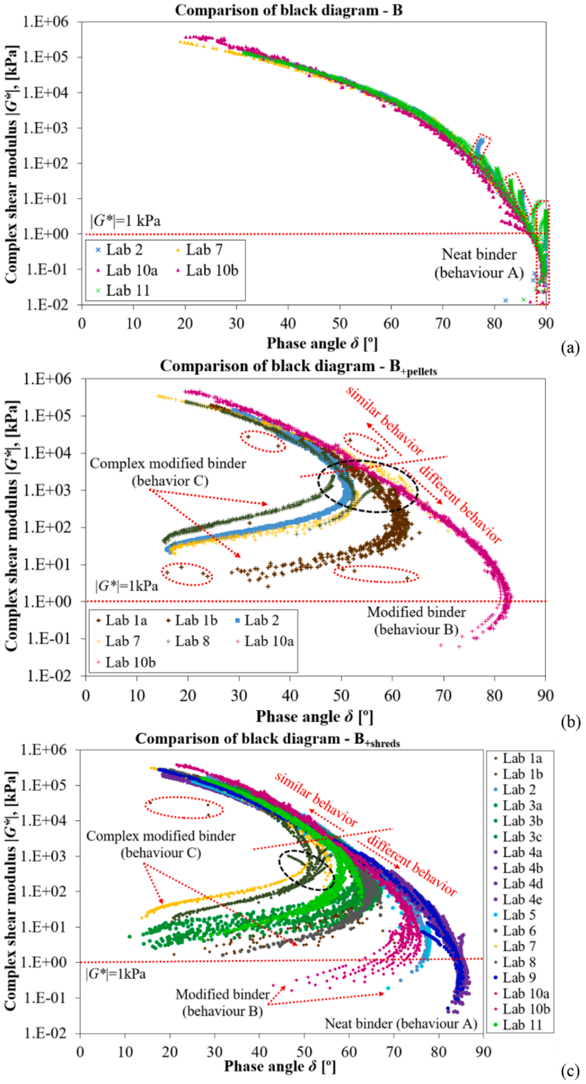

Fig. 6. Black diagram comparison: a) B; b) $B_{+pellets}$ and c) $B_{+shreds}$. vertical red line indicates minimum complex shear modulus of 1 kPa, outliers are marked with red circles.

highlighted by the black ellipses); however, in the overlapping temperatures (between PP08 and PP25 measurements), PP25′s results seemed to match the overall trend of the curve; hence, only the results of PP08 measured higher than 28 °C were discarded.

In Fig. 6b and c, laboratories 4 and 9 exhibited neat binder response (behavior X); laboratories 2, 5, and 10, evidenced a modified binder response (behavior Y); laboratories 1.1, 1.2, 3, 6, 7, 8, and 11, showed complex modified binder response (behavior Z). Such a classification group is further used to evaluate the reproducibility among laboratories in the next section. Due to the limited available results, only the modified binder behavior was observed for $B_{+pellets}$ (Fig. 6b), while all three behaviors were found for $B_{+shreds}$ (Fig. 6c). It is clear that the blends did not behave as expected for bituminous binders and displayed a wide heterogeneity. This deviation is primarily due to the heterogeneity of the blends and low viscosity from the neat binder at high temperatures. This divergence can be clearly seen in the phase angle results. At higher temperatures, the solid PE particles control the rheology behavior. These results corroborate others reported in the literature, for example, in the case of other materials such as crumb rubber (Loderer *et al.*, 2018).

4.3. Reproducibility analysis and effect of rheological behaviors

The evaluation of reproducibility among laboratories and the effect of rheological behaviors was carried out based on previous findings. First, the isochronal trend of dynamic shear moduli, $|G^*|$, and phase angles, δ, at 1.59 Hz was plotted as illustrated in Fig. 7. The low and high temperature-frequency sweep tests using PP08 and PP25 geometry were displayed for all the three binders studies. Next, the variation coefficients of all binders and those among different behavior groups (A, B, and C) were calculated and compared. Finally, the effect of two blend batches was also addressed by laboratory 1.

In Fig. 7a and b, it was evident that binder B exhibited the expected

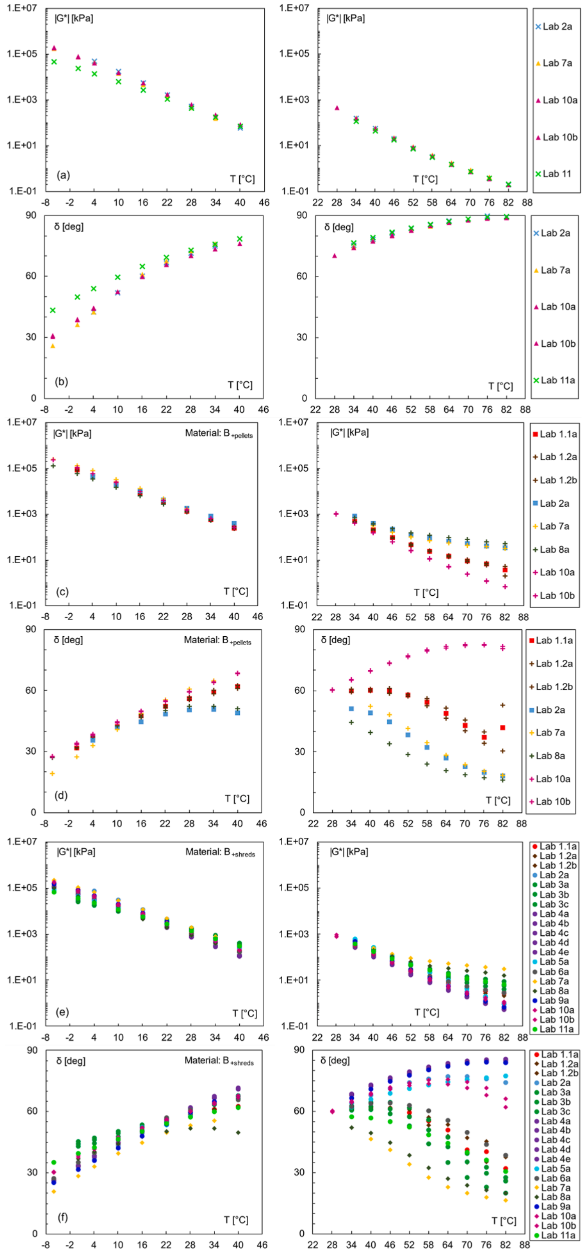

Fig. 7. Isochronal plots: (a) $|G^*|$ for binder B at low (left) and high (right) testing temperature; (b) δ for binder B at low (left) and high (right) testing temperature; (c) $|G^*|$ for blend $B_{+pellets}$ at low (left) and high (right) testing temperature; (d) δ for blend $B_{+pellets}$ at low (left) and high (right) testing temperature; (e) $|G^*|$ for blend $B_{+shreds}$ at low (left) and high (right) testing temperature; (f) δ for blend $B_{+shreds}$ at low (left) and high (right) testing temperature.

behavior of a neat binder. Except for laboratory 11, the interlaboratory tests showed that both $|G^*|$ and δ did not substantially differ, regardless of the test temperature. Hence, laboratory 11′s result was excluded in the following analysis.

For $B_{+pellets}$, $|G^*|$ at low temperatures (Fig. 7c) presented similarities to the neat binder B. On the contrary, phase angles tested with PP08 at low temperatures started to show, in some cases, diverging tendencies concerning B, that were expressed by δ inversions above 34 °C. Based on the presented isochronal plots shown in Fig. 7, a behavior transition located at intermediate temperatures (between 28 and 34 and 46 °C in both PP08 and PP25) could be identified. This phenomenon is also true for $B_{+shreds}$. This finding can be explained by considering the softening of PE blends occurring at higher temperatures (Porcello *et al.*, 2011). It is worth noticing that this is in the range of the softening point temperature of the neat binder, where it will become more liquid. Such an effect most likely resulted in less homogeneous samples causing floating of plastic particles inside the testing specimens and thus variable rheological responses. However, it is worth noting that such intermediate-temperature transition corresponded to the temperature range adopted for overlapping the measurements obtained from the test geometries (PP08 vs. PP25).

Differences between $B_{+pellets}$ and B were visible when tested under the PP25 configuration at higher temperatures (Fig. 7d). In general, a greater stiffness was detected for each $B_{+pellets}$ series, indicating the stiffening effect provided by the pellet's inclusion. Moreover, it can be observed that curves between different laboratories followed two distinct trends. On the one hand (laboratories 1.1, 1.2, 2, 7, 8), evident rheological changes seemed to occur towards a more elastic behavior. This trend can be seen as phase angles within Fig. 7d right were always lower with respect to B and assumed a decreasing trend. On the other hand, it was observed that at high temperatures (70–76 °C), the phase angle of laboratory 10 reached the peak value, and then a slight reduction occurred. The same tendency was more evident for $B_{+shreds}$ (Fig. 7e and f). At low testing temperatures (PP08), the effect of PE seemed to lead to simple stiffening in most cases, whereas at high temperatures (PP25), a spectrum of significantly different trends was observed.

Besides visual comparison, quantitative analysis was carried out for the data reproducibility among laboratories (Sybilski *et al.*, 2004; Poulikakos *et al.*, 2018). The multi-laboratory precision was evaluated according to AASHTO T315-20 standard (AASHTO T315, 2020) for unaged asphalt binders - acceptance limits for $|G^*|/sin\delta$ of 3.6% for *1s%*. Although these acceptance limits were designed for the unmodified neat binder, it is used in this case. Alternative approaches are also available in the literature (EN 14770, 2012). Precision results are presented in Table 5. Regarding binder B, reproducibility obtained from all available data (laboratories 2, 7, 10, 11) both at low- and high-temperature (series were purged of isolated outliers) did not meet the requirements. As anticipated, laboratory 11 exhibited misaligned trends of $|G^*|$ and δ with respect to other laboratories for binder B (see Fig. 5); however, even excluding laboratory 11 from the analysis, the reproducibility was improved at low temperatures, but not enough at the high ones (Table 5). This fact was entirely unexpected, especially taking into account that the original bitumen was obtained from the same single source: nonetheless, it should be noted that the remaining laboratories (2, 7, 10) executed the DSR tests with slight differences in terms of testing protocol (Table 3) (conditioning time, frequency ramps). For $B_{+pellets}$ and $B_{+shreds}$, reproducibility was assessed, including all available data coming from all laboratories (laboratories 1.1, 1.2, 2, 7, 8, 10 for $B_{+pellets}$, laboratories 1.1, 1.2, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11 for $B_{+shreds}$). Based on these results, it was again found that reproducibility did not meet the AASHTO T315-20 requirements, regardless of the testing geometry (PP08 or PP25).

Table 5
Reproducibility statistics of *1s%* for $|G^*|/\sin\delta$ precision [kPa] at the reference frequency f=1.59 Hz (10 rad/s): multi-laboratorty precision: B, $B_{+pellets}$, and $B_{+shreds}$ blends.

Material laboratories	B All available	All (11 excluded)	$B_{+pellets}$ All available	$B_{+shreds}$ All available
-6 (PP08)	45.1*	4.24*	32.0*	45.2*
0 (PP08)	40.7*	*2.52*	28.1*	45.8*
4 (PP08)	35.6*	9.42*	26.2*	44.7*
10 (PP08)	30.1*	6.99*	23.5*	36.8*
16 (PP08)	24.3*	6.44*	18.7*	29.7*
22 (PP08)	18.4*	7.23*	15.1*	26.7*
28 (PP08)	15.7*	12.33*	14.3*	30.7*
34 (PP08)	15.0*	15.59*	17.8*	36.6*
40 (PP08)	14.5*	16.28*	27.8*	41.7*
28 (PP25)	0.3	0.29	0.4	6.4*
34 (PP25)	11.2*	2.31	36.4*	28.2*
40 (PP25)	10.2*	3.53	48.0*	36.1*
46 (PP25)	9.3*	4.86*	68.0*	50.5*
52 (PP25)	7.7*	5.52*	89.9*	77.5*
58 (PP25)	7.2*	6.36*	109.2*	116.9*
64 (PP25)	6.4*	6.08*	119.7*	154.3*
70 (PP25)	6.6*	6.94*	124.2*	177.7*
76 (PP25)	6.7*	7.20*	123.9*	196.6*
82 (PP25)	2.4	0.81	127.8*	204.1*

- : non-available data; * : data not matching AASHTO T315-20 criteria – max(1s %): 3.6.

Results in Table 5 show that reproducibility was always poor regardless of the considered materials group. It is not surprising that the neat binder B (excluding laboratory 11) indicates the best reproducibility among the three materials. $B_{+shreds}$ materials have an overall worse *1s%* value compared to $B_{+pellets}$; this can be attributed to the PE shreds, which are the by-product of the production process of the pellets. Hence, a more diverse distribution of the PE shreds may occur than PE pellets. Indeed, it should be noticed that fewer laboratories tested B+pellets with respect to shreds ones. However, at the actual stage of the research, it could be concluded that the test reliability was not strictly related to the type and magnitude of rheological modification of the blends. As shown in Table 3, all the relevant experimental parameters were adopted by all laboratories. For both blends, it was impossible to establish a clear correlation between data trends and protocol details (e.g., temperature, frequency sweep order, or conditioning time).

Regarding $B_{+shreds}$, reproducibility precision was evaluated, also taking into account different groups of laboratories, given the other rheological behaviors identified previously (Table 6). Depending on the identified groups (X, Y, and Z), the behavior X group indicates a relatively narrow range of scattering in the reproducibility. In contrast, a large range of spreading in the reproducibility is experienced for behaviors Y and Z, particularly for the complex modified binder group. Based on the isochronal analysis, the PE modified binders did not meet the reproducibility requirements considering the standard for neat binders.

Given the data's initial analysis, the Black diagram's visualization, and the reproducibility values, an additional evaluation was conducted to study the effect of blending batches by using the results from the two batches performed by a single laboratory. Fig. 8 compares the results obtained by laboratory 1 (laboratory 1.1 vs. laboratory 1.2), which tested pellets and shreds blends from Batch 1 and Batch 2. Plots indicated very limited variation in the trends with respect to the source of the blend for the same shape of plastic, regardless of the testing geometry (variability was very low for $|G^*|$ and δ, for both pellets and shreds blends). Hence, only the results from batch 1 from laboratory 1 will be further analyzed in the following section.

4.4. *Master curves and rheological parameters*

In this section, a visual comparison of the fitted master curve of $|G^*|$ and δ are first conducted to understand the rheological response of the material better. The fitting parameter of Williams–Landel–Ferry (WLF)

Table 6
Reproducibility statistics of *1s%* for $|G^*|/\sin\delta$ precision [kPa] at the reference frequency f=1.59 Hz (10 rad/s) for $B_{+shreds}$ blend as a function of the rheological behavior.

Material Behavior laboratories	$B_{+shreds}$ X (neat binder) 4, 9	Y (modified binder) 2, 5, 10	Z (complex modified binder) 1, 3, 6, 7, 8, 11
-6 (PP08)	***15.59****	***48.59****	***61.00****
0 (PP08)	***16.47****	***36.01****	***58.51****
4 (PP08)	***18.21****	***46.37****	***51.23****
10 (PP08)	***20.16****	***35.12****	***39.87****
16 (PP08)	***22.01****	***26.18****	***29.89****
22 (PP08)	***24.40****	***16.15****	***25.32****
28 (PP08)	***25.40****	***11.75****	***26.69****
34 (PP08)	***26.44****	***10.90****	***28.85****
40 (PP08)	***5.28****	***8.81****	***30.27****
28 (PP25)	-	**6.43***	-
34 (PP25)	**21.85***	**25.44***	**27.04***
40 (PP25)	**20.05***	**29.92***	**32.50***
46 (PP25)	**16.94***	**31.03***	**44.50***
52 (PP25)	**17.21***	**32.23***	**64.80***
58 (PP25)	**14.92***	**32.86***	**90.28***
64 (PP25)	**13.69***	**28.49***	**110.41***
70 (PP25)	**11.34***	**20.66***	**119.86***
76 (PP25)	**9.18***	**10.52***	**129.60***
82 (PP25)	**8.65***	**11.76***	**132.37***

- : non-available data; * : data not matching AASHTO T315-20 criteria – max(1s %): 3.6.

(Williams, Landel and Ferry, 1955) model together with rheological parameters, including Glover-Rowe (*G-R*) parameter (Glover *et al.*, 2005; Anderson *et al.*, 2011; Rowe, 2014) and crossover temperature (Garcia Cucalon *et al.*, 2019; Wang *et al.*, 2019b), are calculated, compared and discussed. Finally, a statistical analysis was conducted on the *G-R* parameter to evaluate the difference in the three rheological behaviors. *G-R* thresholds for each rheological behavior are also proposed.

4.4.1. Master curve and rheological parameters

Master curves provide a convenient visualization of the rheological behavior of bituminous materials. Over the years, researchers have developed different master curve models to address the variety of asphalt materials responses, from thermo-rheologically simple binders to more complex asphalt mixture behaviors (Bahia *et al.*, 2001; Moon *et al.*, 2020; Cannone Falchetto *et al.*, 2021), commonly exhibiting a U-turn curve in the black diagram (Brinson *et al.*, 1991; Kim, 2009; Yusoff *et al.*, 2013). All these models rely on the shifting approach of the measurements obtained at different temperatures to generate a single data representation. The related fitting parameters have the potential to evaluate the mechanical and rheological properties over a wide range of temperatures. This study adopted the model proposed in NCHRP 459 Report (Bahia et al., 2001), suitable for both binder and mastic, to generate the master curves for all binders. The following formulation is used to fit the experimental data of $|G^*|$ and δ:

$$|G*(f,T)| = |G_e| + \frac{|G_\infty| - |G_e|}{\left[1 + (f_c/a_T f)^k\right]^{m_e/k}} \tag{1}$$

$$\delta = 90I - (90I - \delta_m)\left\{1 + \left[\frac{\log(f_d/a_T f)}{R_d}\right]^2\right\}^{-md/2} \tag{2}$$

$$I = \begin{cases} 1 & \text{for mixture and mastic} \\ \left\{\begin{matrix} 0 & \text{if } f' > f_d \\ 1 & \text{if } f' \le f_d \end{matrix}\right\} & \text{for binders} \end{cases} \tag{3}$$

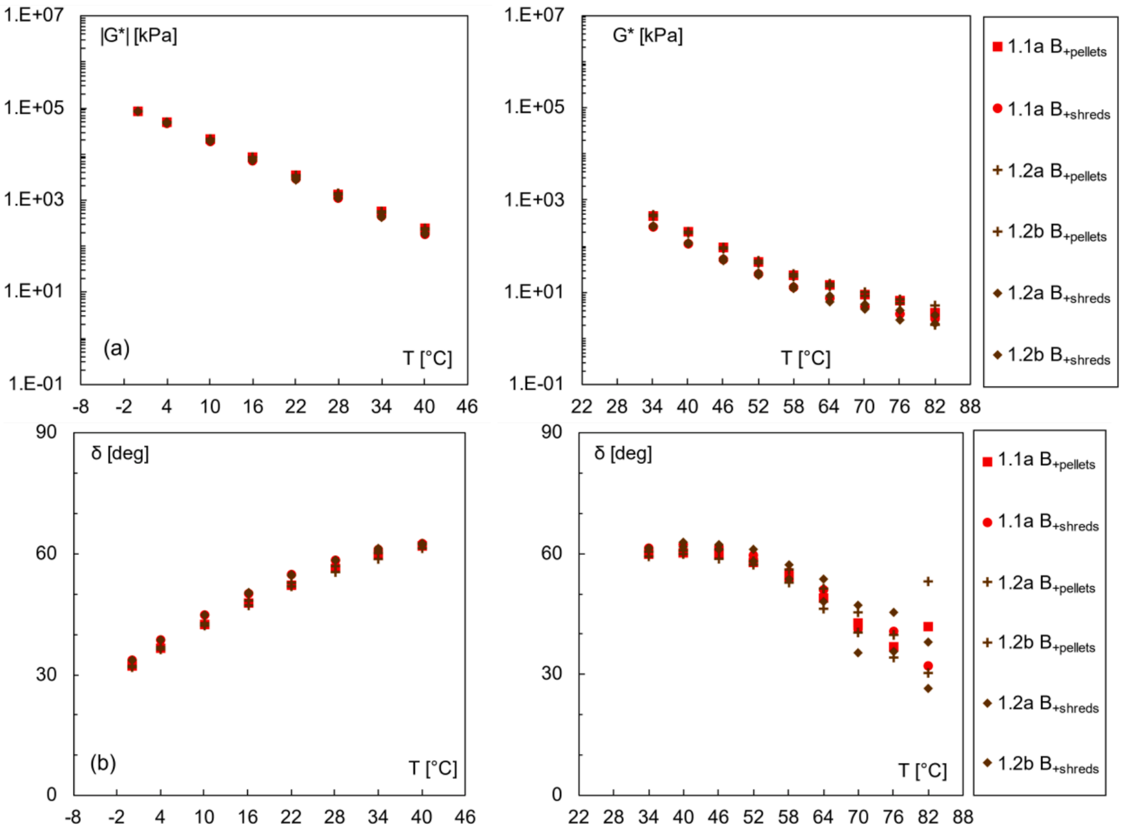

Fig. 8. Isochronal plots of blends $B_{+pellets}$ and $B_{+shreds}$ in terms of: a) $|G^*|$ at low (left) and high (right) testing temperature; b) δ at low (left) and high (right) testing temperature.

where, G_e is the equilibrium complex shear modulus; G_e equals to zero for neat binders; G_∞ is the glassy shear modulus when frequency tends to infinite; f is the frequency (Hz); f_c is the crossover frequency (where $G' = G''$); a_T is the horizontal shift factor at temperature T; δ_m is the phase angle constant; f' is reduced frequency; f_d is the location parameter with dimensions of frequency and k, m_e, R_d and m_d are dimensionless shape parameters. Since different rheological behaviors were observed in this study, different values of I were selected for the related binders.

To generate master curves of complex shear modulus and phase angle measurements are commonly shifted horizontally at a selected reference temperature T_0. For this purpose, the WLF formulation was adopted to determine the horizontal shift factor, a_T. The vertical shift factor is not needed due to the relatively high testing temperatures (Wang et al., 2020). a_T can be calculated by:

$$\log a_T(T) = -\frac{c_1(T - T_0)}{c_2 + (T - T_0)} \tag{4}$$

where, T_0 is the reference temperature, T_0=22°C in this study; c_1 and c_2 are the temperature constants. In Fig. 9, the master curves of $|G^*|$ and δ are illustrated for the entire set of three asphalt binders investigated, the fitted c_1 and c_2 are listed in Table 7. It should be noted that a horizontal line of 45° was marked in the master curves of δ to indicate the location of crossover temperature, $T_{\delta=45^\circ}$ (Garcia Cucalon et al., 2019; Wang et al., 2019b).

The crossover temperature is where loss and storage moduli are equal at a given frequency ($f = 1.59$ Hz in this paper); it can be used to evaluate the cracking and aging resistance properties at intermediate temperature (Porot et al., 2016, Garcia Cucalon et al., 2019). Hence, both $T_{\delta=45^\circ}$ and $|G^*|@T_{\delta=45^\circ}$ were calculated to estimate the effect of PE additives on the rheological properties of binders.

In Fig. 9a and b, overall similar trends were found in the neat binder's master curves (G^* and δ) among the three different laboratories. However, this was not the case in $B_{+\text{pellets}}$ (Fig. 9c and d) and $B_{+\text{shreds}}$ (Fig. 9e and f). Similar trends and curves were observed for both master curves at low temperatures and high frequencies, while significant differences are found at high temperatures and low frequencies. In particular, for the phase angle curves, very scattered distributions were found. Moreover, no clear correlations were observed between results and affecting factors, including plastic shapes, batches, and experimental conditions. From Fig. 9c and d, similar trends were noticed, namely two different rheological behaviors in both master curves.

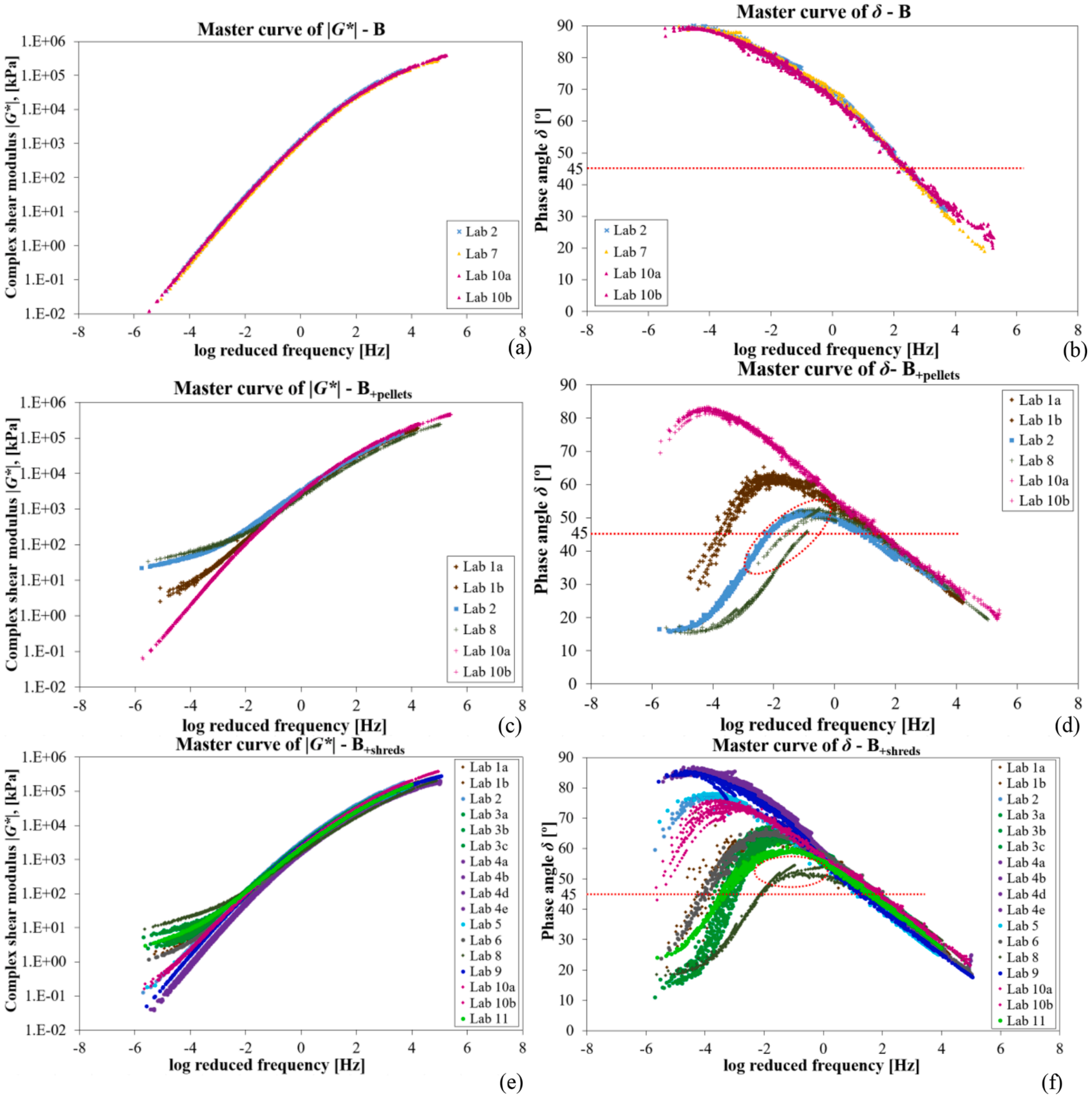

Fig. 9. Master curve comparison among laboratories at reference temperature 22 °C: (a) $|G^*|$ of binder B; (b) δ of binder B; (c) $|G^*|$ of blend $B_{+pellets}$; (d) δ of blend $B_{+pellets}$; (e) $|G^*|$ of blend $B_{+shreds}$; (f) δ of blend $B_{+shreds}$.

Table 7
Rheological parameters of all the materials.

Materials	Laboratories	c_1	c_2	Rheological behavior	$T_{\delta=45^\circ}$ (°C)	$\|G^*\|$@ $T_{\delta=45^\circ}$ (kPa)	*G-R* (kPa)
B	2	12.84	112.52	X*	5.9	37444.4	0.184
	7	13.08	119.87	X	4.6	35527.1	0.188
	10a	14.31	129.75	X	4.7	39847.2	0.301
	10b	14.66	133.15	X	4.6	39939.6	0.299
$B_{+pellet}$	1a	16.03	139.23	Z*	11.5 / 68.2	16666.5 / 9.0	26.833
	1b	16.39	143.05	Z	14.3 / 63.8	11110.2 / 15.3	33.033
	2	15.50	134.17	Z	14.8 / 46.6	12078.9 / 217.9	171.587
	8	14.42	134.77	Z	13.4 / 33.3	8709.5 / 738.9	213.691
	10a	15.88	137.16	Y*	12.56	18600.7	5.681
	10b	15.79	136.70	Y	11.44	21993.0	5.089
$B_{+shreds}$	1a	18.07	152.34	Z	14.3 / 60.8	9863.6 / 10.8	21.180
	1b	19.51	164.26	Z	10.2 / 71.5	19486.9 / 3.5	18.388
	2	14.88	125.70	Y	12.5	21361.9	8.755
	3a	18.34	168.73	Z	5.8 / 62.1	27661.6 / 20.8	17.931
	3b	19.08	171.76	Z	7.1 / 61.5	22076.7 / 26.4	31.241
	3c	17.49	153.95	Z	6.4 / 67.2	25475.5 / 10.3	13.521
	4a	14.58	135.86	X	10.1	13197.5	1.240
	4b	15.00	137.70	X	10.9	12424.5	1.578
	4d	13.77	129.14	X	10.6	11333.3	1.084
	4e	14.03	132.69	X	10.4	11297.1	0.886
	5	20.50	184.94	Y	10.3	12545.0	4.190
	6	16.05	147.14	Z	9.4 / 75.4	18472.0 / 3.5	13.586
	9	16.18	147.19	X	13.3	12478.6	3.093
	10a	17.75	165.13	Y	8.1	26596.9	6.014
	10b	17.20	155.58	Y	8.5	27127.8	5.679
	11	23.57	218.78	Z	8.9 / 65.1	14060.9 / 16.9	28.587

*X: neat binder; *Y: modified binders; Z: complex modified binder.
c_1 and c_2 are the fitted parameters of WLF model.

Smooth curves were obtained in most cases, while poor overlapping (red ellipses) was detected in the phase angle master curve measured by laboratory 8. In the case of $B_{+shreds}$, three different rheological behavior were observed and marked in Fig. 9e and f. The classification of rheological behaviors is presented in Table 7. In addition, two crossover temperatures were detected in several laboratories for both PE modified binders. Both $T_{\delta=45^\circ}$ and $|G^*|@T_{\delta=45^\circ}$ are calculated for all binders and listed in Table 7.

Glover-Rowe (*G-R*) parameter was also introduced to evaluate the rheological behavior of all the binders. *G-R* parameter was originally developed as an alternative to the failure tensile strain obtained from the ductility test (Anderson *et al.*, 2011; Rowe *et al.*, 2014). Its value relies on the $|G^*|$ and δ at 15°C and 0.005 rad/s, which can be derived from the fitted master curves. The equation can be expressed according to Eq. 5; the calculated results are shown in Table 7.

$$G - R = \frac{G^* \times (cos\delta)^2}{\sin\delta} \tag{5}$$

To gain further insight on the contribution of plastic additives and experimentation, a visual comparison of crossover temperatures and *G-R* parameters is proposed in Fig. 10. The relationship of T and $|G^*|$ at δ=45° is shown in Fig. 10a. Hence, only the lower $T_{\delta=45^\circ}$ and the corresponding $|G^*|$ are used in the analysis. In Fig. 10b, the *G-R* parameters were plotted, along with the two thresholds curves: the damage onset curve of 180 kPa and the failure curve at 600 kPa (Anderson *et al.*, 2011; Rowe *et al.*, 2014).

For neat binder B, it is not surprising that similar WLM coefficients (c_1 and c_2) and rheological parameters were observed among all laboratories, while slightly higher *G-R* parameters were exhibited for laboratory 10 than the others. It should be noted that even though the relative difference in the *G-R* parameter is more than 50%, the absolute difference is only around 0.1 kPa. Moreover, in both Fig. 10a and b, a small green box encloses the consistent results of neat binder B. Hence, the measured rheological behaviors in different laboratories only showed minimal differences. Concerning $B_{+pellet}$ and $B_{+shreds}$, Table 7 suggests that the WLF fitting coefficients and crossover temperature are within a narrow range. No clear correlations were observed between the values of the parameters and the rheological behaviors. Visually, both PE modified binders exhibited a higher scatter in the *G-R* parameter. The cyan and red boxes associated with $B_{+pellet}$ and $B_{+shreds}$ show a larger spread in both $T_{\delta=45^\circ}$ and *G-R* parameters. For $B_{+shreds}$, larger rectangles are obtained for both parameters. However, this does not indicate the plastic shreds may lead to more scattered results than the plastic pellets since more data points are used in $B_{+shreds}$. Further studies should be conducted to evaluate the effect of plastic types as PE may not be entirely compatible with binder, although in this case, due to the more

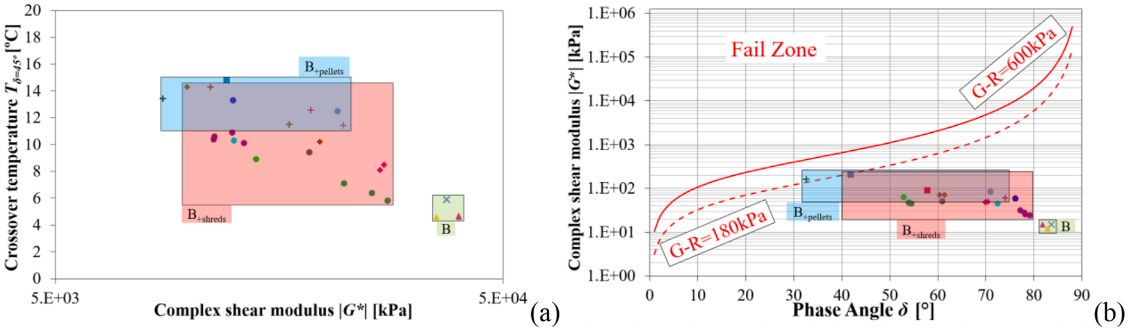

Fig. 10. Relationship of: (a) $T_{\delta=45^\circ}$ vs. $|G^*|$ in crossover temperature for all binders; (b) $|G^*|$ vs. δ in *G-R* parameter for all binders.

inhomogeneous plastic shreds characteristics, it should lead to more heterogeneous results for its modified binder. Overall, from the data presented in Table 7 it can be assumed that the addition of the two shapes of PE has: 1) increased the *G-R* parameter to higher values, similarly to softening point temperature or high temperature DSR parameters, 2) moved the cross over temperature to higher values implying that the material remains elastic at higher temperature and 3) reduced the crossover modulus to higher values indicating stiffer behavior.

In order to advance the analysis of the data presented in Table 7 and exploit the larger number of measurements on $B_{+shreds}$, the behavior groups (A-light yellow, B-light orange, and C-light grey) were introduced into the plots of $T_{\delta=45^\circ}$ and *G-R* parameter to determine any potential capability of the respective parameter in discriminating across material behavior. Fig. 11 visualizes the two cases.

In Fig. 11a, the three boxes representing the three materials' behavior (X, Y, and Z) are substantially overlapping. While districts trend in the material response could be seen from the overall shapes of the master curves and black diagram, the single crossover temperature value is apparently unable to distinguish the different rheological responses of the materials. On the other hand, Fig. 11b exhibits three distinct boxes corresponding to the investigated materials' previously identified behaviors: behavior X in light yellow, behavior Y in light orange, and behavior Z in light grey. Therefore, the *G-R* parameter appears to be more sensitive to detecting diverse rheological characteristics than the crossover temperature. In addition, it should be remarked that the *G-R* values are associated with lower temperatures than those where more considerable differences occur, as shown in the high temperature - low frequency domain both in the master curves and black diagram. This observed capability further supports the potentiality of this parameter in discriminating the rheological behaviors of PE modified binders. Hence, the next section presents a statistical analysis of the G-R parameter.

4.4.2. *Statistical analysis of G-R parameter*

The considerable number of results obtained by 11 participating laboratories provides a solid sample of data for performing the statistical analysis. Therefore, *G-R* parameter of $B_{+shreds}$ is investigated in this section. Based on the rheological behavior classification shown in Table 7, three groups of data were obtained. 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.

Statistical software SPSS (Hinton *et al.*, 2014) was used for the present analysis. First, a Shapiro-Wilk Test was used to validate the normal distribution within groups. All three groups passed the validation. Then, analysis of variance (ANOVA) with a significance level α=0.05 was performed to analyze the *G-R* parameters by setting the rheological behavior as a factor at three levels: X, Y, and Z. The statistical analysis outcome shows a *p*-value equal to $0.000013 < 0.05$, which means that the rheological behaviors are statistically significant. Next, a multiple comparison statistical test based on the Tukey's HSD (honestly significant difference) method was conducted to investigate the difference between each pair of rheological behaviors. The *p*-value of pairwise comparisons between each pair is X:Y (0.26276), X:Z (0.00002), and Y:Z (0.00039). Such results indicate that only behavior X vs. Z and behavior Y vs. Z are significantly different, while behaviors X vs. Y are not statistically different.

Based on the current results, the average value $\bar{x}$ can be easily calculated for each group. However, the mean value μ of the sample can be different; a 95% confidence interval of μ can be calculated as:

$$\mu = \bar{x} \pm 2 \times \sigma_n \tag{6}$$

where, σ is the standard deviation, $\sigma_n = \sigma / \sqrt{n}$, n is the number of samples. Based on Eq. 6, two μ values can be calculated, where μ_1 and μ_2 are the lower and upper bounds, respectively. With these two μ values, the boundary of each rheological behavior with 90% confidence interval can be calculated as: (μ_1-1$\times\sigma_n$, μ_2+1$\times\sigma_n$). The calculated results are shown in Table 8.

Based on the results listed in Table 8, the thresholds of these three different rheological behaviors can be defined. Considering the variety of results of the PE modified materials observed in previous studies (Glover *et al.*, 2005; Anderson *et al.*, 2011; Rowe, 2014; Wang *et al.*, 2019b), the lower boundary of neat binder (behavior X) can be located as above 0, while the upper threshold of complex modified binder (behavior Z) corresponds to the damage onset value 180 kPa of the *G-R* parameter. Hence, the following boundaries for the different behaviors were defined for the PE modified binders based on the results obtained in this study: behavior X: (0, 3.25]; behavior Y: [3.04, 9.28], and behavior Z: [8.45, 180), the unit being kPa.

5. Discussion

The recycling and reuse of waste plastics could lead to less consumption of nonrenewable fossil-based products and bring significant environmental benefits. Several scientists investigated the possibility of adding plastics to asphalt materials (Al-Hadidy *et al.*, 2009a, 2009b; Attaelmanan *et al.*, 2011; Gawande *et al.*, 2012; Kalantar *et al.*, 2012; Rokade, 2012; Gawande, 2013; Zhang and Hu, 2016; Nouali *et al.*, 2020). However, to the authors' knowledge, the present research is the first large interlaboratory study on the rheological properties of PE modified binders. The experimentation was based on a common and

Table 8
boundaries of three different rheological behaviors.

Behaviors	$\bar{x}$	σ	n	σ_n	μ_1	μ_2	μ_1-1$\times\sigma_n$	$\mu_2+1\times\sigma_n$
A	1.58	0.88	5	0.40	0.78	2.37	0.39	3.254
B	6.16	1.65	5	0.74	4.69	7.637	3.04	9.284
C	20.63	6.9	7	2.62	15.39	25.88	8.457	28.50

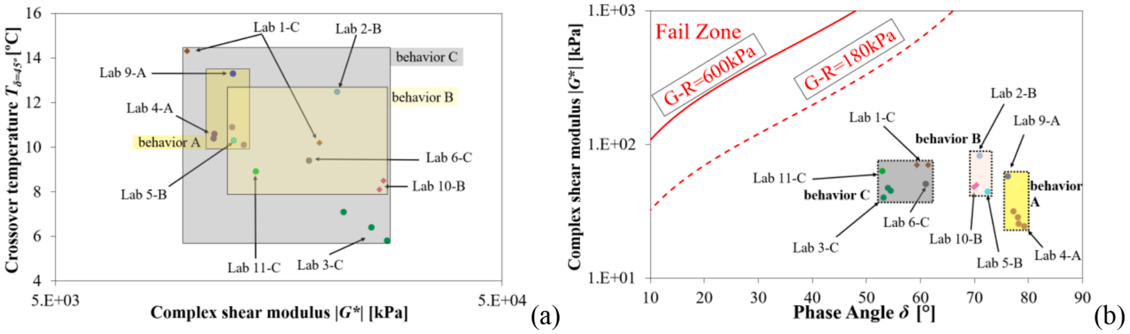

Fig. 11. Relationship of: (a)$T_{\delta=45^\circ}$ vs. $|G^*|$ in crossover temperature for $B_{+shreds}$ with different behaviors; (b) $|G^*|$ vs. δ in *G-R* parameter for $B_{+shreds}$ with different behaviors.

well-established testing protocol for DSR measurements developed by a different RILEM technical committee (Cannone Falchetto et al., 2022; Hugener et al., 2022). Various rheological parameters were carefully selected to obtain a broad spectrum of information on the materials' behavior. This approach resulted in a large amount of data not common in specific studies performed by a single research group, ultimately providing a solid set of measurements capable of representing the data population. Such a multi-laboratory contribution captured three different rheological responses (neat binder, modified binder, and complex modified binder) of the PE-modified binders. This diverse material behavior was not previously observed in single research efforts. These results prompted the need to identify sensitive tools that can discriminate across behaviors. Based on the literature, conventional and rheological parameters potentially able to determine essential changes in material response were screened (Al-Hadidy *et al.*, 2009a; Köfteci *et al.*, 2014; Garcia Cucalon *et al.*, 2019). The list includes softening point temperature, penetration value and index, ductility values, G^*, δ, $T_{\delta=45^\circ}$, $|G^*|@T_{\delta=45^\circ}$, and the *G-R* parameter. This study observed that *G-R* parameter (representing the plastic deformation ductility properties at 15°C) is particularly effective. Previous research found that the addition of polymer additive could significantly improve the plastic deformation resistance of bituminous materials (Moreno-Navarro *et al.*, 2014). Hence, the results of this paper seem to support further the idea that the *G-R* parameter function as a sensitive tool to detect such modifications. For crossover temperature (Garcia Cucalon *et al.*, 2019), it was observed that this parameter is useful for evaluating long-term durability and early rutting (pre-permanent deformation stage). Hence, such parameters may not be sensitive enough for plastic modified binders. The rheological behavior and identification of sensitive tools for discriminating plastic modified bituminous materials would need further investigation on a larger set of materials while extending the study to additional parameters beyond rheology to the mixture level. Potential limitations, including inhomogeneity of the plastic and the resulting blended binders, were discussed in detail in previous publications (Kakar *et al.*, 2021; Tušar *et al.*, 2021). In the authors' future work, the influence of available devices, including DSR calibration and instrument compliance correction, will be further considered, as they might partially influence the results in a large interlaboratory study. In addition, the repeatability within the same laboratory and the reproducibility among laboratories will be addressed more in-depth to overcome potential experimental limitations.

In this current study, an inter-laboratory study was performed instead of a conventional round-robin test. Each of the eleven laboratories followed its internal practice (see Table 3). Such an experimental plan was designed to better evaluate the effect of the experimental conditions, including conditioning time, testing duration, the order of temperature and frequency application, and replicates for blended binders. However, as explained in section 4.3, no clear trend was observed among different testing protocols. Hence, it is to say that these testing conditions only have limited influences on the rheological response of PE modified binders. More experimental conditions will be considered in future research.

6. Conclusions, recommendations, and outlook

As part of the RILEM technical committee TC-279 WMR Task Group (TG 1), Round Robin experiments were performed using conventional tests and the Dynamic Shear Rheometer (DSR) for characterizing asphalt binders modified with polyethylene (PE). The tests were conducted on a neat binder and two blended binders consisting of the neat binder blended with two types (pellets and shreds) of waste PE. Two batches of binder blends were prepared, and eleven laboratories participated in the experiments following a common protocol developed at the Task Group level. The following conclusions can be drawn from the experimental results.

- Conventional tests indicated a decrease in penetration and an increase in softening point and Fraass breaking point temperatures with the addition of PE, leading overall to a stiffening effect.
- The two material batches had limited influence on the rheological result. Repeatability within laboratory did not seem to be influenced by the plastic type (B+pellets vs. B+shreds).
- The batches modified with plastics experienced higher heterogeneity at higher testing temperatures, i.e., the effect of inhomogeneity becomes more pronounced as the binder becomes less viscous. The inhomogeneous distribution of PE plastic particles hypothesized to occur in the samples at higher temperatures seems to affect the measurements considerably.
- In the cold temperature regime, the PE modified binders showed a similar complex shear modulus to the neat binder. All laboratories could observe an increase in the complex shear modulus compared to the neat binder in the high temperature regime.
- The phase angle of the PE modified binder in the high temperature domain showed a range of trends with a tendency to reduce with respect to the neat bitumen, suggesting a complex behavior. In this temperature range, considerable differences were observed across laboratories results.
- Based on the presented isochronal plots, a transition in behavior located close to the softening point temperature of the neat binder (between 28–34 and 46 °C in both PP08 and PP25) could be identified. This finding can be explained by considering the change of consistency of the neat binder towards the liquid state occurring as the temperature increases. This phenomenon led to less homogeneous samples causing uneven distribution of plastic particles inside the binder matrix and thus variable rheological responses.
- The curves of PE modified binders among different laboratories followed three distinct trends: neat binder (behavior X), modified binder (behavior Y), and complex modified binder (behavior Z). Such trends are visible in the black diagram.
- The addition of the two types of waste PE moved the crossover temperature to higher values and increased the G-R parameter. However, the latter remains below the threshold of the damage onset curve.
- The crossover temperature appears to be unable to discriminate across binder behaviors X, Y, and Z. On the other hand, the G-R parameter proved to be a more sensitive parameter that can be used to identify the rheological behaviors of plastic modified binders.

In view of the above conclusions, the following recommendations are proposed for future research:

- The testing protocol used may need further optimization for these plastic-modified binders in the Dynamic Shear Rheometer experiments.
- The present study has experimentally shown the potential for enhancing the asphalt binder performance by incorporating PE plastic. The impact on mixture performance should be evaluated to upscale the benefits of the PE material. An ongoing project on the effect of various percentages of PE additive on mixtures' performance properties is being conducted under the framework of RILEM WMR TG 1.
- This study has shown potential for improving the mechanical performance of binders when using waste PE. Complete environmental assessment, including life cycle assessment (LCA) and life cycle cost assessment (LCCA), is necessary for a holistic evaluation of this material.

CRediT author statement

The authors confirm contributions to the paper as follows: LP, MT, MRK, LPo, ACF, and DW conceived and designed the experiments; LP, MT, and CR wrote the original introduction and background; DW and LP wrote the original objective and research approach; MRK, LPo, DW, and CR wrote the original materials and testing; AB, EP, MP, DW, ACF, LPo, GG, and DLP wrote the results and analysis; LP, KV, and MT wrote the conclusion and recommendations. All authors reviewed the results and approved the final version of the original manuscript. All authors contributed to the review editing and response to reviewers' comments.

Declaration of Competing Interest

We wish to confirm that there are no known conflicts of interest associated with this publication and there has been no significant financial support for this work that could have influenced its outcome.

Data Availability

No data was used for the research described in the article.

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