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Modification of bitumen with PE waste plastic

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Abstract

Various kinds of plastics contribute to a considerable portion of global waste, on both land and in the oceans. This waste may be better managed if more uses are introduced for these plastics or their derivatives. The purpose of this study is to evaluate the use of waste plastics as bitumen modifiers. Waste plastics in the form of PE-pellet and PE-shredded were added by 5% mass of bitumen and the stability of the plastics in the bitumen was evaluated using stability test. The rheological properties of the waste plastic modified bitumen were evaluated by a Dynamic Shear Rheometer (DSR) in order to determine if the modification can contribute to desirable properties. Finally, Scanning Electron Microscopy (SEM) was used to determine the microstructural properties and how well the plastic blended with the bitumen. The results show that modification of 5% (by mass of bitumen) improved the high temperature resistance to deformation, which is an indicator of better rutting resistance. However, the thermal stability test results showed that both the PE-pellet and PE-shredded waste plastic are susceptible to phase separation.

1. INTRODUCTION

Roads are the primary means of transportation worldwide with more than 34 million kilometres around the globe and hence play a very important role in the overall economy and social development [1]. A large amount of resources that includes materials, energy, and manpower are invested not only for the construction of new pavements, but also for the maintenance and restoration of the existing ones [2].

The performance enhancement of asphalt pavements by the modification of the bitumen involve various kind of polymers types, such as block styrene-butadiene-styrene copolymers (SBS) [3], styrene butadiene rubber (SBR), and ethylene-vinyl acetate copolymers (EVA) [4] [5]. From an environmental and economical prospective, polymer additives derived from recycled solid wastes have drawn more attention of researcher due its combined advantage of both cost effectiveness and as a solution for waste management problems [6]. At a European level, out of the 50 million tons the plastic waste production in 2016, around 27 million tons of plastic were collected, of which 30% were recycled, whereas the other 70% were incinerated to recover energy or sent to a landfill [7]. Aiming to improve these numbers, most European regulations and directives encourage the reuse, recycling and recovery of these wastes, with disposal as the last option.

The idea of the circular economy refers to the use of more secondary raw materials and the approach of reducing waste generation by retaining and repairing products that are in use [8]. A review by the authors has demonstrated how a considerable amount of waste produced in the urban and suburb environment can be recycled in asphalt roads. Various type of waste materials such as glass, wood, concrete, asphalt, tires or plastics technically have a potential for re-use in asphalt roads. Considering the available amounts of the European target waste materials, that would otherwise be incinerated or disposed of in landfills.

Given the prevalence of asphalt, a significant amount of waste produced in the urban and suburban environment can be potentially recycled in asphalt roads. Nevertheless, although technically feasible, the waste materials should be selected for its use in the roads only if this superior use for them. As an example, the amount of waste concrete from construction will grow and continue to be available in large quantities in the near future [9] [10], and the efficient use of such materials may indeed be road construction.

It was revealed that there is high possibility in Europe for recycling in road construction, in particular, under the hypothetical scenario where 33% of new roads would be made of the target waste materials (excluding reclaimed asphalt pavement RAP which is already recycled). It is estimated that 16% of the available waste quantities could be recycled in roads. Analysis has shown considerable savings in costs, CO₂ and energy in comparison to conventional asphalt mixtures using all virgin components [11]. In this research two types of waste plastic were used to modify the bitumen. The rheological performance along with materials thermal characteristics of waste plastic modified bitumen was assessed. The thermal properties of the waste plastics where investigated by Differential Scanning Calorimetry (DSC) and Thermogravimetric Analysis (TGA) and the chemical properties with Fourier Transform Infrared (FTIR) Spectrometry. The rheological performance of plastic modified bitumen was tested with Dynamic Shear Rheology (DSR) Master Curves. The stability at high temperatures was also tested using a stability tube test followed by viscosity measurements. Finally, the blending of the bitumen and plastics were observed on a microscale with Environmental Scanning Electron Microscopy (ESEM).

2. MATERIALS

Bitumen 70/100 supplied by Kuwait Petroleum Corporation was used as the base material for modification with waste plastic. The plastics were PE-pellet and PE-shredded acquired from Inno-recycling (Switzerland). In this study, polymer modified bitumen Pmb 45/80-65 was used as the reference material. The engineering properties such as softening point are given in Table 1. The melting temperature and density of PE-pellet and PE-shredded are given in Table 2.

In order to facilitate the physical blending of waste plastic (PE-pellet and PE-shredded) with bitumen, the waste plastic was grinded into sizes of about 2 mm and under before blending it with bitumen. A high shear mixer (Silverson L5M Laboratory Rotor/Stator Batch Mixer) with a Vertically Slotted Lower Stator (Stainless steel) was used to grind the PE-pellet and PE-shredded. The grinding process was carried out in a container filled with 2/3rd of tap water at room temperature. The grinding process continued for 5 min at speed of 5000 rpm.

The process of blending PE-pellet and PE-shredded with bitumen is controlled by factors such as type of mixer/blender, speed and time of mixing, temperature and shear/speed rate and as well as mixing conditions (temperature control). The blending process of PE-pellet and PE-shredded with bitumen was executed using high shear mixer (Silverson L5M Laboratory Rotor/Stator Batch Mixer, England) with a square hole high shear lower stator as shown in Figure 1. The blending speed of 3500 rpm for 60 min at temperature of 170°C was controlled using an oil bath. The amount of waste plastic (PE-pellet and PE-shredded) was 5% by mass of the base bitumen 70/100. In parallel, to simulate the effect of

ageing in bitumen during blending process, the base bitumen 70/100 was also "blended" using high shear mixer at speed of 3500 rpm for 60 min without adding plastic and termed as control material.

3. METHODS

The Dynamic shear rheometer DSR (Physica MCR 301 DSR, Anton Paar® GmbH., Austria) in parallel plate configuration was used to analyze the rheological performance of the control and modified bitumen. The time and temperature dependent mechanical performance of the modified bitumen was evaluated by carrying out the frequency sweep tests. To control the temperature of the DSR parallel plates, Peltier Systems H-PTD200 and P-PTD200 (Anton Paar® GmbH., Austria) was used. The magnitude of the complex shear modulus, $|G^*|$ and phase angle, δ of the modified and unmodified binders were obtained from dynamic shear loading. In this study, the effect of the physical presence of the waste plastic (PE-pellet and PE-shredded) were analysed as per EN 14770 [12] using master curves. The parallel plate configuration of 8 mm and 25 mm diameters to the corresponding 2 mm and 1 mm thicknesses of test specimens was used respectively. Constant strain amplitude of 0.1% and 0.05% for high temperature range between 40 °C to 80 °C and low temperature range of 40 °C to -10 °C was used respectively. The testing frequencies were between 0.1 and 20 Hz at each temperature.

Based on the time-temperatures superposition principle for linear viscoelastic materials, the complex modulus master curve were obtained at a reference temperature (T_{ref}) of 20 °C. The shifting the individual modulus-frequency curves are obtained at different temperatures from -10 °C to 80 °C with increment of 10 °C along the logarithmic frequency axis. The shift factor a_T for shifting the complex moduli curves at a certain temperature T to a master curve for a reference temperature T_{ref} was expressed in equation (1) using the Williams-Landel-Ferry relation [13]:

$$\log a_T = \frac{-C_1 (T - T_{ref})}{C_2 + (T - T_{ref})} \quad (1)$$

Where C_1 and C_2 are material related constants and T_{ref} is the reference temperature.

The morphology of the waste plastic modified and unmodified bitumen was investigated by using environmental scanning electron microscopy (REM Quanta 650). According to a previously used protocol [14], the ESEM specimens were prepared by pouring it in 8 mm moulds after heating the specimen at 110 °C for 2 h. Meanwhile, in order to flatten the specimen surface, they were placed on a hot plate (110 °C) for 2 min. The PEP-5% and PES-5% specimens required higher temperature of 150 °C for 10 min to flatten the specimen surfaces.

The thermal transitions unmodified bitumen and bitumen modified with of PE-pellet and PE-shredded were analyzed using a DSC (Perkin Elmer®, USA). A specimen of approximately 10 to 13 mg was prepared for the DSC analysis. The specimens were firstly cooled from 25 °C to 20 °C and were then maintained at this temperature for 5 min. The specimens were heated up to 200 °C and kept at this temperature for 5 min before cooling again to 20 °C where it remained for 5 min until finally being heated up to 200 °C. The cooling and heating ramps were conducted at a rate of 20 °C/min. For reproducibility purposes and standard deviation calculation, each specimen was tested at least three times. The thermal stability of PE-pellet and PE-shredded blends with bitumen were examined by thermogravimetric analysis (TGA) under a nitrogen atmosphere using TGA 209 from NETZSCH (Germany).

In order to investigate and monitor the effect of PE-pellet and PE-shredded on the chemical structure of base bitumen 70/100, FTIR spectroscopy was used to test the modified specimens. The analysis was carried out using Bruker Vector 22 / Digilab BioRad FTS 6000 FTIR spectrometer (Germany).

Finally, to evaluate the phase separation of the PE-pellet and PE-shredded waste plastic in base bitumen 70/100, a high temperature storage stability test was conducted in accordance with the standard protocol EN-13399 [15]. The specimens modified with PE-pellet and PE-shredded were poured into aluminium tubes of 25 mm diameter and 120 mm height. To minimize oxidation by air, after filling, the aluminium tubes were subsequently sealed tightly. The filled aluminium tubes were storage vertically for 72 h at the temperature of 180 °C in a forced draft oven and were left to cool down at room temperature, still in the same vertical position. To facilitate the cutting/peeling of the tube or the removal of aluminium foil, the tubes were cooled at -20 °C for 30 min and cut horizontally into three equal parts. The samples belonging to the top and lowermost segments of the tube were used to assess the storage stability of the binders based on their measured viscosity values. The viscosity was measured using dynamic shear rheometer DSR (Physica MCR 301 DSR, Anton Paar® GmbH., Austria) in rotational mode. Approximately 15g of specimens was used in a test container for 30min wait time at test temperature followed by test measurement at a constant speed of 20rpm for the duration of 60min and the average of last 10 readings was taken in the analysis of the results.

4. RESULTS AND DISCUSSION

The thermal transitions (20 °C to 200 °C) and density measurements of waste plastic modified bitumen were conducted by using DSC and a Hilum Pycnometer, respectively. The melting temperature 123.8 °C and 132.7 °C with the overall areas of 813.44 mJ and 1143.92 mJ was observed for PE-shredded and PE-pellet, respectively. It therefore infers that at a mixing temperature of 170 °C, the blending of PE-pellet and PE-shredded with bitumen 70/100 is way above the melting temperature of waste plastic. The TGA analysis of waste plastic PE-pellet and PE-shredded are shown in Figure 2. The results show that both type of waste plastic start disintegrating at around 400°C. It therefore, confirms that both type of waste plastics (PE-pellet and PE-shredded) used in this study remain thermally stable during mixing temperature (170°C) with bitumen.

1. Table 1: Softening point of bitumen modified with waste plastic.

Sr. #	Specimen	Softening Point (°C)
1	Bitumen 70/100 (0%)	46
2	Bitumen 70/100 (Control)	52
3	Pmb 45/80-65	66
4	Bitumen 70/100 (5%, PE-pellet)	63
5	Bitumen 70/100 (5%, PE-shredded)	89

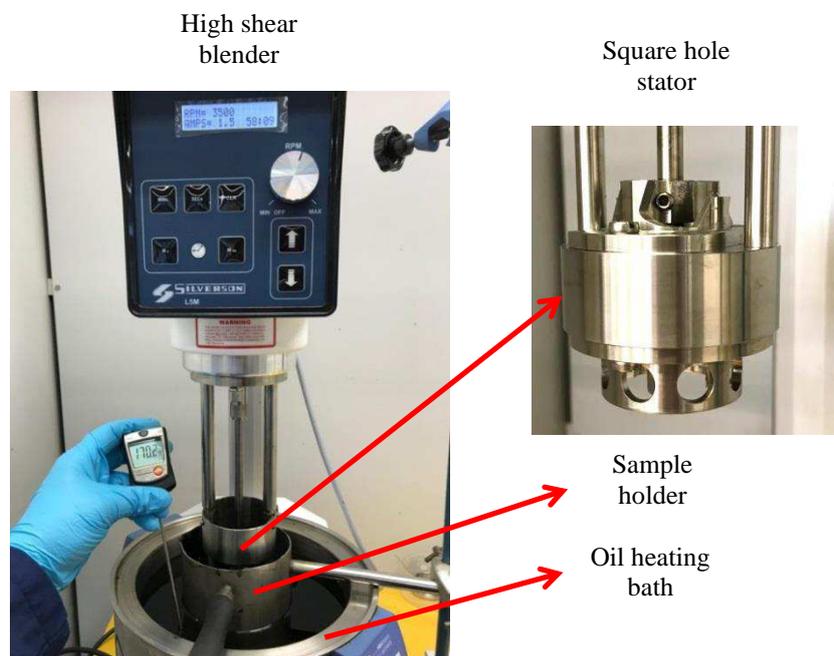


Figure 1: Blending process of waste plastic (PE-pellet and PE-shredded) with bitumen

2. Table 2: DSC results of PE-pellet and PE-shredded waste plastics

Sample	Density (g/cm ³)	Melting Temperature (°C) (DSC)
PE-pellet	0.9454	132.7
PE-shredded	0.9499	123.8

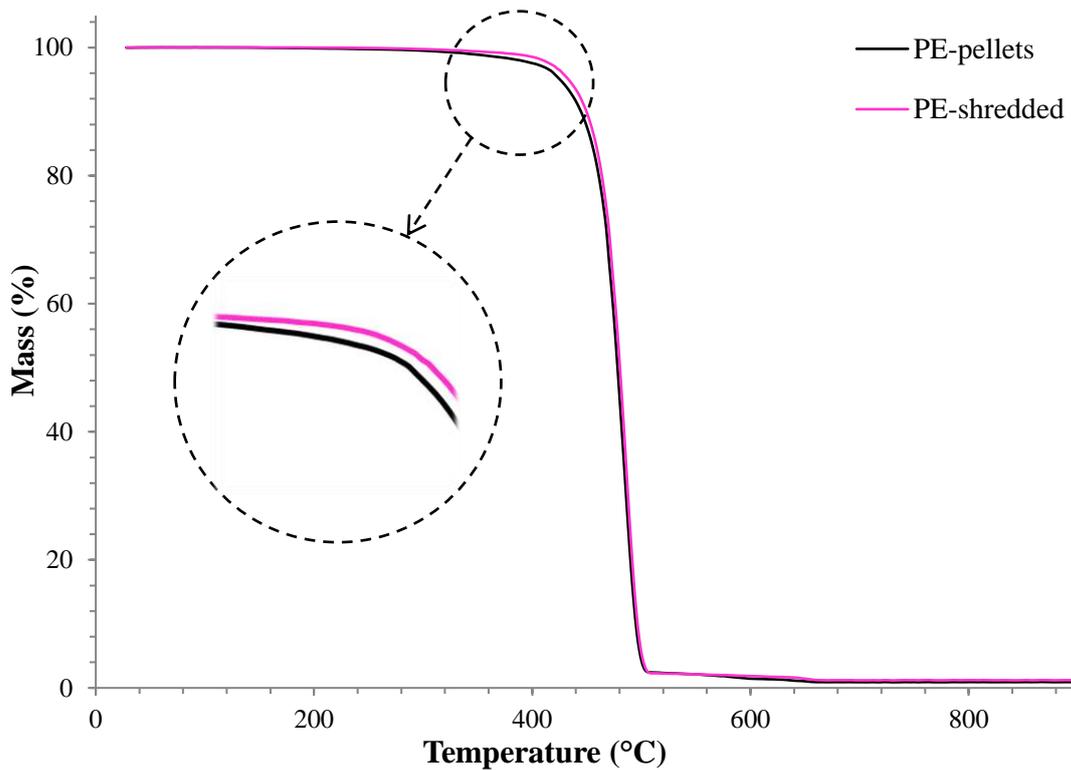
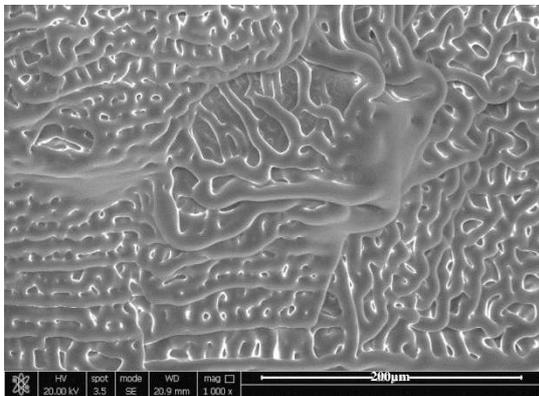
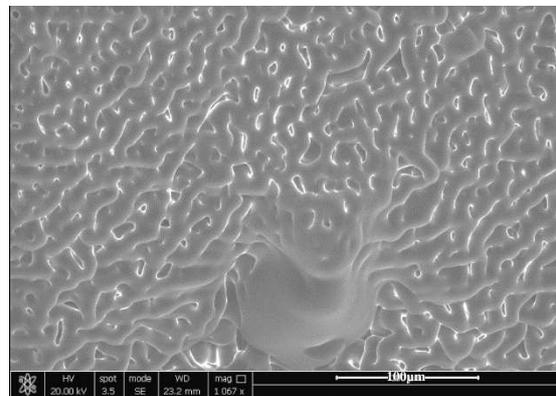


Figure 2: TGA analysis of PE-pellets and PE-shredded

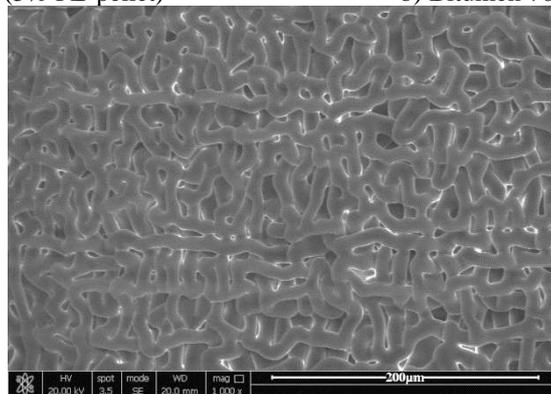
The bitumen samples were observed with ESEM to examine the plastic blending and distribution. ESEM images of bitumen 70/100 and PE modified binders are shown in Figure 3. Looking at the spread of the plastic in the bitumen over all, it can be seen that the plastics are not fully blended in the bitumen, but are spread in fragments around the bitumen matrix. This is consistent with the findings of the stability testing described later. However, looking closer at the individual pellets, a modification of the bitumen (Figure 3a and 3b) compared to the base binder (Figure 3c) can be seen in the bitumen surrounding the pellets, indicating that some blending is occurring and that the pellets are bound to the bitumen.



a) Bitumen 70/100 (5% PE-pellet)



b) Bitumen 70/100 (5% PE-shreds)



c) Bitumen 70/100

Figure 3: Scanning Electron Images

The FTIR analysis of both waste plastics is presented in Figure 4. The spectra from both the plastics are similar to each other and also to the FTIR spectra of bitumen due to all these being hydrocarbons [16]. There are small peaks at 1700 and 1300 peaks in the PE-shredded, which are not present in the PE-pellet. This is due to the PE-pellet being made in a more controlled process than with the PE-shredded, which is a waste product of the PE-pellet production. In comparison with PE-pellets, the peaks observed at 1741, 1240 and 1020 refer to the impurities present in PE-shredded, which coincides with the fact that these shreds are a type of waste without a much control on their composition.

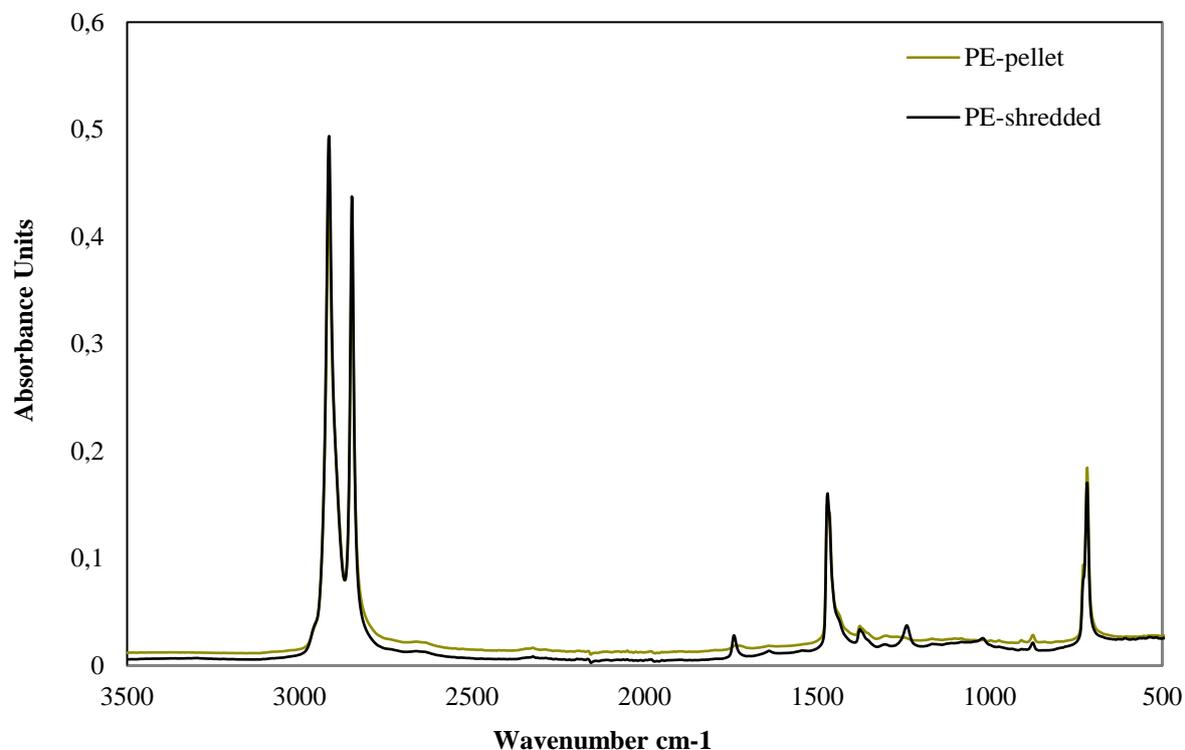


Figure 4: FTIR spectrum of PE-pellet and PE-shredded

The storage stability of polymer modified bitumen has been identified as an important criterion of production and practical usage of modified asphalts. In order to obtain a homogeneous material, the conventional designs of storage tanks used in this field show that most of the modified asphalt binders are stored with continuous agitation [17]. The storage stability test procedure was conducted according to European Standard test method EN-13399 [15] with the only exception of using viscosity measurements on storage stability test tube specimens. The results as given in Table 3 illustrates a substantial difference in the viscosity values of bottom and top portion of the tube specimens of both PE-pellet and PE-shredded modified bitumen. The viscosity measurements suggest that higher values of top portion of the tube specimen are due to the fact that the PE-pellet and PE-shreds are moved upward during the stability test, which is due to their lower density. Moreover, the differences in viscosity measurements of both PE-shredded and PE-pellets are not ascertained based on the individual density values (cf. Table 2) which are not significantly different. On the other hand, the viscosity measurements of polymer modified bitumen Pmb showed that the top and bottom part has almost similar values, indicating a much more stable binder.

3. Table 3: Viscosity results for top and bottom part of storage stability tube

Storage Stability Specimen	Temperature (°C)	Viscosity (Pa-s)		
		Bitumen 70/100 (PE-pellet)	Bitumen 70/100 (PE-shredded)	Pmb
Top	150	7.01	5.33	0.74
	160	5.43	3.54	0.50
	170	3.59	2.58	0.35
Bottom	150	0.37	0.41	0.73
	160	0.25	0.27	0.49
	170	0.17	0.19	0.35

The rheological measurements of waste plastic modified bitumen, unmodified and polymer modified bitumen Pmb were performed using DSR master curves. The results of master curves were analyzed using the Black diagram (Log of complex modulus Vs phase angle) as shown in Figure 5. The results show that the control bitumen 70/100 follows the normal trend for bitumen of the phase angle increasing with increase in temperature, along with the subsequent decrease in complex modulus. On the other hand, the polymer modified bitumen Pmb has shown a deviation from the unmodified bitumen curve, which showed comparatively higher resistance in terms of stiffness during high temperature. More interestingly, both the waste plastics (PE-pellet and PE-shredded) have shown higher deviations compared to unmodified and polymer (Pmb) modified binder curve. It therefore, indicates that the waste plastic modified bitumen has large potential to resist against high temperature rutting. The rheological results further illustrate that at high temperature (80°C), the bitumen modified with PE-shredded has high modulus values compared to PE-pellet modified bitumen. This indicates that the higher softening point value (cf. Table 1) of PE-shredded bitumen is related to such effects. Furthermore, the master curves at low temperature (-10°C) suggest that at frequency 20 Hz, the corresponding values of complex modulus and phase angle of 0% control bitumen, PE-pellet and PE-shredded modified bitumen are $3.56\text{E}+08$ (Pa); 12.6 (°), $3.56\text{E}+08$ (Pa); 12.7 (°) and $3.45\text{E}+08$; 13.0 (°), respectively. Therefore, referring to the Black diagram at low temperature extreme (-10°C), the results represents no obvious differences among all the binders tested. Previous studies [18] [19] also suggests that the use of PE in bitumen has the tendency to improve the low temperature characteristics such as fracture toughness. However, low temperature analyses along with moisture resistance are recommended for future studies.

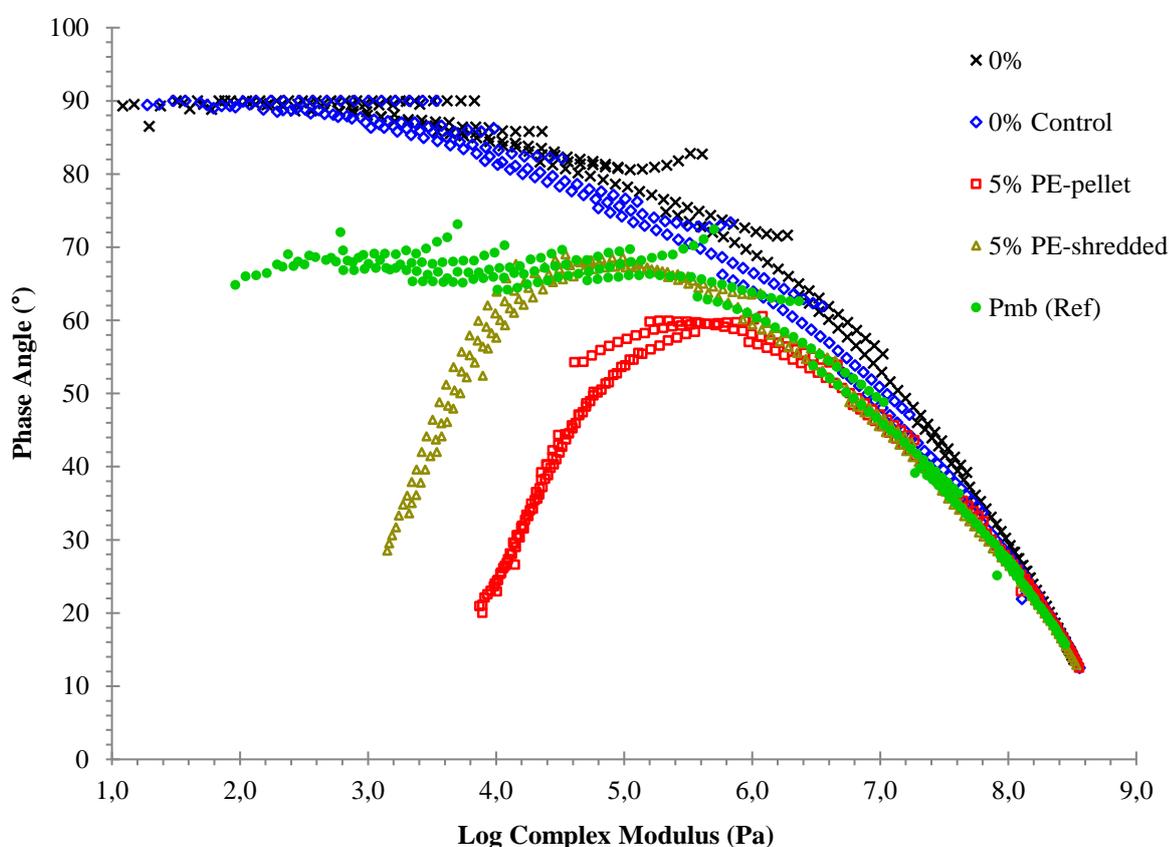


Figure 5: Black diagrams for different bitumen with and without waste plastic.

5. CONCLUSIONS

In this study, preliminary investigations of potential waste plastic materials (PE-pellet and PE-shredded) as bitumen modifiers are summarized. The methods of mixing and blending waste plastic in bitumen are very important and need careful consideration. In this research, to facilitate a homogeneous blend of PE-pellet and PE-shredded with bitumen, both the pellet and shreds were grinded before mixing it with base bitumen 70/100. The rheological test results obtained using DSR master curves showed that PE-pellet and PE-shredded modified bitumen significantly enhanced the high temperature performance related to rut resistance or permanent deformation. However, on the other hand the storage stability or high temperature phase separation related problems were identified in PE-pellet and PE-shredded modified bitumen. Hence, it therefore becomes very important to further investigate the use of additives/modifiers that help in resolving the storage stability problems in waste plastic modified binders. Furthermore, this study also encourages the investigation of using stabilizer, possibly waste oil, for waste plastic materials in bitumen.

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