Chapter 2

Bituminous binder and bituminous mixture modified with waste polyethylene

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Abstract. RILEM TC-279 WMR task group TG 1 studied the performance of waste Polyethylene (PE) in bituminous binders and bituminous mixtures. Several laboratories participated in this study following a common protocol. Locally sources aggregates and bituminous binder and same source of waste PE were utilized. The binder experiments showed that at high temperatures, using MSCR tests, PE modified blends had better resistance to permanent deformation in comparison to the non modified binder. Whereas at low temperatures, using the LAS tests, fatigue performance of the PE blends could withstand more loading cycles under low strains; however, it could sustain less loading cycles under high strains due to the increase in brittleness. Dry process was used for the mixture experiments in order to bypass the stability and inhomogeneity experience that was observed at the binder scale. The PE modified mixtures showed improved workability and increased strength. The higher the PE dosage, the higher the ITS increase with respect to the values measured for the control materials (i.e., without any plastic waste) thanks to the improved cohesion of the plastic modified mastic. The stiffness experiments tended to show an improved performance with a lower time dependence and a higher elasticity when plastic was added. The cyclic compression tests demonstrated a reduced creep rate along with a higher creep modulus thanks to the addition of PE; similar conclusions can be drawn from the experimental findings coming from wheel tracking test. Furthermore, acceptable and often improved moisture resistance was observed for PE modified materials.

Keywords: Bitumen; Asphalt, Bituminous mixture; Waste; Polyethylene

1 Introduction

Discovery of plastic materials in the beginning of the 20th century enabled acceleration in the development of the human civilization. In 1839, Charles Goodyear discovered the process of making plastics by chemically modifying natural polymers - the process of vulcanizing rubber - forming connections between polymer chains of rubber. In 1909, Leo Hendrik Baekeland created Bakelite. Fully synthetic plastic material made of phenol and formaldehyde. By modifying this process later polyvinyl chloride, polystyrene, polyethylenes, polypropylene, polyamide (nylon), polyester, acrylic, silicone and polyurethane were produced. In 1910, Sergei Lebedev synthesized artificial rubber, which led to the development of styrene-butadiene rubber during World War II. Since then, plastic materials are widely used all over the world. However, as plastic materials are cheap and widely used, in the last 50 years enormous production of cheap plastic materials has unfortunately led to large amounts of plastic waste.

Following the principles of a sustainable development, and a circular economy goods need to be recycled or reused at the end of their service life [1]. International research results have shown that many waste materials, including waste plastics, can be successfully used as alternative materials to the virgin materials in roads [2-4]. It was documented by Piao et al. [3], that the Technology Readiness Level (TRL) for the use of waste materials for road construction for many of the viable materials has remained, for the most part, at the research level or limited to use of some materials and countries. This indicates that in order to open up a broad market acceptance, such solutions must be developed and widely demonstrated. The main goal of the present work was to evaluate the use of such alternatives materials for use in pavements and as a result to develop a robust and fundamental understanding for their use, with the aim of delivering at least similar performances to conventional materials. To achieve this target, inter-laboratory tests were used to evaluate the performance of road materials that contained wastes, and standard processes for their selection, preparation, and usage were developed. Several types of waste plastics were considered at the beginning of the work of TG 1. However, it was decided to perform experimental work only with waste Polyethylene (PE) from one source, due to the fact that the softening point of PE is around 125°C which is lower than the temperature of production of hot bituminous mixtures (around 150°C). It was expected that PE would be easily and evenly mixed in the bituminous binder and in the bituminous mixture. In this work, two types of PE applications were performed: first, waste PE was mixed with bituminous binder and with waste PE mixed in bituminous mixture according to dry procedure.

2 Literature review

Various test methods were selected for the characterization of PE modified binders. The literature review below is focused on these tests namely Temperature frequency sweep (*T-f*-sweep), Multiple Stress Creep and Recovery (MSCR), Linear Amplitude Sweep (LAS) rely on Dynamic Shear Rheometer (DSR), Differential scanning calorimetry (DSC), and Fourier Transform Infrared Spectroscopy (FTIR). To characterize the PE modified mixtures workability, strength, stiffness and permanent deformation were investigated. The background of these testing methods will be explained below.

The Dynamic Shear Rheometer (DSR) [5] is frequently used to assess the viscoelastic characteristics of bituminous binders at high and intermediate temperatures. Recent studies also have indicated that the 4mm DSR can also be utilized for low temperature characterization [6,7]. In general, a combination of multiple experimental configurations, such as sample geometries, temperatures, frequencies, complicated shear/stress levels, and static/dynamic loading mode, were applied to assess the rheological properties of bituminous binders by using DSR [5,8]. The T-f-sweep test findings have been proven to be one of the most effective instruments for evaluating the rheological characteristics of bituminous binders and differentiating between non modified and modified binders over a wide range of temperatures and frequencies. The measured data, such as

$|G^*|$ and δ, can be further evaluated in accordance with the Time-Temperature Superposition Principle (TTSP) [9,10], such as by creating master curves and figuring out the associated rheological parameters. These include the crossover temperature [11,12], the Glover-Rowe parameter [13,14], the rheological index R [15], and T_c [16,17].

Nowadays, researchers employed DSR to assess how PE additions affected modified binders. Compared to the reference non modified binders, the binder modified with waste plastic shows generally improved rheological characteristics [18]. Enhanced viscoelastic characteristics, including higher $|G^*|$ and reduced δ [19-22], were discovered. However, high temperatures also revealed poor repeatability and reproducibility [23].

The Multiple Stress Creep and Recovery (MSCR) test, included in the Superpave Performance Graded (PG) specification, aims to characterize the rutting resistance of bituminous binders under application of different stress levels, and enables to assess both the non-recoverable creep compliance (J_{nr}) and the percent recovery (R) parameters [24]. J_{nr} can be calculated by the residual strain in a sample after one creep and recovery cycle divided by the stress applied (kPa^{-1}), while R (%) represents the elastic response and stress dependence of the bituminous binder. The plot of R related to J_{nr} can also identify the presence of a polymer and its crosslinking density as an alternative method for other tests [25].

Recently, several studies used the MSCR tests to evaluate the anti-rutting properties of binders modified with waste PE [26-30]. These investigations reported benefits regarding the elastic response with the addition of PE up to 6% by weight of bituminous binder, lowering the composite's unrecovered strain under high temperatures. At 58 °C and stress level of 3.2 kPa, for example, PE addition provided results of J_{nr} between 0.24 and 0.51 kPa^{-1}, while non-modified bituminous binder presented values between 1.02 and 1.75 kPa^{-1} [26,29]. This favorable condition is justified because of the recycled thermoplastics high ductility and strength properties, even when submitted to high service temperatures.

Additionally, the stress sensitivity of the binders modified with waste PE was analyzed based on the percent difference in non-recoverable compliances ($J_{nr,diff}$). Zhou et al. (2021) indicated that reduced stress sensitivity with the presence of PE in the bituminous binder. This trend was not observed by Nuñez et al. [29]. Nevertheless, all of the tested binders demonstrated reliable stress sensitivity in relation to the specification's maximum limiting value of 75% [31].

Joohari and Giustozzi [27] investigated specifically the effects of adding different types of PE to an SBS (styrene-butadiene-styrene) modified bituminous binder, e.g., low-density Polyethylene (LDPE), linear low-density Polyethylene (LLDPE), and a recycled linear low-density Polyethylene from waste plastics (RLLDPE). The study concluded that the addition of these thermoplastics can increase rutting susceptibility, based on the MSCR test parameters. More studies should be carefully performed in order to assess the impact of additional plastic types, with or without cross-linking agents' incorporation (e.g., sulfur) aiming to enhance the polymers stabilization.

Developed to assess the bituminous binders fatigue performance, the Linear Amplitude Sweep (LAS) test applies cyclic loads with increasing amplitudes to accelerate damage in order to assess the ability of bituminous binders to endure fatigue damage

[32]. Usually, this behavior is interpreted based on two analyses, the viscoelastic continuum damage (VECD) model with test results of both parameters A35 and B, and the damage tolerance parameter, αf. The material characteristics against accumulated damage are represented by the parameter A35, while the material sensitivity to changes of the applied shear loads is represented by the parameter B.

Research, that evaluated fatigue resistance of bituminous binder modified with PE, reported benefits when PE is incorporated into neat bituminous binder, indicating higher values of fatigue life (N_f) independent of the shear strain levels tested and the type of PE [26,27,28]. Overall, tested bituminous binder samples were submitted to short- and long-term aging, with the exception of Zhou et al. [30] and Joohari & Giustozzi [27], that used unaged bituminous binders. Bituminous binders modified with PE are mostly less sensitive and more resistant to fatigue cracking [30].

Differently, Nuñez et al. [29] observed that, for higher shear strain levels (higher than 7%), the binder modified with PE presented worse fatigue performance than the reference neat bituminous binder, for all tested temperature and aging conditions, respectively, at 25 and 35 °C, and both short ASTM D2872-04 [31], EN 12607-1 [32] and long-term aging, ASTM D6521-08 [33], EN 14769 [34]. Thus, further investigations regarding the fatigue behavior at high strain levels should be evaluated, since only two of these studies considered strain levels beyond 10%.

A thermo analytical technique named Differential Scanning Calorimetry (DSC) enables the identification of physical changes brought about by heating and cooling in a substance. As DSC may be used to measure a variety of characteristics and phenomena in the material, therefore, it has been employed extensively in the analysis of bituminous binder [35]. Identification of endothermic or exothermic activities is aided by the DSC analysis. Measurements are made of important characteristics including the glass transition temperature (T_g), and phase transitions like melting and crystallization. Additionally, it is possible to measure chemical processes and heat capacity [36, 37]. The findings might be used to determine the wax content, demonstrate the presence of wax in bituminous binders, and the melting point of the wax [38]. Bituminous material containing many different species, the determination of T_g is not straightforward and may require additional measurement features including modulated transition [39]. Using DSC technique in modified bituminous binder is a powerful tool to identify the morphological structure of the binder. In a perfect blend the glass transition will result in an intermediate T_g, while in a two-phase binder, the glass transitions of both compounds remain distinct.

In order to characterize chemical changes in the binders as a result of the blending with PE, Attenuated Total Reflectance Fourier Transform Infrared Spectroscopy (ATR-FTIR) was used. The ATR-FTIR method for bituminous binder allows to identify certain functional groups. With the FTIR technique, the molecules are excited by electromagnetic waves in the infrared range of 4000 to 400 cm^{-1}. Molecular functional groups will absorb specific electromagnetic radiation leading to vibration and increase in total energy. With bituminous binder the most interesting wavelengths are located between 1800 and 600 cm^{-1} to track structure and modifications. It is often used in research to characterize bituminous binders [40,41]. Especially to address modifications either occurring during aging through oxidation [42] or from blending with specific additives or modifiers [43]. With proper analysis of the spectra it is possible to define typical footprint of conventional bituminous binder [44], which later can help in identifying different modifiers through specify peaks in the spectrum.

Bituminous mixtures can be characterized by various laboratory methods as a means to predict their performance during their service life. The particular methods should be able to identify weaknesses in the material. For example, workability of the material can be assessed by analyzing the gyratory compaction curves [45]. The tensile strength [46] of the material can be determined in both dry and wet conditions and used to evaluate the strength and the moisture resistance, respectively. On the other hand, stiffness and linear visco-elastic characteristics can be studied through pulse indirect tension tests and cyclic tension-compression tests at intermediate service temperatures (25 °C to 35 °C). Finally, Cyclic Compression Tests (CCT) [47] on cylindrical specimens or Wheel Tracking Tests (WTT) [48] on slabs can be used to analyze the resistance of the investigated materials against permanent deformation at 60 °C.

3 Bituminous binder modified with waste PE

3.1 Preparation of modified bituminous binder

The materials used for this inter-laboratory study were straight-run bituminous binder 70/100 with a penetration of 82 mm^{-1} and softening point of 50 °C and two types of polyethylene waste (pellets and shreds). The pellets were produced from waste packaging and the shreds were a secondary waste as a result of the pellets production. The waste PE pellets and shreds were ground to smaller sizes of ca. 2 mm before being blended with bituminous binder. A single 5% content of PE additive was used to prepare the blended binders, for the blending procedure the PE waste was added to hot bituminous binder that was heated to 170°C for 1h and mixed with a speed of 3500 rpm using a high shear mixer for 1h. More details on the process of grinding and blending can be found at Kakar et al., [49]. Three types of blends were used in this work with the following designations: B (bituminous binder B 70/100); $B_{+pellets}$ (bituminous binder B 70/100 blended with PE-pellets), and $B_{+shreds}$ (bituminous binder B 70/100 blended with PE-shreds). The detailed composition of the investigated binders was displayed in Table 2.1.

Table 2.1 Detailed compositions of the investigated binders

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

3.2 Experimental plan

Inter laboratory tests were conducted by laboratories located all over the world. The penetration values and softening point temperature were conducted based on the EN 1426 [50] and EN 1427 [51], respectively. The storage stability was performed based on the EN 13399 [52]. The rheological characterization of the bituminous binder was conducted with the Dynamic Shear Rheometer (DSR) [53]. Temperature-frequency sweep (*T-f*-sweep) tests were used to assess the materials' viscoelastic behaviour over a wide range of temperatures and frequencies. More specifically, for complex binders, the DSR measurements return engineering parameters such as complex modulus, G^*, that provide more insight into the material response than conventional physical properties used for purchase specification. In this study, the *T-f*-sweep tests were run by eleven laboratories with two standard geometries: 8 mm diameter and 2 mm gap for intermediate temperatures ranging from -6 °C to 40 °C (-6, 0, 4, 10, 16, 22, 28, 33, and 40 °C), and 25 mm diameter 1 mm gap for high temperatures from 33 ° to 82 °C with an interval of 6 °C. The imposed frequencies ranged from 0.1 to 20 Hz. Among the eleven participating institutions, four laboratories tested bituminous binder *B*, five tested $B_{+pellets}$, while the entire interlaboratory team worked on $B_{+shreds}$. More information can be accessed in the authors' previous publications [23,54,55].

The MSCR test [56,57] is intended to assess the bituminous binder's sensitivity on permanent deformation. For this test, shear stress is applied using an oscillating load for a period of one second followed by an unloading (recovery) phase of nine seconds. The test starts with the application of a low stress 0.1 kPa for 10 creep/recovery cycles, follows with an increased stress of 3.2 kPa and repeated for an additional 10 cycles.

In this study, the tests were carried out using the 25 mm parallel plates configuration with 1 mm gap at 60 °C. The tests were performed on the neat binder (*B*), the blend with the PE shreds ($B_{+shreds}$) and the blend with PE pellets ($B_{+pellets}$) The tests were performed on freshly prepared samples in contrast to the standard specification request (after Rolling Thin Film Oven, RTFO). At least two replicates were tested per binder type. After 10 cycles using Equation 1, at each stress level the average percent recovery (*R*) is determined. Furthermore, the non-recoverable creep compliance (J_{nr}) is calculated 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 Equation 2.

$$R_\tau = \frac{1}{10}\sum_{N=1}^{10} \frac{(\varepsilon_1^N - \varepsilon_{10}^N)}{\varepsilon_1^N} \times 100\% \ (\%) \tag{1}$$

$$J_{nr}^N = \frac{\varepsilon_{10}^N}{\tau} (kPa^{-1}) \tag{2}$$

where,

τ the applied stress, 0.1kPa and 3.2kPa;

ε_1^N the strain value at the end of creep portion (after 1 s) of each cycle;

ε_{10}^N the strain value at the end of the recovery phase (after 10 s) of *N-th* cycle.

The LAS test [58] is used to assess the materials' resistance to fatigue damage by applying cyclic loading at amplitudes that increase until the damage occurs. Data were collected each second as the shear strain was linearly varied from 0 to 30%. The parallel-plate setup with an 8 mm plate diameter and 2 mm gap was used to record the binder samples at 10 Hz and 20 °C. At least two replicates were tested per binder type. The samples did not undergo any short- or long-term ageing before testing. The material's accumulated damage rate served as a proxy for the bituminous binder blends' fatigue behavior.

The information on the undamaged material characteristics (represented by the parameters α and β; see Equation 3) was determined based on the outcomes of frequency sweep tests [59] in order to complete the fatigue analysis. The following equation was used to apply a straight line best-fit to the data, with loading frequency on the horizontal axis and storage modulus on the vertical axis, in order to get the α and β parameters [60].

$$\log G'(f) = \frac{1}{\beta}\log(f) + \beta \tag{3}$$

where,

G' the storage modulus;

f the reduced frequency;

α, β fitting constants.

Damage intensity (D_t) in the bituminous binder sample was calculated using the following summation.

$$D_t \cong \sum_{i=1}^{t}\left[\pi\gamma^2\frac{(G'_{i-1} - G''_i)}{G'_{initial}}\right]^{\frac{\alpha}{1+\alpha}}(t_i - t_{i-1})^{\frac{1}{1+\alpha}} \tag{4}$$

where,

γ the shear strain;

G'' the loss modulus;

t the time.

Finally, the performance indicator for binder fatigue (N_f) can be expressed according to the following equation.

$$N_f = A(\gamma)^B \tag{5}$$

where,

γ the shear strain;

A, B the viscoelastic continuum damage (VECD) model coefficients that depend on the material properties.

To be specific, A is defined using damage accumulation corresponding to a 35% decrease from the initial loss shear modulus, and B is determined by the linear viscoelastic properties of binder. In general, more fatigue resistant binders tend to have higher A values and lower absolute B values.

FTIR was performed by two laboratories lab7 and lab8, but only one run the test on the base bituminous binder 70/100 and the two blends with plastic shreds and pellets. It was conducted in Attenuated Total Reflectance (ATR) between 600 and 4000 cm^{-1} with 64 scans and a resolution of $4cm^{-1}$. In this present study, FTIR spectroscopy was used to track how plastic pellets and plastic shreds affected the reference binder's chemical composition. The examination of B, $B_{+pellets}$ and $B_{+shreds}$ was conducted by using the Bruker Vector 22 / Digilab BioRad FTS 6000 FTIR spectrometer.

DSC was performed by one lab Lab7 on linear amplitude heat flow. First the sample was heated up at 165 °C and stabilized for 5min to remove any heating history. The sample was then chilled at a rate of 2 °C/minute until it reached -60 °C, kept for 5 minutes and then reheated at the same rate to 165 °C. The thermal properties of B, $B_{+pellets}$ and $B_{+shreds}$ were analyzed by using DSC. The followed testing protocol was followed: the samples were first cooled from 25°C to 20°C and held at this temperature for 5 minutes. Then, they were heated to 200°C and held for 5 minutes before being cooled once more to 20°C and held at that temperature for 5 minutes before being heated to 200 °C. A single cooling and heating rate of 20 °C/min was applied for such processes. For the sake of reproducibility, each sample was tested at least three times.

4 Results and Analysis on bituminous binder tests

4.1 Conventional tests results

The results of penetration values seem similar among different laboratories, a reduction of 50% was observed when the PE additives were blended. However, this is not true for the softening point temperature, consistent results were obtained among non modified binders, while the results measured in the binders modified with PE were quite different. In $B_{+shreds}$, the softening point temperature results ranged from 45 °C to 110 °C which led to a maximum difference of more than 60 °C [54]. This may be attributed to the inhomogeneous distribution of plastic particles in the modified binders [54,55]. More detailed information and discussion about the storage stability can be accessed in the authors' previous publication [49,54].

4.2 Interlaboratory comparison of DSR *T-f-sweep* tests bituminous binder

The results of the DSR measurements conducted on the binder modified with PE are analyzed and discussed in this section. To assess the repeatability within labs and reproducibility across laboratories, the isochronal curves of the complex shear modulus, $|G^*|$, and the phase angle, δ, at the reference frequency of 1.59 Hz (10 rad/s), were first constructed. All three binder types indicate quite a high repeatability in the results, while the binders modified with PE had low reproducibility. This shows that the mixes are not as homogeneous as bituminous binders made for ordinary paving.

Figure 2.1 makes it clear that the testing temperature had a considerable and direct impact on the outcomes. Reduced variability was observed for all the investigated materials at relatively low testing temperatures (PP08). This temperature effect was less pronounced for the binders modified with PE ($B_{+pellets}$ and $B_{+shreds}$). Within this low-temperature range, these blends exhibited a complex shear modulus that was comparable to that of the reference unmodified binder. In addition, all laboratories were able to detect an increase in the complex shear modulus relative to the neat binder in the high temperature regime. A more elastic phase angle behavior was seen in the low temperature domain. The inclusion of plastics, on the other hand, caused the phase angle to decrease in the high temperature regime as compared to the neat binder, indicating a more elastic response. Interestingly, the differences between the findings from the various laboratories were larger at the high temperature range. Due to the weaker binder and lower viscosity at the higher testing temperatures, these findings may indicate that the plastic particles are really being assessed instead of the entire mix, exposing the sample's heterogeneity and resulting variances in results (depending on the amount of plastic particles in the binder samples). A larger diameter and smaller gap of the samples utilized for the high temperature study (PP25) might be another element enhancing such an impact. Regardless of the testing temperature, reasonable sample stability was seen when binder B, $B_{+pellets}$, and $B_{+shreds}$ mixes were tested.

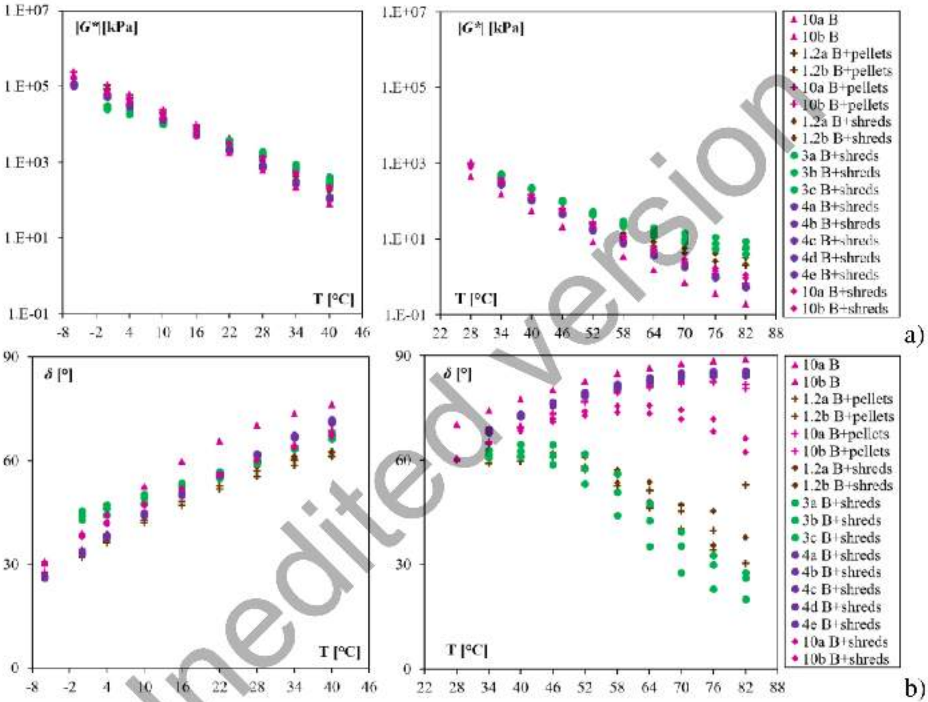


Fig. 2.1 Isochronal plots of binders modified with PE $B_{+pellets}$ and $B_{+shreds}$ at the reference frequency of 1.59 Hz (10 rad/s): a) $|G^*|$ at low (left) and high (right) testing temperature; b) δ at low (left) and high (right) testing temperature [54].

The analysis of the raw DSR data for all frequencies at the different temperatures was made using the black space with complex shear modulus, $|G^*|$, and phase angle, δ. This enables to display all data points for all frequencies and temperatures without any shift of the values.

0shows the plot for the neat binder 70/100 as reported by the three laboratories in frequency sweep at different temperatures including two of them having performed also temperature ramping. Lab2 showed some discontinuity with the 25mm geometry presuming morphology disruption during the test with no time temperature superposition.

The other two labs gave reasonably good reproducibility either in frequency sweep or temperature ramping.

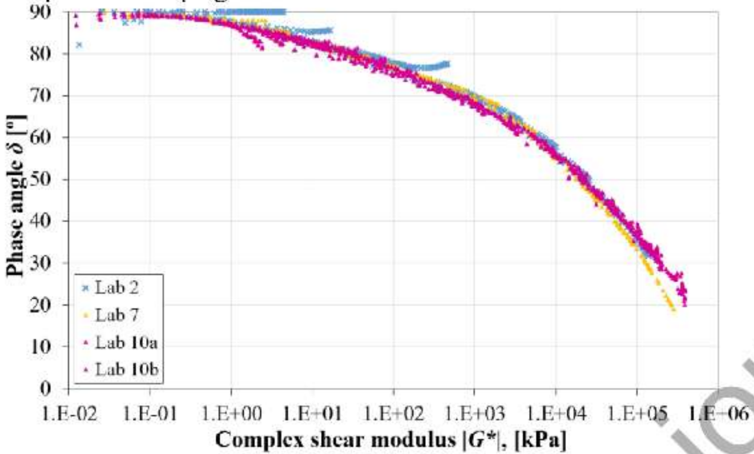


Fig. 2.2 Black diagram showing phase angle vs complex shear modulus for the neat bituminous binder 70/100 [54].

For shreds blends, eleven laboratories performed frequency sweep measurement. The analysis was divided in two parts first the 8mm geometry representing the cold temperature regime and second the 25mm geometry representing the warm temperature regime.

With the pellet blends the same observation was made with poor reproducibility as shown in Figure 2.3. Fig. 2.4a reports the measurement with the 8mm geometry made at intermediate/low temperatures. For Lab3, outlier data points were observed at high temperatures. With the 8mm geometry, the curves follow the same smooth trend despite variability of the results, especially going forward with low shear modulus corresponding to higher temperature measurement. As compared to the neat bituminous binder, the curves were in the same range of magnitude. Fig. 2.4b reports the measurement for the 25 mm made at high temperature regime above 33 °C. all curves displayed high scatter and no trends between laboratories can be identified. There was no smooth overlapping between temperatures, meaning the time temperature superposition can't be applied in this case.

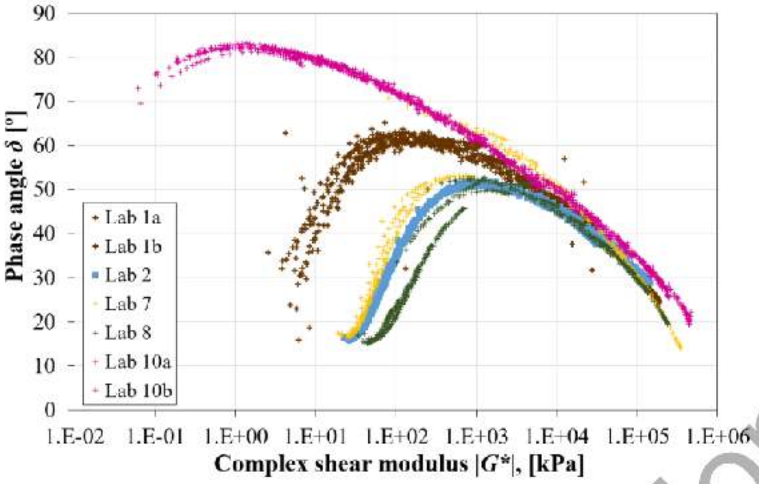
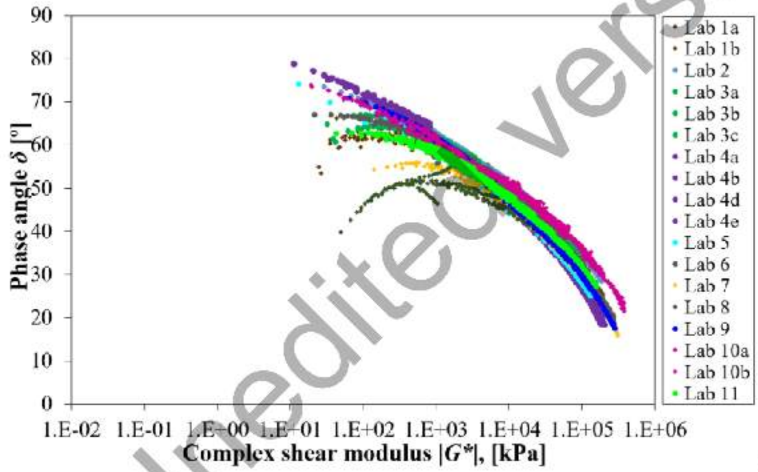


Fig. 2.3 Black space for $B_{+pellets}$



a)

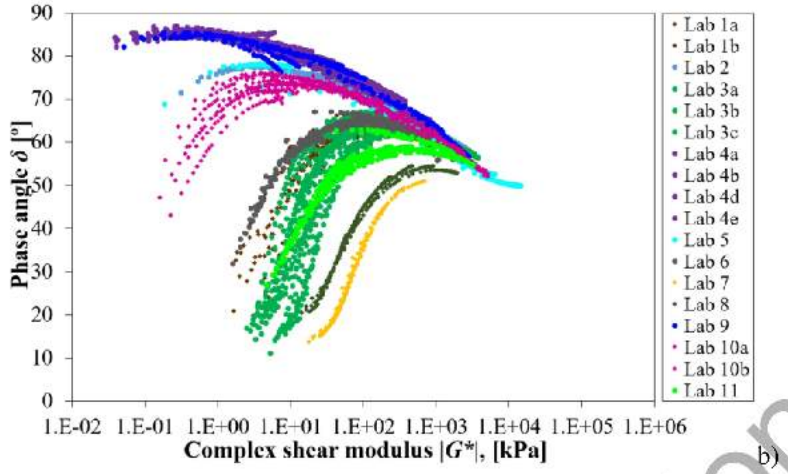


Fig. 2.4 Black diagram for $B_{+shreds}$ measured with: a) 8mm geometry; b) 25mm geometry [54].

4.3 MSCR and LAS tests bituminous binder

Fig. 2.5 shows the MSCR results at 60 °C for both stress levels for binders B, $B_{+pellets}$, and $B_{+shreds}$. It can be observed that as the stress level increases from 0.1 kPa to 3.2 kPa, the R decreases and the J_{nr} increases. Additionally, at both stress levels, the neat binder had the greatest J_{nr} and lowest R values. These findings indicate that, in comparison to mixes with PE shreds and pellets, the clean bituminous binder is more susceptible to persistent deformation. Additionally, because of its greater R and lower J_{nr} values, $B_{+shreds}$ routinely outperform $B_{+pellets}$ in terms of deformation recovery and permanent deformation sensitivity.

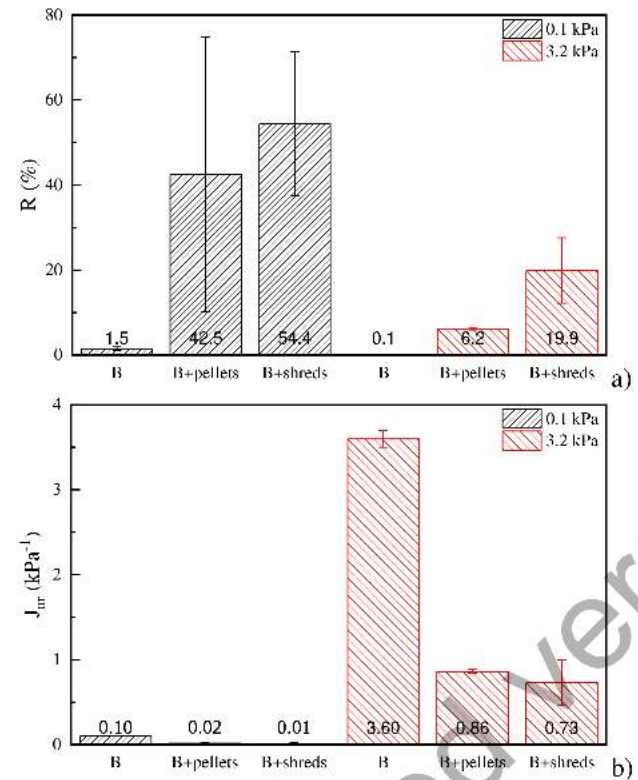


Fig. 2.5 a) Strain recovery, and b) non-recoverable creep compliance at two stress levels.

Fig. 2.6 presents the fatigue results of the neat binder, *B*, the blend with the PE-pellets and PE-shreds, $B_{+pellets}$ and $B_{+shreds}$, at 20 °C. At the low strain level (lower than 2%), the results show that the neat binder has the lowest N_f values compared to the $B_{+shreds}$ and $B_{+pellets}$. It expresses that the binder containing PE shows lower fatigue life than virgin binder. On the other hand, the fatigue life of plastic modified binders ($B_{+shreds}$ and $B_{+pellets}$) was shorter than that of neat binder at high strain level (higher than 4%). These findings indicate that as bituminous binder becomes stiffer and brittle because of plastic modification, it can undertake more loading cycles under low strain levels; on the contrary, it can sustain less loading cycles under high strain levels due to the increase in brittleness. In addition, the $B_{+shreds}$ shows consistently better fatigue resistance $B_{+pellets}$ at any strain level.

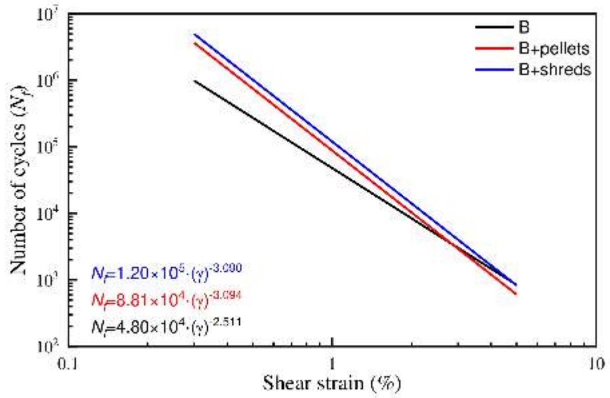


Fig. 2.6 Fatigue performances of three studied binders

The damage characteristics (C-Damage Intensity) curves of the tested binders are compared in Fig. 2.7. C denotes the ratio of the loss shear modulus to the starting value at any given time. By comparing the damage curves, it can be observed that the PEN 70/100 neat binder has the least damage compared with $B_{+shreds}$ and $B_{+pellets}$. When comparing the modified binders, it can be seen that $B_{+pellets}$ display somewhat less damage than $B_{+shreds}$.

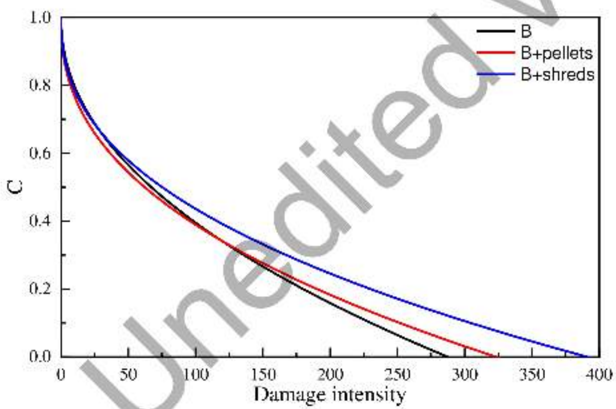


Fig. 2.7 C-Damage Intensity curves of three studied binders

4.4 FTIR and DSC tests

Fig. 2.8 displays the whole FTIR spectrum between 4000 to 600 cm^{-1} for the base bituminous binder 70/100, the blend with plastic shreds and the blend with pellets. Fig. 2.9 provides a close up of most specific wavelength numbers for bituminous binder. From that spectrum it was not possible to identify any differences and as compared to the base bituminous binder 70/100 no additional peaks were detectable in the blends with plastics. This may have been an artifact of the measurement. The plastic having not been fully dispersed homogeneously in the bituminous binder matrix, there was only 5% of probability of having some plastic exactly under the diamond. Would have been the blend homogeneous continuum phase of plastic and bituminous binder, additional peaks should have been identified as the footprint of the plastic. An attend of FTIR on the plastic itself was also made but was not successful due to the difficulty to apply the sample straight on the diamond ensuring proper contact for the measurement.

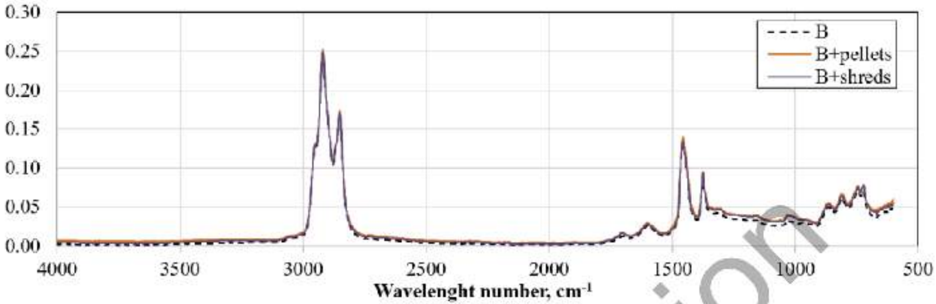


Fig. 2.8 FTIR spectrometry of 70/100 bituminous binder, blenda with shreds and pellets

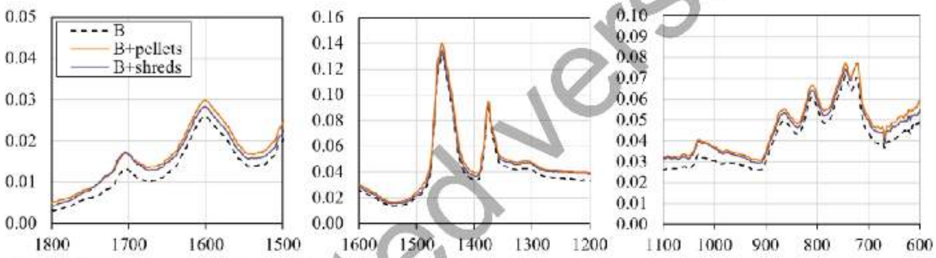


Fig. 2.9 Close up FTIR spectrometry

Differential scanning calorimetry was performed first on the plastic shreds and pellets even on several shreds samples differentiated by color. Fig. 2.10 displays the heat flow during the heating phase from -60 °C and +165 °C. A single melting point was identified around 124 and 130 °C more pronounced for the pellets. No other outside the temperature range of the measurement.

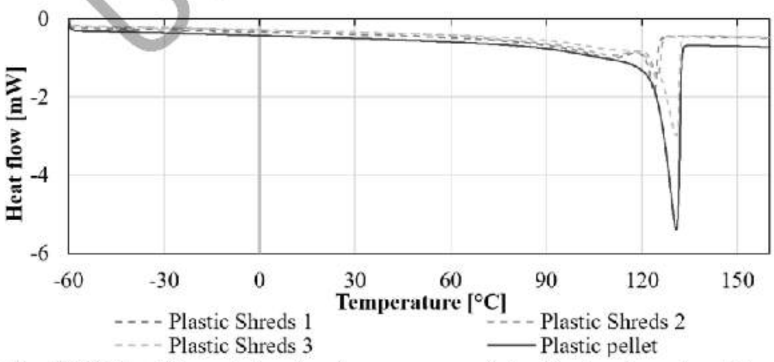


Fig. 2.10 Heat flow during heating process of plastic shreds and pellets

Then DSC was performed on the base bituminous binder 70/100 and the two blends with shreds and pellets plastics. Fig. 2.11 shows the heat flow during the heating phase for the three binders. The glass transition that can be observed for the neat bituminous binder around -25 °C is also visible with the shreds and pellets blends. However, the latter clearly distinguishes the melting point of the plastic around 120 °C. Should have been the blend homogeneous with continuum phase morphology, the resulting glass transition would have been in between the base bituminous binder and the plastic. Both components maintained their specific calorimetric behaviour without any interaction, meaning the material has a two-phase morphology.

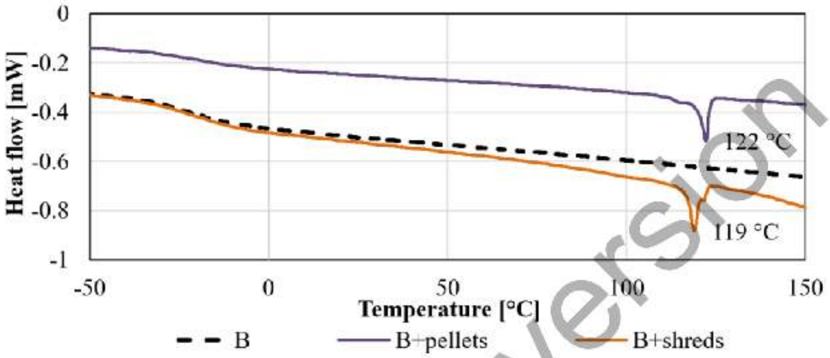


Fig. 2.11 Heat flow during heating process of 70/100 and plastic blends

The storage stability of the binder blends was assessed by one lab using the binder stability test [52]. These reveal that both PE blended binders show a significant difference in viscosity of the top and bottom samples and therefore, have a storage stability problem [49]. Softening point results not reliable, DSC can be a better tool to differentiate them.

5 Bituminous mixture modified with waste PE

Given the promising results as well as the challenging issues observed at binder level and described in detail above, an interlaboratory exercise on bituminous mixtures modified with waste PE was carried out. This research activity was carried out by a selection of eleven laboratories from nine countries already involved in TG1 and/or TG3 (i.e., waste aggregates in bituminous mixtures) activities. This experimental study was aimed at determining if performance enhancement can be also observed at mixture level while limiting/hindering the abovementioned issues concerning stability and homogeneity.

To accomplish this objective, the selected laboratories were asked to prepare and characterize a dense graded bituminous mixture with 16 mm maximum aggregate size and containing different amounts of waste PE. In particular, each laboratory used its own aggregates and bituminous binder whereas waste PE was the same shreds used in the binder modification and was sampled and provided by a single source. Fig. 2.12 shows particle size distribution of waste PE used by the different labs.

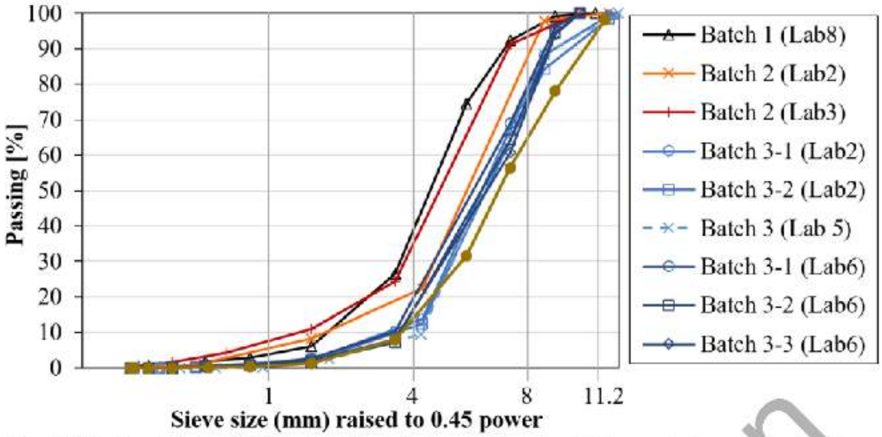


Fig. 2.12 Gradation of PE waste batches used by the different laboratories [60]

A common target mixture gradation (Fig. 2.13) as well as aggregate and bituminous binder type (ie., limestone aggregates and 50/70 penetration grade bituminous binder, respectively) were suggested in order to minimize variations among laboratories. Only two laboratories used aggregates of different origin (i.e., granite for Lab2 and natural river sand for Lab6) while Lab 8 used a 70/100 pen grade bituminous binder.

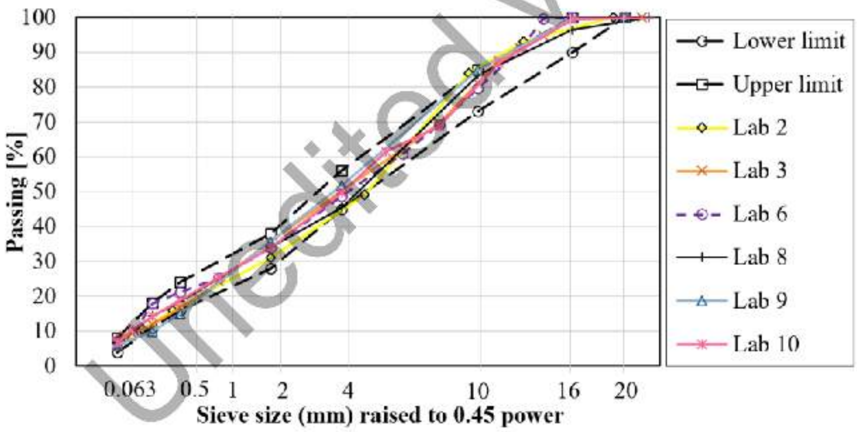


Fig. 2.13 Aggregate mix gradation adopted by participating labs [60]

A binder content between 4% and 6% by aggregate weight was used in order to maximize mix properties and obtain a void content of 4-6% when compacted with impact (i.e., Marshall) compactor or using a gyratory compactor. As far as the content of waste plastic concerns, all the laboratories investigated 1.5% plastic dosage by mix weight along with the reference materials (i.e., the mixture prepared without any plastic addition). Moreover, 0.25 (corresponding to 5.0% by bituminous binder weight as established for wet modification of binder discussed in the previous sections), 0.5, 1.0 and 5.0% waste PE were also used by some selected laboratories as detailed in Table 2.2.

Table 2.2 Bituminous mixture properties

PE %	lab1	lab2	lab3	lab4	lab5	lab6	lab7	lab8	lab9	lab10	lab11
	Void content (vol. % in asphalt sample)										
0	5.30	3.60	3.54	5.60	4.30	5.54	6.59	4.50	6.00	3.70	5.54
0.25	5.10	4.00	3.48					4.50	5.80		
0.5											5.14
1.0											5.05
1.5	4.80	5.00	5.93	4.40	4.50	4.59	3.13	4.50	4.70	5.70	
5					5.47				2.70		
	Bituminous binder content (w. % by weight of stone aggregate)										
0	4.00	4.70	4.10	5.00	5.00	4.71	5.4	5.00	5.30	4.88	5.26
0.25	4.00	4.70	4.10					5.00	5.30		
0.5											5.29
1.0											5.32
1.5	4.00	4.70	4.10	5.00	5.00	4.71	5.4	5.00	5.30	4.90	
5					5.00				5.30		

The mixtures were prepared in the laboratory using a protocol which can be considered as a dry process since the plastic at ambient temperature was added to the hot aggregates (160 °C) and premixed for few minutes (1 to 7 minutes depending on the batch size) prior to the addition of the hot bituminous binder. This allowed to achieve a homogeneous mix without a complete melting of the plastic particles. Cylindrical or slab specimens were then compacted at 155 °C using Marshall, gyratory or roller compactor compliant with EN standards.

The reference and PE modified bituminous mixes were characterized in terms of workability, tensile strength, stiffness, permanent deformation as well as moisture resistance. Workability was assessed by analyzing the gyratory compaction curves while the indirect tensile strength test carried out in both dry and wet conditions was used to evaluate the strength and the moisture resistance, respectively. On the other hand, stiffness and linear visco-elastic characteristics were studied through pulse indirect tension tests and cyclic tension-compression tests at intermediate service temperatures (25-35 °C). Finally, cyclic compression tests (CCT) on cylindrical specimens or wheel tracking tests (WTT) on slabs were used to analyze the resistance of the investigated materials against permanent deformation at 60 °C. More details about materials, specimen preparation and testing methods are reported elsewhere [61].

The experimental findings achieved at multi-laboratory level mainly showed that the presence of waste PE did not negatively affect mix workability. In particular, lower construction densification index (CDI) and compactability parameter K were generally found for plastic mixtures as shown in the following Fig. 2.14 and 2.15. The error bars indicate the standard deviation of the results in Figure 2.14 and the following plots.

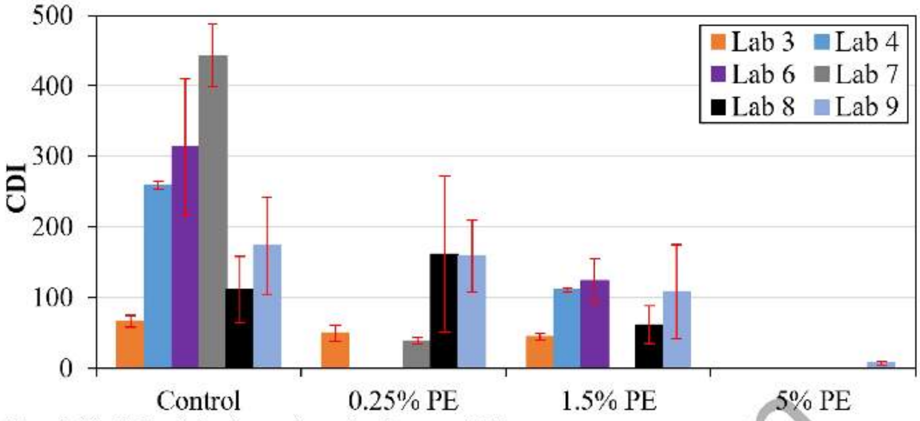


Fig. 2.14 CDI of the investigated mixtures [61]

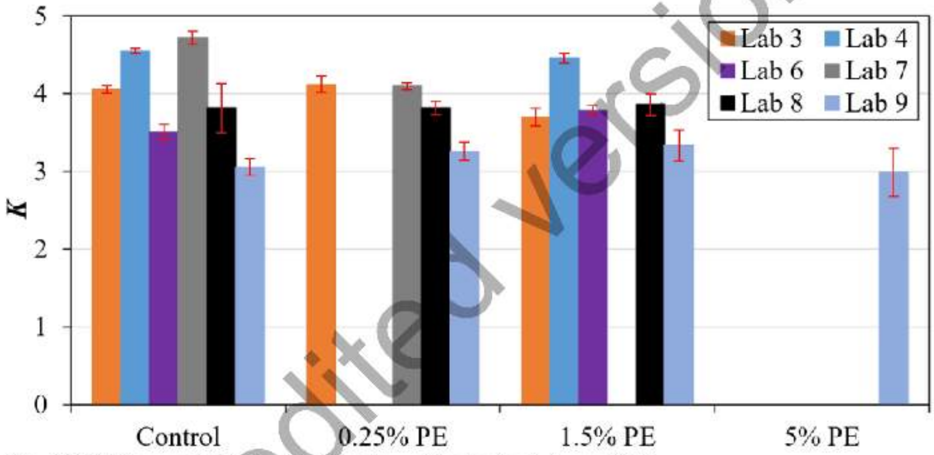


Fig. 2.15 Compactability *K* of the investigated mixtures [61]

Such experimental findings suggest that the investigated amounts of waste plastics would not lead to workability issues thus allowing a proper laying of such materials. Furthermore, enhanced elasticity, stiffness and permanent deformation resistance were also observed by the different laboratories.

In this sense, Fig. 2.16 depicts the higher strength achieved thanks to the inclusion of waste PE; overall, the higher the PE dosage the higher the ITS increase with respect to the values measured for the control materials (i.e., without any plastic waste) likely thanks to the improved cohesion of the plastic modified bituminous mastic. It is worth noting that average absolute ITS values varied between 1.09 and 1.58 MPa.

The dynamic tests at intermediate temperatures mostly confirmed the performance enhancement achievable with the inclusion of the selected waste PE shreds into the bituminous mixtures since an increase in stiffness, a lower time dependence and a higher elasticity were often measured when plastic was added (Figs. 2.17 and 2.18). Furthermore, a higher variability among the different labs was noticed likely due to a higher sensitivity of such properties to the air void content of the tested specimens.

The enhanced stiffness and elasticity of the plastic mixes reflected in a clear improvement of their permanent deformation resistance. In this regard, CCT tests demonstrated a reduced creep rate along with a higher creep modulus thanks to the addition of PE shreds as shown in Fig. 2.19; similar conclusions can be drawn from the experimental findings coming from WTT (Fig. 2.20).

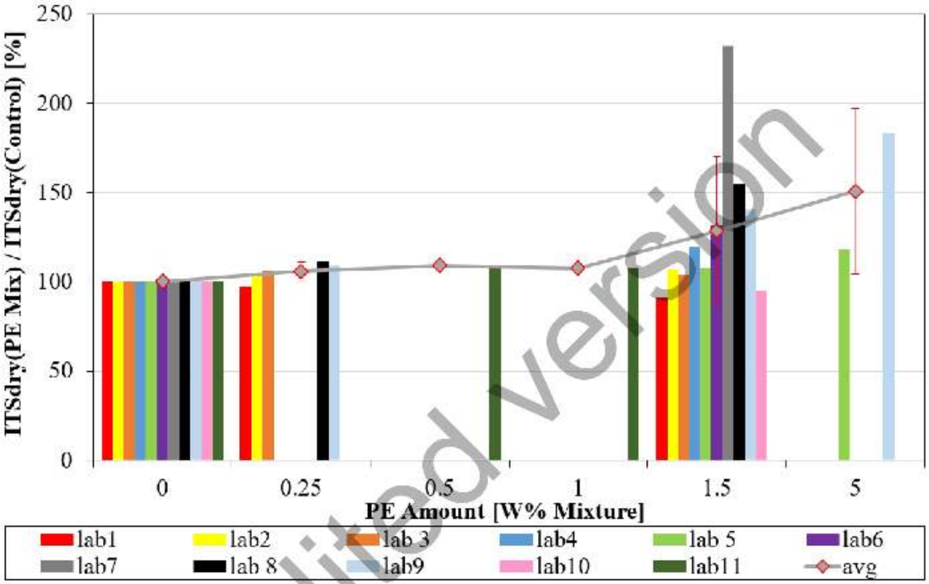


Fig. 2.16 ITS increase as a function of waste PE content [60].

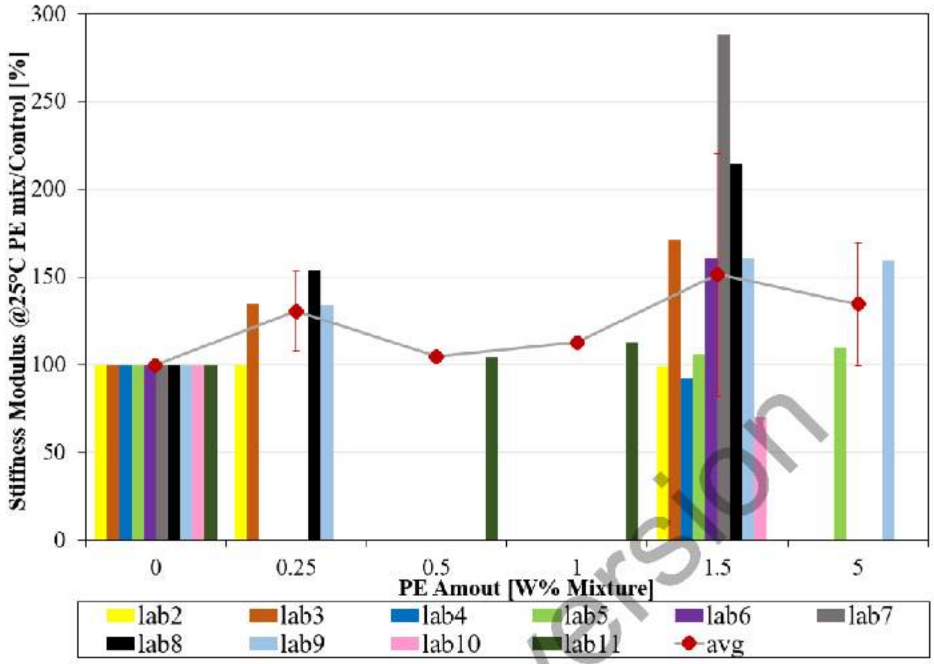


Fig. 2.17 Stiffness characteristics as a function of waste PE content [60].

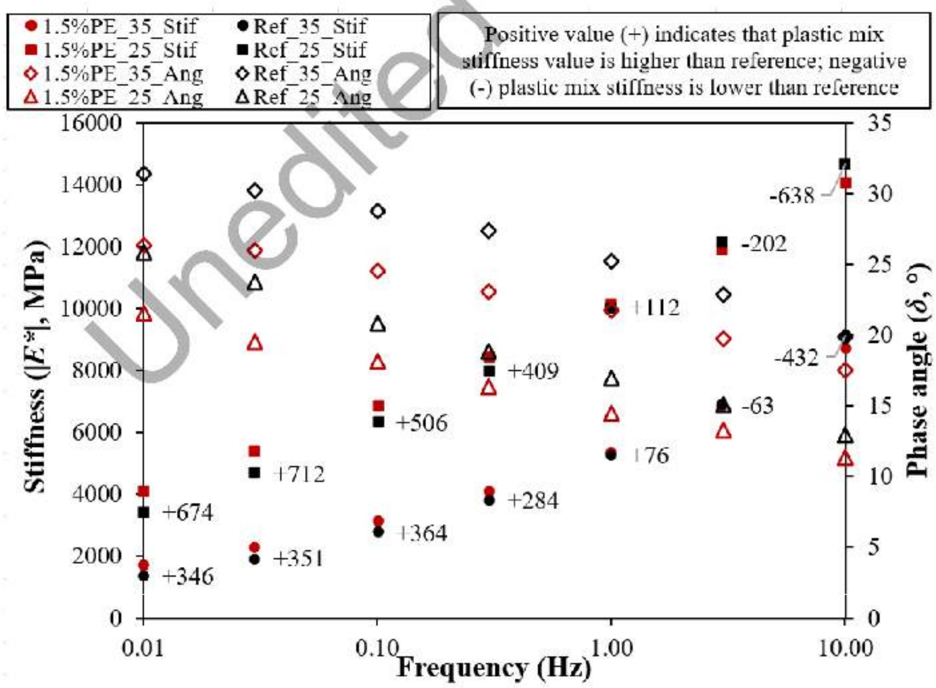


Fig. 2.18 Linear visco-elastic properties as a function of waste PE content [60].

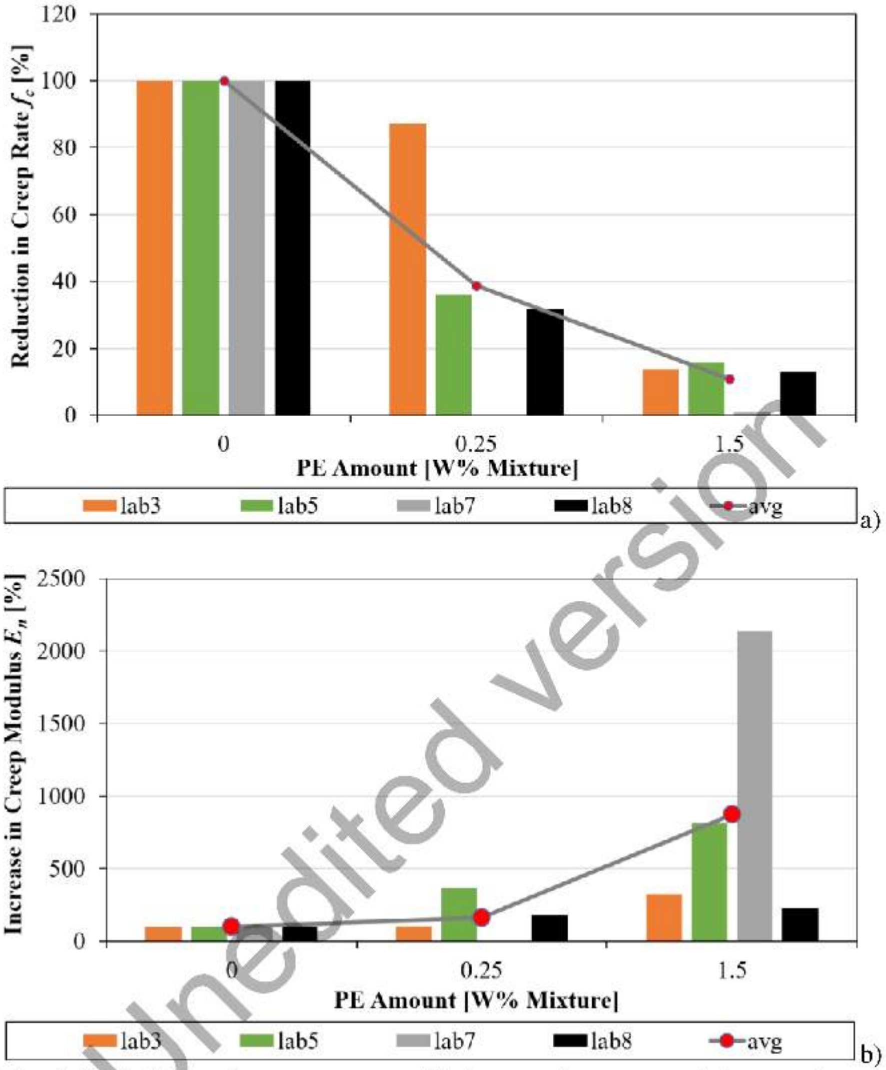


Fig. 2.19 a) Change in creep rate, and b) increase in creep modulus as a function of waste PE content [61]

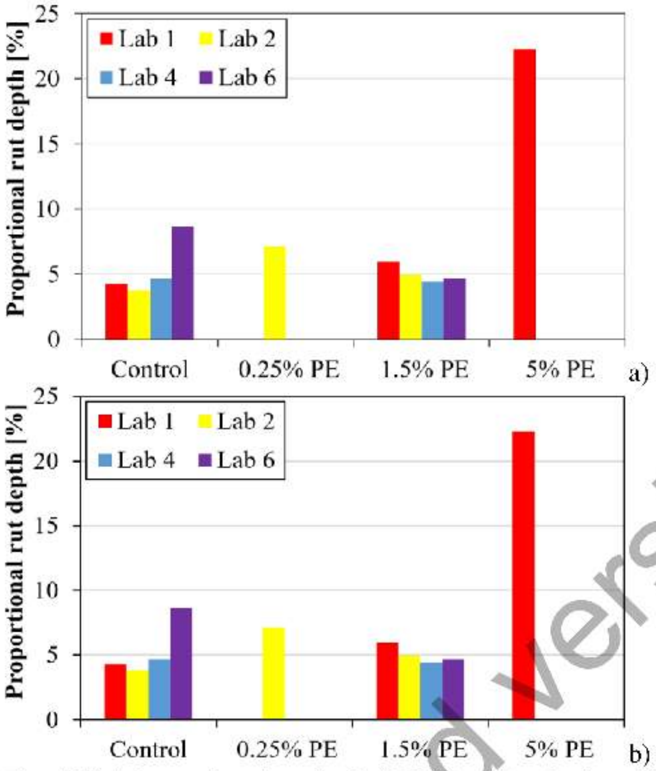


Fig. 2.20 a) Proportional rut depth (PRD), and b) wheel tracking slope (WTS) as a function of waste PE content [61]

Finally, acceptable (often even improved) moisture resistance was also observed for PE modified materials. Indeed, the percentage ITS ratio measured in wet and dry conditions (ITSR) is reported in Fig. 2.21 as a function of the plastic content. In Fig. 2.21, it is worth noting that ITSR absolute values higher than 80% (i.e., value accepted by the most part of highway agencies worldwide) were measured by all the involved laboratories, regardless of the presence of PE waste plastic. The only exception was lab2 which tested bituminous mixtures prepared with water sensitive granite aggregates.

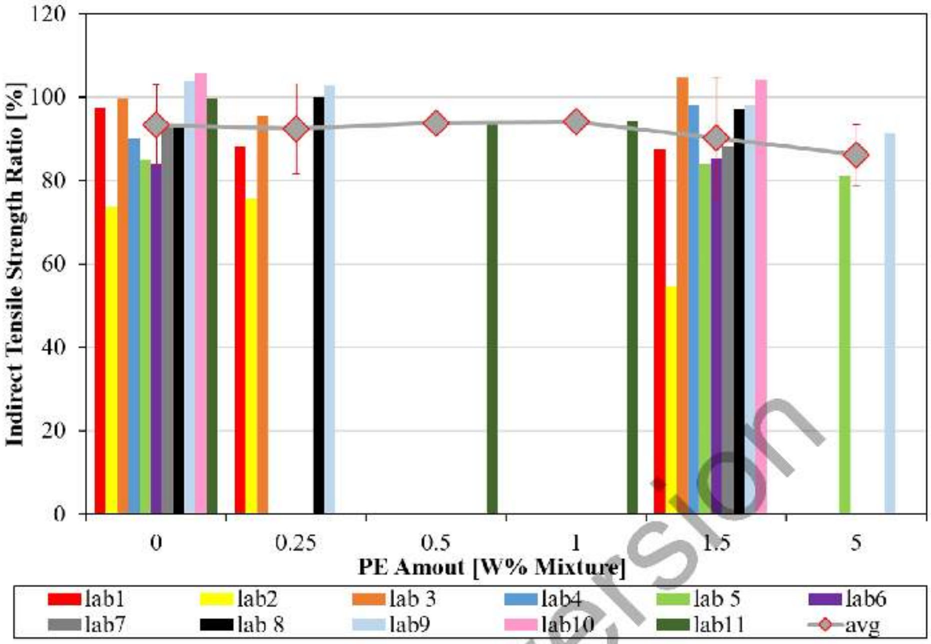


Fig. 2.21 ITSR as a function of waste PE content [60]

6 Summary and Conclusions

The research work performed by TG1 was presented and summarized in this second chapter of the State-of-the-Art Report of the RILEM TC 279-WMR. This investigation was devoted to the combination of waste Polyethylene (PE) plastic in bituminous binder and asphalt mixture due to the characteristics of these materials. Two forms of PE were evaluated: pellets and shreds obtained from packaging and its recycling in the form of pellets and the secondary waste produced as a result, respectively. The research conducted on bituminous binder modified with PE consisted of a comprehensive experimental program supported by an advanced data analysis. The program was performed on plain binder, binders modified with PE shreds and PE pellets, and included conventional characterization, rheological testing with the Dynamic Shear Rheometer (DSR), Differential Scanning Calorimetry (DSC), and Fourier Transform Infrared Spectroscopy (FTIR) for chemical analysis. PE-modified bituminous mixtures were also prepared to expand the understanding of the effect of PE on paving mixtures. The bituminous mixture was designed according to the dry process, and a series of mechanical tests were conducted to determine stiffness, strength, strength ratio, and resistance to permanent deformations. Based on the experimentation and analysis performed, the following conclusions can be drawn:

- At high temperatures, using the Multiple Stress Creep Recovery (MSCR) test, the binder blends modified with PE were less sensitive to permanent deformation compared to the non modified binder.
- At intermediate temperatures using the Linear Amplitude Sweep (LAS) tests, the fatigue performance of the PE blends could withstand more loading cycles under low strain levels; however, it could sustain less loading cycles under high strain levels due to the increase in brittleness.
- The addition of waste PE did not significantly affect the workability of asphalt mixtures.
- The higher the PE dosage, the higher the Indirect Tensile Strength (ITS) increase with respect to the values measured for the control materials (i.e., without any plastic waste) thanks to the improved cohesion of the plastic modified mastic.
- The stiffness experiments showed an improved performance with a lower time dependence and a higher elasticity when plastic was added.
- The cyclic compression tests demonstrated a reduced creep rate along with a higher creep modulus thanks to the addition of PE shreds; similar conclusions can be drawn from the experimental findings from the wheel tracking test.
- Acceptable and often improved moisture resistance was observed for PE modified mixtures.

7 Perspective and outlook

The activities of TG1 of TC-297 WMR summarized in this chapter demonstrate that the use of PE in bituminous mixtures can be a viable option. However, appropriate test methods need to be used to evaluate the performance of the material. As binder modified with PE specimens can have the disadvantage of segregation and inhomogeneity, mixture performance should be preferably used as the deciding procedure. A balanced mix design that considers the low temperature and high temperature performance should be considered. In addition, environmental issues should be part of the evaluation procedure such as, for example, leaching risks. In order to determine the viability of using waste materials throughout their life cycle, a complete life cycle assessment preferably a cradle to grave procedure should be considered. In such analysis the waste management procedure in different geographical areas should be considered as using waste in bituminous mixture can take this material away from another industry (e.g., cement industry where plastic waste is used as fuel [62]) and this would have consequences for that industry that could not be necessarily sustainable.

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