Rheological Characterization of Bitumen/Waste Polyethylene Composites: Part III

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Abstract— *Waste polyethylene (PE) bags were reused in the modification of commercially available bitumen. The bitumen/PE blend was prepared by mixing homogenously commercial bitumen with waste PE in the molten state. The samples of bitumen blend and bitumen base were rheologically analyzied. The rheological properties such as complex modulus, shear compliance, torque and complex viscosity of bitumen and bitumen blend were studied using an ARES-Rheometer (Rheometric Scientific, Co.) in the dynamic mode, plate-plate geometry with diameter 8 mm. The measurements were performed over a wide range of temperatures ranged from-10°C to 60°C and frequencies from 100 to 0.1 radians per second. The results evidence that the complex modulus, shear compliance, torque and viscosity are improved by the incorporation of the waste PE into bitumen. The flow activation energy for neat bitumen and bitumen-9%PE are calculated from η_0 by using the rheometric software.*

Keywords: bitumen; rheology; polymer modification, waste plastics.

1. INTRODUCTION

Bitumen is the black adhesive that binds flexible pavements on roads and airfields together. Bitumen is also used in other areas of application, such as waterproofing, flooring and joint materials. Almost all bitumen originates from crude oil and is the residue of a refining process. It is well-known that bitumen is a very complex and temperature dependent material consisting of hydrocarbon molecules. Naphtenic-base crude oils often give a large yield of bitumen that may be of good quality, while paraffinic crude oils may give bitumen of good quality or yield bitumen not suitable for road construction [1].

Nowadays a very large majority of the roads are constructed using a mixture of bitumen (5 wt %) and mineral aggregates. Notwithstanding this low bitumen content, the performance of the road pavement depends to a large extent on the properties of bitumen itself, since it constitutes the only deformable component. The correlation between the complex colloidal structure of bitumen and its viscoelastic response is therefore a subject of scientific and technical interest [2 -7].

Polymer additives are well-known to improve the rheological properties of bitumen The polymer addition allows an increase in the resistance of the binder to permanent deformation at high temperature.Besides, the fracture properties including critical stress intensity factor (K1C) at low temperature of polymer modified bitumens (PmB's) were shown to be higher than those of the bitumen base.To determine the crack propagation mechanism controlling fracture properties, previous studies have focused on establishing the relationship between the fracture properties and the morphology of polymer modified bitumen [8]

The addition of synthetic polymers to enhance service properties over a wide range of temperatures in road paving applications was considered a long time ago and nowadays has become a real alternative. As has been pointed out in relevant papers about bitumen and polymer/ bitumen blends[9 -18] understanding the interactions of asphaltene and maltene (main components of bitumen) with the polymer, is a crucial point to gain insight into the routes to improve the capacities of these systems.

In recent years, the volume of municipal plastic waste (MPW) has increased greatly, and this has resulted in a critical problem for modern society and future generations. Polyolefins, poly(ethylene terephthalate), Polyvinylchloride (PVC), polystyrene (PS), and high-impact polystyrene (HIPS) are among the most common components of plastic waste because they are among the most frequently used commercial plastics in our daily lives and in industry.Recycling mixed plastic residues in the form of blends is attractive from academic and industrial points of view because of the improvements in the impact strength , dimensional stability, stress cracking, and processability with respect to virgin blends[19].

In the present study, waste plastics are used as a filler for making bituminous roof mastic up to a level of 20 wt %. The viscoelasticity of a bitumen modified with waste 9wt% PE is analysed and compared to bitumen base.

2. EXPERIMENTS PART

Materials and preparation

Waste plastics were collected from the garbage, sorted and shredded into coarse particles. The waste plastics and commercial bitumen were weighted and heated individually in an oven until melt. Molten waste plastic was poured into the molten bitumen and stirred vigorously to give a homogenous sample. The hot mixtures were then cast into a ring stamp with 25 mm diameter and 2 mm thickness for rheology testing .

Measurements

Solid-state dynamic viscoelastic measurements of all the pure bitumen and bitumen blends samples were accomplished in a Solid state institute, Research center Juelich, Germany. In this study we used an ARES- rheometer (Advanced Rheology Expanded System, Rheometric Scientific Co.,) in the dynamic mode, under nitrogen atmosphere, plate-plate geometry with 25 mm in diameter to determine the rheological characteristics of the neat bitumen and bitumen blends. The measurements were performed over a wide range of temperatures ranged from 25°C to 160°C and frequencies from 100 to 0.1 radians per second. A sample (about 1.0 g) was placed in the lower plate then the upper parallel plate was lowered for a tight contact with the sample. All the samples were held at a constant temperature of 50°C for 10 min, cooled to 25°C and measured at temperatures (25, 40, 60, 80, 100, 110, 120, 130°C, 145°C and 160°C). The applied strain were 2%.

3. RESULTS AND DISCUSSIONS

In this section the results are presented in a master curves. The master curve is a single curve at which the experiments data which measured over a wide range of the temperature, T are shifted into this curve at a reference temperature T0 . By using of the time-temperature superposition principle which described by Williams-Landel-Ferry [20] as, log aT = -C1 (T-T0) /(C2+(T-T0)).Where aT is the horizontal shift factor and constants C1 and C2 are material specific. T0 is chosen in this study to be 25°C. aT shifts the data obtained at different temperatures along the log frequency, ω axis as shown in Figure 1. Since aT shifts the high temperatures data toward the right ω-wing at low ω and the low temperatures data toward the left ω-wing at high ω. In vertical direction the experiments data are shifted by bT (bT = $\rho T \ / \ \rho_0 T_0$ [21]). Where ρ is the material density. bT shifts the data up-and-down by a factor related to the temperature. Where high temperatures data are multiplied by a factor and low temperatures data divided by a factor. Because the modulus decreases with increasing the temperatures. This master curve enable us to enlarge the range of the frequencies which it looks like data measured at a single temperature as a function of an extremely wide range of frequencies. As an example ω can be enlarged from 102 –10-1 radians/s to 106 –10-6 radians/s and this range is difficult to measure by a machine until now.

The master curves of the shear complex modulus, G* for bitumen and bitumen blends at 25°C are logarithmically plotted in Figure 2 against the angular frequency. This quantity of G* is a combination of the storage modulus (G') and loss modulus (G'') as, G*= G' + i G'' [38] .Where, G' is the ability of the material to store energy in the cycle of the deformation and G'' is the energy dissipated as heat in this cycle. Master curves of G* display two regions of behavior. These region are : the melt and rubber-like zones from the left side to the right side. The third regime of the dynamic glass transition is observed at very high frequency (ω ~ 1×109) in the case of pure bitumen as shown in Figure 2 because it could measured at low temperature (-10°C). At the flow regime, G* decreases further as shown in Figure 2. As the deformation shear rate increases, G* increases until the intermediate regime of a rubber-like plateau is observed. This rubber-like region is somewhat flat and it is nearly independent on the frequency as shown in Figure 2. Because this rubber like behavior reflects the elasticity at which the loss energy is minimum. But in the melt regime G* falls by many order of magnitude because of the high loss energy per the cycle of deformation due to the effect of the viscosity in this regime. The beginning of the melt regime is shifted toward low frequencies by the addition of waste PE to bitumen as shown in Figure 2. Since the melt regime begins at ω ~ 2.4×104and 30 radian/s in the cases of neat bitumen, bitumen-9%PE , respectively. This because of the high elasticity due to the bitumen-PE network formation. The adding of waste plastics to bitumen showed a high effect as seen in Figure 2 the value of G* modulus for bitumen blend is higher than this of bitumen base.The difference in the values of G* between bitumen-PE and pure bitumen increases with decreasing the frequency. Where at ω ~ 2.3×105 radian/s the values of G* are 6.3×106 and 1.4×107 Pa for pure bitumen and bitumen-9% PE, respectively. At ω ~ 2.3×102 radian/s The values of G* in the case of bitumen-9%PE is higher than this of pure bitumen by 2 times. This factor reaches to 8 at ω ~ 0.02 rad/s. This increase in G* modulus by the addition of waste PE reflects the interaction between polymer and bitumen which increases the stiffness. that leads to high stability for a long time of defromation (low frequencies) than neat bitumen. This interaction is mostly happened between polymer with saturate resins, owing to their closer solubility. Besides, Some authors have attributed this improvement to the existence of a third mixed phase modifying the asphaltene network. [22]. Since bitumen consists of matlenes and asphaltenes and the ratio of the asphaltenes to the maltenes has a significant effect on the viscoelastic properties of bitumen and, consequently, on its performance as road paving binders [23].

The master curves at 25°C of the storage (J') and loss compliance (J'') versus ω are presented in Figures 3 and 4, respectively. $J'(\omega)$ is a measure of the energy stored and recovered per deformation cycle, therefore is called the storage compliance. And $J''(\omega)$ is a measure of the energy dissipated as heat per cycle of the sinusoidal deformation, for that is called the loss compliance. Figures 3-4 show the dynamic shear compliances moduli for bitumen blends compared to pure bitumen. These Figures show clearly two regions reflect the behavior of the material under the measured temperatures as a function of frequency as explained earlier in the case of G* (ω) curves. As shown in Figures 3-4 the incorporation of the waste PE into bitumen affect on the $J'(\omega)$ and $J''(\omega)$ moduli. Since these moduli are enhanced for the bitumen blends. This is clear at $\omega \sim 5.2 \times 10^3$ radians/s (Fig.2) where the value of J' changed from 1.5×10^{-7} for neat bitumen to 1.0×10^{-7} Pa^{-1} in the case of bitumen/PE blend. But at low frequencies the effect is more noticeable. since at $\omega \sim 5.9 \times 10^{-2}$ radians/s, the values of J' become 4.0×10^{-5} and 1.3×10^{-6} Pa^{-1} for pure bitumen, and bitumen blend, respectively. The same effect is observed also for J'' moduli in Figure 4. In this Figure at $\omega \sim 91$ radians/s the values of J'' are 4.3×10^{-7} and 1.9×10^{-8} Pa^{-1} for neat bitumen, bitumen-PE, respectively. This because of the matrix formation between the PE chains and bitumen which leads to increase the strength and stiffness. This increase in the stiffness and strength by the introduction of the waste PE to bitumen is confirmed also in the shear creep stress, J(t) in Figure 5. This Figure show J(t) as a function of time for pure bitumen and bitumen blends. As shown in Figure 5 J(t) increases with time and the difference between the values of J(t) in the case of neat bitumen and bitumen blends is higher at long times than at short times. Since at t $\sim$ 200 seconds, the values of J(t) are, 1.1×10^{-3} and 1.4×10^{-5} Pa^{-1} for neat bitumen and bitumen-PE, respectively. This indicates the improvement of the stability of the bitumen for a long time by the addition of the waste PE due to network of bitumen-PE.

Figure 6 presents log-log plot of the complex viscosity (η^*) as a function of ω for neat bitumen and bitumen blends at 100°C. In this Figure η^* increases rapidly with decreasing ω up to $\omega = 0.001$ radian/s after that it becomes independent on ω. At this very low shear rates η^* is called the melt viscosity, η_0 (zero-shear viscosity). As shown in this Figure the incorporation of the waste plastics into the bitumen increases the viscosity over all the frequencies range. The difference between the viscosity of bitumen/PE blend and bitumen base is more noticeable at very low frequency (at η_0). Since the values of η_0 increase from 5.0×10^4 to 1.3×10^5 , 2.7×10^5 , 3.2×10^5, 4.3×10^5 and 5.7×10^5 Pa.s by the addition of 3, 7, 9,13 and 20 % PET , respectively. This because the viscosity of the melt plastic is higher than this of bitumen and that is related to the length of the chains.

The flow activation energy for neat bitumen and bitumen blends (bitumen-10%PET) are calculated from η_0 by using the rheometric software and it is found to be 1.063×10^5 and 1.46×10^5 J/mol.K, respectively.

The torque in NM as a function of frequency for neat bitumen and bitumen blends are logarithmically plotted at 100°C in Figure 7. The torque decreases with decreasing the shear rates as shown in Figure 7.This Figure evidences also that the addition of waste plastics to bitumen enhances the torque. Since the torque increases from 5.0×10^{-4} to 1.3×10^{-3} , 2.7×10^{-2} , 3.2×10^{-1}, 4.3×10^{0} and 5.7×10^{1} NM by the addition of 3, 7, 9,13 and 20 % PET , respectively. This because the formation of the bitumen-plastic matrix which leads to increase stiffness of the bitumen blends.

4. CONCLUSION

Waste PE bags were used as a modifier in making improved bitumen. The blend compared to bitumen base was subjected to evaluate their response against temperature and frequency sweeps. By using an ARES- Rheometer (Rheometric Scientific, Co.) in the dynamic mode and parallel plate geometry with diameter 8 mm. The measurements were performed over a wide range of temperatures ranged from -10 to 60°C and frequencies from 100 to 0.1 radians per second. The flow activation energy for neat bitumen and bitumen blend are calculated from η_0 by using the rheometric software and it is found to be 1.063×10^5 and 1.46×10^5 J/mol.K, respectively. It was found that, the complex modulus, shear compliance, torque and complex viscosity are enhanced by the incorporation of waste 9wt%PE.

5. ACKNOWLEDGMENTS

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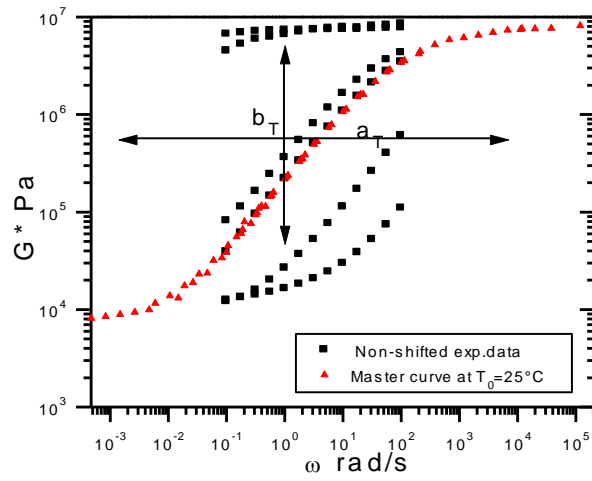23. F.Martinez-Boza, P. Partal, B.Conde, C.Gallegos, *Energy & Fuels, V14, P.* 131-137, 2000.

Figure 1: Shifting the experiments data of G* for bitumen/PE measured at different temperatures into master curve at T$_0$ =25°C by using a$_T$ and b$_T$.
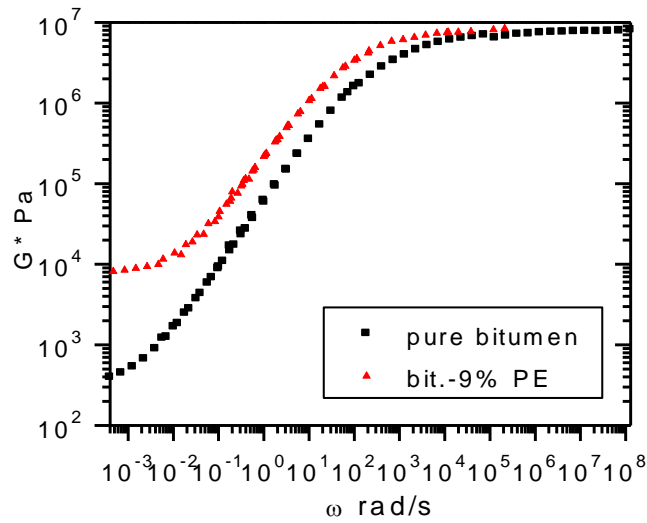

Figure 2: Master curves of G* for bitumen/PE blend and bitumen base at T$_0$ =25°C
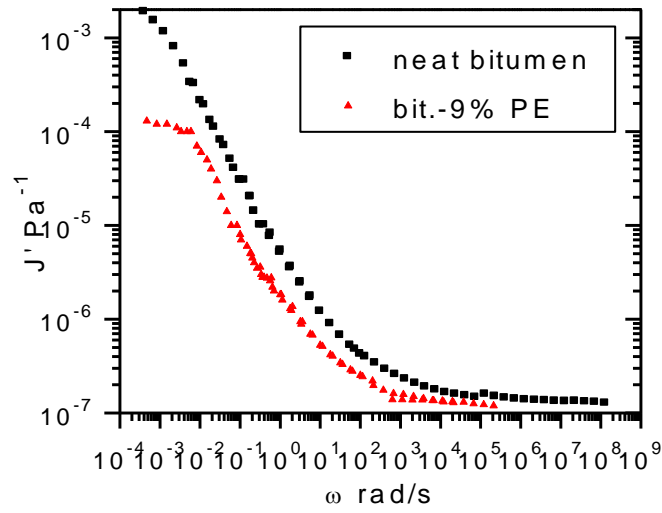
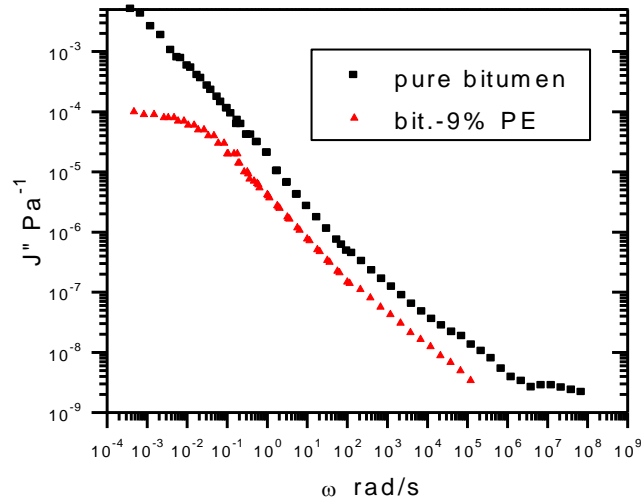Figure 3: Master curves of J' for bitumen/PE blend and bitumen base at $T_0 = 25°C$

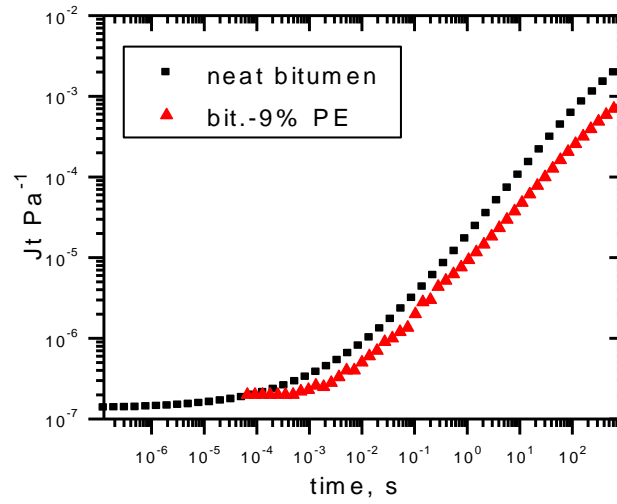Figure 4: Master curves of J'' for bitumen/PE blend and bitumen base at $T_0 = 25°C$

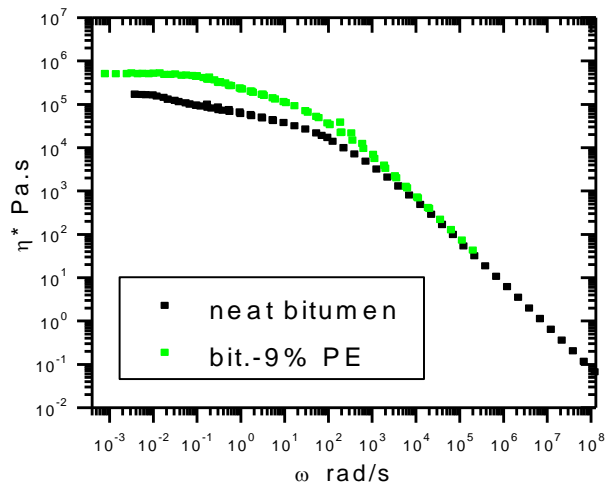Figure 5: The shear creep for bitumen/PE blend and bitumen base at $T_0 = 25°C$

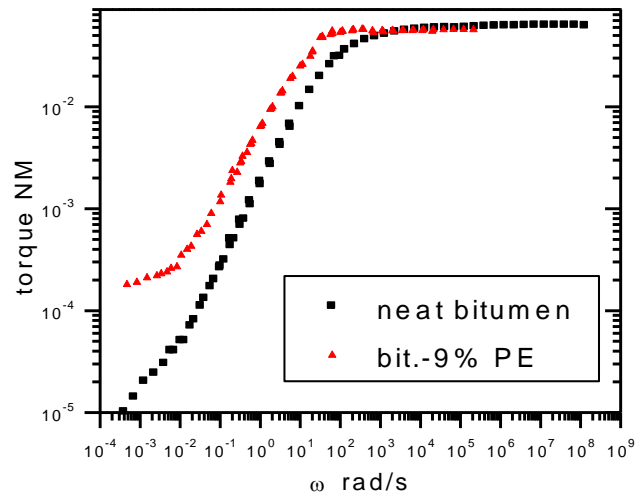Figure 6: Master curves of η^* for bitumen/PE blend and bitumen base at $T_0 = 25°C$

Figure 7: Master curves of the torque for bitumen/PE blend and bitumen base at $T_0 = 25°C$