# RILEM TC 279 WMR round robin study on waste polyethylene modified bituminous binders: advantages and challenges

*Original*
RILEM TC 279 WMR round robin study on waste polyethylene modified bituminous binders: advantages and challenges / Tusar, M., Kakar, M.R., Poulikakos, L.D., Pasquini, E., Baliello, A., Pasetto, M., Porot, L., Wang, D., Cannone Falchetto, A., Dalmazzo, D., Lo Presti, D., Giancontieri, G., Varveri, A., Veropalumbo, R., Viscione, N., Vasconcelos, K., Carter, A.. - In: ROAD MATERIALS AND PAVEMENT DESIGN. - ISSN 1468-0629. - ELETTRONICO. - 24:2(2023), pp. 311-339. [10.1080/14680629.2021.2017330]

*Publisher:*
Taylor and Francis Ltd.

*Published*
DOI:10.1080/14680629.2021.2017330

This document is the accepted manuscript version of the following article:
Tušar, M., Kakar, M. R., Poulikakos, L. D., Pasquini, E., Baliello, A., Pasetto, M., … Carter, A. (2022). RILEM TC 279 WMR round robin study on waste polyethylene modified bituminous binders: advantages and challenges. Road Materials and Pavement Design, 1-29. https://doi.org/10.1080/14680629.2021.2017330

# RILEM TC 279 WMR Round Robin study on waste polyethylene modified bituminous binders: advantages and challenges

**Marjan Tušar * [1,2], Muhammad Rafiq Kakar [3,4], Lily D. Poulikakos [3], Emiliano Pasquini [5], Andrea Baliello [5], Marco Pasetto [5], Laurent Porot [6], Di Wang [7], Augusto Cannone Falchetto [8], Davide Dalmazzo [9], Davide Lo Presti [10], Gaspare Giancontieri [10], Aikaterini Varveri [11], Rosa Veropalumbo [12], Nunzio Viscione [12], Kamilla Vasconcelos [13], Alan Carter [14]**

*** Corresponding author**

**[1] Slovenian national building and civil engineering institute, Dimičeva ulica 12, SI- 1000 Ljubljana, Slovenia**

**[2] National Institute of Chemistry, Hajdrihova 19, SI- 1001 Ljubljana, Slovenia**

**[3] Empa, Swiss Federal Laboratories for Materials Science and Technology, Überlandstrasse 129, 8600 Dübendorf, Switzerland**

**[4] Department of Architecture, Wood and Civil Engineering, Bern University of Applied Sciences (BFH), Switzerland**

**[5] Department of CIvil, Environmental and Architectural Engineering (ICEA), University of Padova, –Via Marzolo, 9 - 35131 Padova, Italy**

**[6] Kraton Chemical B.V., Transistorstraat 16, 1322 CE, Almere, the Netherlands**

**[7] Technical University of Braunschweig, Braunschweig Pavement Engineering Centre(ISBS), Beethovenstraße 51b, 38106 Braunschweig, Germany**

**[8] University of Alaska Fairbanks, 1760 Tanana Loop, Fairbanks, AK 99775, US**

**[9] Politecnico di Torino, Corso Duca degli Abruzzi 24, 10129 Torino, Italy**

**[10] Università degli Studi di Palermo, Piazza Marina, 61 90133 – Palermo, Italy**

**[11] Delft University of Technology, Postbus 5, 2600 AA Delft, the Netherlands**

**[12] Department of Civil, Construction and Environmental Engineering University of Naples Federico II, Via Claudio,21, 80125 Napoli NA, Italy**

**[13] Escola Politécnica da Universidade de São Paulo, Av. Prof. Luciano Gualberto, 380 - Butantã, São Paulo - SP, 05508-010, Brazil**

**[14] École de technologie supérieure, ÉTS, 1100 Notre-Dame St W, Montreal, Quebec H3C 1K3, Canada**

## Abbreviations

| | |
|---|---|
| B | Bitumen B 70/100 |
| $B_{+pellets}$ | Bitumen B 70/100 modified with PE pellets |
| $B_{+shreds}$ | Bitumen B 70/100 modified with PE shreds |
| BTSV | Binder fast characterisation test |
| DHR | Dual helical ribbon |

| | |
|---|---|
| DSR | Dynamic shear rheometer |
| EVA | Ethylene vinyl acetate |
| $J_{nr}$ | Non-recoverable creep compliance for MSCR test |
| LAS | Linear amplitude sweep |
| LVE | Linear viscoelastic |
| MSCR | Multiple stress creep-recovery |
| PE | Polyethylene |
| PET | Polyethylene terephthalate |
| %R | Average percent recovery for MSCR test |
| PP | Polypropylene |
| PU | Polyurethane |
| PVC | Polyvinyl chloride |
| TC | Technical committees |
| TG | Task group |
| TRL | Technology readiness level |
| VECD | Viscoelastic continuum damage |

## Abstract:

Inter-laboratory experiments were designed to evaluate the impact of plastic waste blended directly in bitumen and to assess the properties, using conventional and advanced bituminous binder testing. The blends targeted 5 % of plastic waste in 95 % bitumen, using two types of polyethylene (PE) primary (pellets) and secondary (shreds) waste. In order to facilitate the blending with bitumen, a first processing was made to reduce the size of the plastics by shredding and sieving to 1 mm before blending it with bitumen. The experiments showed that the addition of PE waste to bitumen does not alter the chemistry of the bitumen, the blending is physical. The results indicate a strong dependency on the testing temperature as at low temperatures the composite material bitumen and PE behave both elastically whereas, at higher temperatures, the bitumen becomes viscoelastic. The round-robin results show that the results from the labs diverge in the high temperature regime where the bitumen becomes less viscous and the effect of plastic additives is more prominent. Also, the effect of inhomogeneity is more apparent at high temperature, and the PE blends were not stable. Using the dynamic shear rheometer, for both rheological modifications vs. filler-like behaviour begun to be more evident after a threshold temperature region between 34°C and 40 °C. A helical spindle was used and it provides a more stable trend for viscosity measurements due to improved mixing of the specimens. The MSCR tests indicated that the neat binder is more sensitive to permanent deformation compared to the blends with PE. The fatigue performance using the Linear Amplitude Sweep (LAS) test showed a better performance in terms of stress and fatigue life for the PE blends. In general, considering all the experimental results, a significant difference between the blends with shreds and pellets was not seen implying that the secondary waste (shreds) can be used to achieve the same performance as the primary waste (Pellets).

**Keywords: Asphalt; Waste; Polyethylene; Plastics; Performance**

## 1. Introduction

It is well established that a considerable amount of waste is produced currently and it is a significant societal challenge to recycle these materials avoiding disposal in landfills and incinerators. This challenge can be met through the concept of “circular economy” where goods at the end of their service life are used or recycled as resources for the same or other industries with clear implications in terms of resource efficiency and reduction in climate and environmental impacts. This is part of the waste management hierarchy through prevention by reducing waste, preparing for re-use, recycling, recovery including energy recovery and disposal; the earlier the stage on the hierarchy, the higher the value [EU Waste]. Various types of waste materials such as plastics, crumb rubber, glass and construction & demolition waste have been successfully recycled in road pavements [Lo Presti 2013; Poulikakos et al. 2017; Piao et al. 2021]. However, as shown by Piao et al. (2021), the technology readiness level (TRL) for these products has remained, for the most part, at the research level or limited to some countries and therefore, there is an urgent need to develop and broadly demonstrate such solutions in order to open widespread market uptake. RILEM technical committees (TC) present an optimal international cooperative opportunity to address all aspects pertaining to the recycling of waste and marginal materials in roads. This can span from the materials choice to laboratory performance up to in situ performance of selected materials. The goal of the technical committee TC-279 WMR (Valorisation of Waste and Secondary Materials for Roads) is to develop a robust and fundamental understanding for evaluation of the use of such materials as valuable alternatives for the traditional road materials, aiming to deliver at least similar performances to conventional materials. The use of waste, marginal and secondary materials is sometimes limited based on the perception that there could be lower performances associated with their use for road construction. Using inter-laboratory and interdisciplinary research beyond the traditional civil engineering and materials science fields, the above-mentioned TC gives an overview of methods used to characterize asphalt binders and mixtures modified with waste and marginal materials. To this end, the TC aims to evaluate the use of waste, marginal and secondary materials for roads by investigating the performance of road materials containing waste through round robin tests and through the development of standard procedures for their selection, preparation and use. These objectives will be accomplished through the coordinate efforts of five task groups (TG) addressing different aspects of TC main topic (www.rlem.org).

### TG 1 – Asphalt Binder Additives

Given this background, TG1 is investigating the feasibility of using waste plastics as a binder additive. It is known, from the literature, that more waste plastic materials can be used as additives in asphalt materials than currently used [Austroads 2019; Kalantar et al. 2003]. It has long been known that the most promising bituminous additive is chemically processed PET (polyethylene terephthalate) [Ledesma et al. 1991], which is traditionally used as an antistripping agent [Padhan et al. 2003], and it is also known that PET can be fully recycled [Awaja & Pavel 2005]. Therefore, the prices of waste PET are relatively high. Studies on various waste polymers such as polyethylene (PE), polypropylene (PP), polyurethane (PU), ethylene vinyl acetate (EVA) and various fibers are also available. As for any waste or secondary materials, health and safety have to be considered to qualify the validity of their recycling without known harmful effects. If plastics are to be used in asphalt mixtures, they must be safe to use, recyclable, cost effective, and provide equivalent or improved service life [Willis et al. 2020]. In most articles, the research seems to focus on modification with PE [Ho et al. 2006] and EVA [Garcia-Morales et al. 2004].

Due to availability and promising results in the literature, waste PE was selected as the first additive for investigation under TG 1. In particular, TG1 studies the use of waste PE as a binder additive and its effects on the physical and chemical properties of the modified binder.

In the first part of the characterization of PE modified binder, the following main activities were performed:

- Study the use of PE in bituminous binders with several laboratories round robin tests;
- Determine repeatability and reproducibility using the round robin test results;
- Recommend appropriate blending methods and test methods for characterising the properties of PE-blend bituminous binder.

This paper intends to provide a general overview of the experimental activities carried out by all the laboratories involved in the round robin test campaign. Specific attention is dedicated to sample preparation and materials characterization. Then, the main chemical and mechanical observed properties are illustrated and analysed highlighting benefits and challenges related to waste PE bitumen modification.

## 2. Materials and Specimen Preparation

### 2.1. Materials

Waste materials from a Swiss plastic recycling centre were used for this study. The company Innorecycling (https://innorecycling.ch/en/) collects waste packaging made primarily of polyethylene (PE) and produces pellets (primary waste), of approximately 5 mm, that are further sold and used for other purposes such as the production of pipes. This production process also produces waste (secondary waste) in the form of shreds that is burned as fuel in cement plants. Both types of materials have been investigated here designated as PE-pellets and PE-shreds. A single neat bitumen, 70/100 penetration grade as per EN 12591, was supplied by an European bitumen supplier as a straight run bitumen and used in this research program.

The inter-laboratory experiments were designed to evaluate the impact of plastic waste blended directly in bitumen and to assess the properties, using conventional and advanced bituminous binder testing. The blend targeted 5 % (by weight) of plastic waste and 95 % bitumen. Figure 1 shows pictures of both PE-shreds and PE-pellets. The PE-shreds were plastic having been cleaned and shredded. Different types of plastics (PE) could be identified by colour and degree of hardness.

The materials used in this work have the following designations:

- B: bitumen B 70/100
- $B_{+pellets}$: bitumen B 70/100 blended with PE-pellets
- $B_{+shreds}$: bitumen B 70/100 blended with PE-shreds

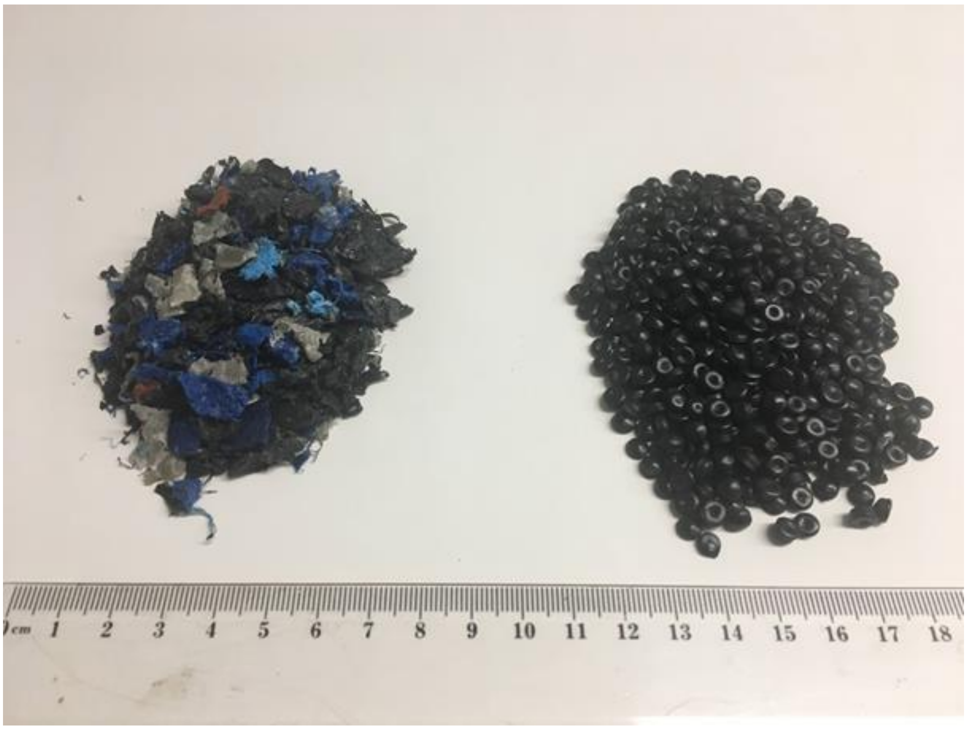


Figure 1. Photographs of the two types of plastic waste. PE-shreds left and PE-pellets right as obtained from the waste recycling plant Innorecycling

## 2.2. Grinding

In order to facilitate the blending with bitumen, a first processing was made to reduce the size of the plastics (shreds and pellets) by shredding and sieving to circa 1 mm before it was blended with bitumen. The following protocol was applied:

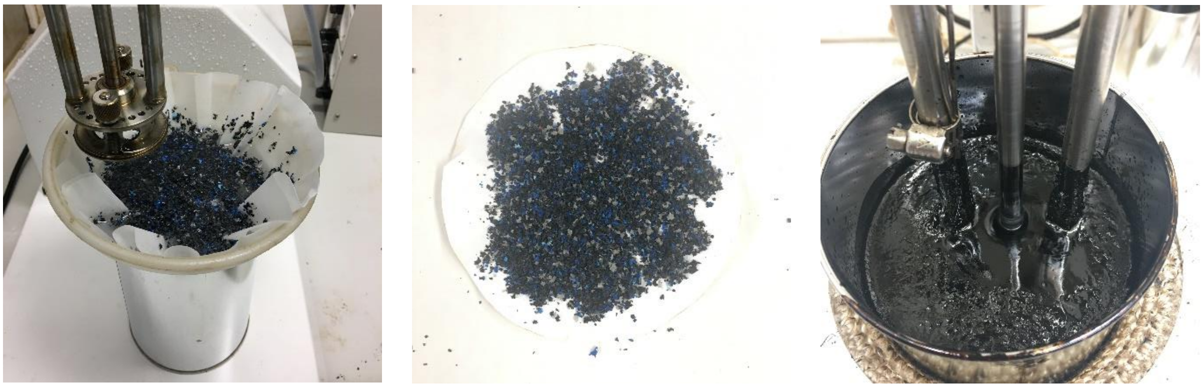
- Use shredding head for high-speed mixer;
- Measure 5 g plastic;
- Fill the recipient with cold water up to ¾ depth;
- Start stirring, add plastics, mix for 5 min at 5000 rpm;
- Pour water with shredded plastics on the 1 mm sieve;
- Dry in oven 110 °C for 1h.

## 2.3. Blending

The waste plastics, PE-shreds and PE-pellets, were blended, in the lab with neat bitumen 70/100. In order to avoid potential deviation between the labs while making the blends, the blends were initially made by a single lab, Lab1 (Blend1). As the number of participants increased, a second lab, Lab7, made another batch of blends to share with three other labs (Blend2). Both Lab1 and Lab7 used the same protocol to produce the blends as listed below:

- Use mixing head of a high shear mixer;
- Heat bitumen at 170 °C for 1 h;
- Add plastic shreds to bitumen;
- Mix at 3500 rpm for 1 h.

For the blends prepared at Lab1, the blends were visually homogeneous and no special observation was reported. For Blend2, a homogeneity problem was observed during the blending process. The plastic shreds were first chopped up using the mixing head and left to dry. Afterwards, the plastic was added to the hot bitumen, but it did not dissolve in the bitumen. When the mixing was paused, particles could be seen floating on top of the blend. It was then decided to raise the temperature to 180°C, but this showed no improvement. Figure 2 shows the steps from the shredding and blending process.

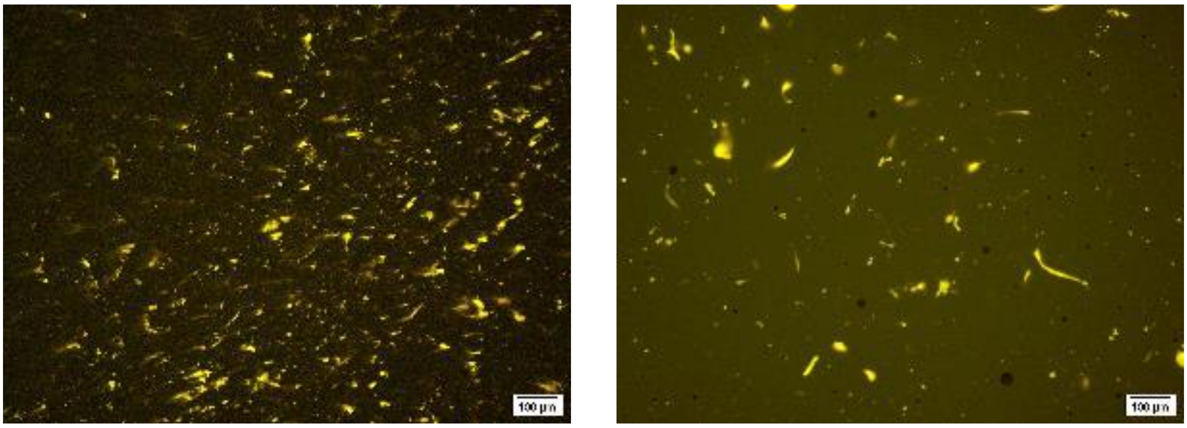


Shredding the plastic Drying and sieving Blending in bitumen

Figure 2. Sample preparation for plastic shreds in bitumen for Blend 2

Each blend was transferred to a can. Once cooled, the materials did not look visually homogeneous with particles still floating on top. Furthermore, the cleaning process for the high shear heads was rather difficult with some lumps of sticky plastic still present.

At the end of the blending process, samples were collected for fluorescence microscopy and assessment of the morphology of the blends. Figure 3 shows the images for each PE-shreds and PE-Pellets produced by the second batch of Blend2. For both materials, plastic shreds or pellets, the plastic is well dispersed within the bitumen, and a coarse morphology with no clear homogeneous phase or lumps of plastics is observed.



a) Blend with shreds b) Blend with pellets

Figure 3. Fluorescence microscopy of Blend 2 with shreds (a) and pellets (b)

The batches were split in equal quantities to be further dispatched to the 14 participating labs. Table 1 shows the type of blends and the type of plastics that each lab received. Lab1 was able to have both blends for comparison. Due to the inhomogeneity of the blends, instructions were given to each participating lab to heat the received blends to 180°C then stir them manually with a rod before sample preparation.

Table 1. Dispatch of blends between labs

| Lab# | | 1 | 2 | 3 | 4 | 5 | 6 | 7 | 8 | 9 | 10 | 11 |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Blend1 (2018) | Shreds | X | X | X | X | X | X | | | X | | X |
| | Pellets | X | X | X | X | X | X | | | | | |
| Blend2 (2019) | Shreds | X | | | | | | X | X | | X | |
| | Pellets | X | | | | | | X | X | | X | |

## 3. Measurements

### 3.1. Basic properties

A wide range of tests were performed by the different laboratories including measurement of physical properties and analysis of the chemical structure. The activities started in 2017 and were completed in 2021.

The standard properties of the blends were measured with penetration value at 25 °C, according to EN 1426, and softening point temperature, according to EN 1427, by two laboratories (7 and 10). *Table 2* shows the basic properties reported for the two labs. Both labs evaluated the second batch of blends (Blend2).

Table 2. Penetration and softening Point properties for the binder blends using Blend2

| | Bitumen 70/100 | | Blend2 PE-shreds | | Blend2 - PE-pellets | |
|---|---|---|---|---|---|---|
| | Pen value, 0.1 mm | Soft Point, °C | Pen value, 0.1 mm | Soft Point, °C | Pen value, 0.1 mm | Soft Point, °C |
| Lab7 | 88 | 45.0 | 40 | 109.0 | 36 | 63.4 |
| Lab10 | 81 | 45.8 | 39 | 79.0 | 38 | 60.8 |

The penetration values for the three binders were consistent between the two labs. The addition of 5% of plastic waste decreased the penetration by about 50 %, from 70/100 to 35/50 range.

The softening point temperature increased with greater effect for the PE-shreds but with higher scatter. As shown in Figure 4, other laboratories than 7 and 10 did measure the softening points. In figure 4, the filled marks denote Blend1 and unfilled marks Blend2. For the neat bitumen, the results were consistent with a mean value of 45.6 °C, varying between 44.6 °C and 46.6 °C, and within the limits of standard repeatability of +/- 2 °C (EN 1427). With the blends either with PE-shreds or with PE-pellets, a wide scatter of results was observed with a mean value of 78.6 °C, varying between 44.6 °C and 108.8 °C, for the PE-shred blend and a mean value of 66.8 °C, varying between 60.8 °C and 76.4 °C, for the PE-pellet blend. This may come from the difficulty to pour a small specimen in the ring, as the materials were not homogeneous. During softening point experiments, it was observed that the ball did not drop straight through the ring, but sometimes descending at an angle. Again attesting to the inhomogeneity of the samples putting to question the suitability of this test for such blends. In addition, the large variability of the softening point results for the PE-shreds is also attributed to the inhomogeneity in the shreds as discussed in the next sections.

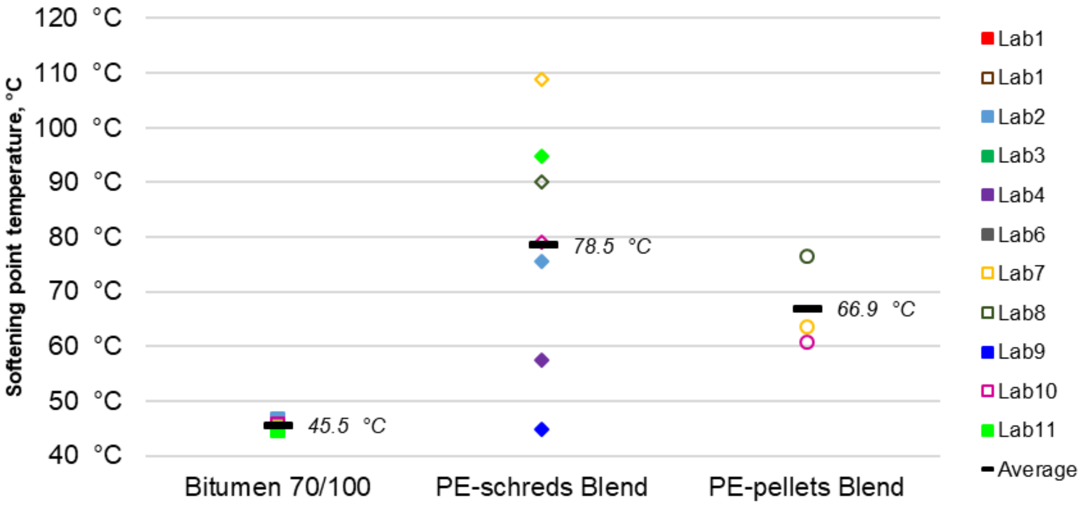


Figure 4. Softening point temperatures of binders between labs. Filled marks denote Blend1 and unfilled marks Blend2

In addition to these basic properties, one lab determined the Fraass breaking point temperature according to EN 12593 on the original binders from blend2 as reported in *Table 3*. While the measurement came from one single lab, it provides an indication of the effect of plastic waste on low temperature properties. The Fraass breaking temperature increased by at least 5 °C, showing a significant effect on the low temperature properties.

Table 3. Fraass breaking point temperature for the three binder blends from blend2

| | Bitumen 70/100 | Blend shred | Blend - pellet |
|---|---|---|---|
| Lab7 | -17 °C | -12 °C | -10 °C |

Storage stability was also evaluated by Lab7 according to EN 13399 at 180 °C for three days. However, the test failed since there was a clear separation in the tube with the entire plastic particle agglomerating on top. Only bottom samples were measured with softening point temperature of 54 °C for both blends, far below the measured softening point temperature of the blends and much closer to the reference bitumen. The blends were not stable at all, with visible segregation of the plastic.

This test was also performed by Lab1 with similar results using a different 70/100 binder mixed with the recycled PE. The results showed a clear storage stability problem with the viscosity of the sample from the top of the tube being much greater than the one measured from the bottom of the tube [Kakar et al. 2021].

### 3.2. DSR (low and high temperatures)

The principal viscoelastic parameters of the neat bitumen and the corresponding blends containing plastic shreds or pellets were determined by dynamic mechanical analysis using oscillatory shear mode tests. Temperature and frequency sweep tests were made by several laboratories to measure the norm (|G*|) of the complex shear modulus and the phase angle (δ). The tests were run in frequency sweep from 0.1 Hz to 20 Hz, with 10 logarithmic increments, at different temperatures between -6 °C and 82 °C, with 6 °C increments. To cover the temperature interval, different testing geometries were used by the laboratories depending upon their availability and the equipment characteristics, while following these general instructions: 25-mm parallel plates (PP25) with 1-mm gap between 34 °C and 82 °C, 8–mm parallel plates (PP08) with 2-mm gap between -6 °C and 40 °C. All dynamic measurements were made within the linear viscoelastic domain by applying a shear strain amplitude of 0.1% and 0.05% for the PP25 and PP08 respectively.

### 3.2.1 *Isochronal shear complex modulus and phase angle at 1.59 Hz (10 rad/s)*

To allow a simple representation and comparison of the results obtained by the different laboratories, it has been chosen to present the temperature and frequency sweep tests in the form of isochronal plots. Figure 5 to Figure 10 show the isochronal plots, in which |*G**| and *δ* raw data recorded at 1.59 Hz are plotted against the test temperature, for all three materials (B, $B_{+pellets}$ and $B_{+shreds}$). The results for neat bitumen (Figure 5 and 6), where four labs participated, show the rheological behaviour that is typical for non-polymer modified bitumen. Except for Lab 11, comparable results between Labs were observed. Looking at the tests repeatability, a numerical comparison found that, when testing different replicates in the same Lab, the differences of |*G**| are less than 2%, and those for *δ* are less than 0.25°. For simplicity, Figures 5 and 6 report the average data for each Lab. This value, 2%, is much lower than the reported variability (10%) for neat bitumen. This very good consistency was expected considering that neat bitumen came from the same batch.

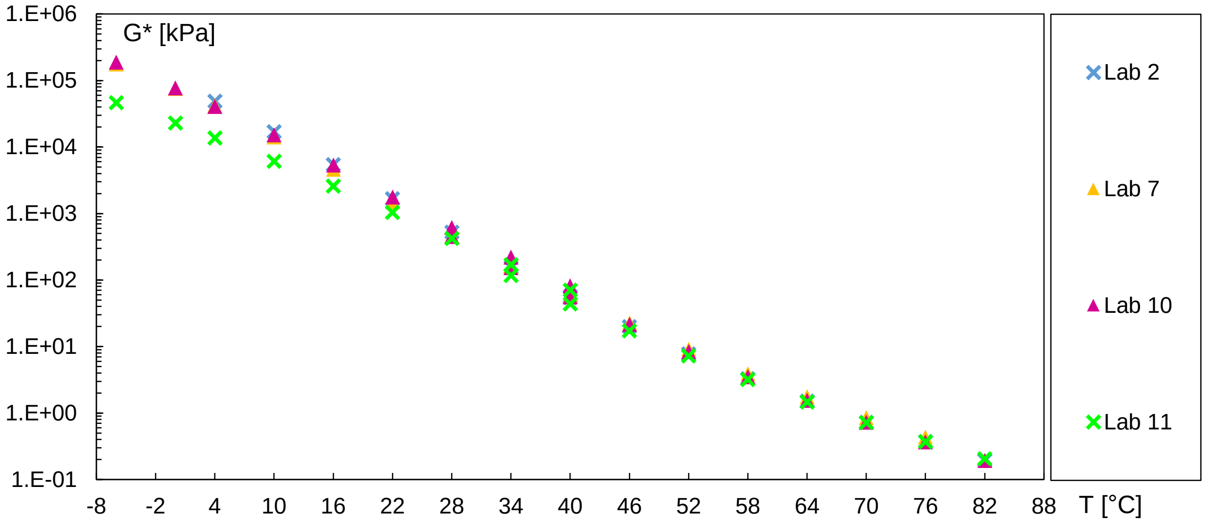


Figure 5. Modulus isochronal plot (|*G**| vs. *T*) at 1.59 Hz for straight run binder B

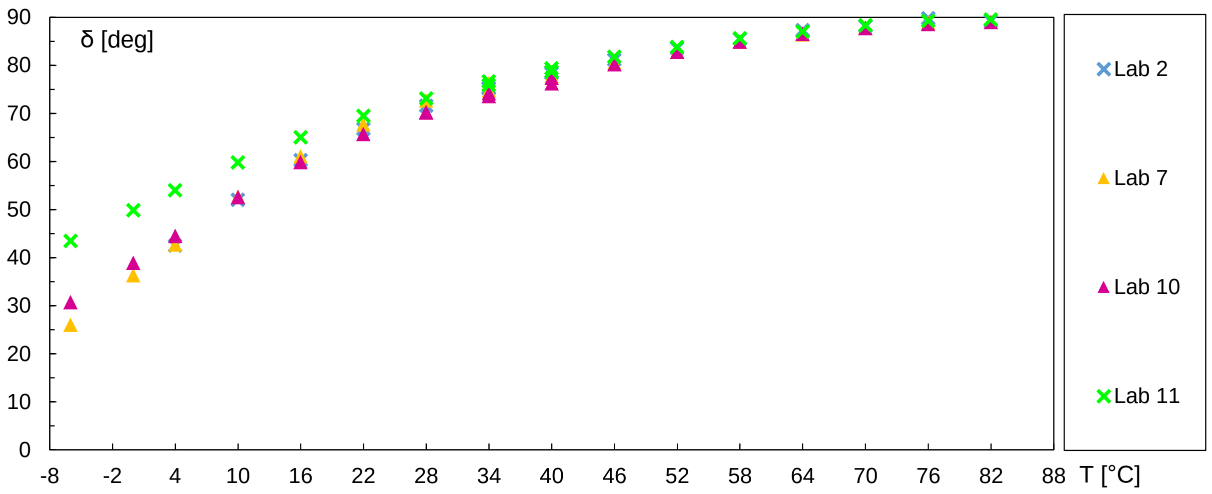


Figure 6. Phase angle isochronal plot (*δ* vs. *T*) at 1.59 Hz for straight run binder B

In the case of plastic modification ($B_{+pellets}$ or $B_{+shreds}$), where 5 and 11 labs participated respectively, the reported scattering of the results is more significant. As shown in Figure 7 to 10, the results from the different labos follow two different trends. On the one hand, deep rheological changes seemed to occur in favour of a significantly higher elastic behaviour at higher temperatures as seen with lower phase angles (for instance, note Labs 1.1, 1.2, 2, 7, 8 for $B_{+pellets}$, or Labs 1.1, 1.2, 3, 6, 7, 8, 11 for $B_{+shreds}$). On the other hand, some Labs reported iso-frequency curves with similar shapes to that of the neat bitumen. In this sense, plastic inclusion could be intended as a sort of filler-like particles addition since it results only in a slight stiffening effect, without a real rheological modification. It can be stressed that, for both $B_{+pellets}$ and $B_{+shreds}$, the curve divergency (rheological modification vs. filler-like behaviour) starts to be more evident after a threshold temperature region ($T_{TRSH}$) located between 34 °C and 40 °C (see red box in Figures 7 to 10). At temperatures higher than $T_{TRSH}$, the plastic particles likely started to separate from the binder matrix which is becoming more viscous. This “transition” temperature was the one used to shift from the 8 mm plate to 25 mm plate DSR geometry. Therefore, the relevance of experimental parameters for DSR testing should be carefully chosen when similar blends are tested. In this sense, using a higher measurement gap and/or investigation scale (e.g., mastics) could represent an interesting challenge for the DSR approach. Data scattering becoming greater for high-temperature testing was also visible analysing the maximum variation coefficient related to |G*|/sinδ at 1.59 Hz (1s% value): in this regard, Table 4 illustrates such concern considering, as instance, the single-operator variability for Lab 10 (it internally tested both B+pellets and B+shreds blends). Evidently, the scattering started to be greater when using PP25 geometry, overpassing the typical acceptance limit of 1.6 (AASHTO T315-20). Really, in the case of B+shreds, 1s% confidence threshold was not matched even at lower testing temperatures (PP08), and this fact was ascribed to the poorer homogeneity of the shreds-modified binders.

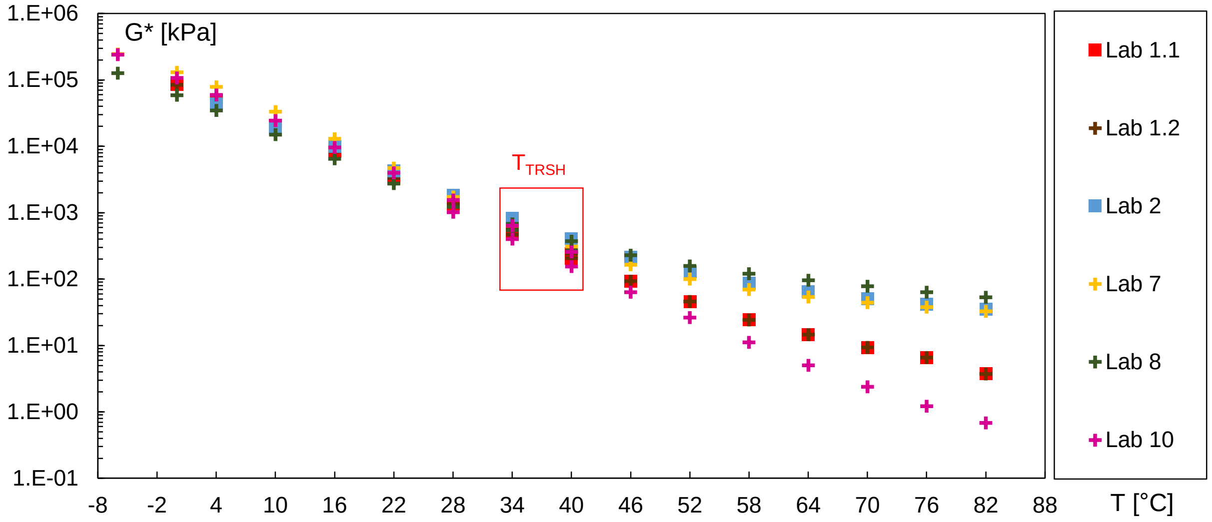


Figure 7. Modulus isochronal plot (|*G**| vs. *T*) at 1.59 Hz for $B_{+pellets}$

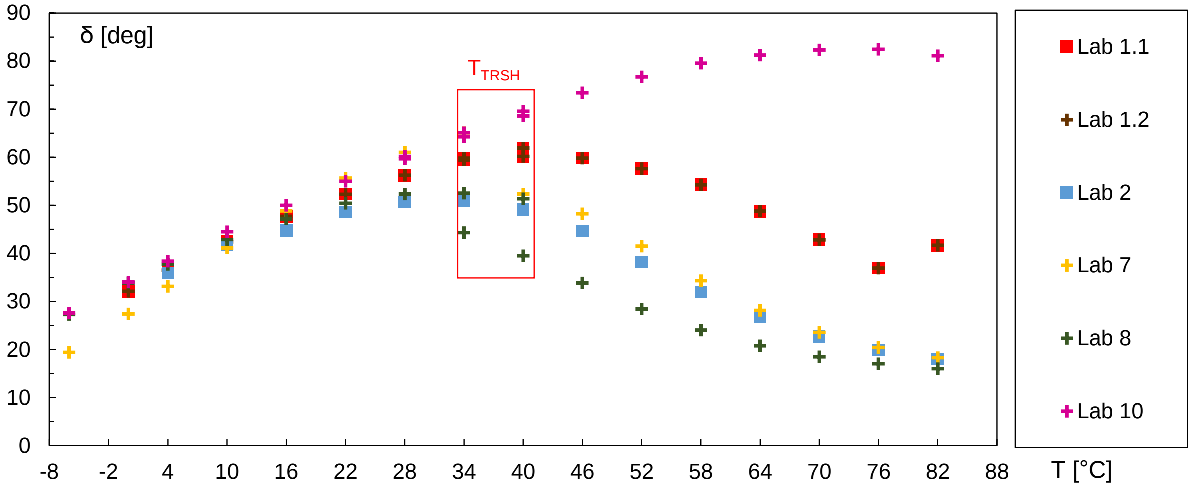


Figure 8. Phase angle isochronal plot (*δ* vs. *T*) at 1.59 Hz for $B_{+pellets}$

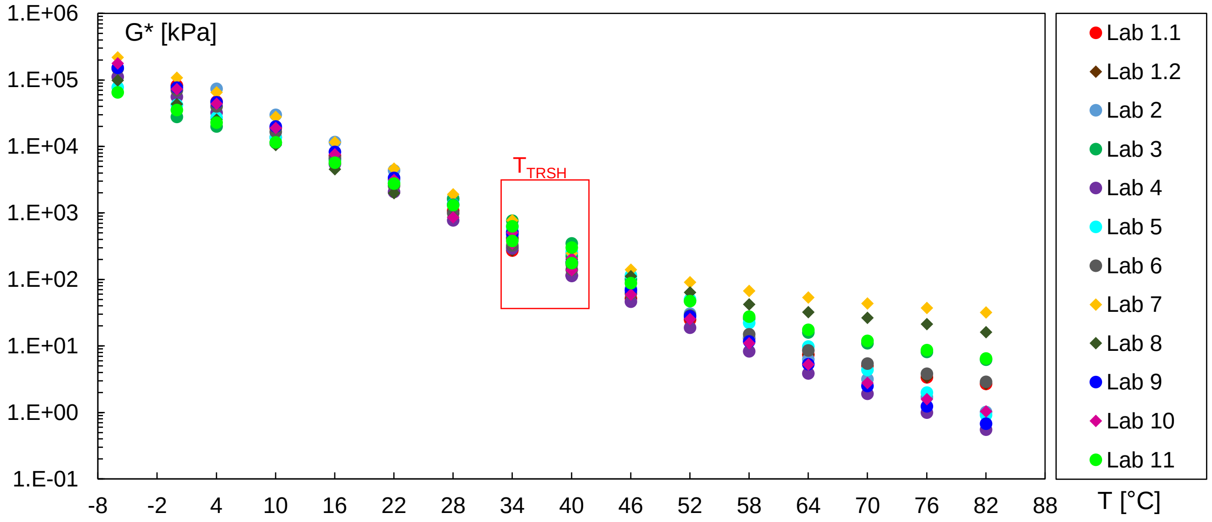


Figure 9. Modulus isochronal plot (|*G**| vs. *T*) at 1.59 Hz for $B_{+shreds}$

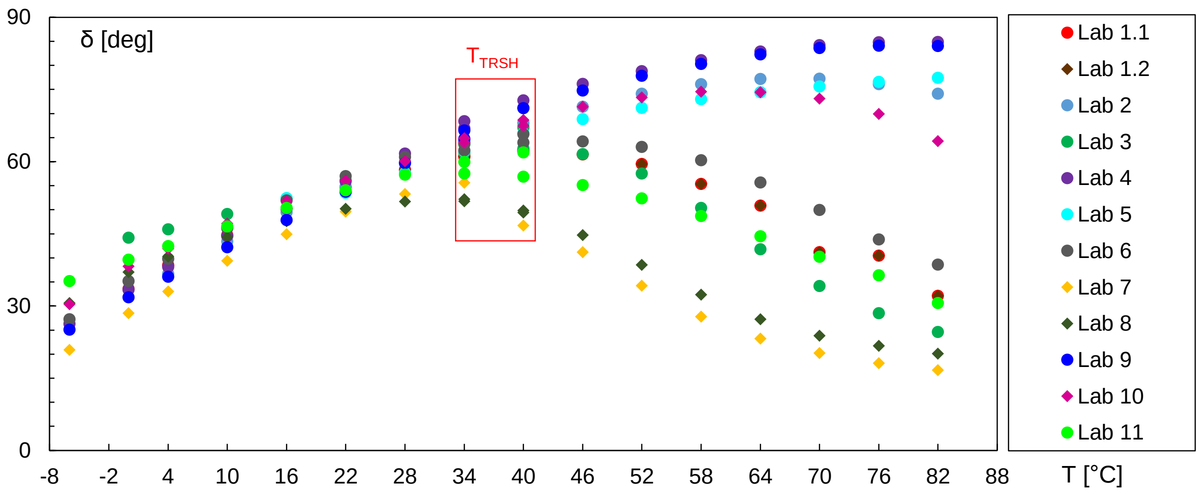


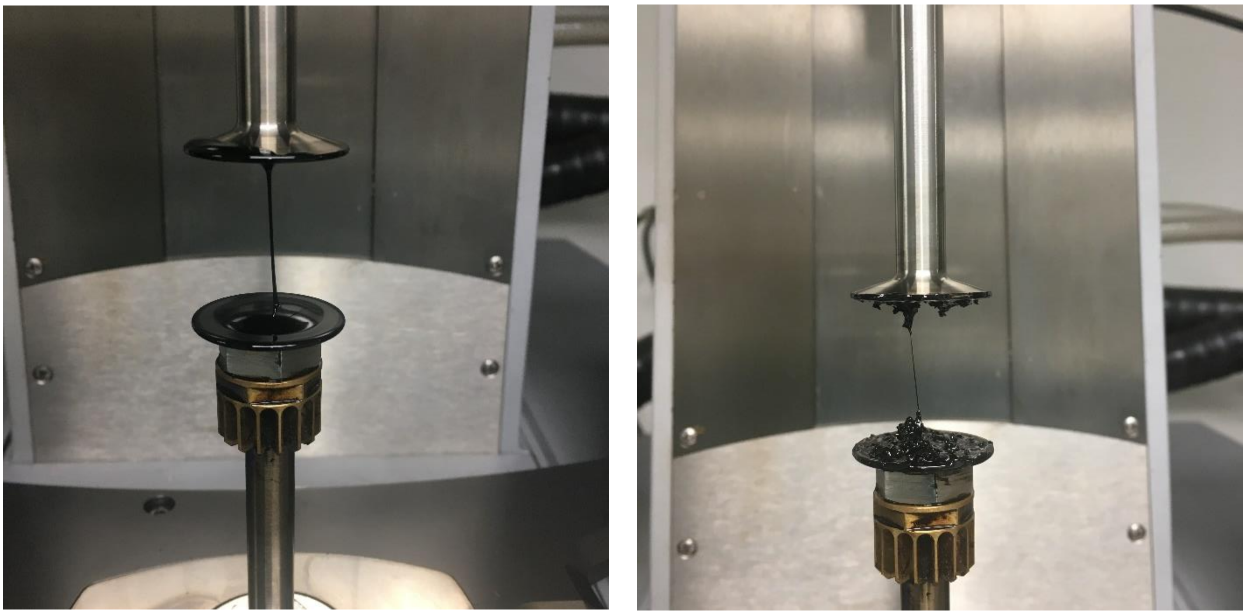
Figure 10. Phase angle isochronal plot ($\delta$ vs. $T$) at 1.59 Hz for $B_{+shreds}$

Table 4. Single-operator |G*|/sinδ precision at 1.59 Hz for Lab 10 ($B_{+pellets}$ and $B_{+shreds}$)

| Maximum variation coefficient 1s% | | | | | | | | | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Geom. | PP08 | | | | | | | | | PP25 | | | | | | | | | |
| T [°C] | -6 | 0 | 4 | 10 | 16 | 22 | 28 | 34 | 40 | 28 | 34 | 40 | 46 | 52 | 58 | 64 | 70 | 76 | 82 |
| $B_{+pellets}$ | 0.3 | 0.7 | 0.8 | 0.9 | 1.1 | 1.2 | 1.0 | 1.2 | 1.2 | 0.4 | 0.6 | 0.6 | 0.3 | **3.3** | **3.0** | **2.6** | **2.1** | 1.4 | **2.2** |
| $B_{+pellets}$ | **5.4** | **6.4** | **7.1** | **7.6** | **8.0** | 0.2 | **3.9** | **4.0** | **4.5** | **6.4** | **5.0** | **4.7** | **6.4** | **6.5** | **8.8** | **8.5** | **8.6** | **9.8** | **11.2** |

* **Bold** values represent data not in agreement with AASHTO T315-29 criteria

The observed variability among laboratories DSR results for plastic modified binders could be attributed to the inhomogeneous distribution of plastic particles, and to the binder morphology. During DSR measurement, some laboratories experienced great difficulty in performing the test on $B_{+pellets}$ and $B_{+shreds}$. Some pictures, made at end of the test when plates were lifted at 100 °C, clearly showed the existence of a coarse and inhomogeneous structure in binders containing plastic. An example is given in Figure 11, in which comparing plastic shred binder with neat bitumen, it is easy to observe some solid-like particle.



a) Standard 70/100 bitumen

b) Plastic shred binder

Figure 11. DSR sample at end of the test with 25 mm plate and 100 °C, for neat bitumen (a) and $B_{+shreds}$ bitumen (b)

To better understand the effect of inhomogeneity on the rheological behaviour of plastic modified bitumen, replicates of the same materials were tested by several labs. As explained in the previous paragraph, neat bitumen *B* was tested twice by Lab 10, this was also done for $B_{+pellets}$ and $B_{+shreds}$ with Lab 1 and Lab 10. In Lab 3 and Lab 4, three and five replicates were made for $B_{+shreds}$, respectively. In Figure 12 and 13, the isochronal plots of $|G^*|$ and $\delta$ raw data at 1.59 Hz are given for $B_{+shreds}$. Very similar $|G^*|$ curves can be found for both Lab 4 and Lab 10 over the entire testing temperatures. For Lab 1 and Lab 3, the differences between replicates start to be significant when the testing temperatures are higher than $T_{TRSH}$ between 34 °C and 40 °C, which is the shift between testing geometries. In the case of δ, high repeatability was found between five specimens from Lab 4. The discrete dots start to separate after $T_{TRSH}$ for both Lab 3 and Lab 10 blends. This was expected because of the softening of the bitumen at higher temperature (fluidity has a direct effect on homogeneity and the related material responses).

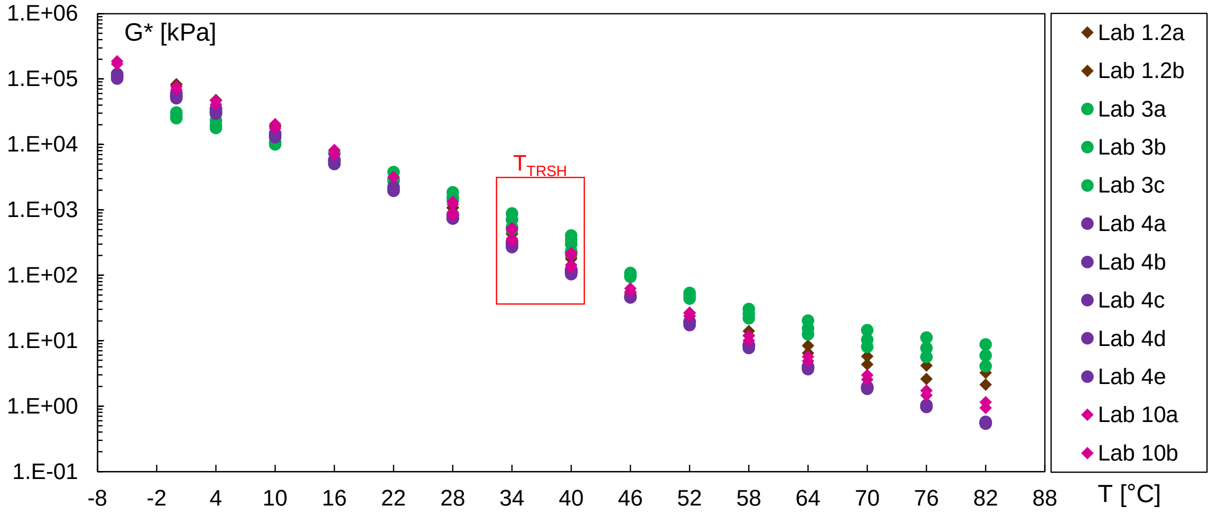


Figure 12. Evaluation of repeatability of Modulus isochronal plot ($|G^*|$ vs. $T$) at 1.59 Hz (10 rad/s) for Lab 1, 3, 4 and 10

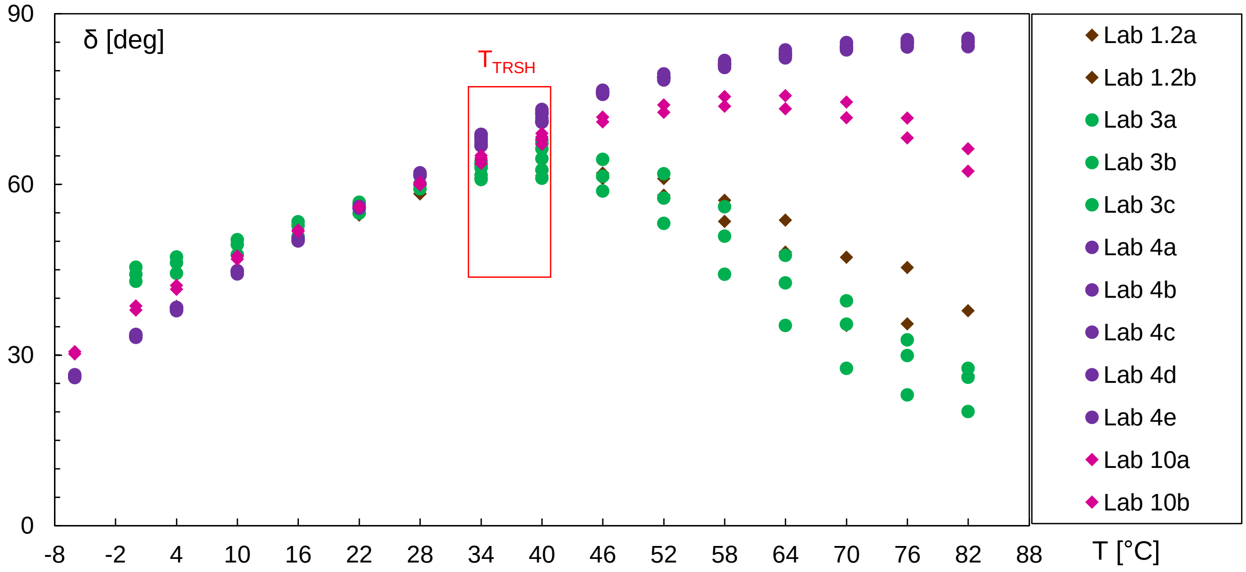


Figure 13. Evaluation of repeatability of Phase angle isochronal plot (*δ* vs. *T*) at 1.59 Hz (10 rad/s) for Lab 1, 3, 4 and 10

In Figure 12 and Figure 13, results for a single frequency is provided. To better understand the effect of experimental conditions (frequency and temperature), numerical comparisons are conducted using all the raw data of plastic modified bitumen (excluding outlier points by visual inspection), results are summarised in Table 5. It can be seen that, in most cases, the maximum differences between samples are higher than the acceptable tolerance of 10%. For plastic modified binders, the difference can be attributed to the heterogeneity of the blends. It is very interesting that at higher testing temperature, and/or lower frequency, the differences are larger. i.e., the effect of inhomogeneity becomes more pronounced as the bitumen becomes more viscous.

Table 5 Repeatability evaluation and influenced experimental conditions (outliers excluded)

| Materials | Lab No. | max. differences in \|G*\| (%) | max. differences in δ (°) | max. differences with frequency | max. differences with temperature |
|---|---|---|---|---|---|
| $B_{+pellets}$ | Lab 1 | 20 | 9.95 | low frequency | High T |
| | Lab 10 | 5 | 1.76 | No trend | No trend |
| $B_{+shreds}$ | Lab 1 | 25 | 12.85 | low frequency | High T |
| | Lab 3 | 36 | 6.97 | low frequency | High T |
| | Lab 4 | 5 | 3.22 | No trend | No trend |
| | Lab 10 | 16 | 2.38 | low frequency | High T |

### 3.2.1 Effect of plastic source and plastic type (at 1.59 Hz )

A better evaluation of the influence of blending of plastic material was done by comparing the results obtained by Lab 1, which tested materials from Blend 1 and Blend 2 with the same equipment (Figure 14 and 15). In this case, plots clearly indicate that, when fixing the plastic type, negligible differences in curves were due to the blend source (see the correspondence between square-cross or circular-rhomboidal markers). Therefore, the curves' scattering between Labs could be rather ascribed to different plastic dispersion in binders, thus strengthening the hypothesis of non-homogeneous blending of particles. In this perspective, a promising task could be addressed to increase of the compatibility between the selected bitumen and plastic particles (e.g., solvents) to enhance the plastic dispersion and the material homogeneity. On the other hand, further extrapolations of iso-frequency curves permitted to assess the influence of plastic types ($B_{+pellets}$ or $B_{+shreds}$), when both blends were tested by the same Lab (see Figure 16 and Figure 17 For the sake of clarity, a singular source (Blend 2) was plotted.

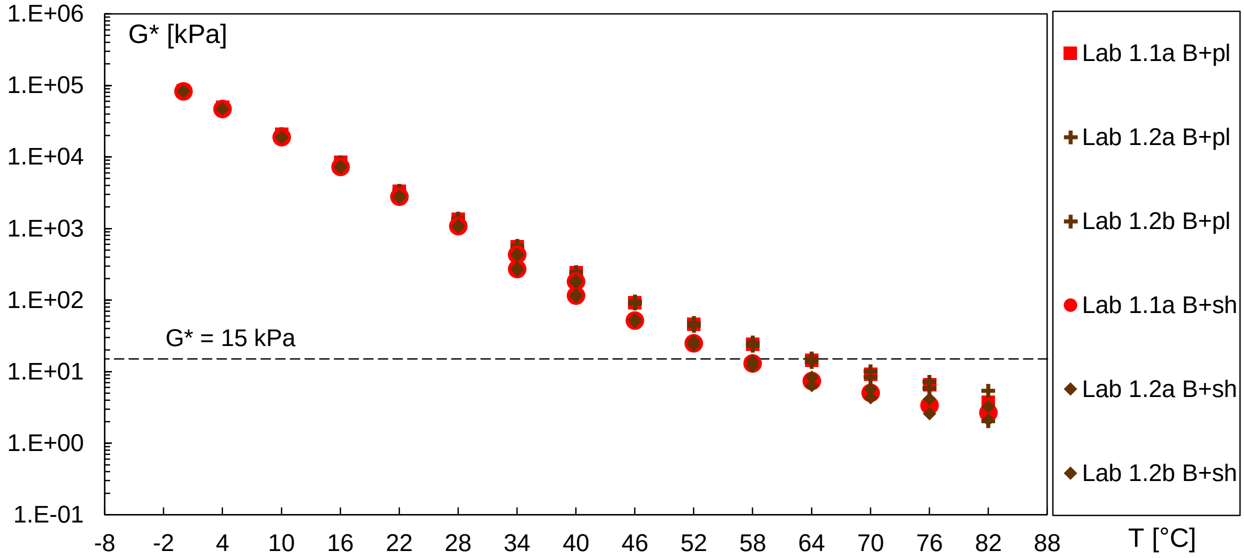


Figure 14. Influence of plastic material source: Modulus isochronal plot (|*G**| vs. *T*) at 1.59 Hz (10 rad/s) for Lab 1

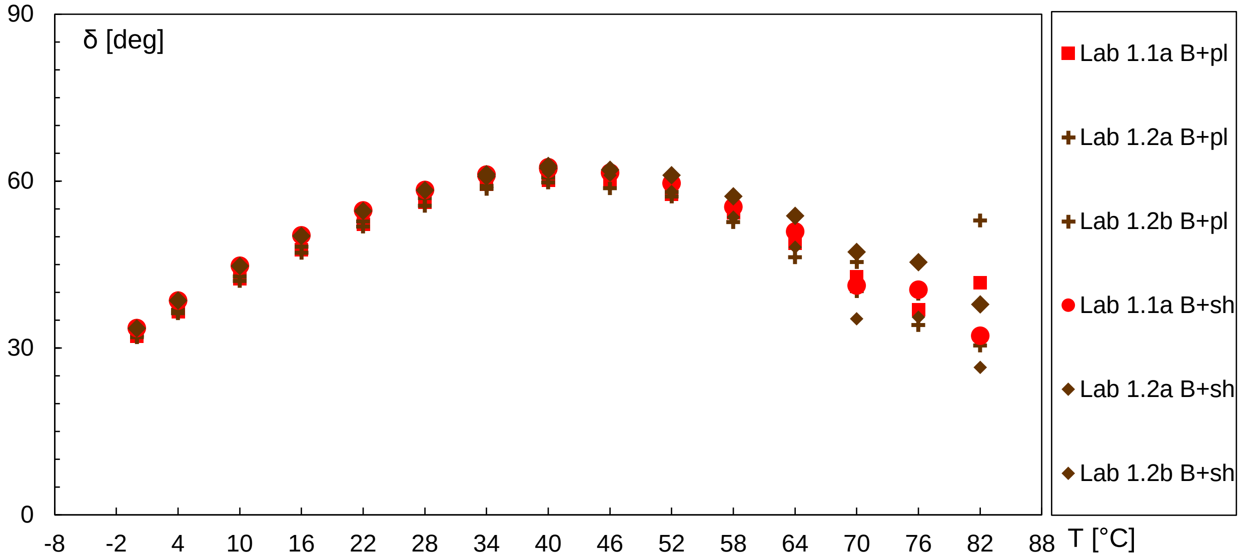


Figure 15. Influence of plastic material source: Phase angle isochronal plot ($\delta$ vs. *T*) at 1.59 Hz (10 rad/s) for Lab 1

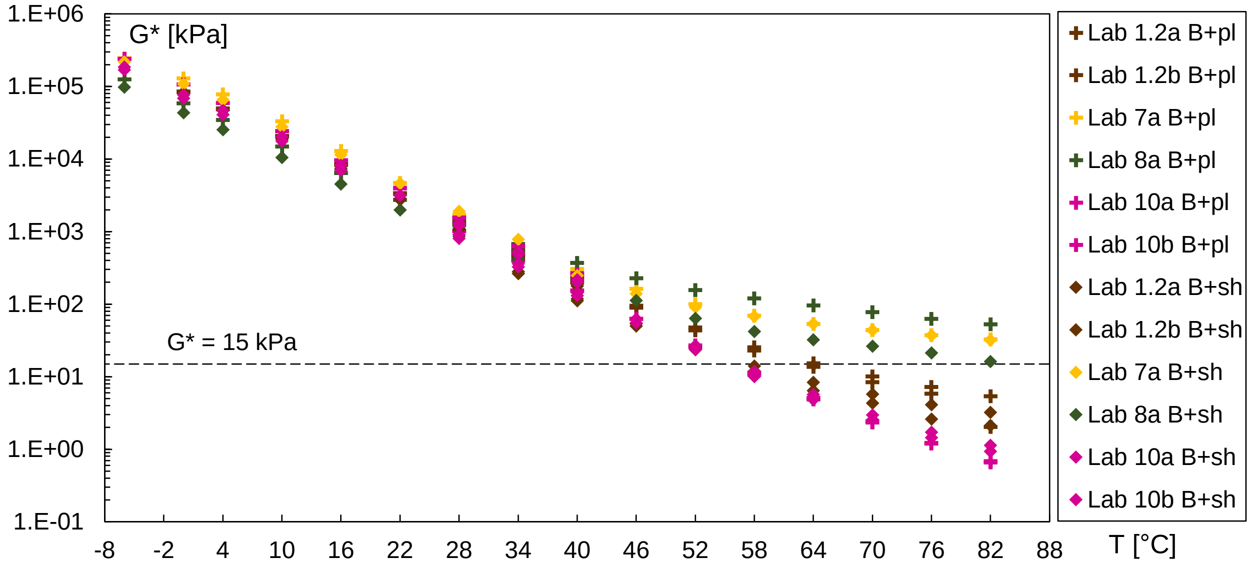


Figure 16 Influence plastic type (Blend 2): Modulus isochronal plot (|*G**| vs. *T*) at 1.59 Hz

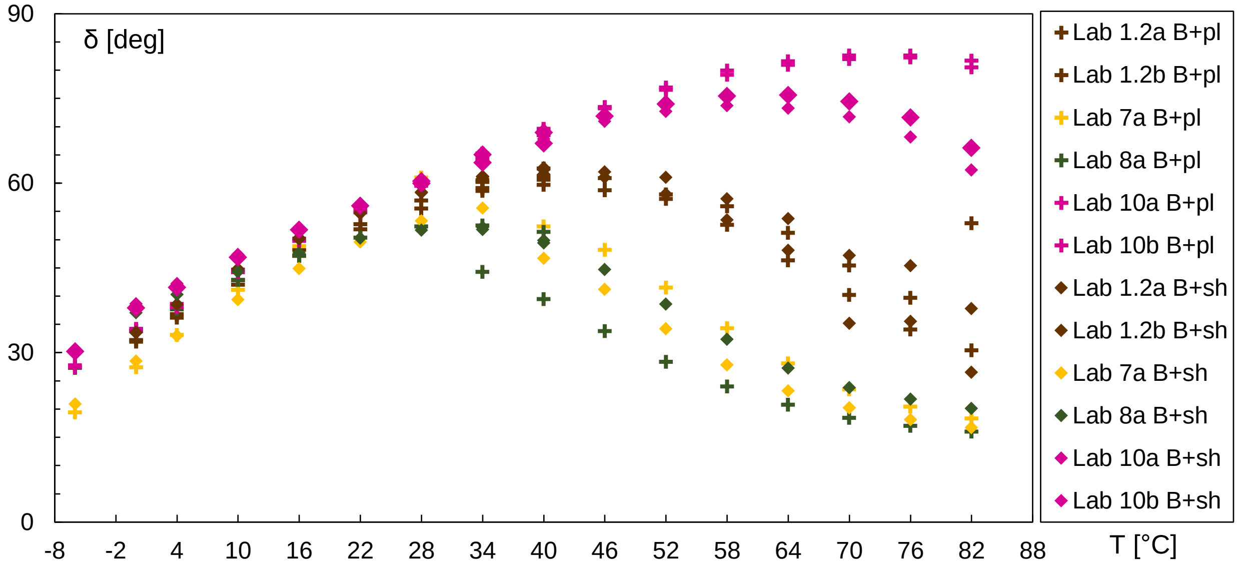


Figure 17. Influence blend sources (Blend 2): Phase angle isochronal plot (*δ* vs. *T*) at 1.59 Hz

### *3.2.3 Rheological parameters*

A recent approach was developed in Germany known as Binder-Fast-Characterization-Test (*Bitumen Typisierungs Schnell Verfahren* in German) BTSV (Alisov et al., 2018) and consider parameters such as iso-modulus of 15 kPa with corresponding temperature ($T_{BTSV}$), and the corresponding phase angle ($\delta_{BTSV}$). The value of 15 kPa has been selected to correlate with softening point temperature for neat bitumen. More information about this method can be found in previous publications (Alisov et al., 2018; Walther et al., 2019).

In the round-robin test, a comparison was made between the iso-modulus temperature at |G*|=15kPa and softening point temperature. As only seven laboratories performed the later, the analysis is made within this reduced lab panel. Results are calculated (Alisov et al., 2018) and summarised in Table 6. Compared to the neat bitumen, an overall increase in $T_{G^*=15kPa}$ and softening point temperature is found for the blends, while a decrease in phase angle is observed. Within the same materials, except for the softening point temperature of $B_{+shreds}$, very similar $T_{G^*=15KPA}$, $\delta_{G^*=15KPA}$ and softening point temperature between each lab are obtained. There are no significant differences between different blend sources. For neat bitumen *B* and $B_{+pellet}$, $T_{G^*=15KPA}$ is similar to softening point temperature (less than 5 °C difference). It should be noticed that, in the majority of the cases, for binder *B*, softening point temperature is lower than $T_{G^*=15KPA}$. However, the opposite trend was found for $B_{+pellet}$. In the case of $B_{+shreds}$, Lab 2 and Lab 10 measured a relatively high softening point temperature. This can be attributed to the heterogeneity of the plastic blends. Overall, while a trend is observed between the softening point temperature and the temperature at 15 kPa for the neat bitumen, no correlation is found for the blends with plastics.

Based on the raw data, the high temperatures of Performance Grade (PG) (AASHTO M320) were calculated for the listed labs (Table 6). For all labs that tested the neat bitumen, a consistent high PG of 64 is obtained. For both $B_{+pellet}$ and $B_{+shreds}$, a relatively higher high PG is observed. Except for specimens a to c of Lab 4 (PG 70), all the other materials indicate PG 76. It should be noted that in specimens a to c of Lab 4, the real high PG is very close to 76 (less than 1.3 °C). Hence, it is possible to say that the use of plastic can increase the PG of two PG grades.

Table 6. Comparison of softening point temperature and DSR parameters

| Materials | Lab No. | Softening point, ºC | $T_{G^*=15KPA}$, ºC | $\delta_{G^*=15KPA}$, º | high PG ($G^*/\sin\delta$=1kPa), °C |
|---|---|---|---|---|---|
| B | Lab 2 | 47.0 | 48.52 | 83.7 | 67.4 (64) |
| | Lab 7 | 45.0 | 48.74 | 82.6 | 68.1 (64) |
| | Lab 10 | 45.8 | 49.19 | 81.9 | 67.7 (64) |
| | Lab 11 | 44.6 | 49.47 | 84.1 | 67.8 (64) |
| $B_{+pellet}$ | Lab 10 | 60.8 | 56.93 | 77.4 | 77.2 (76) |
| $B_{+shreds}$ | Lab 2 | 76.2 | 58.35 | 73.9 | 80.4 (76) |
| | Lab 4 | 57.2 | 56.61 | 78.1 | 77.1 (76) |
| | Lab 9 | - | 57.34 | 79.1 | 77.3 (76) |
| | Lab 10 | 79.0 | 57.55 | 70.3 | 79.6 (76) |

### 3.3. Viscosity

For pavement engineering applications, the dynamic viscosity, with a rotating spindle, is used to ensure that the viscosity of the bitumen at normal processing temperatures is low and therefore the binder is liquid enough to coat the aggregates and to enable satisfactory compaction of the asphalt (Hunter *et al.*, 2015). Limit for pumpability is usually around 1000 mPa.s depending on the pump system. Rheologists are used to control the high temperature dynamic viscosity of bituminous binders for product development and quality control purposes (Caltrans, 2011). Usually, this exercise is carried out in a laboratory with a rotational viscometer with coaxial cylinders testing geometries (spindle), such as a Brookfield viscometer. The dynamic viscosity is therefore a key engineering parameter that asphalt technologists need to carefully control and target.

Viscosity measurements were performed using two spindles: the standard spindle per EN 13302 and a helical spindle specially designed for inhomogeneous binder blends. Using the standard spindle, three labs reported results for the neat binder (B), six labs for the blends with PE-shreds ($B_{+Shreds}$) and two for blends with PE-pellets ($B_{+Pellets}$) as shown in Table 7 and *Figure 18*. The mean values of dynamic viscosity increased with the addition of both types of PE at all three tested temperatures. The PE blends show higher viscosities compared to the neat binder, indicating a stiffening effect. Although the number of samples is small, the standard deviation (SD) gives a good indication of the spread in the data. As shown, the SD of the measurements was considerably higher for the blends with PE-shreds and this value decreased with increasing temperature. This is not surprising as the PE shreds Blends has higher heterogeneity compared to the Pellets blends. The results show that when mixing temperature of 160°C and 170°C, the viscosity is below the 1000mPas that is recommended for all binders tested. Additionally, from *Figure 18* it can be observed that the blends with PE Shreds and PE Pellets have very similar viscosities at the same temperature and the exponential trends with respect to temperature are very similar.

Table 7. Dynamic viscosity measurements using the conventional spindle for neat binder and PE modified (Blend1)

| Materials | Lab No. | T=135°C<br>η [mPa.s] | | | | T=160°C<br>η [mPa.s] | | | | T=170°C<br>η [mPa.s] | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | Mean | Max | Min | SD | Mean | Max | Min | SD | Mean | Max | Min | SD |
| B | 3 | 588 | 705 | 375 | 185 | 194 | 237 | 128 | 58 | 130 | 149 | 92 | 33 |
| $B_{+Shreds}$ | 6 | 1596 | 2675 | 1049 | 557 | 650 | 1241 | 503 | 314 | 457 | 1022 | 257 | 281 |
| $B_{+Pellets}$ | 2 | 1623 | 1754 | 1492 | 186 | 541 | 548 | 534 | 10 | 370 | 414 | 325 | 63 |

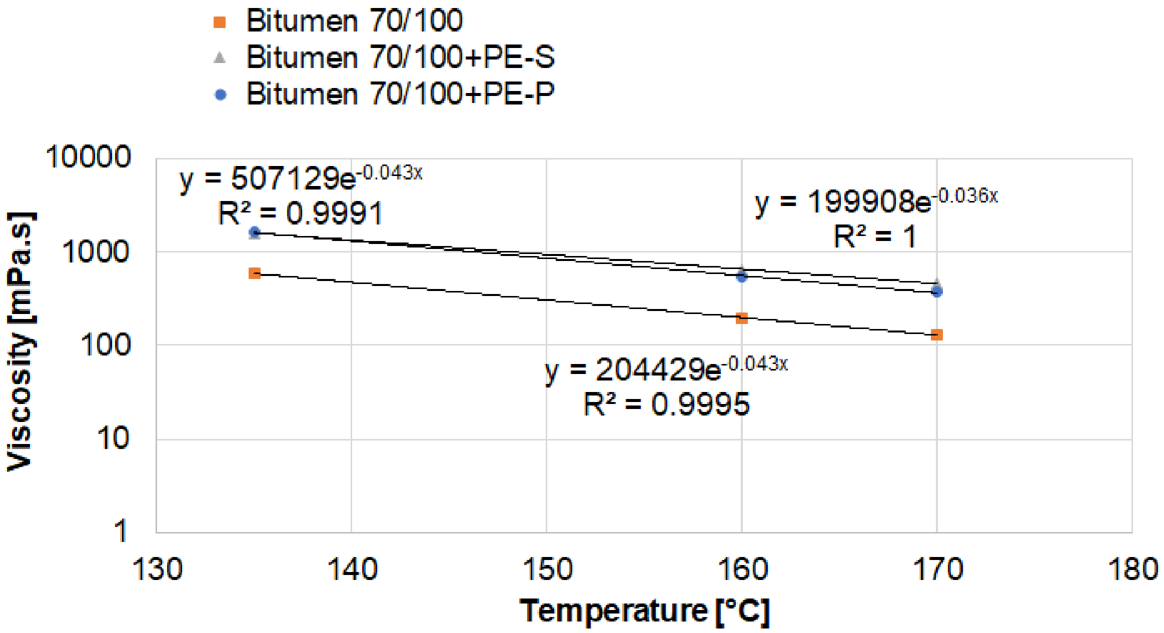


Figure 18. Viscosity vs temperature for neat binder and PE modified (Blend 1)

Due to the high level of heterogeneity of (PE) blends when tested using the rotational spindle, the sample may be affected by sample instability issues (e.g. phase separation, sedimentation, agglomeration) leading to results that are not representative. This investigation introduces the adoption of a Dual Helical Ribbon (DHR) presented in Figure *19*, which is a mixing/measuring device for rotational viscometers specifically designed to address these issues and in turns providing more realistic viscosity measurements (Giancontieri *et al.*, 2018).

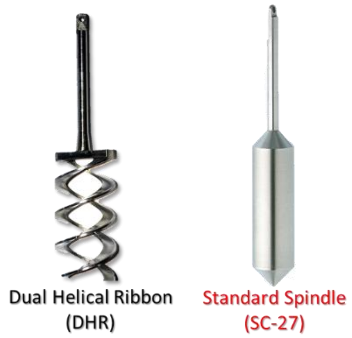


Figure 19. The dual helical ribbon (DHR) and the standard coaxial geometry (SC-27) used in this study

Dynamic viscosity measurements were undertaken with the DHR and a standard coaxial cylinder geometry (SC-27), as a reference, to record eventual differences in viscosity measurements of PE blends. The material was tested at three temperatures, 135, 160 and 170 °C, with a rotational speed of 100 rpm using a Brookfield DV II PRO Digital Rheometer. Tests were carried out for a total duration of 90 min, where the first 30 min were allowed to reach the thermal equilibrium (0 rpm). Then, impellers were quickly submerged in the blend and the viscometer was turned on to carry out the test. Each of the reported results was obtained as the average of three replicates.

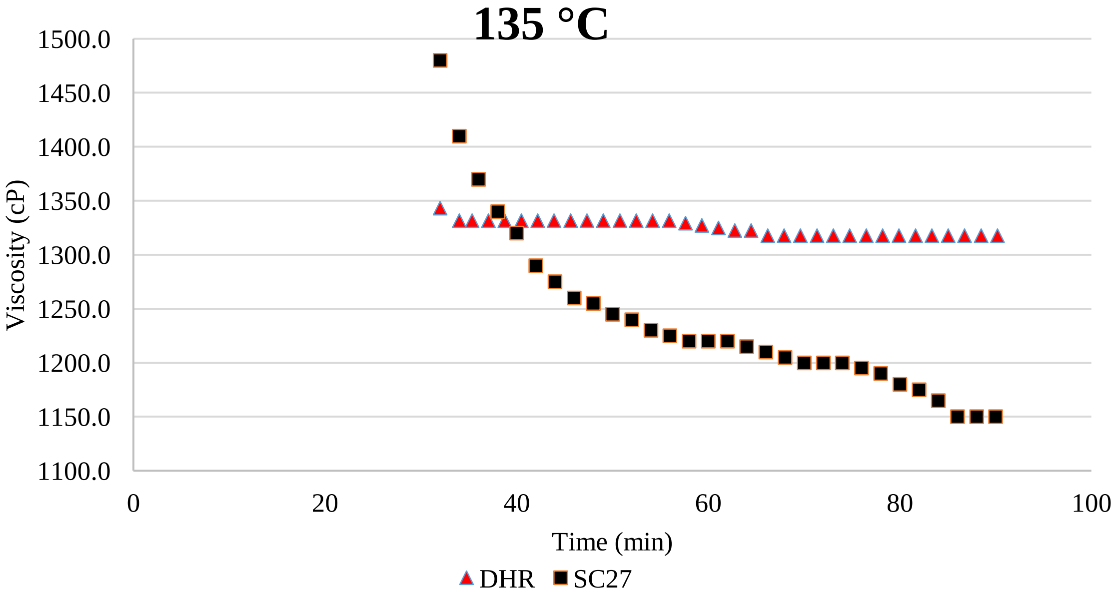


Figure 20. Viscosity measurements over time at 135 °C and 100 rpm. Blend1 with shreds

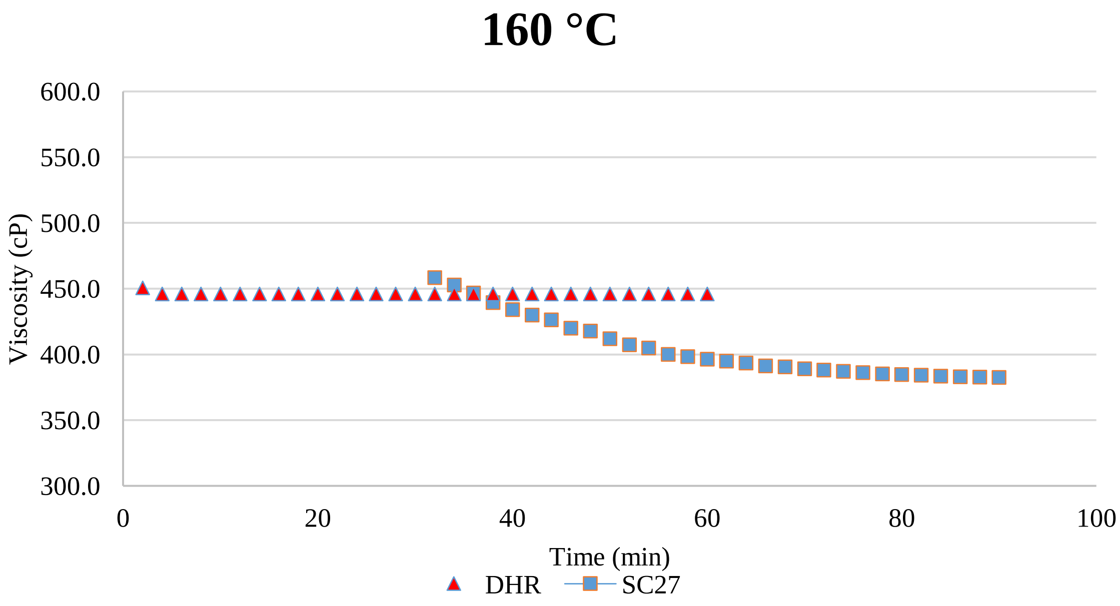


Figure 21. Viscosity measurements over time at 160 °C and 100 rpm. Blend1 with shreds

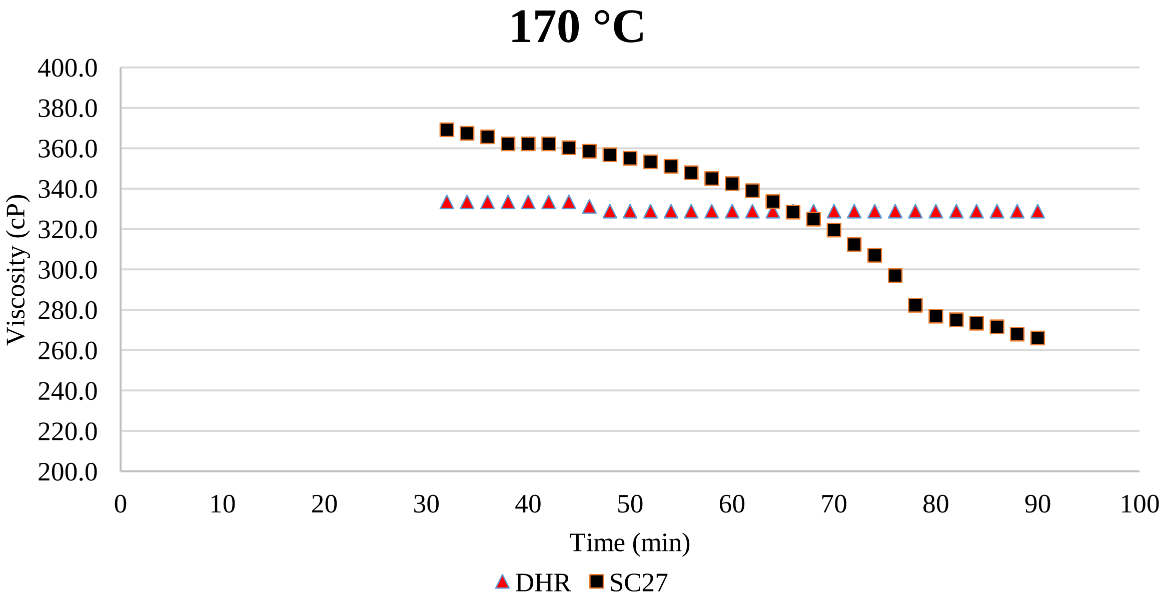


Figure 22. Viscosity measurements over time at 170 °C and 100 rpm. Blend1 with shreds

Rotational test results highlighted differences in viscosity measurements and prove that when PE blends are tested, the conventional coaxial cylinder setup might not be adequate, while the DHR may solve the issue. Figure *20* to Figure *22* show that at all the tested temperatures, the SC-27 was not able to provide a stable trend of viscosity measurements. The DHR not only allows to continuously measure the viscosity of the sample but also provides improved mixing performance which guarantees the stability of the sample during measurements.

As a result of the work done, this study helped to produce evidence to support the following conclusions:

- Rotational tests suggested that due to the high level of heterogeneity of PE modified asphalt binder, sample instability phenomena may occur;
- Compared to the conventional SC-27, the DHR provides a more stable trend of dynamic viscosity measurements. Another study has shown that this is due to improved mixing of the sample (Giancontieri *et al.*, 2018);
- Rotational viscosity measurements obtained by using the SC-27 testing geometry are time-dependent while results obtained with the DHR are not;
- The differences in viscosity measurement results, at all considered temperatures, are between 12% and 18%.

## 3.4. Other DSR tests (MSCR, LAS, and BTSV)

### *3.4.1 Multiple Stress Creep Recovery (MSCR) Test*

The multiple stress creep recovery (MSCR) test is a relatively new test methodology, developed within the framework of the NCHRP 9-10 research program (Bahia & Hanson 2001), that is used to evaluate the nonlinear properties of binders. Past studies have shown that the MSCR test relates well with rutting predictions and is able to characterize the behaviour of both unmodified and modified binders (D'Angelo, 2009; Masad, Huang, D'Angelo, & Little, 2009; Wasage, Statsna, & Zanzotto, 2011). The test is performed on asphalt binder samples, after being subjected to the Rolling Thin Film Oven Test (RTFOT), using the Dynamic Shear Rheometer (DSR) device and it is performed at two stress levels of 0.1 kPa and 3.2 kPa (AASHTO TP70-10, 2013; EN 16659:2016), that represent loading within and beyond the linear viscoelastic (LVE) domain, respectively (Wasage, Statsna, & Zanzotto, 2011). The difference between the response at high and low stress levels provides an indication of the stress dependence of binders (Soenen et al. 2013). The testing procedure involves the application of a shear stress using a haversine load for a period of 1 second followed by an unloading (recovery) phase of 9 seconds. A total of 20 loading-unloading cycles are performed at 0.1 kPa stress (with the last 10 cycles used for calculating the test parameters) and 10 loading-unloading cycles at 3.2 kPa stress, resulting in a total test duration of 200 s. The test is carried out using the 25 mm parallel plates configuration with a 1 mm gap. The average percent recovery (%R) after 10 cycles is determined at each stress level following Eq. 1. Moreover, the non-recoverable creep compliance ($J_{nr}$) is determined by dividing the non-recoverable shear strain by the shear stress. The average of the $J_{nr}$ values after 10 loading cycles at each stress level is calculated using Eq. 2.

Eq. 1 $$\%R_\tau = \frac{1}{10}\sum_{N=1}^{10}\frac{(\varepsilon_1^N - \varepsilon_{10}^N)\cdot 100}{\varepsilon_1^N} \quad (\%)$$

Eq. 2 $$J_{nr}^N = {\varepsilon_{10}^N}/{\tau} \quad (kPa^{-1})$$

where $\tau$ is the applied stress (0.1kPa and 3.2kPa), $\varepsilon_1^N$ is the strain value at the end of the creep portion (after 1 s) of each cycle, and $\varepsilon_{10}^N$ is the strain value at the end of the recovery phase (after 10 s) of N-th cycle.

Soenen et al. (Soenen et al., 2013) reported better reproducibility and repeatability of the MSCR results for unmodified binders than for neat bitumen. The high variation in the tests was attributed to poor specimen preparation procedures and the difficulty of specific rheometers to transition between the loading and the unloading phases.

Here, MSCR tests were carried out for the neat binder (B), the blends with the PE Shreds ($B_{+Shreds}$) and with Pellets ($B_{+Pellets}$) by Lab8 and Lab10. The testing conditions for each lab are summarized in Table 8.

Table 8. Test conditions for MSCR tests used by each laboratory

| | **Lab8** | **Lab10** |
|---|---|---|
| Repetition(s) | 1 | 2 |
| PP system (mm) | PP25 | PP25 |
| Stress level (kPa) | 0.1, 3.2 | 0.1, 3.2 |
| Loading scheme | 1 s load, 9s recover | 1 s load, 9s recover |
| Temperature (°C) | 60 | 60 |
| Conditioning time (min) | 5 | 30 |

The samples (Blend 1) tested in Lab8 and Lab10 were produced by the same laboratory (Lab1). However, the handling of the samples prior to testing varied among the two laboratories, but the samples were not subjected to RTFOT ageing prior to the MSCR testing.

- Lab8: From production to testing, the binder samples were stored at 8°C. The amount of material required to perform a single test was cut from the stored binder, placed on the preheated lower plate before lowering the upper plate and perform the trimming operations. The sample was conditioned for 5 minutes after reaching the test temperature;
- Lab10: Around 0.9 g of binder was cut with a hot knife from the stored binder, then the sample was reshaped by hand using the 25 mm silicon mold, and subsequently was loaded on the DSR machine. The sample was conditioned for 20 minutes after reaching the test temperature.

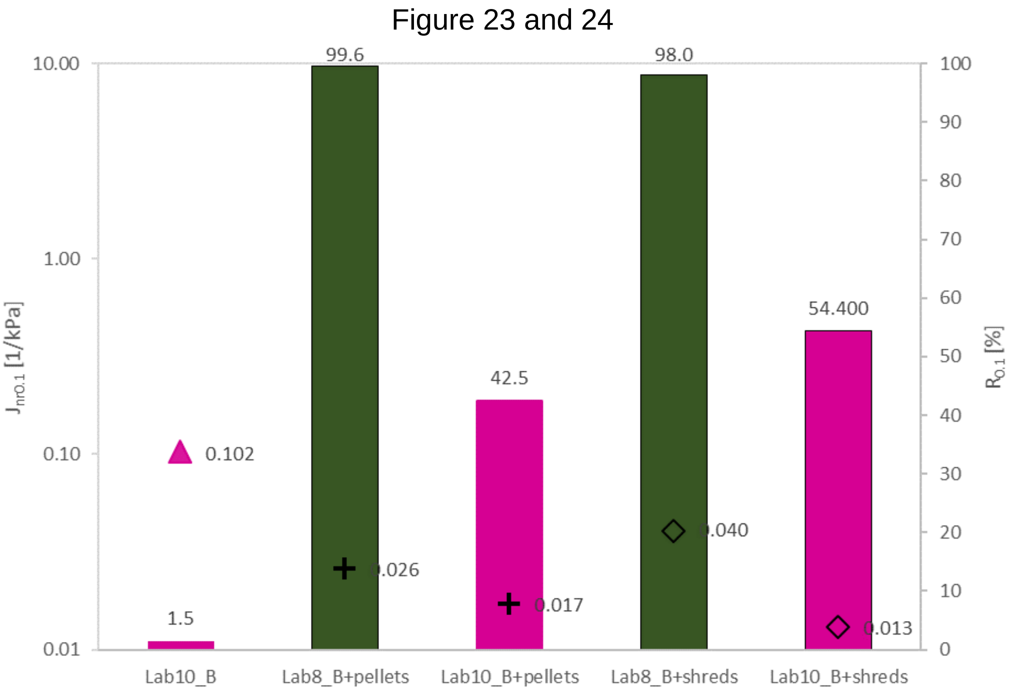


*Figure 23* show the MSCR test results of the neat binder (B), the blend with the PE Shreds ($B_{+Shreds}$) and the blend with Pellets ($B_{+Pellets}$) at 60 °C for 0.1 kPa and 3.2 kPa, respectively. Two replicates for each sample were tested by Lab10 and therefore the mean value of $J_{nr}$ and %R is reported. Overall, for all binders, the %R reduces and the $J_{nr}$ increases as the stress level increases from 0.1 kPa to 3.2 kPa. At both stress levels, the neat binder has the lowest %R among the various binders. On the other hand, the $J_{nr}$ value of neat binder do not show any specific trend when compared with the modified binders. At a stress level of 0.1kPa, the $J_{nr}$ value of the neat binder is higher than the modified binders, which is also the case for the higher stress level of 3.2 kPa, with the exemption of the $B_{+Shreds}$ for which the $J_{nr}$ value has substantially increased. In general, the results suggest that the neat binder is more sensitive to permanent deformation compared to the blends with PE shreds and pellets.

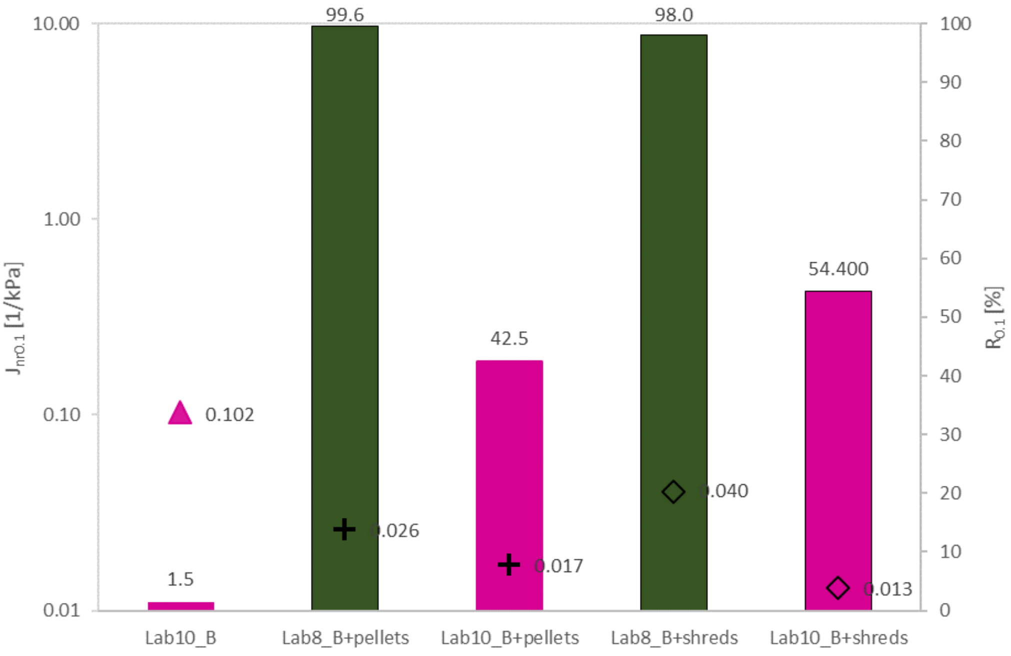


Figure 23. Strain recovery (%R) and non-recoverable ($J_{nr}$ )creep compliance at 0.1 kPa

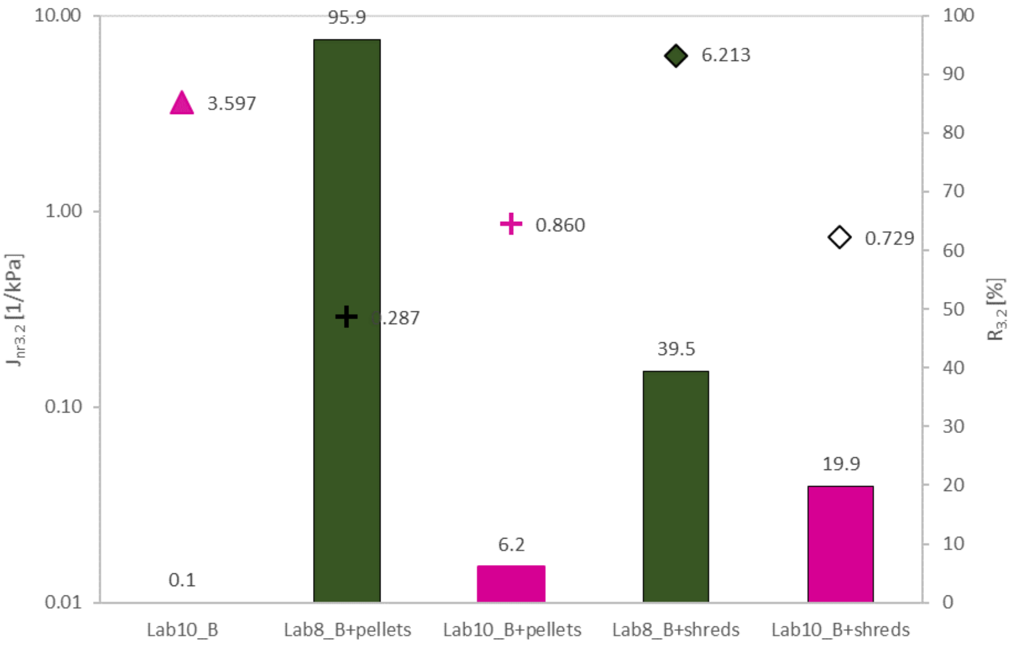


Figure 24. Strain recovery (%R) and non-recoverable ($J_{nr}$) creep compliance at 3.2 kPa

The recovery values obtained from Lab8 are systematically higher than those from Lab10. At low stress level, the recovery of the modified binders tested in Lab8 is almost twice the %R values reported for Lab10. A great difference in the recovery values of the modified binders between the two laboratories is observed at the higher stress level, with an %R of 95.9% for Lab8 and a %R equal to 6.2% for Lab10. Interestingly, the difference for the $B_{+Shreds}$ binders is at similar levels with the results at 0.1 kPa applied stress. These differences could possibly be explained by the handling of the material from the production of the blends until testing. The blends were produced in the same laboratory (Blend 1) and neither of the laboratories used thermal treatment during sample handling prior to testing. However, the storage conditions and the duration of the temperature conditioning stage, after the sample had reached the testing temperature, were different for the two labs as reported in Table 8. Moreover, the sampling of the material from the stored binder was not performed in the same way and this could possibly lead to the heterogeneity of the tested samples. Nevertheless, on the basis of the %R values, $B_{+Shreds}$ have consistently a better deformation recovery ability than $B_{+Pellets}$. Furthermore, as seen in the other tests reported in this work, the results of the labs diverge at the high temperature regime where bitumen becomes more viscous and the effect of plastic additives is more prominent, and hence the effect of inhomogeneity more apparent.

### *3.4.2 Linear Amplitude Sweep (LAS) Test*

The LAS test is used to evaluate the capability of bituminous binder to resist fatigue damage according to AASHTO 101-14 standard. Basically, the test is an oscillatory strain sweep test that produces damage to the binder by applying a linear increment of load amplitudes. The LAS test consists of two steps:

- First, a frequency sweep is performed to get information about the properties of undamaged material and to estimate the rheological characteristics of the bituminous binder;
- Second, the damage characteristics of the binder are measured using a linear amplitude strain sweep test.

In this study, frequency sweeps were done at 0.1% strain amplitude in a frequency range from 0.2 to 30 Hz, and the amplitude sweep test was conducted maintaining a constant test frequency of 10 Hz. The test temperature was fixed at 20 °C for Lab8 and Lab10 and to 22 °C for Lab2. All tests were conducted using a DSR with an 8-mm diameter parallel plate and a 2-mm gap. The tests were carried out on different types of binders, with and without plastics waste ($B_{+Shreds}$ and $B_{+Pellets}$ from Blend1), and without any prior ageing as described in the sections above. One replicate was tested for each binder. Before the testing phase, two different sample preparation procedures were adopted:

a) The binder was heated for 1 hour in a pre-heated oven at 170 °C. After this first conditioning process, the bitumen blend was stored before being directly poured into molds for testing;
b) The quantity of blended material needed to perform a single test was cut from the binder disks stored at low temperature. Then it was placed on the preheated lower plate before lowering the upper plate and perform the trimming operations. (This procedure was adopted from the Lab10 because this is a standard procedure adopted in their lab to store and test binders).

The number of cycles to failure was calculated using Eq. 3.

Eq. 3 $$N_f = A(\gamma_{max})^B$$

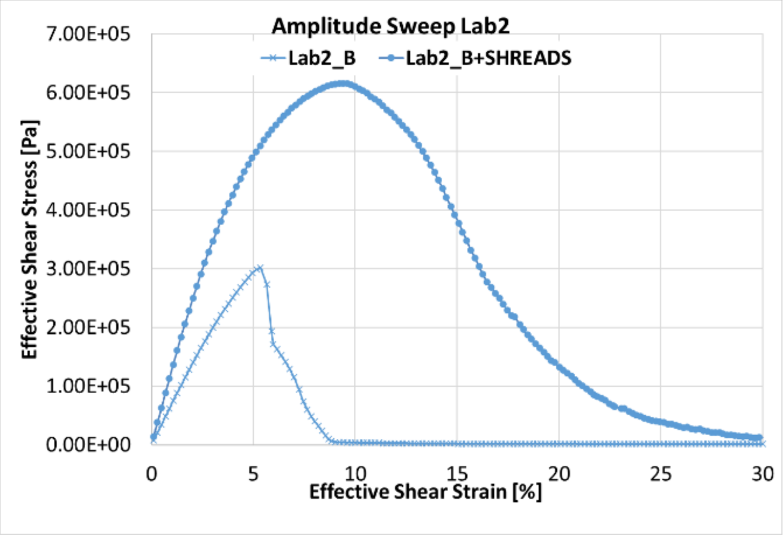
The failure definition in the LAS test is defined for all laboratories as 35% reduction in the initial modulus, where A and B are the viscoelastic continuum damage (VECD) model coefficients that depend on the material characteristics. Details of A and B parameters formulations can be found elsewhere (Chen & Bahia, 2020). In summary, parameter A represents the material's ability to keep its integrity during loading cycles and due to the accumulated damage and it is directly related to the storage modulus. By decreasing the storage modulus through loading cycles, the A parameter decreases, which indicates the low binder resistance in maintaining its integrity during loading and due to accumulated damage. From Eq. 3, when the strain level is equal to 1, the fatigue life will be equal to the A parameter, hence, A parameter can be considered as the fatigue life of the binder at a strain level of 1 (100%). The sensitivity of the asphalt binder to the strain level modification is described by the B parameter. More precisely, a higher absolute value of the B parameter indicates that the fatigue life decreases at a higher rate when the strain level amplitude increases. In general, higher fatigue resistant binders tend to have higher A values and lower absolute B values. Within the Inter-laboratory research program, the LAS test was conducted by Lab2, Lab8 and Lab10 by adopting the test conditions reported in Table 9. Only Lab2 followed the AASHTO specification (0-30% strain level at 20 °C), while Lab8 and Lab10 have considered a strain level equal to 0-40% at a test temperature of 20 °C.

Table 9. LAS test condition for each laboratory

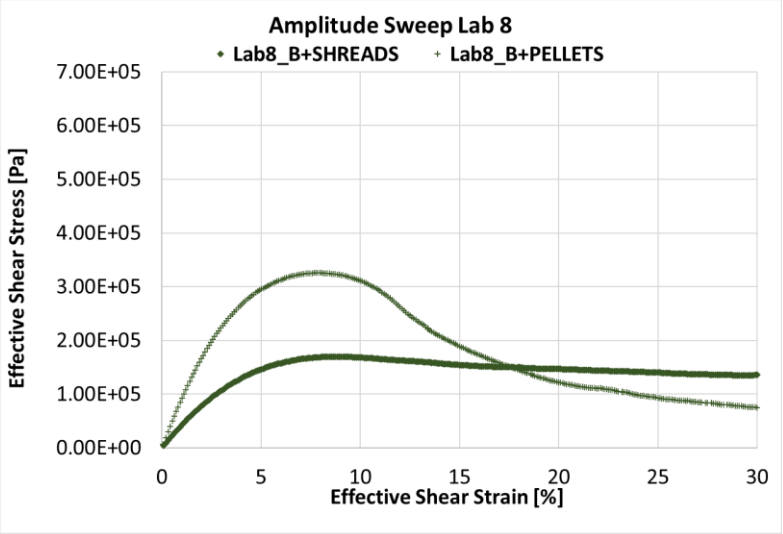
| Test Condition | Laboratory label | | |
|---|---|---|---|
| Name | Lab2 | Lab8 | Lab10 |
| Repetition | 2 | 1 | 2 |
| PP system (mm) | 8 | 8 | 8 |
| Strain (%) | 0-30 | 0-40 | 0-40 |
| Stress (kPa) | - | - | - |
| Temperature (°C) | 22 | 20 | 20 |
| Frequency (Hz) | 10 | 10 | 10 |
| Standard | TP 101-14 | TP 101-14 | TP 101-14 |
| Conditioning time (min) | 30 | 31 | 32 |

Figure 25 shows the effective stress-strain relationship for the three labs. It can be seen in Figure 25 (a) and (c), the peak stresses of the samples containing shreds ($B_{+Shreds}$) are higher than those of the neat bitumen (B). It is observed that the plastics modified binders have delayed falling curves compared to the base binder. From the analysis of the graphical results, it is possible to observe how the stress values are much higher and closer to each other for Lab2 and Lab10, whereas Lab8 exhibits much lower values (almost half of the other two). This condition can be related to the different production process adopted by the labs. Specifically, in the case of Lab8 and Lab10 the bitumen with plastics (both shred and pellet) were prepared by heating bitumen at 170°C and stirring it for 1h at 3500 rpm. However, during the process, as the plastic did not really melt, temperature was increased to 180 °C. The procedure adopted was the same for Lab8 and Lab10, but the temperature was kept constant to 170 °C in the case of Lab8. This variation in the mixing temperature can influence the stress level.

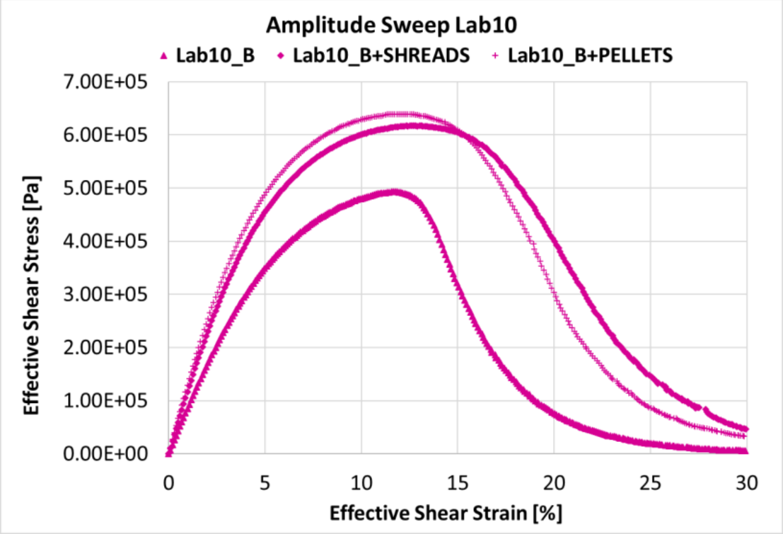
This condition confirms that not only the type of plastic affects the bitumen properties but also the associated homogeneity of the plastic adopted (the PE pellets are more homogeneous than the SE shreds).



a)



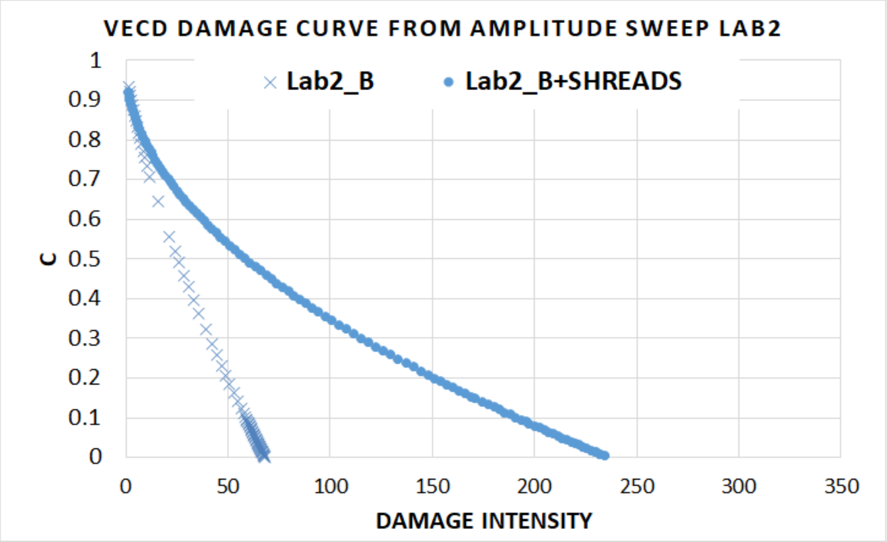
b)



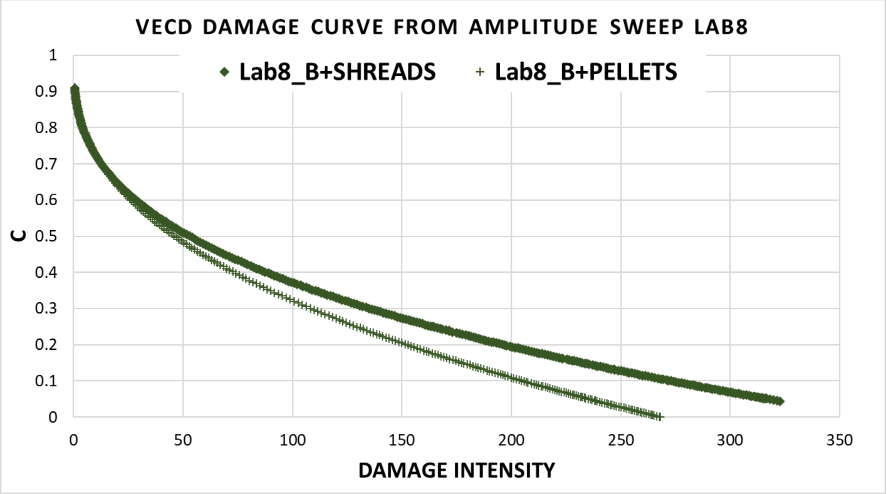
c)

Figure 25. Plotting shear strain vs shear stress: a) Lab 2, b) Lab 8 and c) Lab10.

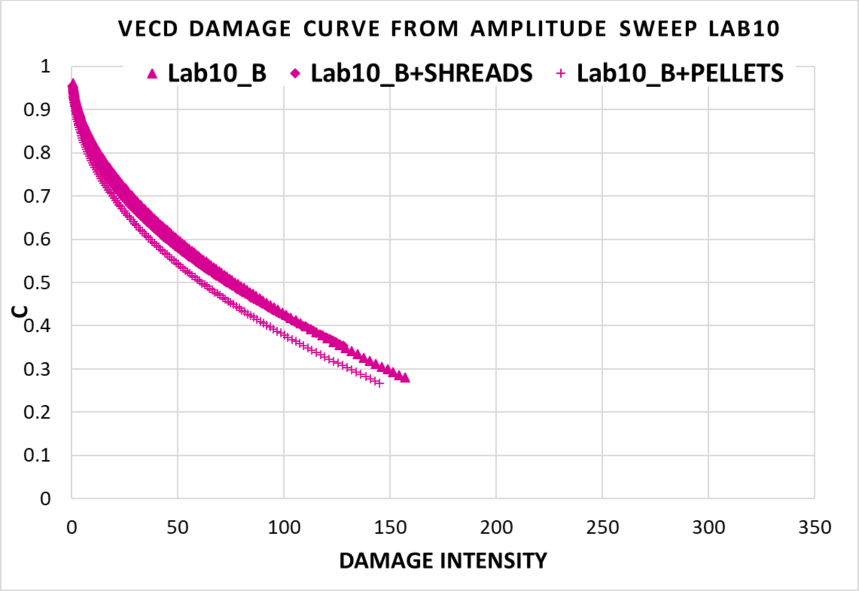
Figure 26 compares damage characteristics (C-Damage Intensity) curves of the tested binders. From the results plotted in Figure 26 (a)-(c), it can be observed that the base binder (B) shows an unfavourable damage curve compared to the binder samples containing shreds when considering the results of Lab2 (*Figure 26* (a)). However, from the results shown in Figure 26 (c), it can be observed that the base binder has the least damage when compared to the damage curves of the PE-modified blends. Between the two modified binders, it can be observed that the $B_{+Shreds}$ exhibit slightly higher damage than the $B_{+PELLETS}$.



a)



b)



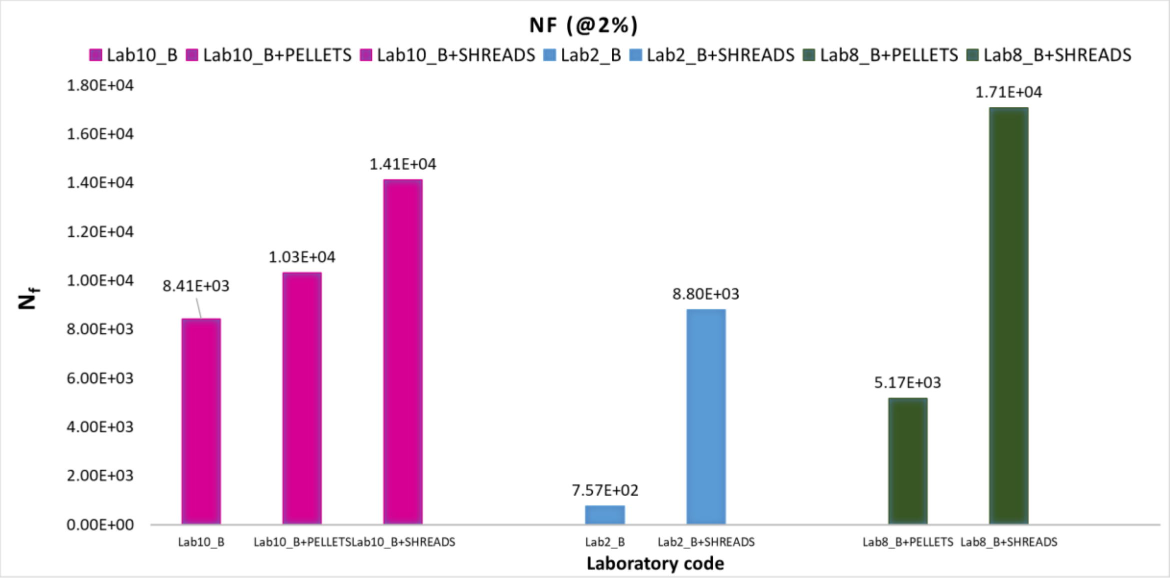
c)

*Figure 26. C-Damage Intensity: a) Lab 2, b) Lab 8 and c) Lab10*

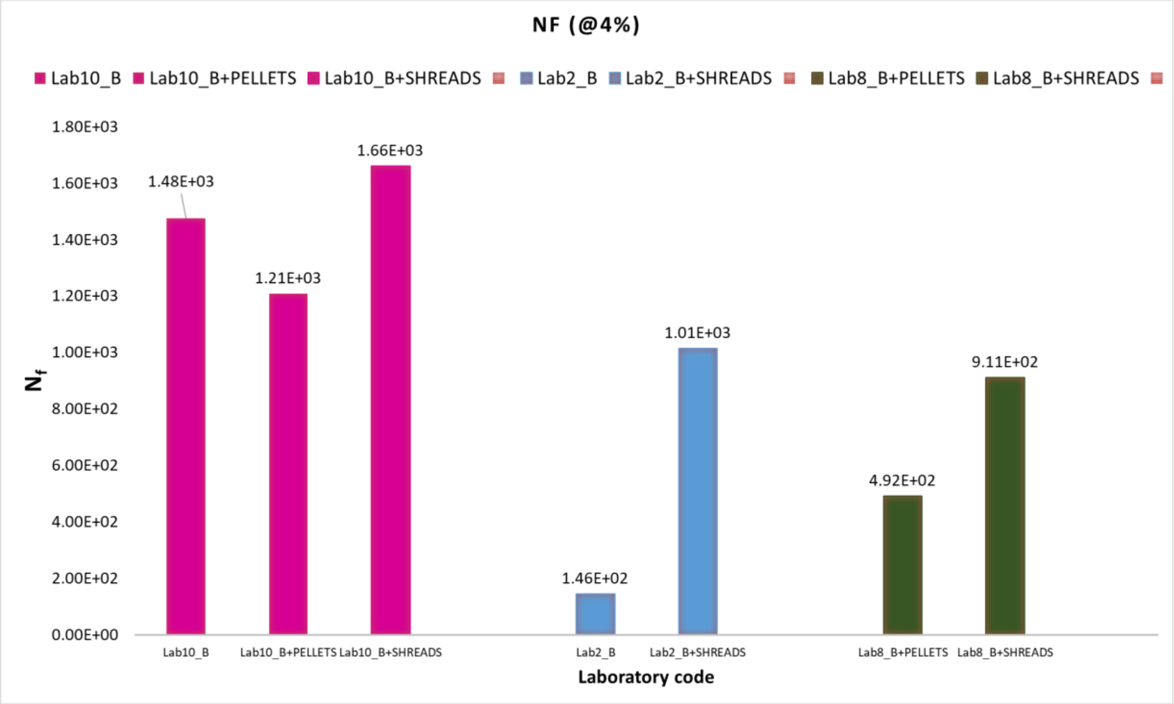
Figure 27 compares the calculated $N_f$ values (eq. 3) of all tested binders at 2% and 4% strain level. The results show that the base binder has the lowest $N_f$ compared to the tested modified binders at 2% strain as shown in Figure 27 (a). At strain levels of 4%, the unfavourable results are explained by the bitumen prepared using plastic waste in pellets form ($B_{+Pellets}$) as shown in Figure 27 (b). It is also possible to observe how passing from 2% to 4% of stress level bitumen prepared with plastics exhibits a lower variation than neat bitumen (12% vs 18% for Lab10, 12% vs 19% for Lab2 and 5 to 10% for Lab8). In any case, the binder containing plastic in shreds' form ($B_{+Shreds}$) shows consistently better results in terms of $N_f$ compared to the rest of the binders and the results are independent of the testing lab. The LAS test parameters (VECD model coefficients, number of cycles) are summarized in Table 10.

Table 10. LAS test condition for each laboratory

| Sample code | Parameter A | Parameter B | $N_f$ (@2%) | $N_f$ (@4%) | Variation |
|---|---|---|---|---|---|
| Lab10_B | 4.80E+04 | -2.511 | 8.41E+03 | 1.48E+03 | 18% |
| Lab10_ $\mathbf{B}_{\mathbf{+Pellets}}$ | 8.81E+04 | -3.094 | 1.03E+04 | 1.21E+03 | 12% |
| Lab10_ $\mathbf{B}_{\mathbf{+Shreds}}$ | 1.20E+05 | -3.090 | 1.41E+04 | 1.66E+03 | 12% |
| | | | | | |
| Lab2_B | 3.93E+03 | -2.377 | 7.57E+02 | 1.46E+02 | 19% |
| Lab2_ $\mathbf{B}_{\mathbf{+Shreds}}$ | 7.63E+04 | -3.116 | 8.80E+03 | 1.01E+03 | 12% |
| | | | | | |
| Lab8_ $\mathbf{B}_{\mathbf{+Pellets}}$ | 5.44E+04 | -3.395 | 5.17E+03 | 4.92E+02 | 10% |
| Lab8_ $\mathbf{B}_{\mathbf{+Shreds}}$ | 3.20E+05 | -4.228 | 1.71E+04 | 9.11E+02 | 5% |



a)



b)

Figure 27. Nf value comparing the three binders: a) strain level of 2% and b) strain level of 4%

In conclusion, from the analysis of LAS test results, the shreds-modified binder gives better results in terms of the number of cycles to failure ($N_f$) and damage curves. Furthermore, shreds-modified binder displays a higher peak stress value than the base binder. The PE-modified binder prepared with plastic pellets, exhibits a higher peak stress value than the base binder, but this is not supported by the $N_f$ values and the damage curves, which are mainly comparable or unfavourable if compared with those of the base binder.

## 3.5. Other tests (FTIR, DSC, and microscopy)

### *3.5.1 Fourier Transform Infrared, FTIR*

In addition to physical properties, Lab7 and Lab8 performed Fourier-Transform-Infra-Red spectroscopy in ATR mode using Blend2. The profile gave consistent spectrum between the two labs. The two blends were compared with the reference bitumen. Figure 28 shows the FTIR spectrum from Lab7 between 2000 and 600 cm$^{-1}$. There are hardly any differences, at least no additional peaks, between the reference and the two blends indicating no new chemical bonds forming due to these blends.

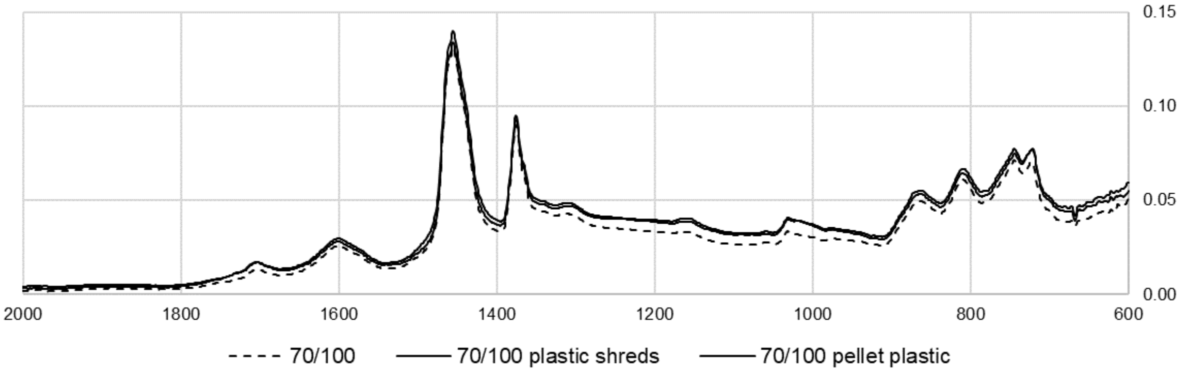


Figure 28. Infrared spectrometry on the three binders (B), B+Shreds, and B+Pellets using Blend2

### 3.5.2 Differential Scanning Calorimetry, DSC

DSC was also performed on the plastics themselves, the blends and the reference bitumen. The method provides information on glass transition temperature and melting point. After equilibrium at 165 °C, the experiment was run by cooling from 165 °C to -60 °C at a cooling rate of 2.00 °C per min followed by heating in the same condition.

Figure 29 shows the DSC heat flow diagrams on the plastic samples themselves. As the plastic shreds were of different types, with blue, white or black shreds, DSC was run on each of them to identify any difference. Two plastic shreds had a melting point around 124 °C and the other plastic shred, at around 130 °C. Those results are in the same range of the melting point for the plastic pellets. It was difficult to have an exact value for the plastic shreds as it depends on the proportion of the different shred types. Outside this temperature, there were no glass transition nor additional melting point that were identified.

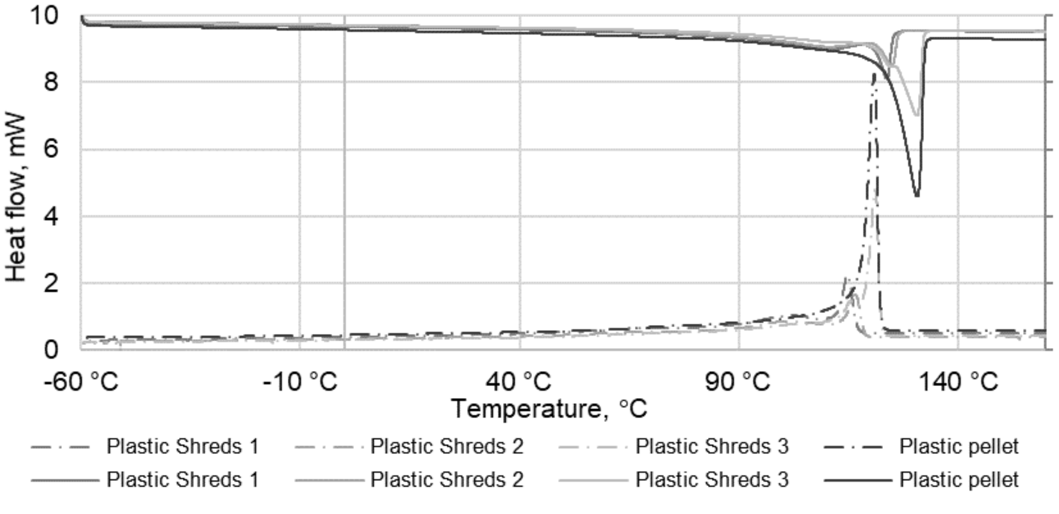


Figure 29. DSC on plastic samples B, B+Shreds and B+Pellets using Blend2 The continuous lines are for heating process and the dotted lines for cooling

Figure 30 shows the heat flow results on the reference base bitumen and the two plastic blends. Between -30 °C and -10 °C a smooth glass transition was observed for the bitumen. This part was similar for both the neat bitumen and the plastic blends, meaning that the plastic did not interact with the bitumen matrix to influence the glass transition. Contrary to the low temperature results, at elevated temperatures, the melting points, observed from the plastics, are clearly visible and located around 119 °C for the plastic shred blend and 122 °C for the plastic pellet blend aligned with the ones from the pure plastics. Very limited interaction was observed presuming non-unique phase morphology.

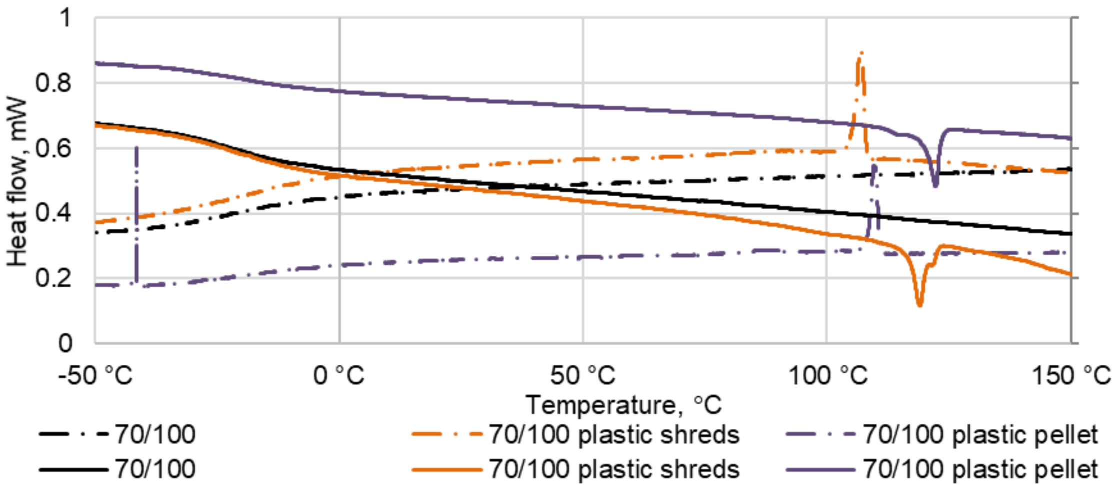


Figure 30 DSC on binders. The continuous lines are for heating process and the dotted lines for cooling

## 4. Conclusions

Round robin experiments were performed on PE blended bituminous binders. As a result of these experiments, the following conclusions can be drawn:

- In order to add PE to bitumen, plastic samples need to be grounded to around 1mm in order to facilitate the blending and homogeneity;
- Addition of PE waste to bitumen does not alter the chemistry of the bitumen. The blending results in a physical rather than a chemical modification. This can be seen also from FTIR results;
- The results obtained from different labs with conventional tests show high variability at high temperature, as recorded by the softening point, and lower variability at 25 °C, as shown by the penetration results. This can be attributed to the heterogeneity of the binder blends, especially at high temperature;
- The Fraass breaking temperature of the PE modified blends increased by at least 5 °C, showing a significant negative effect on the low temperature properties;
- The PE blended samples were not stable with most of the PE particles moving to the top of the tube;
- Using the dynamic shear rheometer, for both for $B_{+pellets}$ and $B_{+shreds}$, the curve divergence (rheological modification vs. filler-like behaviour) begin to be more evident after a threshold temperature region ($T_{TRSH}$) located between 34 °C and 40 °C, after which the plastic particles likely started to separate from the binder matrix. Therefore, in future studies, the relevance of experimental parameters for DSR testing should be carefully considered. Furthermore, other solutions such as using a higher

measurement gap and/or investigation scale (e.g., mastics) could represent an interesting challenge for the DSR approach;

- The PE modification increased the $T_{G^*=15kPa}$, and decreased the corresponding phase angle ($\delta_{G^*=15kPa}$). At the same time, the high temperature PG grading was increased. This parameter does not correlate with softening point temperature for the different tested blends.
- Dynamic viscosity test, using a rotating spindle, suggested that due to the high level of heterogeneity of PE modified asphalt binder, sample instability phenomena may occur;
- Compared to the conventional spindle, the helical spindle provides a more stable trend for viscosity measurements due to improved mixing of the sample. PE blends in bitumen suffer from storage stability with most of the PE floating on top. The use of storage stabilizers or a dry process in mix design are examples of possible solution for this problem;
- Alternative spindle like Dual Helical Ribbon could be proper solution for DSR measurements of bitumen modified with waste PE.
- The MSCR tests indicated that the neat binder is more sensitive to permanent deformation compared to the blends with PE shreds and pellets. A significant difference between the blends with shreds and pellets was not observed;
- The Linear Amplitude Sweep (LAS) Test was used to characterize the fatigue performance of the binder blends. The results showed a better performance in terms of stress and fatigue life for the PE blends;
- Differential scanning calorimetry clearly show distinct glass transition temperature and melting points coming from both neat bitumen and plastic, meaning two separate phases;
- The round-robin results show that the results from the labs diverge in the high temperature regime where the bitumen becomes less viscous and the effect of plastic additives more prominent, and the effect of heterogeneity more apparent;
- In general, considering all the experimental results, a significant difference between the blends with shreds and pellets was not seen implying that the secondary waste (shreds) can be used to achieve the same performance as the primary waste (Pellets).

## Acknowledgements

The Swiss company Innorecycling and Nynas of Sweden are gratefully acknowledged for providing the waste plastics and bitumen for this work. The contribution of the Swiss partners was supported by a grant from the Swiss National Science Foundation grant number 205121_178991.

## References

AASHTO TP70-10 (2013). *Multiple stress creep recovery (MSCR) test of asphalt binder using a dynamic shear rheometer (DSR)*. American Association of State and Highway Transportation Officials.

Alisov, A., Riccardi, C., Schrader, J., Cannone Falchetto, A., & Wistuba, M. P. (2020). A novel method to characterise asphalt binder at high temperature. *Road Materials and Pavement Design, 21(1), 143-155*.

Alisov, A., Riccardi, C., Schrader, J., Cannone Falchetto, A., & Wistuba, M. P. (2018). A novel method to characterise asphalt binder at high temperature. *Road Materials and Pavement Design*, 1-13. doi: 10.1080/14680629.2018.1483258

Austroads, Viability of using recycled plastics in asphalt and sprayed sealing applications, AP-T351-19, 2019, https://austroads.com.au/publications/pavement/ap-t351-19

Awaja, F., Pavel, D. Recycling of PET (2005) European Polymer Journal, 41 (7), pp. 1453-1477. DOI: 10.1016/j.eurpolymj.2005.02.005

Bahia, H., & Hanson, D. (2001). *Characterization of modified asphalt bitumens in Superpave mix design* (NCHRP Report 459). Washington, DC: National Academy Press.

Chen, H., Zhang, Y., & Bahia, H. U. (2020). Estimating asphalt binder fatigue at multiple temperatures using a simplified pseudo-strain energy analysis approach in the LAS test. *Construction and Building Materials*, *26(A), 120911.*

D'Angelo, J.A. (2009). The relationship of the MSCR test to rutting. *Road Materials and Pavement Design, 10, 61–80.*

DIN 52050. (2018). Bitumen und bitumenhaltige Bindemittel – *BTSV-Prüfung. Bitumen and bituminous binders – BTSV test*. German Standard. (In German)

EN 16659 (2016). *Bitumen and Bituminous Binders – Multiple Stress Creep and Recovery Test (MSCRT)*. European Committee for Standardization, Brussels.

[EU waste] European commission, Waste management hierarchy https://ec.europa.eu/environment/green-growth/waste-prevention-and-management/index_en.htm

Garcia-Morales, M., P. Partal, F.J. Navarro, F. Martinez-Boza and C. Gallegos, Linear viscoelasticity of recycled EVA-modified bitumens. Energy & Fuels, 2004. 18(2): p. 357-364. (8): p. 968-981.

Ho, S., R. Church, K. Klassen, B. Law, D. MacLeod and L. Zanzotto, Study of recycled polyethylene materials as asphalt modifiers. Canadian Journal of Civil Engineering, 2006.

Kakar, M R, Mikhailenko, P, Piao, Z, Bueno, M, Poulikakos, L D. Analysis of waste polyethylene (PE) and its by-products in asphalt binder. Construction and Building Materials 280 (2021) 122492. https://doi.org/10.1016/j.conbuildmat.2021.122492

Kalantar, Z.N., M.R. Karim and A. Mahrez, A review of using waste and virgin polymer in pavement. Construction and Building Materials, 2012. 33: p. 55-62.

Ledesma, R. and L.L. Isaacs, Effects of Polyethylene Terephthalate (Pet) from Plastics Waste on the Thermal-Properties of Asphalt. Abstracts of Papers of the American Chemical Society, 1991. 202: p. 53-Fuel.

Lo Presti, D. Recycled Tyre Rubber Modified Bitumens for road asphalt mixtures: A literature review (2013) Construction and Building Materials, 49, pp. 863-881. DOI: 10.1016/j.conbuildmat.2013.09.007

Masad, E., Huang, C., D'Angelo, J., & Little, D. (2009). Characterization of asphalt binder resistance to permanent deformation based on nonlinear viscoelastic analysis of multiple stress creep recovery (MSCR) test. Asphalt Paving Technology, 78, 535–562.

Padhan, R.K., et al., Poly(ethylene terephthalate) waste derived chemicals as an antistripping additive for bitumen - An environment friendly approach for disposal of environmentally hazardous material. Polymer Degradation and Stability, 2013. 98(12): p. 2592-2601.

Piao, Z., Mikhailenko, P., Kakar, M R, Bueno, M, Hellweg, S, Poulikakos, L D., Urban mining for asphalt pavements: A review, Journal of Cleaner Production, 2020, https://doi.org/10.1016/j.jclepro.2020.124916.

Poulikakos, L.D., Papadaskalopoulou, C., Hofko, B., Gschösser, F., Cannone Falchetto, A., Bueno, M., Arraigada, M., Sousa, J., Ruiz, R., Petit, C., Loizidou, M., Partl, M.N. Harvesting the unexplored potential of European waste materials for road construction (2017) Resources, Conservation and Recycling, 116, pp. 32-44. C DOI: 10.1016/j.resconrec.2016.09.008

Soenen, H, Blomberg, T, Pellinen, T., & Laukkanen, O-V (2013). The multiple stress creep-recovery test: a detailed analysis of repeatability and reproducibility. *Road Materials and Pavement Design, 14 (1), 2–11.*

Walther, A., Büchler, S., Cannone Falchetto, A., Wang, D., Riccardi, C., & Wistuba, M. P. (2019). Experimental investigation on asphalt mixtures prepared with reclaimed asphalt pavement and rejuvenators based on the BTSV method. *Road Materials and Pavement Design*, 20(7), 1695-1708. doi: 10.1080/14680629.2019.1594053

Wasage, T.L.J., Stastna, J., & Zanzotto, L. (2011). Rheological analysis of multi-stress creep recovery (MSCR) test. *International Journal of Pavement Engineering, 12, 561–568.*

Willis R., Yin F., Moraes R. , Recycled Plastics in Asphalt Part A: State of the Knowledge, NAPA - IS-142, 2020